**OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT
SPECIAL INSTRUCTION SHEET**

QA: N

Page: 1 of: 1

This is a placeholder page for records that cannot be scanned or microfilmed

1. Record Date
09/29/97

10. Accession Number
MOL.19980203.0570

2. Author Name(s)
ROBINSON BA, WOFBERG AV, VISWANATHAN HS

3. Author Organization
LANL

4. Title
MILESTONE SP25BM3: THE SITE-SCALE UNSATURATED ZONE TRANSPORT MODEL OF YUCCA MOUNTAIN

5. Document Number(s)
SP25BM3

6. Version
N/A

7. Document Type
REPORT

8. Medium
PAPER/OPTIC

9. Access Control Code
PUB

11. Traceability Designator
VADD-VOL-03

12. Comments
THIS ON OF A KIND COLOR DOCUMENT CAN BE LOCATED THROUGH THE RPC, THIS IS A VIABILITY
ASSESSMENT RECORD-DO NOT DELETE

The Site-Scale Unsaturated Zone Transport Model of Yucca Mountain

Bruce A. Robinson

Andrew V. Wolfsberg

Hari S. Viswanathan

Gilles Y. Bussod

Carl W. Gable

Arend Meijer

September 29, 1997

Revision Date: 08/26/97

FY 1997 LEVEL 3 BASELINE DELIVERABLE SP25BM3

TO: DWIGHT HOXIE (Office Manager)

DATE: 8/28/97

Reviewer/Tech. Lead (assigned by Office Manager): ROGER HENNING

Due Date for Completion of Review: 8/29/97

SEE INSTRUCTIONS/GUIDANCE ON ATTACHED SHEET.

Initial on
Appropriate Line

- Review complete on 8/29/97 (Date).
- Comments forwarded to Office Manager? Yes RH
No Comments _____
- "Roadmap" showing where deliverable acceptance criteria are
addressed in deliverable is included with deliverable. Yes RH
Hold for Revision _____
- For "synthesis reports," requirements of Jones to Foust letter are
met? N/A RH
Yes _____
Hold for Revision _____
- Deliverable meets all technical and administrative requirements on
the PPS sheet and is of the editorial and technical quality to allow
release to DOE, regulators, the public, & the general scientific
community? Yes RH
Hold for Revision _____

Reviewer Signature Roger Henning Date 8/29/97

Office Manager approval for submittal Dwight Hoxie Date 8-29-97

Baseline Deliverable Due Date 8/29/97 (Date Deliverable should be at TPM)

Deliverable Received by SPO on _____ Date Sent to TPM _____

INSTRUCTIONS: Reviewers are responsible for reviewing the attached deliverable against criteria/ description on the PPS sheet (also attached) and determining whether the deliverable should be forwarded to DOE for the YAP-5.1Q acceptance review. When the review is complete, reviewers complete the questions on the form by initialing on the appropriate line and sign at the bottom. Any comments, concerns, or recommendations arising from the review are forwarded to the Office Manager/Tech. Lead by the reviewer for possible action with the organization responsible for the deliverable. When any comments are resolved and the Office Manager determines the deliverable is ready for submission to DOE, the Office Manager signs the form and returns the form to Terry Grant (MS 423/822C).

- Completed Level 3 deliverables must be submitted in final form, ready to be submitted as an M&O document to DOE. Level 3 documents cannot contain any notations on the title page or on headers or footers that indicate the document is a "draft" or is otherwise not a final M&O product.
- Five copies of the final document are needed on the due date for the submission process.
- If the Level 3 document might be considered a **synthesis report** by DOE, review the requirements in the attached letter (Jones to Foust). The requirements in this letter were cited by DOE in their recent rejection of the Natural Resources report as some of the reasons for rejection. This letter contains some specific requirements (e.g., Executive Summary) and general guidance on content, level of detail, and approach.
- The "roadmap" is usually a table that lists each criterion from the PPS sheet for the deliverable and shows the text location (e.g., page number, section number, and/or figure number) in the deliverable where the criterion is addressed. The roadmap may be incorporated into the deliverable document or submitted as a separate item. If a criterion is not covered in the report for some reason (e.g., submission of a TDIF), the reasons for this should also be explained in the roadmap. Reviewers and Office Managers should carefully review the roadmap to assure that all requirements on the PPS are listed and that they are adequately addressed. YAP-5.1Q requires that the DOE reviewers use the PPS as the source of criteria for their review.
- The purpose of the review is determine whether the document is at a level of quality and completeness that acceptance by the DOE reviewers can reasonably be anticipated in the YAP-5.1Q review process. Editorial and technical quality should be at a level to allow for publication by the Project and/or release to the public.
- The review process does **not** automatically include a publication release review (YAP-30.12 review). If a publication clearance is needed, this must be indicated at the time of submission. Five additional copies of the document are required for publication clearance (it is a separate request and process).

3-27-97
-MOL 19960422-0215

Department of Energy
Office of Civilian Radioactive Waste Management
Yucca Mountain Site Characterization Office
P.O. Box 98608
Las Vegas, NV 89193-8608

NOV 15 1995

L. Dale Foust, M&O, Las Vegas, NV
Robert W. Craig, USGS, Las Vegas, NV

U.S. DEPARTMENT OF ENERGY (DOE) POLICY ON SYNTHESIS REPORTS
(SCPb: N/A)

The production of synthesis reports will represent a major effort for Assistant Manager for Scientific Programs during fiscal year (FY) 1996. The DOE focus is on those synthesis reports that summarize work at the Site Characterization Plan investigation level. However, these same guidelines are relevant to all technical reports produced in Work Breakdown Structure 1.2.3, regardless of milestone level. The FY 1996 planning effort has included attempts to clarify the intended content and use of synthesis reports. Acceptance criteria for each report will be included in the Planning and Control System. However, this letter addresses a need to provide a higher-level guidance on DOE expectations relative to synthesis reports. This guidance is consistent with the draft policy from the Yucca Mountain Quality Assurance Division (enclosure).

The first concern must be with the quality and completeness of the synthesis report. Recent documents produced for DOE, such as the Technical Basis Report for Surface Processes, have been criticized for being too narrow in scope and not including data and analysis from the published literature. While synthesis reports need to focus on the work that has been conducted by the Yucca Mountain Site Characterization Office (YMSCO), it is also necessary to include discussion of other relevant work, including work with which YMSCO disagrees. References to non-YMSCO work should be more than simple citations and should be broad enough in scope to identify conclusions from these studies that are relevant to the YMSCO work. In some cases, this may require reproduction of some or all of the data from the referenced report. In other cases, a simple restatement of the conclusions with a brief discussion of supporting data may be sufficient. As much data as possible should be incorporated into the synthesis report by reference. However, the reports should be stand-alone documents. This may require some repetition of data presentation with accurate citations. It is clear that the authors of synthesis reports will be asked to address many concerns, some of which are conflicting.

NOV 15 1995

Synthesis reports should be written so that a non-YMSCO scientist, knowledgeable in the topic of the report, can reconstruct the path from the basic data to the report's conclusions. This is what is meant by stand-alone document. Different amounts of detail may be required for different studies and reports. In addition to the detailed technical discussion, the synthesis report should contain an executive summary, written for the general reader, that clearly identifies the purpose of the study and the significance of the conclusion for project objectives.

Synthesis reports should include the identification of qualified and non-qualified data. A discussion must be provided that clearly differentiates qualified and non-qualified data, including non-qualified data from the published literature. The discussion must also identify qualified and non-qualified computer software used for modeling or data analysis. One way to do this is to separate the reference lists and tables into "qualified" and "unqualified" sections.

The conclusions of the report must also be separated into two categories, those that can be drawn from qualified data only and those that can be drawn if the entire data set is considered. Following the controls discussed above will enable the reports to be properly classified as "qualified." The requisite traceability should be provided through inclusion of record accession numbers and Automated Technical Data Tracking system numbers for all data cited or used in the synthesis report.

If you have any questions please call me at 794-7613 or Dennis R. Williams at 794-7968.

Susan B. Jones
Assistant Manager
for Scientific Programs

AMSP:RLR-478

Enclosure:
Draft Letter

cc w/encl:

C. T. Statton, M&O, Las Vegas, NV
L. R. Hayes, M&O, Las Vegas, NV
R. D. Rogers, M&O, Las Vegas, NV
W J. Boyle, YMSCO, NV
S. J. Brocoun, YMSCO, NV

Traceability to Criteria for Milestone SP25BMD 3 *ent 8/29/97*

The table below provides a road map between the requirements listed in the Criteria Statement for this milestone and the specific locations in the document where these requirements are met.

This table is in the executive summary of the report.

TECHNICAL REQUIREMENTS

CRITERIA	TEXT LOCATION
This report will include a conceptual model of the transport of radionuclides in the unsaturated zone at Yucca Mountain, including 2- and 3-D integrated transport calculations and sensitivity analyses.	Conceptual model: 4.3, 6.5.2 2D integrated transport calculation: Chapter 8. 2D sensitivity analyses: Sections 8.5, 8.6, 8.7, 8.8, 8.9 3D integrated transport calculations: Chapter 9 3D Sensitivity analyses: Sections 9.4-9.7
Both equivalent continuum and dual permeability calculations will be made, incorporating discrete structural features as available and appropriate.	Equivalent Continuum calculations: Sections 4.6, 11.4, 11.8 Dual Permeability Calculations: Sections 4.4, 4.7, 5.4, 5.5, 6.7, 6.9-6.12, 8.5-8.11, 9.4-9.7, 10.6-10.9, 12.9 Incorporation of discrete structural features: Sections 3.3, 3.4, 6.5, Included in all 2D and 3D dual permeability calculations.
The model will do the following:	
a) Incorporate and consider isotopic data which are indicative of processes and rates of fluid flow or residence times in the unsaturated zone.	Chapter 6. Processes: 6.5, 6.7-6.12 Rates and Residence Times: 6.10-6.12
b) Draw upon the Report on Summary of Geochemistry/Transport Laboratory Tests to provide defensible values for important transport processes and parameters including:	
1) Sorption and Diffusion Coefficients	8.6, 8.7, 8.10, 9.4, 9.6, 9.7, 11.7

2) Radionuclide solubility and speciation	10.5, 11.4, 11.7
3) Colloids as controlled by relevant thermal effects and water chemistry.	8.10
c) Rely heavily on input from the 3-D mineralogical model of Yucca Mountain.	2.2-2.5, 3.4, 6.5, 8.6, 12.5-12.9
Update 3-D transport simulations for radionuclides including, but not limited to:	
Np	5.5, 8.5-8.7, 8.9, 9.4, 9.5, 10.7, 11.8, 12.9
Pu	8.10, 10.9
Tc	5.5, 8.5-8.7, 8.9, 9.4, 9.5, 10.6
under ambient	5.5, 8.5-8.7, 8.9, 9.4, 9.5, 10.6-10.9
and thermally perturbed	11.8
for two future climates and accompanying infiltration fluxes and effects on recharge	8.7, 8.11, 9.4, 9.5, 11.8
The report will be coupled to appropriate	
near-field source term	10.4, 10.5
and thermal models	11.7, 11.8
The deliverable will be prepared in accordance with OCRWM approved quality assurance procedures implementing requirements of the Quality Assurance Requirements Description.	Done
The product shall be developed on the basis of the best technical data, including both Q and non-q data. The Q status of data used and cited in the report shall be appropriately noted.	QA description of software and data in acknowledgments and in each chapter.
Stratigraphic nomenclature used shall be consistent with the Reference Information Base section 1.12(a): Stratigraphy-Geologic Lithologic Stratigraphy.	Throughout milestone, Table 15.1
References to data used in the report shall include record Accession Numbers or Data Tracking Numbers when available.	QA description of data in acknowledgments and in each chapter.

<p>Technical data contained within the deliverable and not already incorporated in the Geographic Nodal Information Study and Evaluation System (GENISES) shall be submitted for incorporation into the GENISES in accordance with YQP-SIII.3Q.</p>	<p>N/A</p>
<p>Verification of technical data submittal compliance shall be demonstrated by including as part of the deliverable:</p> <p>a) A copy of the Technical Data Information Form generated identifying the data in the Automated Technical Data Tracking system, and</p> <p>b) a copy of the transmittal letter attached to the technical data transmittal to the GENISES Administrator.</p>	<p>a) N/A</p> <p>b) done</p>

Los Alamos

NATIONAL LABORATORY

Earth and Environmental Sciences Division
EES-13 — Nuclear Waste Management R&D
Mail Stop J521, Los Alamos, New Mexico 87545
Phone (505) 667-9768, Fax (505) 667-1934

THIS IS A YMP
MILESTONE *8/29/97*
Plt 3
NO. SP25BMD

August 29, 1997

EES-13-08-97-1666

Larry Hayes
TRW Environmental Safety Systems, Inc.
Mail Station 423
1180 Town Center Drive
Las Vegas, NV 89134

ATTENTION: T. A. Grant

3 Plt 8/29/97
Request for approval of milestone SP25BMD, "Draft Unsaturated Zone Site Transport Model," by B. A. Robinson et al.

Dear Mr. Hayes:

Plt 8/29/97
In compliance with YAP-30.12, milestone SP25BMD, "Draft Unsaturated Zone Site Transport Model," by B. A. Robinson et al. is enclosed for programmatic review.

This paper documents work done under WBS 1.2.3.4.1.5.1; does not fulfill a Level II milestone requirement; and has undergone technical and policy review.

In compliance with YAP-SIII.3Q, there are no new data in this milestone. A table, which lists the data tracking numbers associated with the data generated by other researchers and transmitted to Robinson et al., will be found in the milestone.

In compliance with AP 5.1Q, revision 3, I have attached a YAR form.

Sincerely,

Charles Harrington
Charles Harrington

CH/SHK/jr

- Enclosures:
1. Paper (5 copies)
 2. Review Sheets
 3. YAR

Cy w/enc. 1
P. Dixon, M&O/LANL, Las Vegas, NV
M. Staples, EES-13, J521
J. McGoldrick, M&O/TRW, Las Vegas, NV
M. T. Peters, M&O/WCFS, Las Vegas, NV
T. Hiron, EM-DOE-FP, MS J521
J. E. Young, LATA/EES-13, MS J521

Cy w/enc. 1, 2, and 3
EES-13 File, MS J521
S. H. Klein, CIC-1/EES-13, MS J521
A. L. Thompson, EES-13/LV, MS J902
B. A. Robinson, EES-5, MS F665
A. Sanchez-Pope, ESS/EES-13, MS J521
L. Souza, Mactek, MS J521

PUBLICATION TRAVELER

SECTION I. (Author completes)

TITLE: Draft Unsaturated Zone Transport Model

AUTHOR(S): Robinson et al.

WBS NO: 1.2.3.4.1.5 | UNIQUE IDENTIFIER: LA-EE-13-TIP-97-004 LOS ALAMOS DTN NO.: (If applicable) NA

CHECK ONE

☐ STUDY PLAN

☐ TIP

FOR TIPS:

☐ ABSTRACT

☒ MILESTONE (Number 3) ^{alt 8/29/97}

☐ CONFERENCE PROCEEDINGS

☐ LA MANUSCRIPT REPORT

☐ JOURNAL ARTICLE

☐ OTHER

NAME OF CONFERENCE, DATE, NAME OF JOURNAL ARTICLE, ETC.

LAHS report

SECTION II. (TPO Completes)

I HAVE REVIEWED THE TIP OR STUDY PLAN FOR CONTENT, QUALITY ASSURANCE, AND POLICY.

COMMENTS ATTACHED: ☐ YES ☒ NO (If YES, a Review Sheet will be attached)

TPO: Charles D Harrington Charles D Harrington 8-29-97
Print name Signature Date

SECTION III. (Author completes)

ALL REVIEW COMMENTS HAVE BEEN RESOLVED.

AUTHOR: _____
Print name Signature Date

SECTION IV. (TPO Completes)

THIS TIP OR STUDY PLAN IS ACCEPTABLE AND MAY BE PUBLISHED AS APPROPRIATE

TPO: _____
Print name Signature Date

Los Alamos
Yucca Mountain Site
Characterization Project

TIP/STUDY PLANS REVIEW RESULTS

SECTION I. (Preparer completes)

UNIQUE IDENTIFIER: LA-EE3-13-TIP-97-004 REVISION: 0 TITLE: Draft Unsaturated Zone Transport Model

PREPARER'S NAME: Robinson PHONE: _____ MS: _____ DUE BY: _____
Print name

SECTION II. (Reviewer completes)

REVIEWER INSTRUCTIONS:

1. Review the document for applicability, correctness, technical adequacy, and completeness. Additional criteria may be added.
2. For TIPs, data presented are suitable for the intended use as presented in this report.
3. For comments, enter the location of the section and the proposed actions on the review sheet continuation page. If "no comments," check the "No Comments" box.
4. Any changes to original entries must be initialed and dated.
5. After completing the review, return the review sheets to the preparer.
6. After review comments have been addressed, sign, date, and return review sheet(s) to preparer, if applicable.

☐ Comments Attached

I HAVE FOLLOWED THE INSTRUCTIONS FOR REVIEWING THIS DOCUMENT.

☒ No Comments

REVIEWER: Kay Birdsell Charles Harrington for J521 8-29-97 7-1855
Print name Signature MS or location Date Phone

SECTION III. Signature below indicates that all comments have been resolved satisfactorily.

N/A ☐

REVIEWER: Rogers 8/29/97
Signature Date

Acknowledgments

This work was supported by the Yucca Mountain Site Characterization Office as part of the Civilian Radioactive Waste Management Program. This project is managed by the U.S. Department of Energy, Yucca Mountain Site Characterization Project.

Harold Trease, Terry Cherry, and Kathy Bower assisted in the preparation and testing of the numerical grids used in the model calculations. Guy Roemer performed some of the numerical calculations presented in the environmental isotopes chapter; Al Valocchi and Ines Triay contributed to the study in the areas of reactive transport modeling and radionuclide property values, respectively; Stephen Henderson assisted in the development of the original particle tracking code; June Fabryka-Martin provided valuable editorial comments on the ^{36}Cl modeling study; and Cliff Ho performed the one-dimensional flow calculations with the TOUGH2 code that were benchmarked against the FEHM code. George Zyvoloski consulted with us regularly at all stages of the numerical simulation studies. Kay Birdsell performed a thorough review of the technical and editorial content of this report. Susan Klein, Roger Eckhardt, Laura Wolfsberg, and Angela Sanchez-Pope provided considerable assistance in the editorial phase of the writing of this report, and helped compile the QA information. Finally, the authors are grateful to Lynn Trease for her assistance in preparing this document, and to the Los Alamos YMP Project office for their support during the course of this work.

QA Status of Software Used in This Report

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al, 1992, 1995a, 1995b). The petroleum industry standard software packages STRATAMODEL and RC² are used in the geostatistical analyses of heterogeneous materials. These are off-the-shelf software packages and are applied only to sensitivity studies in

the present study. They are non-Q with respect to YMP and Los Alamos quality assurance requirements but they follow well documented methods for geostatistical representation of spatially varying attributes. The GEOMESH/X3D grid generation toolkit, (Gable et al., 1995, Gable et al., 1996, Trease et al., 1996) is non-Q with respect to YMP and Los Alamos quality assurance requirements. However, version control software tracking procedures identical to those used for the Q code FEHM are followed. GEOMESH/X3D utilizes well established procedures to insure robust mesh generation results and to insure traceability of changes to the software. A suite of test problems has been developed and all new releases are verified against the test suite.

Many sources of data are used in this report. Often, results from other modeling and synthesis studies such as Bodvarsson et al. (1997) are used as input to our modeling studies. If the results of those studies can not be certified as Q, then our results can not be certified as Q either. There are also numerous references to personal communication: they are non-Q.

Data Quality Summary

REFERENCE	DATA STATUS (Q /non-Q)	DTN	CHAPTER(S) WHERE REFERENCE CITED	COMMENT
Altman et. al., 1996 , "Flow Calculations for Yucca Mountain Ground Water Travel Time (GWTT-95). Technical Report SAND96-0819, Sandia National Laboratories, 1996	non-Q	No DTN Assigned	12	No DTN Assigned
Bertetti et. al., 1996 "Neptunium (V) Sorption Behavior on clinoptilolite, Quartz, and Montmorillonite, Mat. Res. Soc. Symp., Proc., Vol. 412, 631-638, 1996			11	

Bodvarsson et. al., 1997 "The Site-Scale Unsaturated Zone model of Yucca Mountain, Nevada, For the Viability Assessment, LBL Technical Report LBNL-40378	Q	LB970601233129.001	3, 5, 6, 8, 9, 10,12	No DTN assigned. Hydrologic property sets computed in this report are used throughout the present study.
Buesch et. al., 1996 , "Revised Stratigraphic nomenclature and macroscopic identification of Lithostratigraphic units of the Paintbrush Group exposed at Yucca Mountain, Nevada, USGS Survey Open-File Report 94-496, 1996	Q	GS941208314211.064	6	
Chipera et. al., 1997 , "Preliminary Three Dimensional Mineralogical Model of Yucca Mountain, Nevada, YMP Milestone SP321AM4"	non-Q	Not Assigned	2, 3, 8, 12	No DTN assigned
Chipera et. al., 1997b "Mineralogic Variation in Drill Holes USW NRG-6, NRG-7-7a, SD-7, SD-9, SD-12, and UZ#14: New Data from 1996-1997 Analysis Los alamos National Laboratory YMP Deliverable SP321BM4"	Q	LADV831321AQ97.001	2,8,12	
Clayton, et. al., 1997 , "ISM2.0: A 3-D geological framework and integrated site model of Yucca Mountain: Version ISM1.0, Doc ID B00000000-01717-5700-00004 Rev 0, MOL. 19970122.0053, CRWMS M&O	Q		3	By agreement with DOE and NRC accepted as Q even though it utilizes some non-Q data

Levy et. al., 1997 , "Investigations of Structural Controls and Mineralogic Associations of Chlorine-36 fast pathways in the ESF,. Yucca Mountain Project milestone report SP2301M4	Q	LASL831222AQ97.001	6	
Nitsche et. al., 1993 , "Measured Solubilities and Speciations of Neptunium , Plutonium, and Americium in a Typical Groundwater (J-13) from the Yucca Mountain Region", Los Alamos National Laboratory Report Number LA-12562-MS, July 1993	non-Q non-Q	LA000000000012.001 LA000000000012.002	11	Accession Nos. MOL.19950705. 0241 NNA.19930507. 0136, and MOL.19950621. 0265, and MOL.19950711. 0329
Ogard and Kerrisk, 1984 , "Groundwater Chemistry along flow paths between a proposed repository site and the accessible environment, Los Alamos National Laboratory Report LA-10188-MS, 1984	non-Q	LA000000000038.001	11	Accession No. NNA.19870406. 0021
Tait et. al., 1996 , "Sorption as a function of groundwater chemistry to elucidate sorption mechanisms, Los Alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3219, 1996	Q Q	LAIT831341AQ95.001 LAIT831341DQ95.002	11	
Triay et. al., 1996a , "Summary and Synthesis Report on Radionuclide Retardation for the Yucca Mountain Site Characterization Project, Los Alamos National Laboratory YMP Milestone Report 3784	Q	LAIT831341AQ96.001	8,9, 10, 11, 12	
Triay et. al, 1996b , "Radionuclide Sorption in Yucca Mountain Tuffs with J-13 Well Water,: neptunium, Uranium, and Plutonium, Los alamos National Laboratory Yucca Mountain Site Characterization Project" Milestone 3338, Technical Report LA-12956-MS	Q	LAIT831341AQ96.001 *	8, 9,10, 11, 12	* DTN super- sedes DTN LA000000000010 4.001
Triay et. al., 1996c , "Batch Sorption Results for neptunium transport through Yucca Mountain tuffs, Los alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3349, Technical Report LA-12961-MS	Q	LAIT831341AQ96.001 *	8,9, 10, 11, 12	*DTN super- sedes DTN LA000000000009 0.001

Triay et. al., 1996d , "Comparison of Neptunium Sorption Results using batch and column techniques, Los Alamos National laboratory Yucca Mountain Site Characterization Project Milestone 3041, Technical Report LA-12958-MS	Q	LA000000000106.001	8, 9, 10, 11, 12	DTN Accession No. MOL.19950630.0300
Triay et. al., 1997 , "Summary Report on Geochemistry/Transport Lab Tests, Los Alamos National Laboratory Milestone SP23QMD	Q	LAIT831341DQ97.003	10	
Yang et. al., 1992 , "Flow and Transport Through Unsaturated Rock - Data From Two Test Holes, Yucca Mountain, Nevada, proc. Third International Conference on High-Level Radioactive Waste Management, Las Vegas, NV, 1992	Q		6	
Yang, et. al., 1996 , Interpretations of Chemical and Isotopic Data From Boreholes in the Unsaturated Zone at Yucca Mountain, Nevada, USGS Water-Resources Investigations Report WRIR 96-4058	Q		4, 6, 11	
Yang et. al., 1997 , Hydrochemical investigations and geochemical modeling in characterizing the unsaturated zone at Yucca Mountain, Nevada, (<i>in preparation</i>).	non-Q	Indeterminate	6, 7	Not Available Yet
Zelinski and Clayton, 1996 , "A 3-D geological framework and integrated site model of Yucca Mountain: Version ISM1.0, Doc ID B00000000-01717-5700-00002 Rev 1", CRWMS M&O	non-Q		2, 3	

Table of Contents

Chapter 1 - Executive Summary	1-1
1.1 Overview	1-1
1.2 Incorporation of Mineralogic Model into Flow and Transport Simulations	1-2
1.3 Mesh Generation	1-3
1.4 Particle Tracking	1-4
1.5 Hydrologic Property Sets	1-5
1.6 Environmental Isotopes and the Hydrologic System	1-7
1.7 Water Chemistry	1-8
1.8 Two-Dimensional Radionuclide Transport Simulations	1-10
1.9 Three-Dimensional Radionuclide Transport	1-11
1.10 Unsaturated Zone Performance Predictions	1-12
1.11 Reactive Transport of ²³⁷ Np with Repository Heat	1-13
1.12 Incorporation of Geostatistics into Flow and Transport Simulations	1-14
1.13 Traceability to Criteria for Milestone SP25BM3	1-16
Chapter 2 - Incorporation of the Mineralogic Model into the Site-Scale Flow and Transport	
Model	2-1
2.1 Abstract	2-1
2.2 Mesh Generation	2-2
2.3 Software and Data QA Status	2-2
2.4 Motivation for the Current Mineralogic and Hydrologic Model	2-3
2.5 Mineralogic Model	2-6
2.6 Conclusions	2-11
Chapter 3 - Mesh Generation	3-1
3.1 Abstract	3-1
3.2 QA Status of Software and Data	3-1
3.3 Unsaturated Zone Computational Mesh from Stratigraphic Model	3-3
3.3.1 Mesh Generation from ISM Input	3-4
3.3.1.1 Import ISM Data into GEOMESH/X3D	3-4
3.3.2 Quality Testing and Correcting of ISM TINs	3-4
3.3.2.1 Identify Faults	3-6
3.3.2.2 Define polygonal subset of TIN surface	3-6
3.3.2.3 Refine TIN surface grids in and adjacent to faults	3-7
3.3.2.4 Define upper and lower boundary truncation surfaces	3-7
3.3.2.5 Define minimum layer thickness	3-9
3.3.2.6 TIN Sheets Stacked and Converted to Prisms	3-9
3.3.2.7 Convert Prisms to Tetrahedra	3-10
3.3.2.8 Extract a Two-Dimensional Plane	3-10
3.3.2.9 Convert Hybrid Grid to Triangles	3-10
3.3.2.10 Clip Mesh to Area of Interest	3-11
3.3.2.11 Optimize Mesh For Computation	3-11
3.3.2.12 Mapping Properties From Three-Dimensional Mineral	

Table of Contents

	Distribution	3-15
3.3.3	Generation of FEHM Input Files	3-16
3.3.3.1	Compute Finite Volume Matrix Coefficients	3-16
3.3.3.2	Compute Area Coefficients of Surface	3-16
3.3.3.3	Output Information for Boundary Conditions and Material Properties	3-16
3.3.4	Mesh Quality Assurance Steps	3-16
3.4	Computational Grids Used in the Present Study	3-17
3.4.1	Three-Dimensional Unsaturated Zone Grid	3-17
3.4.2	Two-Dimensional Unsaturated Zone Grid	3-17
3.5	Conclusions	3-19
Chapter 4 -	Particle Tracking	4-1
4.1	Abstract	4-1
4.2	Software and Data QA Status	4-2
4.3	Introduction: Conceptual Model For Particle Transport	4-3
4.4	Mathematical Development	4-6
4.4.1	Dispersion	4-9
4.4.2	Matrix Diffusion	4-11
4.4.3	Radioactive Decay	4-14
4.4.4	Particle Sources and Sinks	4-14
4.4.5	Transient Flow Fields	4-15
4.4.6	Dual-Permeability Formulation	4-16
4.5	Code Implementation	4-16
4.6	Validation	4-19
4.6.1	One-Dimensional Transport with Dispersion and Sorption	4-19
4.6.2	One-Dimensional Transport with Matrix Diffusion	4-21
4.6.3	Transient Wetting of a 1-D Column	4-22
4.6.4	Benchmarking Calculation for an Interwell Tracer Simulation	4-24
4.7	Applications	4-27
4.7.1	Dual-Permeability Transport for the Unsaturated Zone	4-27
4.7.2	Simulations of Environmental Isotopes in the Unsaturated Zone	4-30
4.7.3	Sorbing tracers with matrix diffusion	4-34
4.8	Conclusions	4-38
Chapter 5 -	Property Sets	5-1
5.1	Abstract	5-1
5.2	Software and Data QA Status	5-2
5.3	Introduction	5-3
5.4	Comparison of TOUGH2 and FEHM Codes	5-5
5.4.1	Flow Results	5-8
5.4.2	Transport result	5-10
5.5	Screening of Hydrologic Property Sets	5-14
5.5.1	Case 6412a	5-26
5.5.2	Case 6412b	5-36
5.5.3	Case 6511	5-36
5.5.4	Case 6521	5-37

Table of Contents

5.5.5 Case 6522.....	5-38
5.5.6 Case 6531a.....	5-38
5.5.7 Case 6531b.....	5-39
5.5.8 Case 6541.....	5-39
5.6 Conclusions	5-41
Chapter 6 - Environmental Isotopes and The Hydrologic System	6-1
6.1 Abstract	6-1
6.2 Software and Data QA Status	6-2
6.3 Introduction	6-4
6.3.1 Implications	6-5
6.4 Background	6-6
6.4.1 Previous Chlorine-36 Studies at Yucca Mountain and Their Implication for Flow and Transport Modeling	6-6
6.4.2 Infiltration Studies	6-9
6.4.3 Present Study Summary.....	6-10
6.5 Hydrogeologic Setting	6-11
6.5.1 Stratigraphy	6-11
6.5.2 Conceptual model of flow and transport	6-12
6.6 Chlorine-36 Study	6-15
6.6.1 Background	6-15
6.6.2 Chlorine-36 input.....	6-15
6.6.3 ESF Data	6-18
6.7 Flow and Transport Model	6-18
6.7.1 Numerical Flow and Transport Model: FEHM	6-18
6.7.2 Transient Signals of Isotopic Sources	6-19
6.8 Simulated Systems	6-20
6.8.1 Overview	6-20
6.9 Carbon-14 Study	6-22
6.9.1 Background	6-22
6.9.2 Carbon-14 Data	6-23
6.9.3 Carbon-14 Input.....	6-25
6.10 Chlorine-36 Modeling: Two- and Three- Dimensional Studies	6-25
6.10.1 Model Description	6-25
6.10.2 Two-Dimensional Chlorine-36 Simulation Results	6-27
6.10.3 Three-Dimensional Chlorine-36 Simulation Results	6-34
6.10.4 South to North Trends in Potential Repository Block Region	6-44
6.11 Carbon-14 Modeling	6-51
6.12 One-Dimensional Process Level Modeling	6-54
6.12.1 Column Descriptions	6-54
6.12.2 One-Dimensional Modeling Approach.....	6-56
6.12.3 Steady-State Results	6-58
6.12.4 Comparisons: Three Property Sets for Steady-State and Transient Flow	6-65
6.13 Conclusions	6-71
Chapter 7 - Water Chemistry	7-1

Table of Contents

7.1	Abstract	7-1
7.2	Software and Data QA Status	7-2
7.3	Introduction	7-5
7.4	Hydrochemical setting	7-5
	7.4.1 TCw: Surface Infiltration.....	7-6
	7.4.2 PTn: Transition from fracture dominated flow to matrix dominated flow.....	7-6
	7.4.3 TSw: Transition from matrix dominated flow to fracture dominated flow.....	7-6
	7.4.4 CHn: composite vertical flow and lateral diversion	7-7
	7.4.5 Water Table: Seepage via fractures and matrix flow from the base of the CHn into the saturated zone.....	7-9
7.5	Perched Water Chemistry	7-9
	7.5.1 The Origin and Significance of Perched Water.....	7-9
	7.5.2 Hydrologic evidence for perched waters	7-11
	7.5.3 Chemical Composition of Perched Waters	7-14
7.6	Conceptual Models for Perched Water	7-15
	7.6.1 Fast-Pathway Fracture Flow Hypothesis.....	7-15
	7.6.2 Transient Mixing Hypothesis.....	7-18
	7.6.2.1 Climatic Evidence	7-18
	7.6.2.2 Structural and Mineralogical Setting.....	7-19
	7.6.2.3 Hydro-geochemical Model	7-23
	7.6.3 Evaluation of Alternate Conceptual Models	7-28
	7.6.3.1 The Radioactive Isotopes of Hydrogen, Chlorine and Carbon	7-29
	7.6.3.2 The Conservative Environmental Tracers Chloride and Cl/Br.....	7-30
	7.6.3.3 The Stable Isotopes	7-32
7.7	Simulation Results	7-33
7.8	Summary and Discussion	7-40
	7.8.1 The Hydrologic Data	7-42
	7.8.2 The Hydrochemical Data.....	7-42
	7.8.3 The Model Results.....	7-43
7.9	Conclusions	7-45
Chapter 8 -	Two-Dimensional Radionuclide Transport Simulations	8-1
8.1	Abstract	8-1
8.2	Software and Data QA Status	8-2
8.3	Introduction	8-3
8.4	Methodology for Radionuclide Migration Calculations	8-4
8.5	The Base Case	8-5
8.6	The Influence of Hydrologic and Geochemical Parameter Sets	8-13
8.7	The Effect of Percolation Flux	8-16
8.8	Grid Refinement Study	8-21
8.9	Uncertainties in Zeolite Distributions and Property Variations	8-26
8.10	Plutonium Colloid Transport Analysis	8-30

Table of Contents

8.10.1	Model Development	8-31
8.10.2	Results of Colloid Transport Simulations	8-36
8.10.3	Discussion of Colloid-Facilitated Transport Results	8-42
8.11	Transient Analyses	8-47
8.12	Conclusions	8-55
Chapter 9 -	Three-Dimensional Radionuclide Transport	9-1
9.1	Abstract	9-1
9.2	Software and Data QA Status	9-1
9.3	Introduction	9-2
9.4	Radionuclide Simulations, Base Infiltration Rate	9-3
9.5	Radionuclide Simulations, 1/3 Infiltration Map	9-5
9.6	Large Scale Spatial Variability in Radionuclide Transport	9-7
9.7	Transport parameter sensitivity studies	9-10
9.8	Conclusions	9-15
Chapter 10 -	Unsaturated Zone Performance Predictions	10-1
10.1	Abstract	10-1
10.2	Software and Data QA Status	10-2
10.3	Introduction	10-3
10.4	Radionuclide Transport Properties	10-4
10.5	Abstracted Models for NFE Source Term	10-6
10.6	Transport Predictions for ⁹⁹ Tc	10-9
10.7	Transport Predictions for ²³⁷ Np	10-16
10.8	Transport Predictions for Uranium Isotopes	10-21
10.9	Transport Predictions for Plutonium Isotopes	10-24
10.10	Conclusions	10-27
Chapter 11 -	Reactive Transport of ²³⁷ Np with Repository Heat	11-1
11.1	Abstract	11-1
11.2	Software and Data QA Status	11-1
11.3	Introduction	11-3
11.4	Model Description	11-5
11.4.1	Flow and Energy Formulations	11-5
11.4.2	Reactive Transport Equations	11-6
11.4.2.1	Governing Equations for the PDVs	11-8
11.4.2.2	Governing Equations for the SDVs	11-9
11.5	Solution Procedure	11-10
11.5.1	Solution Method for the PDVs:	11-11
11.5.2	Solution Method for the SDVs	11-14
11.6	Yucca Mountain Unsaturated Zone Hydrology and Geochemistry	11-15
11.6.1	Hydrogeologic Setting	11-15
11.6.1.1	Unsaturated Zone Geochemistry	11-16
11.6.2	Fluid Percolation Rate	11-19
11.6.3	Computational Domain	11-21
11.7	Neptunium Chemistry	11-21
11.7.1	Solubility and ²³⁷ Np Inventory	11-23

Table of Contents

11.7.2	Speciation	11-24
11.7.3	Sorption Data	11-27
11.7.4	Sorption Models	11-27
11.7.5	Column Studies	11-31
11.8	Model Results	11-32
11.8.1	Preliminary One-Dimensional Flow and Transport Simulations ..	11-32
11.8.2	Thermohydrologic Simulations	11-35
11.8.3	Neptunium Transport Simulations	11-41
11.9	Discussion and Conclusions	11-50
11.10	Conclusions	11-55
Chapter 12	- Incorporation of Geostatistics into Flow and Transport Simulations	12-1
12.1	Abstract	12-1
12.2	Software and Data QA Status	12-2
12.3	Motivation	12-3
12.4	Geostatistical Integration	12-7
12.5	Implications of Zeolite Threshold Cutoff	12-7
12.6	Relationships Between Physical and Chemical Heterogeneity at Yucca Mountain	12-12
12.6.1	Correlation between Permeability and Zeolitic Abundance	12-14
12.6.2	Correlation between Porosity and Permeability	12-14
12.6.3	Correlation Between Permeability and Reactivity	12-15
12.6.4	Correlation Between Permeability and Relative Permeability	12-17
12.6.5	Summary of Field Data	12-17
12.7	Generating Realistic Zeolite Distributions	12-18
12.8	Generating Correlated Properties from Maps of Zeolitic Abundance ...	12-23
12.9	Flow and Transport Simulations with Geostatistics	12-26
12.10	Conclusions	12-32
Chapter 13	- Conclusions	13-1
Chapter 14	- References	14-1
Chapter 15	- Appendix	15-1

List of Figures

- 2-1 Antler Ridge cross section (a) Continuous zeolitic layers (in red) of the Site-Scale Model used in Robinson et al. (1996), (b) Zeolitic distributions are mapped from the 3-D mineralogical model (Chipera et al, 1997a) to the FEHM finite element mesh. The profiles look different because (b) accounts for fault offsets. 2-5
- 2-2 Locations of the 21 wells in plan view of mineralogical model. 2-8
- 2-3 Comparison of the (a) Chipera et al. (1997a) model and (b) the updated mineralogical model (Chipera et al., 1997b) for the Antler Ridge cross section. 2-9
- 2-4 Comparison of Chipera et al. (1997a) and updated mineralogical model (Chipera, 1997b) at locations close to wells (a) zeolite abundance at G1 (G1 existed in both models), (b) zeolite abundance at UZ-14 (UZ-14 only exists in the updated model but is close to G1), (c) smectite abundance at NRG6 (NRG6 only exists in the updated model). 2-10
- 3-1 Layer 9 of ISM shown with colors representing elevation. The area in which a three-dimensional tetrahedral grid is build is shown by the wire frame in which each line represents a stratigraphic layer boundary. The two-dimensional grid used for computations is shown with zeolite abundance shown with red for high zeolite abundance and blue for low zeolite abundance. 3-3
- 3-2 Top view of one TIN from ISM. TIN consists of 4976 nodes and 9823 triangles. Coordinates are Nevada State Plane in meters. Each triangle vertex is assigned an elevation, resulting in a piecewise linear representation of stratigraphic surfaces. 3-5
- 3-3 Layer 38 from ISM at 3X vertical exaggeration. Stratigraphic layer boundary is colored in yellow. West facing faults are green, south facing faults are blue. 3-7
- 3-4 Top view of one TIN illustrating the subregion for the three-dimensional computational grid. TIN consists of 4557 nodes and 6165 triangles. Coordinates are Nevada State Plane in meters. Note the increased resolution near faults in comparison to the original TIN surface (see Figure 3-2). 3-8
- 3-5 Cross section at NSP coordinate 232,100 meters extracted from ISM. In the figure, the cross section is split in two near the Ghost Dance Fault (GDF). The east and west extent of the computational model at NSP 168,274 meters and 174,176 meters are marked by arrows. Colors represent hydrogeologic units incorporated into the computational mesh. 3-11
- 3-6 Two-dimensional mesh generated from YMP model ISM showing faults as part of the computational grid. The faults from west (left) to east (right) are Solitario Fault, Ghost Dance Fault and Bow Ridge Fault. The cross section is bounded on the west and east by unnamed faults in ISM. Colors represent different hydrogeologic units. Close-up views of the grid are shown at each of the faults. 3-13
- 3-7 An example of how elliptic smoothing modifies the mesh. The mesh on the left has the topology that results from extracting a two-dimensional grid from a three-dimensional tetrahe-

List of Figures

- dral mesh. Material interfaces are maintained but the node distribution is very uneven. The mesh on the right has been smoothed by keeping interface nodes fixed but allowing nodes between material interfaces to move. The mesh on the right has a flat bottom by truncating the mesh at elevation 720 meters. 3-14
- 3-8 Zeolite distributions are mapped from a three-dimensional model onto the computational mesh. The upper figure show zeolite distributions on a grid extracted from the Stratamodel three-dimensional mineralogy model. The lower figure shows the zeolite distributions after being mapped onto the computational mesh. 3-15
- 3-9 Three-dimensional unsaturated zone computational grid with 71,781 nodes, 143,562 dual permeability nodes and 414,313 tetrahedral elements. 3-18
- 3-10 Faults are represented as distinct materials. Layer 9 from ISM is shown. There are 14,473 nodes and 45,386 tetrahedral elements used to represent the faults. Faults that dip to the west are filled in red, faults that dip to the east are filled with green. 3-19
- 3-11 A portion of the two-dimensional cross section is shown bounded on the west by the Solitario fault and on the east by the Ghost Dance fault. a) Stratigraphic layer boundaries, b) low resolution computational grid, c) high resolution computational grid. 3-20
- 3-12 A small section of the high resolution two-dimensional grid near the Solitario fault showing the triangulation and the Voronoi cells associated with the triangles. 3-21
- 4-1 Use of the transfer function to determine the residence time of a particle in a cell for the RTTF particle tracking technique. 4-8
- 4-2 Conceptual model for matrix diffusion. One dimensional flow occurs in the fracture, with molecular diffusion into the rock matrix. 4-12
- 4-3 Flow chart illustrating the computation sequence for the particle tracking algorithm and its relationship to the fluid flow calculations in a finite element flow code. 4-18
- 4-4 Cumulative RTD curves produced by the RTTF particle tracking technique for one-dimensional dispersion (filled circles), compared to analytical solution (solid curves). In the curves without sorption, was varied. For the dashed curve, linear sorption was included. ... 4-20
- 4-5 Residence time distribution (RTD) curves calculated using the RTTF particle tracking technique (symbols) for for different numbers of particles, compared to the analytical solution (solid curve). The RTD is a solute concentration normalized so that the area under the curve is unity. 4-21
- 4-6 Cumulative RTD curves produced by the RTTF particle tracking technique for the matrix diffusion model (filled circles), compared to the analytical solution of Tang et al. (1981) (solid curves). Curve a) small amount of matrix diffusion; Curve b) moderate amount of matrix diffusion; Curve c) Matrix diffusion and sorption on fracture and in matrix; Curve d) Dispersion, matrix diffusion, and sorption on fracture and in matrix. 4-22
- 4-7 Movement of particles with the saturation front for the transient wetting case. The curves are

List of Figures

- the saturation profiles at regular time intervals. Simulations are depicted in which the particles start at the entrance to the flow path (the filled circles) and at a position part way down the column (the X's). For the latter case, the particles do not move until the saturation front reaches the particles. 4-23
- 4-8 Model geometry and finite element grid for the two-dimensional models of interwell tracer tests at the C-Wells, Yucca Mountain, Nevada. The solid circles are an injector and producer for the case of wells aligned with the grid, and the open circles are the same for the case of wells misaligned with respect to the grid. 4-25
- 4-9 Comparison of RTTF particle tracking simulation (solid curves) and finite element CD equation solution (dotted curve). The two particle tracking curves show the effect of varying the dispersivity (0 and 1 m). 4-26
- 4-10 Schematic of the one-dimensional model system of the vadose zone at Yucca Mountain used for the dual permeability particle tracking example. 4-28
- 4-11 Composite solute breakthrough curves (fracture plus matrix contributions) at the bottom of each hydrogeologic unit at 1 mm/y infiltration rate using the dual permeability particle tracking model (solid curves). The dashed curves are comparable results assuming an equivalent continuum model. 4-29
- 4-12 Reconstructed $^{36}\text{Cl}/\text{Cl}$ input ratio used in the particle tracking simulations (from Fabryka-Martin et al., 1996c). 4-31
- 4-13 Simulated concentrations of $^{36}\text{Cl}/\text{Cl}$ ratio (a) and ^{14}C in the two-dimensional model domain. For ^{36}Cl , the bomb-pulse signal is red, and the values at the ESF horizon are in agreement with field measurements. For ^{14}C , values in the PTn (above ESF) and CHn (below ESF) are in qualitative agreement with field measurements. 4-33
- 4-14 Simulations with and without matrix diffusion for the two-dimensional interwell tracer experiments modeled in Figure 4-8. Also shown is a simulation of a tracer that sorbs to the matrix rock (the dotted curve). 4-35
- 4-15 Breakthrough curves for a homogeneous permeability field examining the effect of grid orientation on the transport behavior of the RTTF particle tracking method. Solid curves: wells aligned with the grid; curves with symbols: particle tracking solutions. 4-37
- 5-1 Comparison of computed matrix and fracture saturations for the FEHM and TOUGH2 simulations. 5-8
- 5-2 Normalized flux of water in the fracture and matrix continua: FEHM and TOUGH2 simulations. 5-10
- 5-3 Normalized flux of water in the fracture and matrix continua: FEHM and TOUGH2 simulations on a scale expanded to examine the CHn units. Also included is an FEHM simulation with a 4 m thick vitric CHn layer, instead of the original 10 m thick layer. 5-11
- 5-4 Breakthrough of ^{99}Tc at the water table for the one-dimensional SD-9 model: FEHM and

List of Figures

	TOUGH2 generated flow fields, FEHM particle tracking for transport.	5-12
5-5	Breakthrough of ²³⁷ Np at the water table for the one-dimensional SD-9 model: FEHM and TOUGH2 generated flow fields, FEHM particle tracking for transport.	5-13
5-6	Case 6412a hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-18
5-7	Case 6412b hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-19
5-8	Case 6511 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-20
5-9	Case 6521 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-21
5-10	Case 6522 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-22
5-11	Case 6531a hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-23
5-12	Case 6531b hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-24
5-13	Case 6541 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.	5-25
5-14	Case 6412a transport results for the one-dimensional SD-9 model. a) ⁹⁹ Tc; b) ²³⁷ Np. One mole of radionuclide is released at the repository horizon at time = 0.	5-27
5-15	Case 6412b transport results for the one-dimensional SD-9 model. a) ⁹⁹ Tc; b) ²³⁷ Np. One mole of radionuclide is released at the repository horizon at time = 0.	5-28
5-16	Case 6511 transport results for the one-dimensional SD-9 model. a) ⁹⁹ Tc; b) ²³⁷ Np. One mole of radionuclide is released at the repository horizon at time = 0.	5-29
5-17	Case 6521 transport results for the one-dimensional SD-9 model. a) ⁹⁹ Tc; b) ²³⁷ Np. One mole of radionuclide is released at the repository horizon at time = 0.	5-30
5-18	Case 6522 transport results for the one-dimensional SD-9 model. a) ⁹⁹ Tc; b) ²³⁷ Np. One	

List of Figures

mole of radionuclide is released at the repository horizon at time = 0.	5-31
5-19 Case 6531a transport results for the one-dimensional SD-9 model. a) 99Tc; b) 237Np. One mole of radionuclide is released at the repository horizon at time = 0.	5-32
5-20 Case 6531b transport results for the one-dimensional SD-9 model. a) 99Tc; b) 237Np. One mole of radionuclide is released at the repository horizon at time = 0.	5-33
5-21 Case 6541 transport results for the one-dimensional SD-9 model. a) 99Tc; b) 237Np. One mole of radionuclide is released at the repository horizon at time = 0.	5-34
6-1 Conceptual model of hydrologic pathways at Yucca Mountain.	6-13
6-2 Reconstructed $^{36}\text{Cl}/\text{Cl}$ ratio in infiltrating water (Fabryka-Martin et al., 1996c, Plummer et al., 1996), a) last 500,000 years, b) last 50,000 years, c) last 50 years.	6-17
6-3 Plan view of three-dimensional model domain with locations of ESF, ESF stations, two-dimensional east-west model transect, north-south transect, and one-dimensional vertical column studies.	6-21
6-4 Carbon-14 decay function resulting from a 5730 year half-life.	6-25
6-5 Finite-element grid for the two-dimensional cross section at Antler Ridge. Northing location in NSP coordinates is 232100 m. Red arrows show easting location for potential repository horizon breakthrough study.	6-27
6-6 Infiltration rates extracted from Flint et al. (1996) map along Antler Ridge cross section. I map is directly extracted, I/3 map is the I map scaled by 3. Green arrows show easting location for potential repository horizon breakthrough study.	6-28
6-7 Plan view of three-dimensional domain outline on top of Flint et al. (1996) infiltration map. Also shown for reference are the ESF, the east-west two-dimensional cross section and the north-south transect (used to interpret the three-dimensional model results).	6-29
6-8 Simulated $^{36}\text{Cl}/\text{Cl}$ ratios in Antler Ridge 2-D cross section with 6541a material property set and Flint (1996) infiltration map.	6-31
6-9 Simulated $^{36}\text{Cl}/\text{Cl}$ ratios in Antler Ridge 2-D cross section with 6541b material property set (no weighting of fracture-matrix connection in PTn) and Flint (1996) infiltration map. Note the reduced amount of bomb-pulse in the top of the CHn.	6-31
6-10 Difference in simulated $^{36}\text{Cl}/\text{Cl}$ ratios for property sets 6541a and 6541b.	6-31
6-11 Simulated saturations in Antler Ridge 2-D cross section with 6541a material property set and Flint (1996) infiltration map.	6-32
6-12 Simulated saturations in Antler Ridge 2-D cross section with 6541b material property set and Flint (1996) infiltration map.	6-32
6-13 Difference in simulated saturations for property sets 6541a and 6541b.	6-32

List of Figures

- 6-14 Particle pathways from 5 different surface locations. a) pathways in the matrix for locations near faults, b) pathways in the fractures for locations near faults, c) pathways in the matrix for locations in faults, d) pathways in the fractures for locations in faults. 6-35
- 6-15 Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541a set. Infiltration rate at surface is the Flint et al. (1996) map. Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m). 6-36
- 6-16 Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541b set (no weighting of fracture-matrix connection in PTn). Infiltration rate at surface is the Flint et al. (1996) map. Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m) 6-36
- 6-17 Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541 set. Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m). 6-37
- 6-18 Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541b set (no weighting of fracture-matrix connection in PTn). Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m). 6-37
- 6-19 Three-dimensional simulation of $^{36}\text{Cl}/\text{Cl}$ ratios using property set 6541a and infiltration map of Flint et al. (1996). 6-38
- 6-20 Simulated $^{36}\text{Cl}/\text{Cl}$ ratios at ESF horizon with three-dimensional transport model. Uses material property set 6541a and infiltration map from Flint et al. (1996). 6-39
- 6-21 Simulated $^{36}\text{Cl}/\text{Cl}$ ratio in ESF with 3-D transport model and measured ratios from Levy et al. (1997). Simulations performed using 6541 parameter set and Flint et al. (1996) infiltration map 6-40
- 6-22 Solute travel path in the matrix from the top of Yucca Crest into the UZ system. . . . 6-41
- 6-23 Solute travel path in the fractures from the top of Yucca Crest into the UZ system. . . 6-41
- 6-24 Breakthrough (or cumulative age distribution of a porewater sample) in northern ESF simulated with 3D model. Material properties are the 6541a set. Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters). 6-45
- 6-25 Breakthrough (or cumulative age distribution of a porewater sample) in southern ESF simulated with 3D model. Material properties are the 6541a set. Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters). 6-45

List of Figures

- 6-26 Breakthrough (or cumulative age distribution of a porewater sample) in northern ESF simulated with 3D model. Material properties are the 6541b set (no weighting of fracture-matrix connection in PTn). Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters). 6-46
- 6-27 Breakthrough (or cumulative age distribution of a porewater sample) in southern ESF simulated with 3D model. Material properties are the 6541b set (no weighting of fracture-matrix connection in PTn). Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters). 6-46
- 6-28 Breakthrough (or cumulative age distribution of a porewater sample) in northern ESF simulated with 3D model. Material properties are the 6541b set (no weighting of fracture-matrix connection in PTn). Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters). 6-47
- 6-29 Breakthrough (or cumulative age distribution of a porewater sample) in southern ESF simulated with 3D model. Material properties are the 6541b set (no weighting of fracture-matrix connection in PTn). Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters) 6-47
- 6-30 North-South transect cumulative age distributions at potential repository horizon using I infiltration map. a) Property set 6541a b) Property set 6541b. Legend indicates northing coordinate. Easting coordinate is 170600 m. 6-49
- 6-31 North-South transect cumulative age distributions at potential repository horizon using I/3 infiltration map. a) Property set 6541a b) Property set 6541b. Legend indicates northing coordinate. Easting coordinate is 170600 m. 6-49
- 6-32 Carbon-14 simulation with Flint et al. (1996) infiltration map and 6541a properties. Values plotted are percent modern carbon (PMC) 6-52
- 6-33 Carbon-14 simulation with Flint et al. (1996) infiltration map and 6541b properties. Values plotted are percent modern carbon (PMC) 6-52
- 6-34 Carbon-14 simulation for perched water simulation. Values plotted are percent modern carbon (PMC). a) Matrix PMC with I map, b) Fracture PMV with I map, c) Matrix PMC with I/3 map, d) Fracture PMC with I/3 map 6-53
- 6-35 Schematics of three columns used in one-dimensional model studies. 6-55
- 6-36 Breakthrough curves at ESF/potential repository level for a) Station 35, b) Station 57, c) the repository column, and d) a comparison of the three at 5 mm/yr. Steady state infiltration varying between 0.5 and 20 mm/yr, and parameter set 6541a 6-60
- 6-37 Simulated saturations in the Station 35 column for parameter set 6541a. 6-61
- 6-38 Simulated flux distribution in the Station 35 Column for parameter set 6541a at 1 and 10 mm/yr infiltration rate. 6-61
- 6-39 Simulated flux distribution in the Station 57 column for parameter set 6541a at 1 and 10 mm/yr infiltration rate. 6-61

List of Figures

- yr infiltration rate. Also shown is lowest 5% of total flow to highlight non-zero fracture fluxes in PTn. Layer resolution in Calico Hills not provided due to focus above ESF in these simulations. 6-62
- 6-40 Breakthrough curve at ESF for property set 6412b at Station 35. Also shown is flux distribution for 5 mm/yr case. 6-63
- 6-41 Comparison of saturations simulated in upper section of repository column with property sets 6541a and 6541b for 10 mm/yr. 6-64
- 6-42 Station 35 breakthrough comparison for 10 mm/yr average annual infiltration rate. Three property sets (identified in legend) compared for steady-state (ss) and transient (trans) infiltration. Transient model, 10-5-10, is 10 mm/yr accumulation applied over 10 days every 5 years. Log plot on right illustrates component of early arrival 6-67
- 6-43 Station 57 breakthrough comparison for 3 property sets at steady state (ss) and in transient (trans) infiltration model, each with mean annual infiltration rate of 10 mm/yr. Property sets are 6541a, 6541b, and 6541ft. Transient mode is 10 mm/yr accumulated over 10 days every 5 years (10-5-10). 6-67
- 6-44 .Repository column breakthrough comparison for 3 property sets at steady state (ss) and in transient (trans) infiltration model, each with mean annual infiltration rate of 10 mm/yr. Property sets are 6541a, 6541b, and 6541ft. Transient mode is 10 mm/yr accumulated over 10 days every 5 years (10-5-10). 6-68
- 6-45 Comparison of breakthrough curves at potential repository horizon for transient simulations with a) 20 mm/yr average and b) 50 mm/yr average. All water is applied for 10 days every 5 years. 6-69
- 6-46 Comparison of breakthrough curves for 10 day duration transient and 90 day duration transients. Average annual infiltration for all cases is 10 mm/yr. 6-69
- 6-47 Simulated flux variation responses at potential repository for 10-5-10 transient infiltration rate at the surface. 300 years of transient followed by 700 years of uniform 10 mm/yr infiltration. The shaded portion of the 6541ft curve represents the short-term transients at the repository horizon. 6-70
- 7-1 Map of Yucca Mountain site area with boreholes (i.e., G-2, UZ-14, NRG-7/7a, SD-9, SD-12, and SD-7) where perched water has been observed. Green line: site-scale model boundary. Blue: ESF. Red line: outline of potential repository. 7-8
- 7-2 Reconstructed ^{14}C and ^{36}Cl activities for the last 50 ka. From Fabryka-Martin et al. (1996c). 7-25
- 7-3 Reconstructed ^{14}C and ^{36}Cl activities in the atmosphere for the last 20 ka, from Fabryka-Martin et al. (1996c). USW UZ-14A and UZ-14C were shown to be contaminated with modern water. Perched water data from UZ-14, SD-7 and NRG-7/7a. 7-26
- 7-4 Chloride concentration predicted in the three-dimensional model. Top figure: map of chlo-

List of Figures

- ride concentrations at the surface. Bottom figure: concentrations at and elevation of 900 m. Top of page is North. 7-35
- 7-5 EW cross-section of transport site-scale model looking North. Chloride concentrations for the base infiltration map (upper figure) and the I/3 infiltration scenario (bottom figure). 7-36
- 7-6 Detail of EW cross-section of transport site-scale model in area of relatively low infiltration. Close-up of chloride concentrations to the East of the Ghost Dance fault for the I/3 scenario. 7-37
- 7-7 EW cross-section of full site scale transport model, looking North through Yucca Crest. Chloride to bromide ratio (top figure) and sulfate to chloride ratio (bottom figure) simulated using the base case property set and infiltration map. 7-38
- 7-8 Detail of EW cross-sections from the site scale transport model through Yucca Crest, looking North. Fracture saturations predicted using the basal vitrophyre lateral diversion model. Top figure: base infiltration map; bottom figure: I/3 scaled map. Blue areas represent zones of perching with lateral flow along the top of the basal TSw vitrophyre. Principal source of water is from Yucca Crest in both cases. 7-40
- 7-9 Detail of EW cross-section (looking North) of transport site-scale model through Yucca Crest. Chloride concentration (top), sulfate concentration (middle), and sulfate to chloride ratio (bottom) for the basal vitrophyre lateral diversion hydrologic scenario. 7-41
- 7-10 Three-dimensional simulation of transport for particles injected at the surface above the potential repository. Image represents a cut-away fence diagram from the three-dimensional grid. Represented are the Solitario (S.C.) and Ghost Dance (G.D.) faults and the ESF. Particles undergoing matrix flow are represented with the red colors corresponding to a high particle concentration. Significant lateral diversion is predicted in the vitric Calico Hills matrix (beneath the TSw basal vitrophyre) and to the Ghost Dance fault. 7-44
- 8-1 Matrix (top) and fracture (bottom) saturations for the two-dimensional base-case simulation. Parameter set 6541, base infiltration map (I). Also shown are the I and I/3 infiltration map values across the model domain. 8-7
- 8-2 Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map. 8-8
- 8-3 Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map. 8-10
- 8-4 Water table breakthrough of ^{237}Np for the base case. Parameter set 6541, base infiltration map, $k_d = 2 \text{ cc/g}$, constant release rate from 1 ky to 31 ky. 8-11
- 8-5 Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6412b, base infiltration map. 8-14
- 8-6 Cumulative residence time distributions for transport of radionuclides in the two-dimensional

List of Figures

- al site scale model. Property set 6412b, base infiltration map. 8-14
- 8-7 Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6531b, base infiltration map. 8-15
- 8-8 Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6531b, base infiltration map. 8-15
- 8-9 Difference in predicted fluid saturations from an increase in the base-case percolation flux map by a factor of three at all locations. Top figure: matrix saturation difference; bottom figure: fracture saturation difference. 8-17
- 8-10 Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, 3 x I infiltration map. 8-18
- 8-11 Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, 3 x I infiltration map. 8-19
- 8-12 Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, I/3 infiltration map. 8-19
- 8-13 Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, I/3 infiltration map. 8-20
- 8-14 Green's function (left column of figures) and cumulative residence time distributions for the I/3 infiltration map for various parameter sets. a, b) 6412b; c, d) 6412c. 8-22
- 8-15 Green's function (left column of figures) and cumulative residence time distributions for the I/3 infiltration map for various parameter sets. a, b) 6522; c, d) 6531b. 8-23
- 8-16 Comparison of matrix saturations for cases 6412b and 6412c. I/3 infiltration map. Top figure: 6412b; middle figure: 6412c; bottom figure: difference in saturations (6412c - 6412b). 8-24
- 8-17 Comparison of Green's functions (base-case grid and refined grid) for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map. 8-25
- 8-18 Comparison of cumulative residence time distributions (base-case grid and refined grid) for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map. 8-26
- 8-19 Green's function for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), I/3 infiltration map. 8-28
- 8-20 Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), I/3 infiltration map. 8-28
- 8-21 Green's function for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), base infiltration map. 8-29

List of Figures

- 8-22 Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), base infiltration map. 8-29
- 8-23 Schematic of the fracture transport model (after Tang et al., 1981) 8-32
- 8-24 Influence of fluid residence time and dispersivity on breakthrough at the water table. No matrix diffusion or sorption. 8-37
- 8-25 Influence of fracture sorption on breakthrough at the water table. No matrix diffusion or sorption. Fluid travel time = 0.1 y. 8-38
- 8-26 Influence of matrix diffusion on breakthrough at the water table. Fluid travel time = 1 y. No matrix or fracture sorption. 8-39
- 8-27 Influence of matrix sorption on breakthrough at the water table. Fluid travel time = 1 y. = 10-12 m²/s. No fracture sorption. 8-40
- 8-28 Influence of colloid-facilitated transport on breakthrough at the water table. Fluid travel time = 1 y. = 10-12 m²/s. = 1000. No fracture sorption. 8-41
- 8-29 Influence of colloid-facilitated transport on breakthrough at the water table. Fluid travel time = 1 y. = 10-12 m²/s. = 1000. Varying fracture sorption, colloid attachment, colloid sorption. 8-42
- 8-30 Green's function for Plutonium transport through the unsaturated zone for different values of K_c. Case 6541, base infiltration map, aqueous plutonium K_d = 100 cc/g, all units. . . 8-45
- 8-31 Cumulative breakthrough curves for plutonium transport through the unsaturated zone for different values of K_c. Case 6541, base infiltration map, aqueous plutonium K_d = 100 cc/g, all units. Dotted curves are simulations with no matrix diffusion. 8-46
- 8-32 Water mass flow rate versus time exiting the two-dimensional model domain for the three transient cases considered. Changes in percolation rate occur at t = 0. 8-49
- 8-33 Matrix saturations in the ptn3 and ch4v units in response to the long-term transients in percolation flux. Black curves: ptn3; Blue curves: ch4v. 8-50
- 8-34 Matrix saturations in the tsw5 and bf2z units in response to the long-term transients in percolation flux. Black curves: bf2z; Red curves: tsw5. 8-51
- 8-35 Breakthrough curve of ²³⁷Np at the water table for the long-term transient scenario. Also shown are the steady state curves for the two infiltration rates, and the quasi-steady state method. 8-52
- 8-36 Breakthrough curve of ²³⁷Np at the water table for the long-term transient scenario, shown on an expanded scale near the time of the change in percolation flux. 8-53
- 8-37 Green's function for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map, elevated water table. 8-54
- 8-38 Cumulative residence time distributions for transport of radionuclides in the two-dimension-

List of Figures

	al site scale model. Property set 6541, base infiltration map, elevated water table. . .	8-55
9-1	Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map.	9-4
9-2	Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map.	9-5
9-3	Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.	9-6
9-4	Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.	9-7
9-5	Green's function for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, base infiltration map.	9-8
9-6	Cumulative breakthrough curves for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, base infiltration map.	9-9
9-7	Green's function for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.	9-10
9-8	Cumulative breakthrough curves for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.	9-11
9-9	Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, no sorption, different diffusion coefficients. . . .	9-12
9-10	Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, no sorption, different diffusion coefficients.	9-13
9-11	Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, zeolite $K_d = 2$ cc/g, different diffusion coefficients.	9-14
9-12	Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, zeolite $K_d = 2$ cc/g, different diffusion coefficients.	9-15
9-13	Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, no sorption, different dispersivity values.	9-16
10-1	Schematic of abstracted rate models for radionuclide release from the NFE. a) constant-release-rate model; b) exponential-release-rate model.	10-8
10-2	Breakthrough of ^{99}Tc at the water table for different hydrologic parameter sets. Base infil-	

List of Figures

tration map, tcan=1000 y, trel=3000 y.	10-10
10-3 Breakthrough of 99Tc at the water table for different hydrologic parameter sets. Base infiltration map, tcan=1000 y, trel=10000 y.	10-11
10-4 Breakthrough of 99Tc at the water table for different durations of release. Base infiltration map, tcan=1000 y, parameter set 6541.	10-12
10-5 Breakthrough of 99Tc at the water table as a function of infiltration rate. tcan=1000 y, trel = 3000 y, parameter set 6541.	10-13
10-6 Breakthrough of 99Tc at the water table as a function of infiltration rate. tcan=1000 y, trel = 3000 y, parameter set 6412d.	10-13
10-7 Breakthrough of 99Tc at the water table for different values of krel for the exponential release scenario. Base infiltration map, tcan=1000 y, parameter set 6541.	10-14
10-8 Breakthrough of 99Tc at the water table comparing the two-dimensional and three-dimensional models. Base infiltration map and I/3 map, tcan=1000 y, parameter set 6541	10-15
10-9 Breakthrough of 237Np at the water table for different hydrologic parameter sets. Base infiltration map, tcan=1000 y, trel = 10 ky, parameter set 6541.	10-16
10-10 Breakthrough of 237Np at the water table for different hydrologic parameter sets. Base infiltration map, tcan=1000 y, trel = 30 ky, parameter set 6541.	10-17
10-11 Breakthrough of 237Np at the water table for different hydrologic parameter sets. Base infiltration map, tcan=1000 y, trel = 100 ky, parameter set 6541.	10-17
10-12 Breakthrough of 237Np at the water table as a function of duration of release. tcan=1000 y, base infiltration map, parameter set 6541.	10-18
10-13 Breakthrough of 237Np at the water table as a function of infiltration rate. tcan=1000 y, trel = 30 ky, parameter set 6541.	10-19
10-14 Breakthrough of 237Np at the water table as a function of infiltration rate. tcan=1000 y, trel = 30 ky, parameter set 6412d.	10-19
10-15 Breakthrough of 237Np at the water table for different values of krel for the exponential release scenario. Base infiltration map, tcan=1000 y, parameter set 6541.	10-20
10-16 Breakthrough of 237Np at the water table comparing the two-dimensional and three-dimensional models. Base infiltration map and I/3 map, tcan=1000 y, parameter set 6541.	10-21
10-17 Uranium isotope inventory as a function of time for a 70,000 MTU repository and --- add description of inventory assumptions (initial inventory data from Wilson et al., 1994).	10-22
10-18 Water table breakthrough curves for Uranium isotopes. Property set 6541, base infiltration map, release rate similar the 237Np case with 30 ky release scenario (for Uranium only --% is released after one million years).	10-24

List of Figures

- 10-19 Plutonium isotope inventory as a function of time for a 70,000 MTU repository and --- add description of inventory assumptions (initial inventory data from Wilson et al., 1994). 10-25
- 10-20 Water table breakthrough curves for total plutonium. Property set 6541, base infiltration map, different durations of release. Duration of release is directly proportional to effective solubility, which in turn is proportional to K_c 10-27
- 10-21 Water table breakthrough curves for individual isotopes of Plutonium. Property set 6541, base infiltration map, durations of release corresponding to $K_c = 99$ 10-28
- 11-1 Geochemistry and stratigraphy of well UZ-14 (aqueous chemical data from Yang et al., 1996). (a) Chloride and bicarbonate concentration, (b) sodium, calcium, and magnesium concentration, (c) pH and (d) stratigraphy and zeolite abundance 11-17
- 11-2 Stratigraphy and finite element grid used for the two-dimensional model cross section. (a) stratigraphy, including locations of zeolitic rocks, (b) finite element grid, (c) close-up of a small portion of the grid near the repository. 11-18
- 11-3 ^{237}Np speciation in J-13 water at 25°C with a solubility of 5×10^{-5} mol/l. 11-26
- 11-4 Predicted fraction of ^{237}Np as cation as a function of temperature and fluid composition (J-13 fluid versus UE-25 p#1 fluid). 11-26
- 11-5 ^{237}Np sorption coefficient versus Na^+ concentration in pure sodium bicarbonate solutions (data from Tait et al., 1996). The X's with accompanying curve is the predicted ion exchange model result. 11-30
- 11-6 Tritium and ^{237}Np sorption column study using J-13 well water at a pH of 8.3. The curves represent fits to the data for equilibrium and kinetic sorption models 11-32
- 11-7 The fraction of flow residing in the matrix and fracture for the DKM model. 11-34
- 11-8 Breakthrough of ^{237}Np at the water table in the one-dimensional model. Comparison of the ECM and DKM models for different flow scenarios. 11-36
- 11-9 Influence of repository heat on thermohydrologic and chemical conditions in the unsaturated zone. (a) Fluid saturation profile (500 years), (b) temperature profile (500 years), (c) chloride concentration (2000 years). 11-38
- 11-10 Temperature versus time in the rock near an emplacement drift in the repository. "Center"-drift near the center of the repository, "Edge"-drift near the edge of the repository. The family of curves for the "Center" drift show the influence of infiltration rate on the results. 11-39
- 11-11 Temperature versus time at the top of the Calico Hills zeolitic horizon. "Center"-position located beneath the center of the repository; "Edge"-drift near the edge of the repository. The influence of the assumptions regarding the heat transfer at the water table boundary are also shown. 11-40

List of Figures

11-12	²³⁷ Np transport through the unsaturated zone. (a) ²³⁷ Np total aqueous concentration profile after rewetting of the repository region and mobilization of ²³⁷ Np at all locations (10,000 years), (c) ²³⁷ Np total aqueous concentration profile as ²³⁷ Np reaches the water table (50,000 years), and (d) ²³⁷ Np immobile site concentration profile (50,000 years).	11-42
11-13	Mobilization of ²³⁷ Np from the near field, and breakthrough curves at the water table for different infiltration scenarios. The declining curves starting at early times represent the ²³⁷ Np mass remaining as a solid in the repository region (the left y-axis). The breakthrough curves exhibiting peaks are ²³⁷ Np flux at the water table (the right y-axis).	11-43
11-14	Fracture bypass of the zeolitic Calico Hills units.	11-45
11-15	The effect of CO ₂ degassing on pH at the center of the repository (heat-pipe front) and at the top of the zeolitic layers.	11-46
11-16	Influence of pH on the ²³⁷ Np breakthrough curves at the water table. The “carrier plume” result assumes an elevated pH of 10 at the repository for a period of either 500 or 5,000 years due to cement-water reactions.	11-47
11-17	Influence of transient infiltration and chemical conditions on the ²³⁷ Np breakthrough curves at the water table. The “changing climate” result assumes an increase from 1 mm/y to 4 mm/y, accompanied by a four-fold decrease in cation concentrations. The 1 mm/y and 4 mm/y steady state results without geochemical transients are shown for comparison.	11-49
11-18	Influence of thermal boundary condition at the water table on the ²³⁷ Np breakthrough curves at the water table.	11-50
12-1	Antler Ridge cross section (a) hydrologic zones including rock with greater than 10% zeolitic abundance, (b) hydrologic zones including rock with greater than 20% zeolitic abundance.	12-5
12-2	Two possible zeolitic geometries at the subgrid scale (10 m by 10 m).	12-8
12-3	Comparison of the characteristic curves for zeolitic and vitric tuff.	12-10
12-4	Velocity profile for the zeolitic lens case at three different infiltration rates.	12-11
12-5	Comparison of ²³⁷ Np breakthrough curves for the layer and lens cases.	12-13
12-6	Data relating % clinoptilolite to K _{sat} , (from Flint, 1996).	12-15
12-7	Data from Altman et al. (1996) relating porosity to K _{sat} .	12-16
12-8	Data from Altman et al. (1996) relating α to K _{sat} .	12-18
12-9	Variograms depicting the spatial structure of the zeolites generated from RC2 using data from the Calico Hills and Prow Pass units.	12-20
12-10	Kriged zeolite abundance of full mineralogical model.	12-21

List of Figures

12-11 Variance of Kriged zeolite abundance in full mineralogical model.	12-22
12-12 Three-dimensional block for high resolution study (4000mx2000mx300m).	12-23
12-13 Comparison of the kriged map and three conditional simulations.	12-24
12-14 Conditional simulation of percent zeolitic abundance mapped onto the refined computational mesh between the Solitario Canyon and Ghost Dance faults.	12-25
12-15 Permeability distribution for a conditional simulation. Percent zeolitic abundance-permeability correlation is used between the Solitario Canyon and Ghost Dance faults for all layers below the repository. The rest of the computational domain is assigned default permeability values.	12-27
12-16 Saturation profile of the conditional simulation. (a) matrix saturation, (b) fracture saturation.	12-28
12-17 Comparison of the 10% zeolite cutoff runs of Chapter 8 and a conditional simulation for a conservative tracer.	12-29
12-18 Conditional simulation with different Kds compared to the 10% cutoff case with a Kd of 20.	12-30
12-19 Conditional simulation using different Kds with 1/3 the infiltration rate in Figure 12-18.	12-30
12-20 Modification of the 10% zeolite case to better match the conditional simulation. The dotted lines show the results when all rocks with zeolitic abundance less than 10% are assigned a Kd of $0.1 \cdot K_d$	12-31

List of Tables

2-1	QA Status of Data used in this Chapter	2-3
3-1	QA Status of Data used in this Chapter	3-2
4-1	QA Status of Data used in this Chapter	4-2
5-1	QA Status of Data used in this Chapter	5-2
5-2	Summary of hydrologic parameter sets from Bodvarsson et al. (1997) (shaded rows indicate parameter set not simulated in the present study)	5-16
5-3	Parameter sets to include in multi-dimensional transport simulations. Parameter sets carried forward are unshaded in the table.	5-40
6-1	QA Status of Data used in this Chapter	6-2
6-2	Apparent ¹⁴ C Ages from Yang et al. (1997).	6-24
6-3	Parameter Sets for Environmental Isotope Simulations	6-26
6-4	Infiltration Maps Used in Environmental Isotope Simulations	6-27
6-5	Transient Model Description	6-58
7-1	QA Status of Data used in this Chapter	7-3
8-1	QA Status of Data used in this Chapter	8-2
8-2	Potential ranges of hydrologic and transport parameter values	8-36
9-1	QA Status of Data used in this Chapter	9-2
10-1	QA Status of Data used in this Chapter	10-2
11-1	QA Status of Data used in this Chapter	11-2
11-2	Groundwater Compositions (ppm) of Unsaturated Zone Fluids and Fluids Used in Sorption Studies	11-22
11-3	Solubility of ²³⁷ Np (Efurd et al. (1996))	11-23
11-4	Reactions and Log Equilibrium Coefficients for Np Speciation	11-24
12-1	QA Status of Data used in this Chapter	12-2
12-2	: Hydrologic and chemical properties of vitric and zeolitic tuff	12-9
12-3	: Parameters used in the simulations	12-12
15-1	Correlation of Geologic Formations, Hydrological Units and the Model Layers of Both	

List of Tables

LANL and LBL.	15-1
15-2 Property Set 6412a.	15-4
15-3 Property Set 6412b.	15-5
15-4 Property Set 6412c.	15-6
15-5 Property Set 6522.	15-7
15-6 Property Set 6531b.	15-8
15-7 Property Set 6541.	15-9

Chapter 1 - Executive Summary

1.1 Overview

The unsaturated zone at Yucca Mountain is one of the primary barriers to the migration of radionuclides from the potential repository to the accessible environment, and, as such, has received great attention in site-characterization activities. This milestone report describes the revised transport model for the unsaturated zone at Yucca Mountain. The purpose of the modeling studies is to incorporate the latest hydrologic, mineralogic, and geochemical information into a set of numerical models to predict transport of radionuclides from the potential repository to the water table. Since these are predictions of repository performance after waste emplacement, the influence of repository waste heat and future climate changes must be accounted for.

Integrated transport predictions such as these require input from numerous other Project participants. The Project's databases of geology and mineral distributions are used as the structural and geochemical context in which the transport system operates. Numerical grids are built that honor these databases so that they can be populated with flow and transport parameters. The hydrologic parameters for the model were obtained from the recent report on the unsaturated zone flow system by Bodvarsson et al. (1997). These parameters include unsaturated hydrologic property values for all units defined in the geologic framework model, as well as properties of zeolitic tuffs. Two- and three-dimensional flow simulations, using the spatially varying infiltration map of Flint et al. (1996) and variations of this map, are performed to provide either steady state or transient flow fields that are used to simulate the transport of major ions or environmental isotopes, and to predict radionuclide migration. The major-ion chemistry and environmental isotope modeling studies provide important constraints on the hydrologic and transport system; therefore, we devote considerable effort toward developing models that are consistent with these data sets.

Executive Summary

The dual permeability conceptual model is used extensively in this work, as it has been shown previously to provide a more robust hydrologic setting on which to predict transport of solutes through the unsaturated zone (Robinson et al., 1996). Particle tracking is used as the primary model for the transport predictions because of its ability to capture efficiently the vast difference in travel times depending on whether the radionuclide is in the fracture or matrix continuum. Although these models figure prominently in the simulations in the present study, equivalent continuum flow and reactive transport models are employed in some simulations as well. The choice depends on the application in question and the issues being addressed.

This report is divided into chapters that comprise either preliminary foundational discussions on which the numerical simulations are based, such as grid generation, or distinct topics of investigation using the models developed. The following subsections report the main conclusions of each chapter of the milestone. The final section (Section 1.13) provides a road map between the requirements listed in the Criteria Statement for this milestone and the specific locations in the document where these requirements are met.

1.2 Incorporation of Mineralogic Model into Flow and Transport Simulations

The mineralogy of the unsaturated zone has an important bearing on the prediction of hydrologic and transport processes. The distribution of secondary minerals such as zeolites can strongly influence the flow patterns of percolating fluids through the unsaturated zone. The presence of other minerals such as smectite clays, may also be linked to, or evidence for, flow processes occurring in the unsaturated zone. The sorption of many radionuclides are also strongly controlled by the mineral distribution in the flow pathways between the repository and the water table. For these reasons, we have developed a streamlined method for incorporating mineralogic data directly into our site scale model.

The distribution of major and minor minerals has been organized into a numerical model using the STRATAMODEL software. An interface between this model (reported by Chipera et al., 1997a, b) and the flow and transport simulation models developed in the present study has been developed to make the investigation of mineralogic controls on flow and transport as seamless as possible. In this milestone, the distribution of zeolites in the unsaturated zone is modeled using this information either through the assignment of a cut-off percentage above which the rock is given the properties of zeolitic tuff, or by populating the model with a zeolitic abundance that then is used in assigning flow and transport properties (Chapter 12). An updated version of the mineralogic model is incorporated into the simulations of the present study.

Finally, we compare the mineral distributions measured recently with predicted values using the version of the model before these data were available. For zeolites, we show that the new data do not significantly change the mineral distribution reflected in earlier versions of the model, whereas the distribution of smectite, a less abundant mineral in the unsaturated zone, is more refined after incorporating the new data.

1.3 Mesh Generation

Computational mesh generation for this report involves translating the Integrated Site Model, ISM (Zelinski and Clayton, 1996, Clayton et al., 1997), of geologic and hydrologic structure and stratigraphy to a finite element mesh. This process maintains the geometry defined by the ISM model while meeting additional geometric constraints on element size and geometry for stable and accurate flow and transport solutions.

The GEOMESH/X3D grid generation toolkit, (Gable et al., 1995, Gable et al., 1996a and b, Trease et al., 1996), has been developed to provide an integrated software package for all grid generation steps, from initial ISM model import to quality checking of input data, mesh optimization, mesh post processing and quality checking, mesh interfacing with the FEHM flow

and transport code and property interpolation from the mineralogy and petrology model (Chipera et al., 1997a and b).

Chapter 3 describes the algorithms used for mesh generation and how the algorithms have been applied to the two- and three-dimensional meshes used for flow and transport computations.

1.4 Particle Tracking

A new particle tracking technique is presented for simulating liquid or gas phase solute transport in porous media. The method, called the Residence Time Transfer Function (RTTF) particle tracking technique, uses 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. Transient flow fields are approximated by assuming a non-varying flow field during a time interval of a fluid flow solution. The method is a significant departure from the traditional procedure of mapping a particle trajectory by resolving the velocity vector at a given position by 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. 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. For the present study, the technique is used for unsaturated 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.

1.5 Hydrologic Property Sets

In this section we present a summary of the hydrologic property sets provided by LBNL in the site unsaturated zone flow model (Bodvarsson et al., 1997), along with modifications tested in the present study for their impact on predicted performance. Bodvarsson et al. (1997) provide a logical starting point for numerical simulations of flow and transport in the present study, since models using their parameter sets are based on calibrations to fluid matrix saturation and capillary pressure measured in cores extracted from several wells at the site. Many parameter sets are presented in Bodvarsson et al. (1997), each of which employed a different approach in either the conceptual model (e.g. different fracture/matrix interaction models) or parameter estimation methodology (e.g. different initial guesses in the inversion, different decisions on which parameters to allow to vary to obtain the fit). Since it is not feasible to carry forth with complete transport sensitivity analyses for every parameter set in the LBNL milestone, as well as test variations of these as sensitivity studies, an initial screening was performed to determine which parameter sets are apt to yield significantly different performance predictions.

The “model platform” used to test the hydrologic property sets is a one-dimensional flow and transport simulation with stratigraphy and estimated infiltration rate at SD-9. Steady state, dual permeability fluid flow fields were generated for each property set, after which particle tracking transport simulations of ^{99}Tc and ^{237}Np from the repository to the water table were computed. The transport metrics used to identify which property sets yield unique or distinctively different results are the first arrival time of radionuclide at the water table, the relative amount of radionuclide arriving early, and the degree of sorptive delay of ^{237}Np to zeolitic tuffs. In addition, the relative flow split between the fractures and matrix in the zeolitic tuffs was used as a hydrologic metric for judging the property sets. Because transport of radionuclides escaping the repository is the subject

Executive Summary

of the study, the emphasis is on predicted flow and transport below the repository horizon. Parameter sets may differ in their predictions of hydrologic conditions above the repository; these possible differences are not addressed.

Despite the potentially large spectrum of possible behaviors for different hydrologic property sets, the most striking result of the screening study is the similarity of the flow and transport results. In all cases, fracture-dominated flow in the TSw transitions to almost completely matrix flow in the vitric Calico Hills unit at SD-9. Then, at the transition from the vitric to zeolitic tuff, significant fracture flow through the zeolitic tuffs is predicted for every parameter set, even at the 3.6 mm/y infiltration rate assigned at SD-9. The permeability of the zeolitic tuffs is too low to support the 3.6 mm/y infiltration in the matrix, thus resulting in the fracture flow, which will be even more pronounced in high flux areas within the potential repository area. There are subtle differences from one parameter set to the next which we identify and use as the criteria for selecting sets for the multi-dimensional simulations.

Because of the essential similarities among the various property sets and the possibility that the parameter estimation yielded results that only sample a portion of the range of uncertainty of the actual system, we also propose variations to the calibrated property sets. These variations include higher permeability for the zeolitic rocks, full connection-area weighting for the nonwelded units, and an adjusted property set of Bodvarsson et al. (1997) that they derive as an improved match to Calico Hills fluid saturation data using the three-dimensional model. In all, ten property sets are tested in the screening process, and five are used in subsequent two- and three-dimensional flow and transport simulations.

In this section we also perform a code validation check by comparing the results of FEHM and TOUGH2 for the one-dimensional column. Steady state flow fields were generated with each code, and the flow fields were used by FEHM to compute particle tracking breakthrough curves. The saturation profiles agreed closely. Transport of ^{99}Tc differed somewhat due to a slight, predicted difference in the flow split between fractures and matrix in the two cases. More

importantly, the transitions from matrix flow to partial fracture flow at the vitric/zeolitic interface is predicted to occur abruptly above the interface in FEHM and more gradually within the first zeolitic grid block in TOUGH2. Matrix flow within the zeolite in TOUGH2 gave rise to significantly delayed first arrival of ^{237}Np at the water table. We attribute the TOUGH2 behavior to the combination of upstream weighting of the absolute permeability and the large size of the first zeolitic grid block; FEHM calculations using this same weighting scheme yielded results that approached those of the TOUGH2 run. We therefore recommend that in mountain-scale flow and transport simulations, the absolute permeability be harmonically weighted, and/or the grid be more finely discretized near interfaces of contrasting flow and transport properties.

1.6 Environmental Isotopes and the Hydrologic System

In this chapter we examine the role of studies coupling environmental isotope data and numerical modeling in identifying flow and transport processes as well as constraining the property sets used to model them. Starting with multiple, different hydrologic property sets from LBNL which are all based on fits matching matrix fluid saturation and capillary pressure (Bodvarsson et al., 1997), we perform a series of studies with one-, two-, and three-dimensional transport models and examine how well the additional isotopic data are matched. The most significant finding in this chapter is that minor changes in hydrologic parameters lead to very different flow distributions in the PTn matrix and fractures. Namely, one property set leads to matrix dominated flow in the PTn while another yields significant fracture flow. Reducing the uncertainty in property set choice may have significant implications on repository design and performance modeling. For example, if fracture flow in the PTn is ubiquitous, then repository design may need to account for periodic high fluxes resulting from transient infiltration events that are not damped in the PTn matrix.

We find that property sets that do not lead to significant fracture flow in the PTn, except in fault zones, provide a better match with the substantial set of analyses of $^{36}\text{Cl}/\text{Cl}$ ratios measured for ESF samples (Sweetkind et al., 1997, Fabryka-Martin et al., 1996a, Levy et al., 1997). This

finding supports the assumption that episodic infiltration events in the PTn are damped except in fault zones.

Using a multiple model approach in this study, two- and three-dimensional models are used to assess the effects of spatially distributed infiltration and dipping strata of varying thickness and extent in a faulted system. One-dimensional models are then 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. This modeling approach, coupled with the data analysis, is used to identify significant processes and verify conceptual models.

1.7 Water Chemistry

This chapter explores alternative explanations for the water chemistry variations in perched and pore waters at Yucca Mountain. The chemical databases are integrated with hydrologic databases to evaluate different model conceptualizations of the hydrologic behavior of the mountain at low and high infiltration rates. The premise for this exercise is that chemical data, when combined with hydrologic data, provide the most relevant information for “validating” radionuclide transport models, as the movement of naturally occurring solutes is closely related to the potential migration of radionuclides. Our philosophy for model development and testing is one in which these data are used in the building of conceptual models, rather than as data to “calibrate” these models.

Two end-member hypotheses are evaluated for the origin of perched water at Yucca Mountain: a “fast pathway” fracture flow hypothesis, and a transient-mixing hypothesis. The overall consequence of the “fast pathway” hypothesis is that it predicts poor performance for the unsaturated zone, as the Calico Hills transport barrier is essentially bypassed by fracture flow. Since this model is based on non-calibrated fracture properties, we propose an alternative to this model in the transient-mixing hypothesis, based on the integration of all elements of the hydrologic

Executive Summary

and chemical databases at Yucca Mountain. Its main strength is that it explains simultaneously the inversion in apparent ages between pore waters and perched water in the unsaturated zone, and the low chloride content of perched waters relative to the overlying TSw and underlying CHn pore waters. The apparent disequilibrium in pore water and perched water contents is consistent with a model of transient flow, and we demonstrate that the stable isotope data and apparent ^{14}C and $^{36}\text{Cl}/\text{Cl}$ ages can be interpreted as resulting from a mixture of late Pleistocene/early Holocene water with modern waters. The existence of these mixed ancient/modern perched water bodies therefore implies that an additional retardation mechanism exists above the Calico Hills formation. The ubiquitous young ^{14}C ages in the Calico Hills formation pore waters beneath the perched water bodies and in other areas also imply that a lateral component of flow occurs in the vitric Calico Hills and that this formation is not bypassed. Finally, this model is consistent with the three-dimensional stochastic mineral distribution model in which zeolitic and vitric tuffs are intricately interbedded beneath the repository horizon (see Chapter 12 for a description of the stochastic model).

The implication of this model for Performance Assessment is an improved performance of the Calico Hills Transport barrier beneath the repository by 1) the additional barrier component which regulates the downward flow of water to the saturated zone, and 2) increased water residence times in the unsaturated zone due to the presence of long term perched water bodies at depth (i.e., no flow), and lateral diversion above the basal vitrophyre and/or the underlying Calico Hills vitric units. This should result in increased sorption of radionuclides at those boundaries, as actinides sorb in clays and vitric tuffs as well as in zeolites. Finally, we expect that the transient mixing model may give rise to different performance predictions than are achieved with the current hydrologic properties.

1.8 Two-Dimensional Radionuclide Transport Simulations

In most of the two- and three-dimensional simulations presented in this study, steady state flow fields are generated using FEHM, and then radionuclide transport is computed for various transport parameters. The steady state assumption allows us to compute generic breakthrough curves in response to a pulse of radionuclide injected at time 0. This simulation provides the basis for computing, external to the FEHM simulation, the actual response to a realistic near field release scenario. Numerical convolution is employed for this purpose. In this section we present the results of the generic response curves for a variety of flow and transport parameter sets and scenarios.

Multiple parameter sets from the LBNL site-scale flow model and variations on the USGS infiltration map are used in the simulation of radionuclides from the potential repository to the water table. For most of these parameter sets, radionuclides tend to bypass the zeolites of the Calico Hills via fracture transport when base-case infiltration conditions are simulated. When the infiltration rate is scaled by 1/3, significant fracture flow in the zeolites is still simulated with most parameter sets. A modification increasing CHn zeolite matrix permeabilities by a factor of 5 leads to substantially greater matrix flow and, hence, retardation of sorbing radionuclides. This process leads to dramatic improvement in the performance of the unsaturated zone. These studies demonstrate the overriding importance of proper characterization of the zeolitic units for performance assessment. Presently, there is significant uncertainty in the characteristics of these units due to data limitations. Enhanced characterization efforts, including the planned field test at Busted Butte, would reduce the uncertainty in these calculations and lead to a better understanding of the true nature of the hydrologic and transport behavior of the CHn.

Colloid-facilitated transport of otherwise immobile radionuclides like Plutonium is also addressed in this chapter. As very little data are currently available for Plutonium sorption onto colloids, we present a sensitivity analysis of the significant processes to mobility. In the absence of sorption onto the mobile colloids, Plutonium migration is limited due to large sorption coefficients. Assuming reversible sorption, when partitioning onto colloids approaches 99% or

more, Plutonium mobility becomes potentially important to performance. If sorption of Plutonium onto colloids is irreversible, any colloid transport would directly affect performance assessment.

Long-time transients capturing climate change related phenomena are also simulated in this chapter. The results show that the flux of radionuclide at the water table under transient conditions is closely approximated by a quasi-steady state model. Thus multiple steady-state simulations can be used to predict the effects of wetter or drier future climates on radionuclide flux at the water table. The effect of potential water table rise due to wetter, future climates is also considered. The combined effect of a shorter travel distance to the water table and high fluxes under a wetter climate scenario yield prediction of poorer unsaturated zone performance than for the present climate conditions.

1.9 Three-Dimensional Radionuclide Transport

In this chapter, the two-dimensional approach presented in the last chapter is extended to the three-dimensional site-scale model of radionuclide transport. Variations in additional transport properties such as the diffusion coefficient and the dispersion coefficient are also examined in this chapter. With the full three-dimensional transport model, implications of lateral flow, spatial variability of flux, and underlying strata thickness are examined. Compared to the two-dimensional model, greater spreading in the travel time distribution curve is simulated due to the additional flow paths sampling a greater range of property variability that occur in three dimensions. This variability leads to cumulative breakthrough curves at the water table spanning the range from 10 years to 10,000 years. The three-dimensional model predicts some sorption on to the zeolites, similar to the two-dimensional model. However, pathways bypassing the zeolites still exist and may lead to peaks in the mass flux at the water table which are not affected by retardation in the zeolites.

The three-dimensional transport model was used to examine the difference in travel time distribution as a function of the release position in the potential repository. Radionuclides released

from the northern end of the repository are more likely to experience rapid transport to the water table than radionuclides released in the southern portion of the potential repository. The behavior is attributed to less uniform distribution of zeolites in the southern portion of the domain. This results in slower matrix flow in the nonzeolitic units in the south compared to fracture dominated flow in the zeolitic units in the north. As with the two-dimensional simulations, this finding highlights the critical need to develop greater certainty on the material properties assigned to the zeolitic units. A slightly higher matrix permeability in the zeolites leads to substantially greater retardation of radionuclides.

The diffusion coefficient influences the breakthrough curves simulated with the three-dimensional model, especially for the portion of the inventory with the shortest travel times. Diffusion into the rock matrix and sorption onto zeolites retard the transport of ^{237}Np traveling in fractures. The dispersion coefficient is shown to have very little impact on the unsaturated zone transport.

1.10 Unsaturated Zone Performance Predictions

Predicting the response at the water table to the large variety of near field environment (NFE) release scenarios is done most efficiently by decoupling the far-field and near-field models. This chapter describes the process of convolving NFE models with far-field transport simulations. With this approach, the differences in response due to varying hydrologic conditions near waste emplacement drifts, different engineered barrier designs, time varying thermodynamic effects on canister corrosion, different models of release due to dissolution of waste, and uncertainty in transport processes in backfill and invert material can all be evaluated. The present study focuses on the unsaturated zone transport barrier; simplified NFE source term functions are used.

For nonsorbing radionuclides like ^{99}Tc , there is no retardation in any material between the potential repository and the water table. For all of the different property sets developed by LBNL, fracture flow through the zeolitic Calico Hills and underlying units results in bypassing of most of

the unsaturated zone matrix material for the USGS infiltration estimates. NFE release rates control the maximum concentration flux at the water table. Releases that occur over longer periods of time lead to lower peak mass fluxes at the water table. Reducing the infiltration rate in these predictions also leads to lower peak mass flux rates at the water table, and they occur significantly later in time.

As with nonsorbing radionuclides, those that do sorb to zeolites like ^{237}Np still show early arrivals at the water table after release begins for these parameter sets due to the simulated fracture bypass of the zeolites. This early arrival appears to control the peak mass flux as well. Under these conditions, two factors control the magnitude of the peak mass flux at the water table. First, the NFE release scenario directly influences the actual peak mass flux. For example, rapid releases over short time periods yield higher peaks. Second, any parameter that influences the relative flow fraction through fractures has a direct impact on peak mass flux.

Relatively small changes from the LBNL parameters lead to vastly improved performance with respect to sorbing radionuclides. However, without modification, those parameter sets lead to predictions of very little mitigation of releases from the near field by the unsaturated zone. Reduction in uncertainty of the actual hydrologic properties of the Calico Hills (both fracture and matrix) would clarify whether these predictions of poor far-field barrier performance is indeed warranted or whether they are a by-product of a calibration procedure that was not well constrained by data indicating the degree of fracture flow in the units below the basal vitrophyre.

1.11 Reactive Transport of ^{237}Np with Repository Heat

Characterization and performance assessment studies for the potential high-level nuclear waste repository at Yucca Mountain have identified ^{237}Np as a radionuclide of concern for the proposed repository. To predict the migration of neptunium after a repository breach, an understanding of the relevant hydrologic and geochemical processes is required. The hydrologic flow in the unsaturated zone at Yucca Mountain is dependent on the infiltration rate, stratigraphy of the vadose zone, and the heat generated by the decaying radioactive waste. The geochemical

processes that strongly affect ^{237}Np migration include: solubility-limited release of ^{237}Np from the near field environment, aqueous speciation of neptunium into non-sorbing carbonate/hydroxy complexes and the sorbing NpO_2^+ cation, sorption of neptunium onto the zeolitic tuffs via an ion exchange mechanism, and radioactive decay. We investigate the coupled effects of chemical interactions and heat on neptunium transport from the potential repository to the water table. The selective coupling method is introduced to solve these reactive transport problems. The simulations indicate that in the absence of irreversible changes in the hydrologic and transport properties, the heat pulse does not significantly affect the migration of neptunium, as the time scale of heat pulse propagation is shorter than the time scales associated with neptunium release and migration. Water chemistry, particularly pH, calcium, and sodium concentration significantly affect the retardation of neptunium by the zeolitic rocks between the repository and the water table.

1.12 Incorporation of Geostatistics into Flow and Transport Simulations

Small scale variability of chemical and hydrologic properties can play an important role on the prediction of flow and transport in the unsaturated zone. The variation in chemical and hydrologic properties is strongly dependent on the mineral distribution. Due to the correlation between mineral distribution and hydrologic and chemical properties, the distribution of minerals such as zeolites can strongly influence the flow patterns of percolating fluids and the sorption of many radionuclides. In this chapter, the zeolitic threshold method used in previous chapters is replaced with more rigorous geostatistical modeling. With this more sophisticated modeling approach, we examine whether using a threshold cutoff for zeolitic abundance is an appropriate upscaling method for characterizing small scale heterogeneity.

The methodology for integrating geostatistical techniques into the mineralogical model is first developed. These geostatistical techniques are used to develop variograms, kriging maps, and conditional simulations of zeolitic abundance. The kriging maps are used to estimate the

uncertainty in the mineralogical model. The conditional simulations result in equally probable maps of zeolitic abundance. Eventually, simulating flow and transport for many conditional simulations, we will be able to estimate the uncertainty in ^{237}Np travel times using Monte Carlo techniques.

Using the algorithms discussed in Chapters 2 and 3, a conditional simulation of zeolitic abundance was mapped onto the geologic model. We investigate the relationship between percent alteration, permeability changes due to alteration, sorption due to alteration, and their overall effect on radionuclide transport.

A key conclusion to the study is that the retardation due to sorption predicted by a conditional simulation is much larger than the retardation predicted by the zeolite threshold method. The reason for larger retardation of ^{237}Np for the conditional simulation is a small but significant K_d at locations with zeolite abundance less than 10%. At these locations, the K_d is low (less than 1 cc/g) but permeability is large enough for the flow to be matrix dominated. By contrast, the increased retardation is not due to high K_d values which occur at high zeolite abundance because very little flow travels through these low permeability regions. Modifying the 10% threshold case to include some sorption at locations with zeolite abundances less than 10% closely matches the more rigorous conditional simulation. For the purposes of abstracting these results, setting nonzero K_d 's in the vitric tuffs may be an effective way to mimic the more rigorous conditional simulations.

We therefore 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 also in general have low permeability and contaminants can only 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 and other radionuclides through the unsaturated zone. The planned transport test at Busted Butte should provide results for testing this hypothesis.

In order to draw more substantial conclusions, many more conditional simulations will need to be performed. In addition we plan to refine the correlations and variograms and implement linear coregionalization models. In this study, the general framework has been developed to integrate geostatistical information into the site scale flow and transport calculations. These powerful tools provide a means for determining the uncertainties associated with radionuclide transport through the unsaturated zone at small scales not normally studied in site scale models. In addition, we can use these more rigorous simulations to check the validity of the simplifying assumptions incorporated into other flow and transport simulations.

1.13 Traceability to Criteria for Milestone SP25BM3

The table below provides a road map between the requirements listed in the Criteria Statement for this milestone and the specific locations in the document where these requirements are met.

TECHNICAL REQUIREMENTS

CRITERIA	TEXT LOCATION
This report will include a conceptual model of the transport of radionuclides in the unsaturated zone at Yucca Mountain, including 2- and 3-D integrated transport calculations and sensitivity analyses.	Conceptual model: 4.3, 6.5.2 2D integrated transport calculation: Chapter 8. 2D sensitivity analyses: Sections 8.5, 8.6, 8.7, 8.8, 8.9 3D integrated transport calculations: Chapter 9 3D Sensitivity analyses: Sections 9.4-9.7

Executive Summary

Both equivalent continuum and dual permeability calculations will be made, incorporating discrete structural features as available and appropriate.	<p>Equivalent Continuum calculations: Sections 4.6, 11.4, 11.8</p> <p>Dual Permeability Calculations: Sections 4.4, 4.7, 5.4, 5.5, 6.7, 6.9-6.12, 8.5-8.11, 9.4-9.7, 10.6-10.9, 12.9</p> <p>Incorporation of discrete structural features: Sections 3.3, 3.4, 6.5, Included in all 2D and 3D dual permeability calculations.</p>
The model will do the following:	
a) Incorporate and consider isotopic data which are indicative of processes and rates of fluid flow or residence times in the unsaturated zone.	<p>Chapter 6.</p> <p>Processes: 6.5, 6.7-6.12</p> <p>Rates and Residence Times: 6.10-6.12</p>
b) Draw upon the Report on Summary of Geochemistry/Transport Laboratory Tests to provide defensible values for important transport processes and parameters including:	
1) Sorption and Diffusion Coefficients	8.6, 8.7, 8.10, 9.4, 9.6, 9.7, 11.7
2) Radionuclide solubility and speciation	10.5, 11.4, 11.7
3) Colloids as controlled by relevant thermal effects and water chemistry.	8.10
c) Rely heavily on input from the 3-D mineralogical model of Yucca Mountain.	2.2-2.5, 3.4, 6.5, 8.6, 12.5-12.9
Update 3-D transport simulations for radionuclides including, but not limited to:	
Np	5.5, 8.5-8.7, 8.9, 9.4, 9.5, 10.7, 11.8, 12.9
Pu	8.10, 10.9
Tc	5.5, 8.5-8.7, 8.9, 9.4, 9.5, 10.6
under ambient	5.5, 8.5-8.7, 8.9, 9.4, 9.5, 10.6-10.9
and thermally perturbed	11.8

Executive Summary

for two future climates and accompanying infiltration fluxes and effects on recharge	8.7, 8.11, 9.4, 9.5, 11.8
The report will be coupled to appropriate	
near-field source term	10.4, 10.5
and thermal models	11.7, 11.8
The deliverable will be prepared in accordance with OCRWM approved quality assurance procedures implementing requirements of the Quality Assurance Requirements Description.	Done
The product shall be developed on the basis of the best technical data, including both Q and non-q data. The Q status of data used and cited in the report shall be appropriately noted.	QA description of software and data in acknowledgments and in each chapter.
Stratigraphic nomenclature used shall be consistent with the Reference Information Base section 1.12(a): Stratigraphy-Geologic Lithologic Stratigraphy.	Throughout milestone, Table 15.1
References to data used in the report shall include record Accession Numbers or Data Tracking Numbers when available.	QA description of data in acknowledgments and in each chapter.
Technical data contained within the deliverable and not already incorporated in the Geographic Nodal Information Study and Evaluation System (GENISES) shall be submitted for incorporation into the GENISES in accordance with YQP-SIII.3Q.	N/A

Executive Summary

<p>Verification of technical data submittal compliance shall be demonstrated by including as part of the deliverable:</p> <p>a) A copy of the Technical Data Information Form generated identifying the data in the Automated Technical Data Tracking system, and</p> <p>b) a copy of the transmittal letter attached to the technical data transmittal to the GENIS-ES Administrator.</p>	<p>a) N/A</p> <p>b) done</p>
--	------------------------------

Chapter 2 - Incorporation of the Mineralogic Model into the Site-Scale Flow and Transport Model

2.1 Abstract

The mineralogy of the unsaturated zone has an important bearing on the prediction of hydrologic and transport processes. The distribution of secondary minerals such as zeolites can strongly influence the flow patterns of percolating fluids through the unsaturated zone. The presence of other minerals such as smectite clays, may also be linked to, or evidence for, flow processes occurring in the unsaturated zone. The sorption of many radionuclides are also strongly controlled by the mineral distribution in the flow pathways between the repository and the water table. For these reasons, we have developed a streamlined method for incorporating mineralogic data directly into our site-scale model.

The distribution of minerals has been organized into a numerical model using the STRATAMODEL software. An interface between the mineralogic model (reported by Chipera et al., 1997a, b) and the flow and transport simulation models developed in the present study has been developed to make the investigation of mineralogic controls on flow and transport as seamless as possible. In this milestone, the distribution of zeolites in the unsaturated zone is modeled using this information either through the assignment of a cut-off percentage above which the rock is given the properties of zeolitic tuff, or by populating the model with a zeolitic abundance that then is used in assigning flow and transport properties (Chapter 12). The mineralogic model incorporated into the simulations of the present study is an updated version of Chipera et al. (1997a).

Finally, we compare the mineral distributions measured recently with predicted values using the version of the model before these data were available. For zeolites, we show that the new data do not significantly change the mineral distribution reflected in earlier versions of the model, whereas the distribution of smectite, a less abundant mineral in the unsaturated zone, is more refined after incorporating the new data.

2.2 Mesh Generation

Computational mesh generation for this report involves translating the Integrated Site Model, ISM (Zelinski and Clayton, 1996, Clayton et al., 1997), of geologic and hydrologic structure and stratigraphy to a finite element mesh. The relationship between geologic and hydrologic units is presented in Table 15-1. This process maintains the geometry defined by the ISM model while meeting additional geometric constraints on element size and geometry for stable and accurate flow and transport solutions.

The GEOMESH/X3D grid generation toolkit, (Gable et al., 1995a and b, Gable et al., 1996, Trease et al., 1996), has been developed to provide an integrated software package for all grid generation steps, from initial ISM model import to quality checking of input data, mesh optimization, mesh post processing and quality checking, mesh interfacing with the FEHM flow and transport code and property interpolation from the mineralogy and petrology model (Chipera et al., 1997a and b).

Chapter 3 describes the algorithms used for mesh generation and how the algorithms have been applied to the two- and three-dimensional meshes used for flow and transport computations.

2.3 Software and Data QA Status

The petroleum industry standard software packages STRATAMODEL and RC² are used in the geostatistical analyses of heterogeneous materials. These are off-the-shelf software packages and are applied only to sensitivity studies in the present study. They are non-Q with respect to YMP and Los Alamos quality assurance requirements but they follow well documented methods for geostatistical representation of spatially varying attributes. The status of the data utilized in this Chapter are shown in Table 2-1.

Table 2-1. QA Status of Data used in this Chapter

REFERENCE	Q Status	DTN	COMMENT
Chipera et. al., 1997a, "Preliminary Three Dimensional Mineralogical Model of Yucca Mountain, Nevada, YMP Milestone SP321AM4"	non-Q	Not Assigned	No DTN assigned
Chipera et. al., 1997b "Mineralogic Variation in Drill Holes USW NRG-6, NRG-7-7a, SD-7, SD-9, SD-12, and UZ#14: New Data from 1996-1997 Analysis Los alamos National Laboratory YMP Deliverable SP321BM4"	Q	LADV831321AQ97.00 1	
Clayton, et. al., 1997, "ISM2.0: A 3-D geological framework and integrated site model of Yucca Mountain: Version ISM1.0, Doc ID B00000000-01717-5700-00004 Rev 0", MOL. 19970122.0053, CRWMS M&O	Q		By agreement with DOE and NRC accepted as Q even though it utilizes some non-Q data
Zelinski and Clayton, 1996, "A 3-D geological framework and integrated site model of Yucca Mountain: Version ISM1.0, Doc ID B00000000-01717-5700-00002 Rev 1", CRWMS M&O	non-Q		

2.4 Motivation for the Current Mineralogic and Hydrologic Model

Sorptive zeolitic minerals located between the potential repository and the water table strongly affect travel times of radionuclides such as ^{237}Np through the unsaturated zone. Prior to 1997, the site-scale geologic model for the potential repository at Yucca Mountain was comprised of layers of either zeolitic or non-zeolitic material in order to model the sorptive behavior of the zeolites. This division drew upon knowledge of whether a layer was sparsely or abundantly populated with zeolitic minerals. For purposes of transport calculations, those layers containing abundant zeolites were assigned zeolitic properties while the others were given vitric properties.

This assumption may not be valid; because zeolites are formed through secondary alteration, their distribution may not follow the primary geologic stratigraphy.

The greatest weakness of such a binary framework lies in its effect on species retardation: if a layer is not as completely zeolitic as portrayed, retardation of contaminant species may not be accurately captured. Specifically, zeolitic tuff is less permeable than vitric tuff, and a radionuclide such as ^{237}Np which sorbs to zeolites may therefore bypass portions of heterogeneous zeolitic layers leading to less retardation of ^{237}Np . Therefore, the homogenous zeolitic layers of the site-scale model used in previous studies (Robinson et al., 1995, 1996) may have overpredicted the sorption of the zeolites (Figure 2-1a).

In March, 1997, the stratigraphic framework was revised by integrating mineralogic abundances obtained from drill core data (Chipera et al., 1997a). Incorporation of these data has provided a more accurate representation of the 3-D mineralogic distribution, and therefore zeolite abundance distribution, at Yucca Mountain. Previously simplistic homogeneous zeolitic or vitric layers have now been replaced by spatially distributed mineralogic abundances. Specifically, zeolitic properties extracted from the mineralogical model can now be mapped onto the site-scale model providing a more accurate representation of the zeolites (Figure 2-1b) (for details see section 3). After the mapping process, the percent alteration (% zeolite) is prescribed at each node of the site-scale model and the appropriate flow and transport properties are assigned. In most simulations, we chose a simple threshold of 10% to flag nodes as either zeolitic or unaltered. This threshold was used because, at that time, there were no correlations between the degree of zeolitization and the hydrologic and chemical properties. Hydrologic characterizations for purely altered (zeolitic) and unaltered (vitric) matrix materials have been developed (Flint, 1996). Therefore, the threshold provides a method for assigning either vitric or zeolitic properties to each node (i.e., nodes having less than 10% zeolite are assigned vitric properties and nodes having greater than 10% zeolite are assigned zeolitic properties). In this chapter, we will briefly describe the mineralogical model that is used to construct a distribution of zeolitic minerals, which are used

a

b

Figure 2-1. Antler Ridge cross section (a) Continuous zeolitic layers (in red) of the Site-Scale Model used in Robinson et al. (1996), (b) Zeolitic distributions are mapped from the 3-D mineralogical model (Chipera et al, 1997a) to the FEHM finite element mesh. The profiles look different because (b) accounts for fault offsets.

in the flow and transport simulations. In Chapter 12, we replace the zeolitic threshold method with a more rigorous geostatistical model to determine the applicability of using a threshold cutoff for zeolitic abundance.

2.5 Mineralogic Model

Quantitative mineralogy, as obtained by XRD methods, was used as the mineralogic input into the 3-D mineralogic model. Data from a particular drill hole were included in the mineralogic model if a reasonable sample distribution existed for the drill hole or if the drill hole was located in an important location such as at the edge of the modeling area. The drill holes and the mineralogic data sources used for the 3-D model are listed in Table 2 of Chipera et al. (1997a).

Six classes of geologic materials have been incorporated in the 3-D mineralogic model. These minerals, mineral groups, or glasses used in the model are:

1. Smectite + Illite
2. Zeolites (clinoptilolite, heulandite, mordenite, chabazite, erionite, stellerite)
3. Tridymite
4. Cristobalite + Opal-CT
5. Glass
6. Analcime

Appendix I of Chipera et al. (1997a) lists the mineralogy, stratigraphy and depths of analyzed samples for each of the 17 drill holes included in this model. Data from drill holes is represented as a separate ascii file that is input directly into the model.

The commercially available computer code STRATAMODEL was used to interpolate mineral abundances onto volumes where no measurements are available. A common feature of software available for property interpolation (Stratamodel, RC², GSLIB) is that they operate on a

logically rectangular grid. However, the ISM model (Zelinski and Clayton, 1996, Clayton et al., 1997) is not represented on a logically rectangular grid. Therefore, for property interpolation and distribution, but not during construction of the computational grids, the ISM model is interpolated onto regular grids. The geologic model is generated by importing files of surfaces that contain x (easting), y (northing) and z (elevation in meters) coordinates. STRATAMODEL then assigns a regular grid spacing to these irregular grids and generates continuous surfaces. Using knowledge of the structural relationships between the surfaces specific to this site, a stratigraphic framework model is constructed which captures any pinchouts, onlap, offlap or truncations that are known to exist.

The model volume is then populated with attributes by importing the mineralogic data (obtained from Chipera) that occur at specific x, y, and depth locations within the model.

STRATAMODEL uses a $1/r^2$ interpolation scheme to distribute the properties throughout the three dimensional volume. That is, in a model cell for which there is no mineralogic information, STRATAMODEL assigns mineralogic values by searching within a certain radius from that cell and giving weight to each data point found. The weight given is based on the distance (r) of the data point from the cell being filled. Data points farther from that cell are given less weight. STRATAMODEL is guided in this step by a user-specified search radius which is based on knowledge of the site, such as drill core spacing. The $1/r^2$ interpolation scheme acts to smooth the mineralogic distributions. In Chapter 12, we present mineralogic distributions based on a more rigorous geostatistical package that better captures the spatial structure of the zeolitic mineral distributions.

The 3-D model had a grid spacing of 243.84 m (800 ft.), which covers a ground area of approximately $166 \times 10^6 \text{ m}^2$, and contains information from 17 boreholes (Chipera et al., 1997a). Figure 2-2 shows the location of the wells in the mineralogical model. The model volume is divided into 335 layers to adequately capture the vertical density of mineralogic data. The result is a model with 1,034,470 cells. Each cell of the model contains eleven values, including mineral

Figure 2-2. Locations of the 21 wells in plan view of mineralogical model.

abundance for the six mineral classes, cell location, stratigraphic layer, elevation in meters, cell volume and cell thickness. Any cell within the model volume can be queried to determine any of these values.

The more rigorous geostatistical simulations of Chapter 12 utilize an updated version of the 3-D mineralogical model (data from Chipera et al., 1997b), which incorporates data from 21 wells for a total of 3817 zeolite abundance measurements. The new wells and the location of the updated data are also shown in Figure 2-2. Figure 2-3 compares the Chipera et al. (1997a) model with the updated mineralogical model Chipera et al. (1997b) at the Antler Ridge cross section (the cross section used in the two dimensional flow and transport calculations). The figure shows that the updated model resolves more distinct zeolitic layers, but in general, the two models are similar for the Antler Ridge cross section.

Figure 2-4 compares zeolite and smectite abundance for the Chipera et al. (1997a) and updated mineralogical model (Chipera et al., 1997b) at locations close to several wells. As

Figure 2-3. Comparison of the (a) Chipera et al. (1997a) model and (b) the updated mineralogical model (Chipera et al., 1997b) for the Antler Ridge cross section.

expected, the profiles for zeolitic abundance at a location near G1 are nearly identical because G1 is present in both models (Figure 2-4a). The profiles for zeolitic abundance are only slightly different at a location near UZ-14 even though UZ-14 data was not present in Chipera et al. (1997a) model (Figure 2-4b). The Chipera et al. (1997a) model captures the correct zeolitic profile near UZ-14 for two reasons: G1 is close to UZ-14, and zeolites have large correlation lengths in the horizontal direction (see chapter 12). Therefore, the data from UZ-14 and G1 are consistent and act to decrease the overall uncertainty in the model. The additional well data used in the updated well

Figure 2-4. Comparison of Chipera et al. (1997a) and updated mineralogical model (Chipera, 1997b) at locations close to wells (a) zeolite abundance at G1 (G1 existed in both models), (b) zeolite abundance at UZ-14 (UZ-14 only exists in the updated model but is close to G1), (c) smectite abundance at NRG6 (NRG6 only exists in the updated model).

model buttresses the zeolitic abundance data from Chipera et al. (1997a). In general, the maps for zeolitic abundance are very similar between the models. However, the additional well data affects the distributions of smectite which is not as highly correlated (pers. comm. D. Vaniman). Figure

2-4c shows that the two models show significant differences in smectite abundance at a location close to NRG-6.

2.6 Conclusions

Capturing spatial variability is necessary to assess the site performance as it relates to uncertainty in attribute (permeability, porosity, K_d) distributions. This is particularly important for the zeolitic minerals between the basal vitrophere and the water table due to their role in either retarding migrating radionuclides or enhancing fracture flow. The geologic model of the unsaturated zone is based only on primary formation processes and, hence, does not capture the structure of the secondary alteration of vitric minerals to zeolites. Thus, capturing both the geologic structure and the mineralogic structure requires automatic integration of multiple sources of information. An approach and companion tools have been developed to couple the relevant models and generate appropriate attribute distributions to perform the analyses of the impact of zeolite spatial variability on repository performance. This same approach and tool set can then be applied to studies of the impact of uncertainty in attribute distribution. Chapter 12 takes this process through the development of heterogeneous fields of mineral distributions in an examination of the sensitivity to capturing the coupled chemical and physical heterogeneity in the subsurface at Yucca Mountain.

In this milestone, the distribution of zeolites in the unsaturated zone is modeled using the information integration processes developed here either through the assignment of a cut-off percentage above which the rock is given the properties of zeolitic tuff, or by populating the model with a zeolitic abundance which then is used in assigning flow and transport properties. An updated version of the mineralogic model of Chipera et al. (1997b), is incorporated into the simulations of the present study.

Finally, we compare the mineral distributions measured recently with predicted values using the version of the model before these data were available. For zeolites, we show that the new

Incorporation of the Mineralogic Model into the Flow and Transport Model

data do not significantly change the mineral distribution reflected in earlier versions of the model, whereas the distribution of smectite, a less abundant mineral in the unsaturated zone, is more refined after incorporating the new data.

Chapter 3 - Mesh Generation

3.1 Abstract

Computational mesh generation involves translating the Integrated Site Model, ISM (Zelinski and Clayton, 1996, Clayton et al., 1997), of geologic and hydrologic structure and stratigraphy to a finite element mesh. This process maintains the geometry defined by the ISM model while meeting additional geometric constraints on element size and geometry necessary for stable and accurate flow and transport solutions.

The GEOMESH/X3D grid generation toolkit, (Gable et al., 1995, Gable et al., 1996a and b, Trease et al., 1996), has been developed to provide an integrated software package for all grid generation steps including: import of initial ISM model, quality checking of input data, mesh optimization, mesh post processing and quality checking, mesh interfacing with the FEHM flow and transport code, and property interpolation from the mineralogy and petrology model (Chipera et al., 1997a and b). Chapter 3 describes the algorithms used for mesh generation and how the algorithms have been applied to the two- and three-dimensional meshes used for flow and transport computations.

3.2 QA Status of Software and Data

The GEOMESH/X3D grid generation toolkit, (Gable et al., 1995, Gable et al., 1996a and b, Trease et al., 1996) is non-Q with respect to YMP and Los Alamos quality assurance requirements. However, version control software tracking procedures identical to those used for the Q code FEHM are followed. GEOMESH/X3D utilizes well established procedures to insure robust mesh generation results and to insure traceability of changes to the software. A suite of test problems has been developed and all new releases are verified against the test suite.

Table 3-1. QA Status of Data used in this Chapter

REFERENCE	Q Status	DTN	COMMENT
Buesch et. al., 1996 , "Revised Stratigraphic nomenclature and macroscopic identification of Lithostratigraphic units of the Paintbrush Group exposed at Yucca Mountain, Nevada", USGS Survey Open-File Report 94-496, 1996	Q	GS941208314211.064	
Clayton et. al., 1997 , "ISM2.0: A 3-D geological framework and integrated site model of Yucca Mountain: Version ISM1.0", Doc ID B00000000-01717-5700-00004 Rev 0, MOL. 19970122.0053, CRWMS M&O	Q		By agreement with DOE and NRC accepted as Q even though it utilizes some non-Q data
Zelinski and Clayton, 1996 , "A 3-D geological framework and integrated site model of Yucca Mountain: Version ISM1.0", Doc ID B00000000-01717-5700-00002 Rev 1", CRWMS M&O	non-Q		

3.3 Unsaturated Zone Computational Mesh from Stratigraphic Model

All grids are built from the detailed geologic framework model ISM (Zelinski and Clayton, 1996, Clayton et al., 1997) which is correlated to the Project's hydrologic units and nomenclature in Table 15-1. Faults are represented as vertical features. This is consistent with the model of Bodvarsson et al. (1997), that incorporates a subset of the faults used in this work as vertical features. An example of a two-dimensional cross section, within the framework of the three-dimensional mesh it is extracted from is, shown in Figure 3-1.

The primary criterion used in mesh construction is that the mesh will retain the original

Figure 3-1. Layer 9 of ISM shown with colors representing elevation. The area in which a three-dimensional tetrahedral grid is build is shown by the wire frame in which each line represents a stratigraphic layer boundary. The two-dimensional grid used for computations is shown with zeolite abundance shown with red for high zeolite abundance and blue for low zeolite abundance.

geometry of the ISM model. In the grid generation process some smoothing, upscaling or reinterpretation of the ISM model is done. When approximations are necessary, they are carefully controlled and quantified to insure the final grid matches the ISM geometry.

3.3.1 Mesh Generation from ISM Input

The steps required to create a two or three-dimensional computable mesh from the Triangular Irregular Network (TIN) data are detailed below and include:

- Import ISM Triangular Irregular Network (TIN) data into GEOMESH/X3D
- Test and correct inconsistencies in TIN sheets
- Identify and tag the faults on the TIN surfaces
- Define polygonal subset of TIN surface
- Refine TIN surface grids in and adjacent to faults
- Define upper and lower boundary truncation surfaces
- Define minimum layer thickness
- Stack layers and build prism elements between surfaces
- Convert prism elements into tetrahedral elements
- If two-dimensional: Extract a two-dimensional hybrid grid from the three-dimensional tetrahedral mesh
- If two-dimensional: Convert the two-dimensional hybrid grid into a triangulated mesh
- If two-dimensional: Clip the mesh to area of interest
- Optimize the grid for computation using reconnection, smoothing, refinement and derefinement
- Interpolate attributes from geostatistics or Stratamodel representation of mineralogy
- Compute and output finite volume matrix coefficients
- Compute and output area coefficients of surface nodes
- Define node sets for setting initial conditions, boundary conditions and material properties
- Mesh quality checks

3.3.1.1 Import ISM Data into GEOMESH/X3D

The first step required to integrate the ISM data is to import the ISM model. The ISM data is provided as 31 computer files that each contain a representation of the top surface of a geologic units. Each surface is represented by a TIN surface of 4976 x, y, z coordinates and the connectivity for 9823 triangles (Figure 3-2).

3.3.2 Quality Testing and Correcting of ISM TINs

To insure that the triangular sheets representing the ISM model form a valid triangulation, the tests that must be met are:

Figure 3-2. Top view of one TIN from ISM. TIN consists of 4976 nodes and 9823 triangles. Coordinates are Nevada State Plane in meters. Each triangle vertex is assigned an elevation, resulting in a piecewise linear representation of stratigraphic surfaces.

- TIN sheets have no holes
- No triangles overlap
- All triangles are ordered to have their normal vector on the same side of the TIN.

Examination of these data found that all the TIN sheets suffered from a small number of holes and overlaps. Although these holes and overlaps are inconsequential for creating computer visualizations of the data, these problems would make the subsequent mesh generation steps

impossible if they are not first corrected. Thus software was developed to identify, characterize and repair the inconsistent TINs. This process resulted in the addition of 3 triangles to each of the 31 TINs and some minor to the connectivity to eliminate overlaps.

3.3.2.1 Identify Faults

The triangular representation of each stratigraphic boundary in ISM does not have any attribute flags indicating which parts of the TINs represent faults and which parts of the TINs represent a stratigraphic boundary. It is essential for the flow and transport studies to identify which parts of the model are fault related so that hydrologic properties of faults can be included. An algorithm was developed to identify the portions of the TIN sheets that represent faults. The algorithm is based on the observation that faults can be identified in the TIN sheets by examination of the normal vector of each facet of the TIN. The algorithm is described as follows: The normal vector to each triangle is calculated. For these sheets, all normal vectors are within ninety degrees of vertical. If the normal vector is within 45 degrees of vertical, the triangle is taken to be a stratigraphic boundary. If the normal vector is greater than 45 degrees from vertical and less than 90 degrees, it is labeled as a fault triangle. The fault triangles are further identified by their tilt to the north, east, south or west. The result is that each triangle is identified as either a stratigraphic boundary or as one of four fault types (see Figure 3-3).

3.3.2.2 Define polygonal subset of TIN surface

The area of interest for the current flow and solute transport modeling effort is smaller than the area defined in the geologic framework model ISM (Zelinski and Clayton, 1996). A polygonal subset of each TIN surface is created by removing any elements that fall outside a user defined area. Typically, this area is bounded by faults (Figure 3-4). Where no faults exist and a subset boundary is desired, elements are removed outside a straight line that connects natural fault boundaries. Since the element edges do not coincide exactly with the arbitrary bounding line, some unevenness of the boundary of the subset results. This unevenness does not effect the flow and solute transport modeling results because the boundaries are chosen to be sufficiently far from the model area of interest.

Figure 3-3. Layer 38 from ISM at 3X vertical exaggeration. Stratigraphic layer boundary is colored in yellow. West facing faults are green, south facing faults are blue.

3.3.2.3 Refine TIN surface grids in and adjacent to faults

In order to more accurately model the effect of faults on the groundwater system, the element size in, and adjacent to, identified fault triangles on each surface is decreased by adding nodes to the center of element edges (refinement). To keep the horizontal element configuration of all layers identical, refinement is done to ISM layer 24 which is the base of TSW4. This layer has the most complete identification of fault triangles. The refinement is performed after a polygonal subset of TSW4 is extracted as described in Section 3.3.2.2. Refinement consists of adding a node to the center of any element edge containing at least one node that is identified as a fault when the edge is larger than 20.0 m. This refines the element size in, and immediately adjacent to, fault triangles. The new and old nodes are then reconnected into triangular elements. The resulting mesh of Layer 24 contains 4557 nodes and 6165 elements. The z coordinates of every other layer are then mapped onto the triangular element configuration of Layer 24 to create separate surfaces with the correct depth and identical element configurations.

3.3.2.4 Define upper and lower boundary truncation surfaces

The ISM model defines stratigraphic layers much deeper than were needed for unsaturated zone calculations. Therefore, the ISM surfaces are truncated and no grids are generated below an

Figure 3-4. Top view of one TIN illustrating the subregion for the three-dimensional computational grid. TIN consists of 4557 nodes and 6165 triangles. Coordinates are Nevada State Plane in meters. Note the increased resolution near faults in comparison to the original TIN surface (see Figure 3-2).

elevation of 400 meters. This insures that 200 to 300 meters of the grid are available for calculations with different water table levels and also helps make grids smaller by not creating elements deep into the saturated zone.

The top surface of the grid must be truncated at the topographic surface. The ISM model defines stratigraphic surfaces above the topographic surface. The ISM model surfaces are filtered to check if the stratigraphic surface elevation is greater than the topographic elevation. If it is, it is reset to topographic elevation.

3.3.2.5 Define minimum layer thickness

Practical and numerical considerations make it necessary to set a minimum layer thickness before producing volume grids between the stratigraphic surfaces. In areas where a particular unit pinches out to zero thickness or is truncated by faults or topography, there can be extremely thin layers. It was found that some layers in the ISM model were as small as 4 cm, an artifact of the model building process rather than an actual geological feature. In this work, if a layer thins to less than one meter, its thickness is set to zero. This insures that large aspect ratio tetrahedral and triangular elements are avoided.

3.3.2.6 TIN Sheets Stacked and Converted to Prisms

The TIN sheets are stacked from lowest elevation to highest elevation, via a module from the GEOMESH/X3D system developed specifically for this application. The ISM model consists of a set of stacked triangle sheets that all share the same number of triangles and the same x and y coordinates for the triangle vertices. The only difference between sheets is the z elevation. The stacked sheet data structure is then converted to a finite element node and connectivity list. Now all elements are represented as prisms (6 nodes, 2 triangle faces, 4 quadrilateral faces) with vertical connections between adjacent layers. This module for the GEOMESH/X3D system was also developed specifically for this application.

Once stacked, we found that some sheets in the ISM model cross. Therefore, decisions were required for dealing with crossing TIN sheets. After consultation with William Zelinski, it was agreed that whenever a higher stratigraphic horizon crosses the layer below, the higher elevation sheet should be given an elevation equal to the lower layer. In this method layers pinch out to zero thickness if they cross. This approach exactly maintains the geometry of interfaces and faults

except in the cases where the ISM model represented negative volumes, overlapping or very thin layers.

3.3.2.7 Convert Prisms to Tetrahedra

Tetrahedral elements are fundamental to three-dimensional computations. Therefore, prism elements must be converted into tetrahedra. Another module had to be developed for the GEOMESH/X3D system to do this. Prism elements are converted to tetrahedral elements by one of two methods. The options are to create either eighteen or three tetrahedra from each prism. Eighteen tetrahedra are created by adding a node to the center of each face and a node the center of the prism element. Connections are then made to generate eighteen tetrahedra from each prism. Creating three tetrahedra does not require node addition, it is simply a change in the connectivity of the nodes representing each prism. In this work we chose to convert each prism to three tetrahedra. The resulting representation is now in the form of a tetrahedral mesh.

3.3.2.8 Extract a Two-Dimensional Plane

For two-dimensional calculations, extraction of planes from the tetrahedral mesh follows. A plane through the tetrahedral mesh is defined, which can be at any orientation, and the intersections of the tetrahedral elements with the plane are calculated. The result of intersecting a tetrahedral mesh with a plane is a two-dimensional hybrid mesh consisting of triangles and quadrilaterals.

For this work, the plane chosen was an east-west cross section extracted at Nevada State Plane (NSP) coordinate 232,100 meters. The east and west boundaries of the plane are truncated at unnamed faults in ISM at NSP coordinates 168,274 meters on the west and 174,176 meters on the east (Figure 3-5).

3.3.2.9 Convert Hybrid Grid to Triangles

The hybrid mesh of quadrilaterals and triangles must be converted to a triangular mesh. This requires that quadrilateral elements are split into 4 triangles, resulting in a fully triangular mesh.

Figure 3-5. Cross section at NSP coordinate 232,100 meters extracted from ISM. In the figure, the cross section is split in two near the Ghost Dance Fault (GDF). The east and west extent of the computational model at NSP 168,274 meters and 174,176 meters are marked by arrows. Colors represent hydrogeologic units incorporated into the computational mesh.

3.3.2.10 Clip Mesh to Area of Interest

The two-dimensional computational mesh requires east, west and bottom boundaries different than those in the ISM model. The mesh is clipped at specified east and west boundaries immediately inside of the faults and at $z=400$ meter above sea level. This leaves 220 meters below the water table for applying a variable water table at the base of the model.

3.3.2.11 Optimize Mesh For Computation

Numerical solution techniques for flow and transport calculations with finite volume and integrated finite difference methods place geometric constraints on the quality of a mesh. To optimize the mesh, we use a combination of smoothing, refinement, derefinement and reconnection algorithms to enhance mesh quality while preserving geometry.

- Reconnection

Reconnection algorithms change the connectivity of a mesh without modification of node position or the number of nodes in the grid. Two-dimensional grids use a single reconnection algorithm which will flip the shared edge of two triangles. The result is a two-triangle to two-triangle transformation that can be used to improve the quality of the grid. In three dimensions, there are many more options. In addition to two tetrahedra flipped to two new tetrahedra (2to2), there are 2to3, 3to2 and 4to4 flips. These are used in an automated reconnection algorithm to improve grid quality.

- Smoothing

An elliptic smoothing algorithm is employed to smooth the two-dimensional grid. In this step, nodes on material boundaries are held fixed, while nodes in the interior are allowed to move. This produces a mesh with more equal aspect ratio triangles (Figure 3-7). The algorithm for moving nodes poses the problem of where to move nodes in terms of minimization of a function. An objective function is calculated based on the node positions and connections and a set of linear equations are solved to find which direction of motion each node should have to minimize the function. The smoothing used in this work attempts to smooth the grid so all triangle vertex angles are close to 60 degrees. In addition we must insure that the motion of nodes will not cause any overlapping or negative area/volume elements.

Reconnection can be done without adding points by allowing connections to flip. This is useful for keeping the grid from binding during repeated smoothing applications. For the final mesh, points are added as needed to create a Delaunay grid with positive coupling coefficients (Figure 3-6). This assures that the optimum computational mesh results.

- Refinement

There are many different options for grid refinement. The methods used most often are edge, face, and element refinement. Edge refinement adds a node to an element edge, face refinement adds a node to an element face center and element refinement adds a node to the

Figure 3-6. Two-dimensional mesh generated from YMP model ISM showing faults as part of the computational grid. The faults from west (left) to east (right) are Solitario Fault, Ghost Dance Fault and Bow Ridge Fault. The cross section is bounded on the west and east by unnamed faults in ISM. Colors represent different hydrogeologic units. Close-up views of the grid are shown at each of the faults.

centroid of an element. Nodes can also be added at any location the user desires and the code will figure out which type (edge, face, element) of refinement to use when adding the node.

The refinement algorithms are also closely coupled with grid refinement criteria algorithms. All the refinement methods also need criteria which trigger the grid refinement. Some

Figure 3-7. An example of how elliptic smoothing modifies the mesh. The mesh on the left has the topology that results from extracting a two-dimensional grid from a three-dimensional tetrahedral mesh. Material interfaces are maintained but the node distribution is very uneven. The mesh on the right has been smoothed by keeping interface nodes fixed but allowing nodes between material interfaces to move. The mesh on the right has a flat bottom by truncating the mesh at elevation 720 meters.

of the options for refinement criteria are geometric regions, element aspect ratio, element edge length, field value and field gradient. One or more of these criteria are set, and elements that meet the refinement criteria are refined by one of the above methods. Multiple uses of refinement can be performed on the same mesh.

- **Derefinement**

Derefinement is nearly the opposite of refinement. Derefinement removes nodes from the mesh based on a variety of criteria related to element optimization. The criteria can be specified as edge length, element aspect ratio, or element volume. An example would be to remove one node of every element edge in the mesh that is less than 20 m in length. If the criterion of element aspect ratio or element volume is used, one node is removed from the shortest edge of any elements matching the specified criterion. Material interfaces are preserved except when a geologic material layer has a vertical width less than a user specified tolerance. Then the nodes defining the layer at that point may be removed at the user's option, potentially leaving the geologic layer discontinuous.

element in the source mesh and interpolate a value for the sink node from all node values in source element. Interpolation options are linear, $\log_{10}$, $\log_e$ and hyperbolic sine. For mineral distributions, linear interpolation is used.

3.3.3 Generation of FEHM Input Files

3.3.3.1 Compute Finite Volume Matrix Coefficients

The FEHM flow and transport code uses finite volume control volumes for solution of flow and transport equations. Part of the grid generation process is to calculate the Voronoi control volume associated with each node in the grid and the area of the polygonal faces of the Voronoi control volumes.

3.3.3.2 Compute Area Coefficients of Surface

Each node on the surface of the grid has a an associated surface area. This is computed for use in scaling flux boundary conditions to infiltration maps.

3.3.3.3 Output Information for Boundary Conditions and Material Properties

Lists of node sets are created and output. These lists are used by the FEHM code to set material properties and boundary conditions. The node set lists include outside, top and bottom nodes, stratigraphic unit number and fault number.

3.3.4 Mesh Quality Assurance Steps

The process of producing an optimized mesh for computation requires steps that alter the number of nodes and elements and move grid nodes. A number of tests are employed to ensure that the final grid meets the quality standards necessary for accurate and stable computations. These tests are:

- No holes in the mesh
- No overlapping triangles
- All elements have positive volume/area
- All material interfaces are maintained
- The mesh is a Delaunay triangulation
- All geometric coupling coefficients are positive

After the removal of nodes using the derefinement option, the remaining nodes are reconnected leaving a mesh that contains no holes or overlapping elements. The refine, derefine, and smooth options can be used sequentially and can be repeated until the problem converges and no change would occur if the process were continued. When these steps have converged, we consider the grid to be optimal for our computations.

3.3.2.12 Mapping Properties From Three-Dimensional Mineral Distribution

Zeolite distributions from the three-dimensional mineralogy model (Chipera et al., 1997a and b) have been mapped onto the computational grids. Grid to grid mapping algorithms allow properties from one mesh, (the mineralogy model), to be mapped onto another mesh with a different node distribution, (the computational mesh) (see Figure 3-8).

Figure 3-8. Zeolite distributions are mapped from a three-dimensional model onto the computational mesh. The upper figure show zeolite distributions on a grid extracted from the Stratamodel three-dimensional mineralogy model. The lower figure shows the zeolite distributions after being mapped onto the computational mesh.

The method requires a fully connected source mesh with interpolation onto the nodes of the sink mesh. The method is as follows. For each node in the sink mesh, find the corresponding

These tests were employed and passed for all meshes employed in this report.

3.4 Computational Grids Used in the Present Study

3.4.1 Three-Dimensional Unsaturated Zone Grid

The final grid used for three-dimensional calculations is shown in Figure 3-9. It contains 71,781 nodes and 414,313 tetrahedral elements. Faults represented in the ISM model are included as separate materials (Figure 3-10). Since the calculations are dual permeability, the number of grid point for FEHM calculations is 143,562.

3.4.2 Two-Dimensional Unsaturated Zone Grid

Two-dimensional cross section grids have been produced at two different resolutions. The grid shown in Figure 3-6 is a low resolution triangular mesh with 21,494 nodes and 42,086 triangular elements. The FEHM dual permeability grid has 42,988 nodes.

The mesh used for interpolation of geostatistical mineral distributions has much higher resolution between the Ghost Dance and Solitario faults at and below the potential repository, with 66,984 nodes and 78,312 triangular elements and 133,968 dual permeability grid points. A small portion of the high resolution grid is shown in Figure 3-12. In the high resolution area, the spacing between nodes is approximately 4-6 meters.

Figure 3-9. Three-dimensional unsaturated zone computational grid with 71,781 nodes, 143,562 dual permeability nodes and 414,313 tetrahedral elements.

Figure 3-10. Faults are represented as distinct materials. Layer 9 from ISM is shown. There are 14,473 nodes and 45,386 tetrahedral elements used to represent the faults. Faults that dip to the west are filled in red, faults that dip to the east are filled with green.

3.5 Conclusions

Accurate simulation of flow and transport in the unsaturated zone at Yucca Mountain requires simulation tools capable of representing the complex stratigraphy which includes dipping, faulted geologic layers of varying thickness and extent. The GEOMESH/X3D toolkit was developed specifically to address such issues. Further, once the geometric constraints of the geologic system are resolved, the flow and transport simulations are only as accurate as the mesh upon which they are computed. GEOMESH automatically insures quality finite-element grids. Further, the GEOMESH/X3D toolkit is designed to accept electronic input directly from the geologic and mineralogic models, thus insuring nearly exact replication of the structure and attribute distributions in those models.

Figure 3-11. A portion of the two-dimensional cross section is shown bounded on the west by the Solitario fault and on the east by the Ghost Dance fault. a) Stratigraphic layer boundaries, b) low resolution computational grid, c) high resolution computational grid.

Figure 3-12. A small section of the high resolution two-dimensional grid near the Solitario fault showing the triangulation and the Voronoi cells associated with the triangles.

Chapter 4 - Particle Tracking

4.1 Abstract

A new particle tracking technique is presented for simulating liquid or gas phase solute transport in porous media. The method, called the Residence Time Transfer Function (RTTF) particle tracking technique, uses 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. Transient flow fields are approximated by assuming a non-varying flow field during a time interval of a fluid flow solution. The method is a significant departure from the traditional procedure of mapping a particle trajectory by resolving the velocity vector at a given position by 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. 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. For the present study, the technique is used for unsaturated 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.

4.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al., 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 4-1.

Table 4-1. QA Status of Data used in this Chapter				
REFERENCE	Q Status	DTN		COMMENT
Fabryka-Martin et al., 1996b, "Summary Report of Chlorine-36 Studies: Sampling, Analysis and Simulation for Chlorine-36 in the Exploratory Studies Facility, YMP Milestone number 3783 M"	Q	LAJF831222AQ95.005	4, 5, 6	Accession Nos. MOL.19961015. 0100 MOL.19961015. 0103 * DTN super- sedes DTN: LAJF831222AQ 95.013 USGS DTN USGS DTN USGS DTN USGS DTN
	Q	LAJF831222AQ95.007		
	Q	LAJF831222AQ96.013		
		*		
	Q	LAJF831222AQ96.002		
	Q	LAJF831222AQ96.003		
	Q	LAJF831222AQ96.004		
	Q	LAJF831222AQ96.014		
	Q	LAJF831222AQ96.015		
	Q	LAJF831222AQ96.016		
	Q	GS950708315131.001		
	Q	GS960308315131.001		
	Q	GS950508314224.003		
	Q	GS960408314224.001		
	Q	GS960408314224.003		
	Q	GS960408314224.004		
	Q	GS060708314224.009		
Fabryka-Martin et al., 1996c, "Summary Report of Chlorine-36 Studies, YMP Milestone Report 3782M"	non-Q	LAJF831222AN96.001	4, 6	Accession Nos. MOL.19961015. 0094
	Q	LAJF831222AQ96.002		
	Q	LAJF831222AQ96.003		
	Q	LAJF831222AQ96.004		
	Q	LAJF831222AQ96.005		
	Q	LAJF831222AQ96.006		
	Q	LAJF831222AQ96.007		
	Q	LAJF831222AQ96.008		
	Q	LAJF831222AN96.009		
	Q	LAJF831222AQ96.010		
	Q	LAJF831222AQ96.011		
	Q	LAJF831222AQ96.012		
	Q	LAJF831222AQ96.013		
	Q	LAJF831222AQ96.014		
	Q	LAJF831222AQ96.015		
	Q	LAJF831222AQ96.016		
	Q	LAJF831222AQ95.003		
	non-Q	LAJF831222AN95.004		

Yang, et al., 1996, Interpretations of Chemical and Iso- topic Data From Boreholes in the Unsaturated Zone at Yucca Moun- tain, Nevada, USGS Water-Resourc- es Investigations Report WRIR 96- 4058	Q		4, 6, 11	
--	---	--	----------	--

4.3 Introduction: Conceptual Model For Particle Transport

Prediction of solute transport is a critical element of many groundwater flow studies, particularly when the transport of a contaminant is the focus of the work. Modeling efforts typically are motivated by the need to predict the movement of a pollutant in the subsurface to answer practical questions concerning the rate and direction of contaminant movement and the degree to which the quality of the groundwater may be endangered. In a typical solute transport simulation, a dissolved chemical is introduced into a steady-state or time-varying flow field, and the fate of the chemical is tracked while undergoing physical and chemical processes such as advection, dispersion, chemical and biological reaction, or diffusion into dead-end pore space. Often, a concentration front is established that must be tracked accurately. In addition, many field investigations employ tracers to study the flow and transport system, and these tests also require models to simulate the movement of dissolved species.

Traditional solutions to the advection-dispersion (CD) equation, such as those used in most finite-element or finite-difference flow and transport codes, are versatile and allow the simultaneous solution of multiple interacting species. One drawback of a finite-difference or finite-element solution to the CD 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 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 CD 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 casts the CD 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.

Particle-tracking transport models take a fundamentally different approach. 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 Δt . 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 three-dimensional finite difference grids (Goode, 1990; Schafer-Perini and Wilson, 1991; Zheng, 1993), two- and three-dimensional 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.

In this chapter, we first outline the mathematical basis for this algorithm, developing theory to incorporate the effects of sorption, dispersion, and matrix diffusion into this new particle-tracking framework. Then, after a brief description of the structure of the particle-tracking code, we validate the technique by comparing the results to analytical solutions and by benchmarking the code against a finite-element solution of the advection-dispersion equation for a complex flow system for which no analytical solution is available. Finally, we present two applications that examine solute transport issues relevant to YMP, illustrating the usefulness of the particle-tracking technique for studying complex, large-scale transport problems in both unsaturated and saturated flow systems.

4.4 Mathematical Development

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. Thus, we call this particle-tracking approach the “residence time/transfer function” (RTTF) method. The schematic plot shown in Figure 4-1 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.

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

$$\tau_{part} = \tau_f = \frac{M_f}{\sum \dot{m}_{out}} \quad (\text{Eq. 4-1})$$

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.

Figure 4-1. Use of the transfer function to determine the residence time of a particle in a cell for the RTTF particle tracking technique.

Equilibrium, linear sorption is included by correcting the residence time by a retardation factor R_f .

Thus, $\tau_{part} = R_f \tau_f$, and R_f is given by

$$R_f = 1 + \frac{\rho_b K_d}{\phi S_f \rho_f}, \quad (\text{Eq. 4-2})$$

where K_d is the equilibrium sorption coefficient, ρ_b is the bulk rock density, ϕ is the porosity, S_f is the saturation of the phase in which the particle is traveling, and ρ_f is the density of the fluid.

Once again, in the absence of other transport processes, the transfer function is a Heaviside function.

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.

4.4.1 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 one-dimensional, axial dispersion is valid. The transport equation and boundary conditions for the one-dimensional, advective-dispersion equation are:

$$R_f \frac{\partial C}{\partial t} = D_{eff} \frac{\partial^2 C}{\partial x^2} - u \frac{\partial C}{\partial x}, \quad (\text{Eq. 4-3})$$

$$C = 0, \quad t = 0, \quad (\text{Eq. 4-4})$$

$$C = C_0, \quad x = 0, \quad (\text{Eq. 4-5})$$

$$C = 0, \quad x \rightarrow \infty, \quad (\text{Eq. 4-6})$$

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 we assume that the flow dispersion component of D_{eff} is large compared to the molecular diffusion coefficient D_{AB} . A non-dimensional version of equation (Eq. 4-3) can be obtained by the following transformations: $\hat{C} = C/C_0$, $\hat{x} = x/L$, and $\theta = R_f u t / L = R_f \tau_f$, where L is the flow path length. The solution to equation (Eq. 4-3)-equation (Eq. 4-6) is given by

Freeze and Cherry (1979):

$$\hat{C} = \frac{1}{2} \left[\operatorname{erfc} \left(\frac{\sqrt{Pe}(1-\theta)}{2\sqrt{\theta}} \right) + e^{Pe} \operatorname{erfc} \left(\frac{\sqrt{Pe}(1+\theta)}{2\sqrt{\theta}} \right) \right] \quad (\text{Eq. 4-7})$$

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 characteristic length scale of the computational cell. Note that in contrast to conventional numerical solutions of the CD 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 multi-dimensional flow systems, the dispersion model developed for one-dimensional 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:

$$\alpha = \frac{L}{\sqrt{\Delta x^2/\alpha_x^2 + \Delta y^2/\alpha_y^2 + \Delta z^2/\alpha_z^2}}. \quad (\text{Eq. 4-8})$$

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 (Eq. 4-7) for a randomly determined value of the dimensionless concentration $\hat{C}$. This determination is accomplished numerically in the particle-tracking code by fitting equation (Eq. 4-7) at selected values of Pe between 1 and 1000 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% error) for Peclet numbers between 1 and 1000 are easily obtained using this linear-interpolation method.

4.4.2 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. This effect has recently been demonstrated on the laboratory scale by Reimus (1995) and on the field scale both by Maloszewski and Zuber (1991) 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 4-2 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 (Eq. 4-6), whereas transport between the fracture and matrix is described by the one-dimensional diffusion equation:

Figure 4-2. Conceptual model for matrix diffusion. One dimensional flow occurs in the fracture, with molecular diffusion into the rock matrix.

$$R_{f,m} \frac{\partial C}{\partial t} = D_{AB} \frac{\partial^2 C}{\partial y^2}, \quad (\text{Eq. 4-9})$$

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

$$\frac{C}{C_0} = \text{erf} \left[\frac{\phi \tau_f \sqrt{R_{f,m} D_{AB}}}{b \sqrt{t - R_f \tau_f}} \right] \quad (\text{Eq. 4-10})$$

for $t > R_f \tau_f$, and $C/C_0 = 0$ for $t \leq R_f \tau_f$, where b is the fracture aperture. 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 $\frac{\partial C}{\partial y} = 0$ at $y = S/2$. The extent of matrix diffusion can be parameterized using the diffusion number $Di = (\phi \sqrt{R_{f,m} D_{AB}})/b$.

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 one-dimensional behavior, with the transport time given by the fracture transport time times the ratio of the total porosity to the fracture 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.

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 one-dimensional dispersion (equation (Eq. 4-7)) without sorption. This fracture residence time is then used in the plug-flow equation with matrix diffusion and sorption (equation (Eq. 4-10)) to compute the particle residence time. To use equation (Eq. 4-10) as a transfer function, a numerical algorithm was developed to determine the inverse of the error function, that is, the value of x_d for a given value of y_d , such that

$y_d = \text{erf}(x_d)$. 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. Proof that this numerical technique is acceptable is presented in the Validation section.

4.4.3 Radioactive Decay

To incorporate radioactive decay, we use the concept of particle mass (as is used by Fabriol et al. (1993) to track chemical reactions in a particle-tracking model), applying an irreversible, first-order reaction to reduce the mass of the particle from its initial value. When concentration values are computed from the composite behavior of a large number of particles, this method accurately accounts for radioactive decay.

4.4.4 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 2, an arbitrary number of particles are released at each specified cell, regardless of the source flow rate. In the present study, method 1 is used for ^{36}Cl and major-ion chemical simulations in which the solute is entering with the infiltrating fluid, and method 2 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.

4.4.5 Transient Flow Fields

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 t 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 (Eq. 4-1) 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 (Eq. 4-1), whereas equation (Eq. 4-1) itself is used when there is net drainage of fluid.

4.4.6 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. An example illustrating the use of the dual-permeability particle-tracking formulation is described in the Applications section.

4.5 Code Implementation

The RTTF particle-tracking model developed in the present study has been implemented in the Finite Element Heat and Mass (FEHM) computer code (Zyvoloski et al., 1992), a transient heat and mass transport simulator for porous media problems in two or three dimensions. Although FEHM is a finite-element model, it uses a finite volume formulation, so that fluid mass is specified at each node of the computational grid, as well as internode mass flow rates (for details, see Zyvoloski, 1983). Thus, the requirement that the flow solution be cell-based is satisfied for FEHM. The method can also be adapted to the TOUGH2 (Pruess, 1991) or NUFT (Nitao, 1995) codes, both of which are integrated finite-difference simulators.

One of the attractive features of finite-element simulators is the ability to use computational grids that precisely follow major structural features such as faults or surfaces of contact between distinct hydrostratigraphic units. The grids can be quite complex, with elements of various sizes and types and different numbers of connections for each grid point. These grids would make implementation of a robust velocity interpolation scheme problematic. By contrast, the RTTF particle-tracking algorithm requires only the internode fluid mass flow rates, information that is produced as a matter of course in the fluid flow calculation. This factor made the RTTF particle-tracking technique a good choice for incorporation into FEHM.

FEHM simulates the flow of both the liquid and vapor phases. To provide a full transport model capability, the particle-tracking algorithm was implemented for solutes that reside in either the liquid or vapor phase. Figure 4-3 is a flow chart of the particle-tracking algorithm. The heat and mass transfer simulator controls the time stepping of the simulation, and the particle-tracking is carried out with the flow field established at that time. Each time a fluid flow time step is taken, the migration of all particles is determined up to the new time. Time steps are not required within the particle tracking portion of the calculation. For this algorithm, information that is cell-dependent, such as fluid residence time and an array of normalized outflows to adjacent nodes, is computed only once (until the flow field changes) when a particle enters the cell. This information for the cell is then used for all other particles that pass through the cell. Particle-dependent information, such as the transfer function, particle residence time, and the determination of which adjacent cell to travel to, are calculated for each particle. The use of cell-based computations that are independent of the number of particles used, coupled with a transfer-function approach that does not require time stepping, results in an extremely efficient particle-tracking algorithm. For example, several million particles can easily be tracked on conventional PCs or workstations.

Figure 4-3. Flow chart illustrating the computation sequence for the particle tracking algorithm and its relationship to the fluid flow calculations in a finite element flow code.

4.6 Validation

4.6.1 One-Dimensional Transport with Dispersion and Sorption

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

In the first suite of one-dimensional 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 (Eq. 4-7), given by:

$$C = \frac{1}{2\theta} \exp\left[\frac{-Pe(1-\theta)^2}{4\theta}\right] \sqrt{\frac{Pe}{\pi\theta}}. \quad (\text{Eq. 4-11})$$

The cumulative RTD is also compiled from the particle statistics at the outlet node and compared to equation (Eq. 4-7). Figure 4-4 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 4-5. 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 4-4 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 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

Figure 4-4. Cumulative RTD curves produced by the RTTF particle tracking technique for one-dimensional dispersion (filled circles), compared to analytical solution (solid curves). In the curves without sorption, Pe was varied. For the dashed curve, linear sorption was included for $Pe = 20$.

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 three-dimensional problems, which necessarily result in large spatial discretization due to computational limitations.

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

Figure 4-5. Residence time distribution (RTD) curves calculated using the RTTF particle tracking technique (symbols) for $Pe = 20$ for different numbers of particles, compared to the analytical solution (solid curve). The RTD is a solute concentration normalized so that the area under the curve is unity.

4.6.2 One-Dimensional Transport with Matrix Diffusion

Figure 4-6 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 both 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.

Figure 4-6. Cumulative RTD curves produced by the RTTF particle tracking technique for the matrix diffusion model (filled circles), compared to the analytical solution of Tang et al. (1981) (solid curves). Curve a) small amount of matrix diffusion; Curve b) moderate amount of matrix diffusion; Curve c) Matrix diffusion and sorption on fracture and in matrix; Curve d) Dispersion, matrix diffusion, and sorption on fracture and in matrix.

4.6.3 Transient Wetting of a 1-D Column

In this simulation, a transient, one-dimensional, unsaturated flow system is simulated. Using a relative permeability model proposed by van Genuchten (1980), we model the movement of a water front under the influence of gravity and associated tracer particles. The entire flow path is initially set at a saturation below the residual liquid saturation of the medium, and a constant infiltration rate is applied at the inlet of the column. Two particle-tracking simulations are performed: one in which the particles are injected with the infiltrating fluid, and one in which the particles start at a location halfway down the column (cell 54 out of a total of 107). This validation problem tests the

ability of the code to handle the situation of a transient flow field. The exercise is thus important to justify the use of the particle-tracking method for examining radionuclide migration in transient flow fields (Section 8.8).

When the particles are injected with the infiltrating fluid, the particles should move with the saturation front. Figure 4-7 shows that the expected behavior is approximated (the filled circles

Figure 4-7. Movement of particles with the saturation front for the transient wetting case. The curves are the saturation profiles at regular time intervals. Simulations are depicted in which the particles start at the entrance to the flow path (the filled circles) and at a position part way down the column (the X's). For the latter case, the particles do not move until the saturation front reaches the particles.

in the plot), with the particles lagging the front edge of the wetting front by two cells. This slight lag of the particle movement is caused by a slightly dispersed wetting front that does not maintain a sharp interface. For the situation in which the particles are introduced halfway down the length of the column (the X's), the particles remain stationary until the saturation front reaches the particles, after which time the particles move with the front as before. The figure demonstrates that for this transient flow case, the RTTF particle-tracking approach provides an adequate solute transport simulation.

4.6.4 Benchmarking Calculation for an Interwell Tracer Simulation

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 CD equation using FEHM for a complex, two-dimensional flow field. Although this test problem is for a saturated system, it serves as an appropriate test of the method and, thus, is presented in this report.

Figure 4-8 is a plan view of the model domain and grid. This simulation is designed after the field tracer experiments planned at the C-Wells (Robinson, 1994). These tests are designed to measure the hydrologic and transport properties of the saturated zone beneath Yucca Mountain. A permeability field that varies log-normally was generated with a Gaussian spatial correlation structure. The mean of the distribution is 10^{-12} m^2 , standard deviation is 1 (units of $\ln(\text{m}^2)$), 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 4-9 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% 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

Figure 4-8. Model geometry and finite element grid for the two-dimensional models of interwell tracer tests at the C-Wells, Yucca Mountain, Nevada. The solid circles are an injector and producer for the case of wells aligned with the grid, and the open circles are the same for the case of wells misaligned with respect to the grid.

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

Figure 4-9. Comparison of RTTF particle tracking simulation (solid curves) and finite element CD equation solution (dotted curve). The two particle tracking curves show the effect of varying the dispersivity (0 and 1 m).

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% of the cpu time of the advection-dispersion equation solution.

4.7 Applications

The three example calculations presented in this section illustrate the use of the RTTF particle-tracking method for solute transport problems for which conventional solutions to the CD equation are not well suited. The applications are solute transport problems of interest for YMP. More comprehensive analyses on these topics will be published in separate papers or are present as analyses in this report. They are used here to discuss the particle-tracking methods as applied to simulations of interest.

4.7.1 Dual-Permeability Transport for the Unsaturated Zone

In an equivalent continuum flow model, the permeability of the liquid phase is computed as a weighted average of the fracture and matrix portions of the medium. At high fluid saturations, the fractures begin to transmit the majority of the fluid. However, for solute transport, the porosity of the matrix rock is generally assumed to be available for transport, whereas in reality the much smaller available porosity of the fractures may actually transmit solute when fracture flow dominates. A dual-permeability model formulation is a more preferable way to simulate solute transport in such instances because the porosity and flow rate through each continuum is considered explicitly. In this way, the relative proportions of solute traveling rapidly through fractures and more slowly through the matrix rock can be computed.

To illustrate the use of the particle-tracking model in a dual-permeability system, a simplified one-dimensional flow and transport system representing the vadose zone infiltration problem for Yucca Mountain is considered (Figure 4-10). The fluid flow characteristics of this model problem have been examined by Ho et al. (1995). The hydrologic parameters are set by dividing the system into four hydrogeologic units. The uppermost Tiva Canyon welded (TCw) unit and third unit, the Topopah Springs welded (TSw) unit, are composed of highly fractured, welded

Figure 4-10. Schematic of the one-dimensional model system of the vadose zone at Yucca Mountain used for the dual permeability particle tracking example.

tuffs of low matrix permeability. They are separated by a nonwelded, relatively unfractured tuff of high matrix permeability called the Paintbrush Tuff nonwelded (PTn) unit. The fourth unit, the Calico Hills nonwelded (CHn) unit, is also a relatively unfractured rock of high matrix

permeability extending to the water table in this simulation. Hydrologic property values used in the calculations below are those cited by Ho et al. (1995).

At a high-enough infiltration rate, the flow and transport system will exhibit behavior that is characteristic of a fracture-dominated system, especially in the welded TCw and TSw units.

Figure 4-11 shows the composite solute breakthrough curves at the bottom of each unit obtained

Figure 4-11. Composite solute breakthrough curves (fracture plus matrix contributions) at the bottom of each hydrogeologic unit at 1 mm/y infiltration rate using the dual permeability particle tracking model (solid curves). The dashed curves are comparable results assuming an equivalent continuum model.

by adding the contribution from the fractures and matrix. The response of the system to a step change in concentration at the surface is depicted in the figure. At the assumed infiltration rate of 1 mm/y, most of the solute travels rapidly through the TCw unit, bypassing the matrix altogether.

It then transfers to the matrix of the PTn unit, traveling slowly through this unfractured rock mass. Thus, the breakthrough curve at the bottom of the PTn occurs only after spending at least 5000 y in this unit. Some of the solute transfers back to the fractures in the TSw unit where it exhibits rapid travel times before moving back into the matrix of the CHn unit. This sort of behavior is not captured in the equivalent continuum model simulation shown in the figure for the breakthrough curve at the bottom of the model domain. Travel times in units that exhibit significant fracture flow are severely overestimated with an equivalent-continuum model that assumes the solute is perfectly mixed throughout the matrix rock.

The available field evidence supports some aspects of this simulation, as discussed in greater detail below in the application of the particle-tracking technique to the simulation of environmental isotopes. The equivalent-continuum model using either particle tracking or a finite-element solute transport solver cannot represent rapid transport through welded, highly fractured units, because in equivalent-continuum models, the travel times through the TCw are computed based on the matrix porosity, which results in long travel times even though significant fracture flow occurs (> 2000 y).

4.7.2 Simulations of Environmental Isotopes in the Unsaturated Zone

Naturally occurring radioactive isotopes such as ^{36}Cl and ^{14}C are often used as environmental tracers for dating the age of groundwater samples. These isotopes, produced by cosmic-ray secondaries interacting with atoms in the atmosphere, enter the subsurface in precipitation and as dry fallout. The methods used to estimate fluid travel times in the unsaturated zone at Yucca Mountain differ for these isotopes for reasons related to their half-lives and estimated time-dependent source concentrations at the surface. ^{14}C , with its half-life of 5730 y, can be used to date fluids in the age range of about 1 to 30 ky by comparing its concentration with the value present in modern fluid. By contrast, ^{36}Cl has a half-life of 301 ky, meaning that a similar method for age dating can only be applied for fluids at least 50 ky in age (Liu et al., 1995). However, Plummer et al. (1996) show that the source value of the $^{36}\text{Cl}/\text{Cl}$ ratio has varied substantially in the past, perhaps due to geomagnetic variations or a decrease in stable Cl concentration during past wetter climates. Recon-

structions of the surface signal such as that in Figure 4-12 (from Fabryka-Martin et al., 1996c) can

Figure 4-12. Reconstructed $^{36}\text{Cl}/\text{Cl}$ input ratio used in the particle tracking simulations (from Fabryka-Martin et al., 1996c).

be applied in models to interpret field measurements with fluid ages of 0 to 50 ky. Instead of radioactive decay, the past variation in input signal is used for the age estimate. Furthermore, a man-made signal about 400 times larger than the present-day $^{36}\text{Cl}/\text{Cl}$ ratio was associated with atmospheric testing of more than 70 nuclear weapons in the Pacific Proving Grounds between 1952 and 1958. This forty-year-old spike in the input function, termed “bomb-pulse” ^{36}Cl , provides an indicator for fast-moving fluid in the unsaturated zone.

In principal, if enough measurements of these environmental tracers can be obtained as a function of depth and position, hydrologic flow models can be calibrated, thereby enabling more accurate predictions of radionuclide migration from the potential repository to the accessible environment. In the present study, we demonstrate the use of the RTTF particle-tracking technique for simulating the movement of ^{36}Cl and ^{14}C in a dual-permeability, unsaturated-zone flow and transport model. For this application, the mathematical methods outlined above were augmented to record the age of each particle passing through a given cell, so that in the case of ^{36}Cl , the time-varying input signal could be reflected in the calculation of concentration at that location. In the

simulation, all particles are released at the initial time, and the average concentration at a position in the model is computed as the average concentration of all particles passing through the cell. This approach is valid for steady-state flow systems, the assumed state of the Yucca Mountain unsaturated-zone flow system in these simulations.

Figure 4-13 shows the results of simulations of the $^{36}\text{Cl}/\text{Cl}$ ratio (Figure 4-13a) and ^{14}C concentration (Figure 4-13b) in a two-dimensional, West-East cross section at Yucca Mountain for assumed infiltration rate of 5 mm/y. The bomb-pulse ^{36}Cl signal (the red concentration, greater than 1200×10^{-15} , after Fabryka-Martin et al. (1996b)) is located in the PTn, underlying the fractured TCw unit. This result, in agreement with borehole measurements reported by Fabryka-Martin et al. (1993), is caused by fracture transport through the TCw. The prediction of ratios in excess of the present-day ratio of 500×10^{-15} in the TSw unit also agree with measured values in samples collected in the Exploratory Studies Facility (ESF) (Fabryka-Martin et al., 1996b).

Analyses of ^{14}C in the pore water are available mainly in the PTn and CHn hydrogeologic units, as well as in the perched water samples collected at the site (Yang et al., 1996). The model result of Figure 4-13b agrees in general with the findings of Yang et al. (1996). First, young ages are predicted in the PTn, estimated to be of the order of 1000 y (or 0.9 pmc). As with the ^{36}Cl analysis, rapid travel through fractures in the TCw results in young ages in the PTn. The exact age measured in fluids in the PTn matrix depends on local conditions where the sample was collected, but travel times on the order of a few thousand years through the matrix of this unit are expected. Also, perched water fluid ages on the order of 4 to 7 ky in the CHn measured by Yang et al. (1996) are also reflected in these simulations (the pmc of the matrix fluid at the top of the CHn is of the order of 0.5 in this simulation, corresponding to one half-life, or 5730 y). After spending several thousand years traveling through the PTn matrix, a significant fraction of the fluid (and particles) rapidly traverses the TSw in fractures, and relatively young ages are then recorded in the CHn matrix.

Figure 4-13. Simulated concentrations of $^{36}\text{Cl}/\text{Cl}$ ratio (a) and ^{14}C in the two-dimensional model domain. For ^{36}Cl , the bomb-pulse signal is red, and the values at the ESF horizon are in agreement with field measurements. For ^{14}C , values in the PTn (above ESF) and CHn (below ESF) are in qualitative agreement with field measurements.

There are significant limitations to carrying out a similar analysis using conventional finite-difference or finite-element solute transport simulations, especially in dual-permeability calculations. Most important is the enormous disparity in travel times in fractures and matrix, even at the same position in space. A special form of the Courant condition exists in dual-permeability simulations; for accuracy, the time step is limited to the travel time through the fractures during the

period in which significant concentration gradients exist anywhere in the fractured continuum. Either uniformly small time steps or special front-tracking and embedded time-stepping schemes would be required in certain dual-permeability applications. The cell-based particle-tracking method developed here provides accurate solutions without resorting to these sophisticated and computationally intensive methods.

4.7.3 Sorbing tracers with matrix diffusion

The saturated zone beneath Yucca Mountain is being studied for its potential to retard the movement and dilute the concentrations of radionuclides that might be released from a potential repository and percolate through the vadose zone to the water table. Interwell tracer tests are being planned using conservative and weakly sorbing solutes to study the transport mechanisms in the saturated zone. A key issue being addressed in these studies is the extent to which laboratory-determined sorption parameters for radionuclides can be used to predict migration rates in the field. For regulatory and practical reasons, the radionuclides themselves cannot be used in large-scale, saturated-zone field tests. Instead, the plan is to use a nontoxic, sorbing tracer of known sorption characteristics to test the methodology of using laboratory data in field-scale transport models. If accurate predictions of field transport behavior can be made based on laboratory-determined sorption parameters and a measured conservative tracer breakthrough curve, then we have increased confidence that the radionuclide sorption data are valid when used in field-scale models of radionuclide migration from a potential repository.

The simulations presented here using the RTTF particle-tracking model are on the heterogeneous, two-dimensional flow field presented in the benchmarking exercise in the Validation section. Simulations of two-well recirculating tracer tests will now be presented for this example problem. In contrast to the benchmarking calculations, these simulations will include the effects of matrix diffusion and sorption. Matrix diffusion is expected to be significant over the time scale of a field tracer test. However, this problem is exceedingly difficult to model with finite-difference or finite-element solutions to the advection-dispersion equation, as large concentration gradients would be expected adjacent to fractures. For example, for a transit time of 5 hours, the

characteristic diffusional distance into the rock matrix for a diffusion coefficient of $10^{-10} \text{ m}^2/\text{s}$ is about 1 mm. A discrete fracture model with numerous grid points within this 1 mm distance would be required for an accurate simulation of transport by conventional means. By contrast, the transfer function approach for matrix diffusion is ideally suited for this simulation.

Figure 4-14 compares the simulated breakthrough curves with and without matrix

Figure 4-14. Simulations with and without matrix diffusion for the two-dimensional interwell tracer experiments modeled in Figure 4-8. Also shown is a simulation of a tracer that sorbs to the matrix rock (the dotted curve).

diffusion. Tracers with different diffusion coefficients could in theory be employed to measure the extent to which matrix diffusion affects transport in the saturated zone. Reimus (1995)

demonstrated this concept in laboratory-scale fracture transport tests, showing that polystyrene microspheres can be interpreted as tracers with very low diffusion coefficients.

As detailed in the Mathematical Development section, sorption is also included in the matrix diffusion transfer function. For these Yucca Mountain experiments, Robinson (1994) proposed that the lithium cation (Li^+) be used as a sorbing tracer. The measured K_d determined for lithium is about 0.6 g/cc at a fluid concentration of 50 ppm. If we assume that this K_d value applies in the rock matrix, then the value of $R_{f,m}$ in equation (Eq. 4-10) is calculated to be 25. For the curve marked "Matrix Diffusion and Sorption" in Figure 4-14, this retardation factor is assumed for the matrix, with sorption in the fractures assumed to be negligible. If, however, this retardation factor had been taken for both the fracture and matrix, then the model would have predicted breakthrough times that are roughly 25 times longer than those shown in the figure.

The sensitivity of the model to assumptions about sorption implies that field tests such as these in fractured media can be used to pinpoint the nature of transport in fractured rock more precisely than can conservative tracer tests. The RTTF particle-tracking approach is well equipped to handle this transport problem. The diffusional time scale, when compared to the transport time within the fractures, is such that solutes diffuse only a short distance into the rock. Finite-element and finite-difference models, even using a dual-porosity or discrete-fracture formulation, cannot realistically cope with these large concentration gradients. The transfer function for matrix diffusion allows us to simulate this transport process very precisely.

One potential concern when using the RTTF particle-tracking method that is not present in simple one-dimensional calculations is the possibility of grid orientation effects. When the flow streamlines are not oriented with the grid, this method may require that a particle take a somewhat circuitous route that follows the grid. This topic was the subject of discussion (Goode and Shapiro, 1991; Desbarats, 1991) concerning the original node-to-node routing scheme of Desbarats (1990). To examine this effect, we present two calculations on the two-dimensional grid used in this example problem. In both cases, the medium is assumed to be homogeneous, but the wells are

oriented either along a horizontal grid line (the filled circles in Figure 4-8) or diagonally with respect to the grid lines (the open circles). For comparison, finite-element transport solutions were also carried out. Figure 4-15 shows results of this comparison. First, note that although the finite-

Figure 4-15. Breakthrough curves for a homogeneous permeability field examining the effect of grid orientation on the transport behavior of the RTTF particle tracking method. Solid curves: wells aligned with the grid; curves with symbols: particle tracking solutions.

element solution exhibits the same trend in the breakthrough curve, some numerical dispersion is also experienced. With respect to grid orientation effects, the particle-tracking breakthrough curves show grid orientation effects that are similar both qualitatively and even quantitatively to those experienced with the finite-element solution. The same basic phenomenon causing the effect (flow streamlines that are not aligned with the grid lines) exerts the same influence on the model results for the two methods. This amount of error due to grid orientation effects would almost certainly be

acceptable for analysis of field tracer breakthrough curves, because uncertainties in other model parameters, as well as experimental uncertainties, would likely exert a much stronger effect on the results. Nonetheless, the possibility of grid orientation effects that impact the results significantly should be evaluated for each specific particle-tracking model application.

With respect to unsaturated-zone solute transport, the geometry of the flow and transport system, along with actual dispersion mechanisms that compete with grid effects, are expected to minimize the importance of grid effects. The transport pathways have a distance similar to that of the thickness of the unsaturated zone or, in the case of radionuclide migration, the distance from the repository to the water table. Solute source terms are introduced into the system over the entire surface or the entire length of the repository. This geometry is quite different than the case in which a distinct, confined plume of solute must be tracked accurately over long distances at flow path trajectories that are not aligned with the grid. Also, the vast disparity in travel times between fractures and matrix, and the variations in thickness of nonwelded units such as the PTn and CHn give rise to solute spreading that should far outweigh the small amount of grid-induced dispersion. For this reason, we conclude that the RTTF particle-tracking technique is suitable for unsaturated-zone transport simulations of isotope and major-ion chemistry and for radionuclide migration predictions.

4.8 Conclusions

The particle-tracking method developed in this chapter, called the Residence Time Transfer Function (RTTF) particle-tracking technique, employs a new, efficient particle-tracking algorithm that is suitable for performing large-scale transport simulations. Like most particle-tracking methods, this algorithm provides advection-dominated solutions that are free of numerical dispersion. Thus, the technique can track a sharp front in solute concentration without the usual numerical dispersion or concentration-profile oscillations encountered in conventional finite-difference or finite-element solutions of the advection-dispersion equation.

The development that distinguishes the RTTF particle-tracking method from existing groundwater transport particle-tracking techniques is the conceptualization of transport as the movement of particles from cell to cell. This approach eliminates the need to resolve the velocity vectors by interpolation at all particle positions. Instead, the mean residence time and the probabilities of travel to adjacent cells are computed once for each cell (until the flow solution changes), regardless of how many particles travel through the cell. Another advantage of the cell-to-cell approach is that the information needed to compute particle residence times and pathways is readily available in finite-difference or finite-element solutions of the flow problem. The fluid mass and intercell mass flows rates are a direct result of the fluid flow solution, and thus the method can be implemented without regard to the nature of the numerical grid (structured versus unstructured grids, element shapes, etc.). The particle-tracking technique is also simple to implement for dual-permeability flow models, since the fracture-matrix interaction term is treated as another flow rate to or from a given node.

As particles are tracked through the model domain, no particle transport time stepping is necessary; all particles are tracked from the starting time to the ending time of a segment of the simulation. The only reasons to terminate and restart a simulation are either to update a flow field in a transient flow simulation or to pause to write the particle positions to output files. Thus the method is computationally efficient: simulations of several million particles are practical on conventional workstations without employing parallel processing hardware. Like most particle-tracking techniques, the algorithm would parallelize naturally, as the fate of each particle can be computed independently of the movement of other particles. The ability to track the movement of a large number of particles allows large-scale transport simulations to be carried out.

The concept of particle residence times in each cell has been extended to relax the assumption of pure plug flow without dispersion. Transfer functions have been developed to simulate dispersion, equilibrium sorption, and matrix diffusion. For accuracy, dispersion coefficients must be small enough that the grid Peclet number remains greater than one throughout the grid. This limitation is not viewed as overly restrictive, because systems with large dispersion

coefficients can be simulated accurately on relatively coarse grids with conventional finite-difference or finite-element solutions to the advection-dispersion equation. Thus, this particle-tracking method nicely complements the solute transport capabilities present in existing flow and transport codes such as FEHM. The matrix diffusion transfer function fills a void in our ability to simulate tracer experiments in fractured media using equivalent-continuum approaches. Transport time scales in tracer experiments are such that characteristic diffusional distances into the rock matrix are of the order of millimeters. The transfer function is a logical alternative to the explicit simulation of large concentration gradients in the matrix rock in the vicinity of fractures. Linear, equilibrium sorption is also incorporated into the matrix diffusion transfer function. This feature allows a variety of pollutant transport problems to be solved efficiently. When the assumptions inherent in the linear sorption isotherm are inappropriate, more complex chemical transport models should be employed.

The ability to implement the RTTF particle-tracking technique efficiently in a dual-permeability model framework is another advantage. Numerically, the communication between fractures and matrix is treated as one additional connection with a flow rate known from the fluid flow calculation. Rapid transit times through fractures can be duplicated easily using the method, whereas a typical equivalent-continuum representation will overestimate the travel time by using the matrix porosity in the computation. Finite-element solutions to the advection-dispersion equation in dual permeability, though possible, are problematic because the short travel times in fractures impose a very stringent constraint on the time step. This restriction can be minimized by increasing the diffusion coefficient between the fracture and matrix domains, which serves to reduce transport velocities by allowing solute to spend time in the matrix. However, in many transport problems, that extent of communication with the matrix is expected to be small, and compromises such as this will overestimate transport times. This limitation does not exist in the RTTF particle-tracking technique.

Like all numerical methods, the residence time/transfer function particle-tracking technique has limitations that must be considered when deciding whether its use is appropriate for

a given application. The assumptions of advection-dominated transport and linear, equilibrium sorption have just been discussed. The possibility of grid orientation effects must also be considered. The example problem presented here showed that the grid orientation effects for this technique appear to be similar in character to those encountered in conventional finite-difference or finite-element solutions to the CD equation. If grid orientation errors are too large for a given application, then more complex front-tracking algorithms should be used instead, such as conventional particle tracking or locally refined grids.

As unstructured numerical grid generation techniques become more common, the simplicity of implementing the RTTF particle-tracking technique for these grids should be a great benefit. In the Yucca Mountain Project, for example, unstructured grids are made to follow the complex stratigraphy present at the site, so that units with contrasting hydrologic properties can be represented. We suspect that for these grids, orientation errors are small because the grid is aligned with the hydrostratigraphic units and thus are more likely to be aligned with the flow field. Thus, one of the RTTF particle-tracking technique's possible limitations should be minimized.

The particle-tracking technique developed in the present study is designed to solve the CD equation to simulate the movement of tracers, pollutants, or solutes in subsurface porous media. Though not specifically designed to map fluid streamlines, the technique can also be used to view the movement of fluid from a given position, thereby providing a quick look at the flow patterns of a complex flow system. The positive attributes of the technique are its computational efficiency, its simplicity and compatibility with the information provided by numerical flow solutions, and its ability to handle unstructured grids of arbitrary complexity. Thus the RTTF particle-tracking technique should be a useful addition to the suite of numerical methods used to simulate flow and transport in porous media.

Chapter 5 - Property Sets

5.1 Abstract

In this chapter, we present a summary of the hydrologic property sets provided by LBNL in the site unsaturated-zone flow model (Bodvarsson et al., 1997), along with modifications tested in the present study for their impact on predicted performance. Bodvarsson et al. (1997) provide a logical starting point for numerical simulations of flow and transport in the present study, since models using their parameter sets are based on calibrations to fluid matrix saturation and capillary pressure measured in cores extracted from several wells at the site. In the present study, an initial screening is performed to determine which parameter sets are apt to yield significantly different performance predictions. As part of this study, a code validation check is performed by comparing the results of FEHM and TOUGH2. The results yield important insight regarding grid discretization and use of weighting schemes.

The "model platform" used to test the hydrologic property sets is a one-dimensional flow and transport simulation with stratigraphy and estimated infiltration rate at SD-9. Steady state, dual permeability fluid flow fields were generated for each property set, after which particle tracking transport simulations of ^{99}Tc and ^{237}Np from the repository to the water table were computed. The transport metrics used to identify which property sets yield unique or distinctively different results are the first arrival time of radionuclide at the water table, the relative amount of radionuclide arriving early, the degree of sorptive delay of ^{237}Np to zeolitic tuffs. In addition, the relative flow split between the fractures and matrix in the zeolitic tuffs is used as a hydrologic metric for judging the property sets. In all cases, fracture-dominated flow in the TSw transitions to almost completely matrix flow in the vitric Calico Hills. Then, at the transition from the vitric to zeolitic tuff, significant fracture flow through the zeolitic tuffs is predicted for every parameter set. The permeability of the zeolitic tuffs is too low to support the 3.6 mm/y infiltration in the matrix, thus resulting in the fracture flow, which will be even more pronounced in high flux areas within the

potential repository area. There are subtle differences from one parameter set to the next which we identify and use as the criteria for selecting sets for the multi-dimensional simulations.

Because of the essential similarities among the various property sets and the possibility that the parameter estimation yielded results that only sample a portion of the range of uncertainty of the actual system, we also propose variations to the calibrated property sets. These variations include higher permeability for the zeolitic rocks, full connection-area weighting for the nonwelded units, and an adjusted property set of one Bodvarsson et al. (1997) derived to improve the Calico Hills fluid saturation using the three-dimensional model. In all, ten property sets are tested in the screening process, and five are chosen for continued testing in subsequent two- and three-dimensional flow and transport simulations.

5.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al, 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 5-1.

Table 5-1. QA Status of Data used in this Chapter				
REFERENCE	Q Status	DTN		COMMENT
Bodvarsson et. al., 1997 "The Site-Scale Unsaturated Zone model of Yucca Mountain, Nevada, For the Viability Assessment, LBL Technical Report LBNL-40378	Q	LB970601233129.001	3, 5, 6, 8, 9, 10	No DTN assigned. Hydrologic property sets computed in this report are used throughout the present study.
Flint et. al., 1996, Conceptual and numerical model of infiltration for the Yucca Mountain Area, Nevada",USGS WRIR MOL 19970409.0087	Q	GS960908312211.003/NA	5, 6, 8, 10, 11, 12	USGS DTN 3GUI623M

5.3 Introduction

The LBNL Site-Scale flow model (Bodvarsson et al., 1997) contains numerous hydrologic parameter sets that are presented as fits to the existing hydrologic data. The correlation of the units to which hydrologic properties are assigned with the geologic framework model is shown in Table 15-1. The primary data used to perform the fits were the matrix saturation and, to a lesser extent, the capillary pressure as measured in core samples from several wells near the potential repository block. The methodology used (Bodvarsson et al., 1997) consisted first of simultaneous numerical inversions of the matrix saturation and water potential data at individual wells assuming local one-dimensional flow. This simplification allowed the system to be solved as a series of one-dimensional problems, thereby reducing the computational burden of the analysis. The infiltration rate at a given well was estimated from the proposed spatially dependent infiltration map of Flint et al. (1996). After this primary analysis was completed, these parameters are incorporated into a full three-dimensional site-scale model to check for consistency with the one-dimensional results. The inclusion in the three-dimensional model allowed for considerations of multidimensional flow effects and for incorporation of pneumatic effects. An example of the need to consider multidimensional flow effects is the presence of perched water, hypothesized to be suggestive of lateral flow. Fracture permeabilities were reduced in the vicinity of known occurrences of perched water to reproduce the location and proposed extent of the perched water bodies (Bodvarsson et al., 1997). In addition, some modification to the CHn nonwelded vitric units was performed to improve the model.

Considerations of the nature of the geochemical data (chloride ion and environmental isotope data) also entered into the choice of model formulation and corresponding hydrologic parameter values obtained. For example, numerous lines of evidence suggest that the PTn unit moderates the fracture-dominated, episodic flow through the overlying TCw unit, converting the fracture flow to predominantly matrix flow. Fabryka-Martin et al. (1996b) showed that using current estimated values of the percolation flux, the travel times to the ESF level are probably

controlled by matrix flow in the PTn, with fracture-dominated flow in the TCw and TSw hydrogeologic units. Non-bomb-pulse ^{36}Cl levels measured in ESF samples are consistent with travel times of about 10 ky to the ESF, values that would result from infiltration rates of about 1 to 10 mm/y. Due to these and other considerations, for the parameter sets in Bodvarsson et al. (1997) we chose fracture-matrix interaction models that reflect our conventional wisdom that flow should be matrix-dominated through the nonwelded PTn and CHn units. This choice is accomplished by assuming a large fracture/matrix connection area term for the nonwelded units and a small value for the welded units. For the models in which the reduction factor for connection area is a constant multiplier, the values are large in the nonwelded and small in the welded units. For models in which the weighting factor depends on the upstream relative permeability (Ho, 1997), the reduction factor is automatically large in the welded units because of the relative permeability multiplier. This model is not used in the nonwelded units in any of the LBNL model parameter sets because of the considerations discussed above.

Clearly, the form of the weighting factor model chosen will potentially influence the transport predictions, because this parameter exerts a large influence on the relative flow fractions in the fractures and matrix. Despite the various data sets employed in the numerical inversions, there is considerable uncertainty in the model parameters due to uncertainties in the conceptual model (fracture/matrix connection area and other conceptualizations), the infiltration rate, and the nonuniqueness of the resulting fits even for a given model conceptualization. The latter uncertainty is evidenced by the different parameter sets generated in Bodvarsson et al. (1997) by numerical inversion when the only difference is the starting guess in the inversion process. This result is a classic case of an undersampled system, resulting in nonunique results even for the same conceptual model. Given these uncertainties, it is necessary to examine the entire set of published parameter values to determine which uncertainties are likely to be important for unsaturated-zone radionuclide migration.

In this chapter, we present results of a series of one-dimensional simulations designed to provide a preliminary screening of the hydrologic parameter sets. Although multidimensional simulations would be more appropriate in some ways, we chose a one-dimensional model domain from the ground surface to the water table because of the simple model domain, rapid computational times, and consistency with the model calibration efforts. The model area chosen is SD-9, one of the wellbores used in the calibration. This well has been chosen for previous studies because of its proximity to the potential repository, estimated infiltration rate similar to the average value for the site, and complete data set and well-characterized hydrogeology. This well was also selected by performance assessment (PA) analysts as the one-dimensional numerical “testbed” to compare the hydrologic predictions of the FEHM and TOUGH2 (Pruess, 1991) computer codes. The LBNL hydrogeologic column was used to develop the models, with geologic picks provided by Cliff Ho of Sandia National Laboratories (Ho, pers. comm., 1997), who also ran the TOUGH2 simulations used in the model comparison presented in the next section. For the FEHM simulations, a one-dimensional numerical grid was developed with each hydrogeologic unit divided into approximately 10 grid points to provide a detailed description of the hydrology and transport.

The analyses that follow are divided into two sections:

- A detailed comparison of the flow field and transport characteristics of the one-dimensional column using the TOUGH2 and FEHM codes;
- A comparison of the behavior of the one-dimensional system with respect to flow and transport for the various hydrologic property sets published by Bodvarsson et al. (1997).

5.4 Comparison of TOUGH2 and FEHM Codes

The validity of a flow and transport model in part depends on the correct numerical implementation of the governing differential equations. The YMP has three main computer codes for simulating flow and transport in the unsaturated zone: FEHM, TOUGH2, and NUFT (Nitao,

1995). Analyses of flow and transport under present-day and anticipated hydrologic conditions have been performed with both FEHM and TOUGH2. Both codes have been tested and approved for use by the YMP for carrying out flow and transport simulations. Nevertheless, the need for a comparison between the codes stems from the complexity of the numerical simulations for which the codes are used: there are no analytical solutions to compare the results of the codes for the complex case of flow in unsaturated, dual-permeability media. The confidence that can be placed on the results of either code will be increased if it can be shown that they provide nearly identical results for a test problem of moderate complexity.

In principle, a full three-dimensional model comparison effort could be undertaken. A comparison of three-dimensional models was performed by Robinson et al. (1995), using results provided by LBNL in their 1995 version of their unsaturated-zone flow model. In that comparison, we found, by electronically overlaying the results of the two models, that the fluid saturations throughout the model domain were quite similar to one another, with significant differences occurring only at faults. The hydrostratigraphy provided by LBNL at that time did not include faults, even though their numerical simulations included faults with distinct hydrologic properties. Away from faults, the comparisons were close.

In the present study, we decided that it is more useful to construct a simpler model domain so that the results can be compared more precisely. Though the previously described comparison was successful, the ambiguities of the hydrostratigraphy of the full three-dimensional system and the different approaches of the two modeling groups in model building and grid generating makes detailed comparisons very difficult to carry out. Given the success of the previous effort and the short time frame available we decided that a one-dimensional, dual-permeability comparison was more practical. Previous dual-permeability comparisons of the code have also been successful, as evidenced by the inclusion of such a comparison in the FEHM suite of V&V test problems run whenever code changes are made (Dash et al., 1996a, b). The goal of the effort described in this section was to build on previous model results by incorporating a conceptual model with newly developed fracture/matrix interaction terms and to include transport of radionuclides from the

repository horizon to the water table. For transport, the FEHM particle-tracking module was used (Chapter 4). For the FEHM flow and transport simulation test, a steady-state flow field was computed and transport simulations were carried out, all with FEHM. For the test of FEHM transport simulation using a TOUGH2 flow field, the flow field and other hydrologic results generated with TOUGH2 were read into FEHM, along with connectivity information for the one-dimensional model domain, and FEHM's particle-tracking model was again used for transport. This exercise fulfilled a subsidiary goal of developing a capability requested by PA analysts to read a TOUGH2-generated steady-state flow field into FEHM for transport calculations. Because FEHM is used for transport in both cases, the comparison is not a flow and transport model comparison. It is best viewed as a check of the *hydrologic* models of the two codes, with radionuclides used as numerical tracers for comparing the flow fields. Since radionuclide transport through a dual-permeability medium depends greatly on the flow rates and volumetric water contents of the two media, transport predictions should provide a stringent test of the two codes.

The hydrologic parameter sets developed in Bodvarsson et al. (1997) are summarized in the next section. For this comparison, the model parameter set 6541 was used, with the upstream saturation weighting model for fracture/matrix interaction in the nonwelded units and the upstream relative permeability weighting for the welded units. This model required that the weighting schemes for determining the fracture-matrix coupling be implemented into FEHM. Both the upstream relative permeability weighting and the upstream saturation weighting models have now been incorporated into FEHM and the flow and transport results presented here use these weighting schemes. In FEHM, the weighting factor for the saturation model is S^N , where N is a power of 1 or greater. In this study, $N=1$ is used in the saturation weighting model for the nonwelded units, and the relative permeability weighting is used in the welded units, as specified in the inversion to obtain the parameter sets.

5.4.1 Flow Results

Figure 5-1 shows the computed matrix saturations using FEHM, along with the TOUGH2-

Figure 5-1. Comparison of computed matrix and fracture saturations for the FEHM and TOUGH2 simulations.

computed saturation profile. The very close agreement in the computed matrix saturation profiles using the two codes is evidence of similar implementation of the flow models in the two codes. The breaks in saturation profiles and the corresponding jumps in saturation levels are reproduced using FEHM to a high degree of similarity to the TOUGH2 solution. There are slight differences at the interfaces of some hydrologic units that are explored further below so that they may be more fully understood. However, based on this comparison of matrix saturations (the primary data set used in

the inversions), FEHM simulations will maintain the calibration achieved using the TOUGH2 code as long as the same hydrologic parameter sets are used.

There are other model results such as fracture saturation and relative flow rate through the fractures and matrix that are useful to compare for consistency even though they are solely computed quantities for which no direct comparison to data is possible. Note that these differences are irrelevant to the question of whether the models are calibrated, because we have already demonstrated the equivalence of the models with regard to matrix saturations. Nonetheless, agreement of the codes to this level will provide more confidence in the numerical implementation of both codes, and thus is desirable. Figure 5-1 also shows the computed fracture saturations of the two codes. Over most of the model, the agreement is excellent, with two exceptions. First, at interfaces between units in which the flow is transitioning between fracture and matrix flow, the FEHM simulations predict higher fracture saturations. This result could be caused by slight differences in the implementation of the fracture-matrix weighting schemes or by subtle differences in the weighting schemes within a continuum. These possibilities are described further below.

Second, the fracture saturation throughout the Calico Hills and underlying units are different in the two models. The importance of this difference is seen more easily in Figure 5-2, a plot of normalized flux in the fractures and matrix for the two codes. The fracture saturations and, more importantly, the relative fracture fluxes are different throughout the Calico Hills unit. This difference will give rise to different performance of the unsaturated-zone (UZ) system with respect to radionuclide transport and, thus, is important to understand. The relative fluxes in this region of the model are shown on an expanded scale in Figure 5-3. In addition to differences in the eventual flow splits throughout the CHn, the transition from matrix-dominated to a more evenly divided flux distribution in the zeolitic zone occurs more sharply in the FEHM simulation than in the TOUGH2 run. There was a difference in the thickness of the first vitric Calico Hills unit due to the method used to assign properties to this unit in the TOUGH2 simulation. To check whether this difference accounts for the discrepancy in the models, a FEHM simulation was performed with a 4-m-thick

Figure 5-2. Normalized flux of water in the fracture and matrix continua: FEHM and TOUGH2 simulations.

vitric layer (as opposed to 10 m). The FEHM solution with the 4-m-thick vitric unit had a slightly different behavior (not shown in the figure), but the difference does not change the flow split in the underlying unit. Another possibility that was tested is that the differences are due to grid resolution effects. Rerunning the FEHM calculation on a coarse grid (not shown in the figures) ruled out this explanation as well. Further work was performed to determine the nature of this discrepancy, but transport simulations are presented first to demonstrate the extent to which such differences are likely to matter for radionuclide migration.

5.4.2 Transport result

A comparison of the water table breakthrough curves for the two sets of simulations is shown for ^{99}Tc in Figure 5-4. The two transport simulations are for all intents and purposes equivalent. Both

Figure 5-3. Normalized flux of water in the fracture and matrix continua: FEHM and TOUGH2 simulations on a scale expanded to examine the CHn units. Also included is an FEHM simulation with a 4 m thick vitric CHn layer, instead of the original 10 m thick layer.

exhibit bimodal behavior caused by the flow split in the Calico Hills units between the fractures and matrix, and the breakthrough times and peak values for the two models agree closely. There is slightly more mass reaching the water table in the first peak for the FEHM model due to the higher predicted fracture flow fraction in the Calico Hills. The arrival of the second peak is somewhat retarded relative to the TOUGH2 simulation, a consequence of the slower flow rate through the matrix for the FEHM calculation. Nonetheless, the agreement is quite close. The transport results for ^{237}Np are shown in Figure 5-5. The transport parameters were identical with those for ^{99}Tc except for an assumed K_d of 2.5 cc/g for ^{237}Np in the zeolitic units. The greater flow fraction in the frac-

Figure 5-4. Breakthrough of ^{99}Tc at the water table for the one-dimensional SD-9 model: FEHM and TOUGH2 generated flow fields, FEHM particle tracking for transport.

tures for FEHM is accentuated for ^{237}Np , resulting in an earlier first peak and higher peak value. Once again, a check using a coarser grid showed that grid resolution was not the cause of the differences in the transport results. The cause of the differences at early times is the more gradual transition in the flux distribution at the vitric/zeolitic interface in the TOUGH2 simulation. Flow and transport occurs completely within the matrix of the first zeolitic gridblock, and retardation (with an effective retardation factor of about 20) delays the arrival of the earliest arriving ^{237}Np by roughly 10,000 years. Sorption in this gridblock is the reason the agreement for the early arriving radionuclide is quite good for ^{99}Tc but less so for ^{237}Np . This effect is demonstrated by the second TOUGH2 transport curve in Figure 5-5, obtained by changing the sorption coefficient to 0 in this

Figure 5-5. Breakthrough of ^{237}Np at the water table for the one-dimensional SD-9 model: FEHM and TOUGH2 generated flow fields, FEHM particle tracking for transport.

gridblock (all other properties remained the same). The agreement of this simulation with the FEHM run is quite satisfactory, suggesting that the details of the transition between layers of contrasting hydrologic and transport properties may be important.

The key to these differences lies in the way the codes compute the transition between units of contrasting hydrologic parameters. Specifically, the results are different for the transition of flow from matrix to fracture at the CHn vitric/zeolitic interface, and this fact causes the difference in Np transport. FEHM uses a standard method of harmonically weighted saturated permeabilities, with upstream weighting of the fluid properties and relative permeabilities. The TOUGH2 simulations were run using upstream weighting of all properties, including the saturated permeability (Cliff Ho, pers. comm., 1997), called “full-upstream weighting” in the discussion

below. At the interface in question, full-upstream weighting causes flow to transition more slowly from matrix-dominated to a relatively even split. Therefore, in the TOUGH2 flow field, there is matrix flow in the uppermost zeolitic grid block, which, given the size of this block in the TOUGH2 grid, caused a substantial travel time increase of the first peak. For reference, $K_d=2.5$ slows the transport velocity by about a factor of 20, so if this occurs in just one reasonably large grid block, minimum travel times increase dramatically.

To further confirm this explanation, an additional FEHM simulation was carried out with full upstream weighting to make it comparable to the TOUGH2 simulation. In Figure 5-5, ^{237}Np transport with the FEHM-generated flow field but with upstream weighted permeability (solid red curve) comes much more in line with the original TOUGH2 result. The full-upstream weighting scheme used in the TOUGH2 simulation smooths the transition, causing more matrix flow in the upper zeolitic rock, thus causing longer travel times.

The question to be addressed now is whether the remaining differences are large enough to be important to integrated performance of the site. Our claim is that such differences, though perhaps important to demonstrate a complete understanding of the model results, are not sufficiently great to warrant much more study, given the variability of property values across the site, the differences in unit thicknesses, and the uncertainty in parameter values. Therefore, from the standpoint of maintaining a "calibrated" model that agrees quantitatively with the TOUGH2 results, we conclude that the two codes predict essentially the same results and that FEHM simulations carried out in the present study should be valid with respect to their fidelity in reproducing the results of the LBNL site-scale flow model.

5.5 Screening of Hydrologic Property Sets

In this section, we use the one-dimensional SD-9 model to examine the range of behavior expected due to different calibrated hydrologic property sets. Earlier, we discussed the philosophy of performing this screening on a representative one-dimensional model such as the SD-9

stratigraphy. Before presenting the results, we list the various caveats that must be placed on the analysis. First, the calculations assume that, for screening purposes, SD-9 is representative in terms of stratigraphy and infiltration rate. The stratigraphy contains most of the units specified in the full three-dimensional system, except that the column is almost fully zeolitic from the top of the Calico Hills to the water table. Thus, a unit that plays a key role in transitioning flow from fracture-dominated to matrix-dominated, the vitric Calico Hills, is present but of limited thickness (about 10 m). The infiltration rate obtained from an aerial average of the values near the well from the infiltration map of Flint (1996), is 3.6 mm/y, representative of the average rate near the repository but not nearly as high as values predicted below the crest of Yucca Mountain.

Finally, the calibrated parameter sets all were obtained from one-dimensional model inversions. Although the one-dimensional assumption in the calculations of the present study is consistent with the calibration method, the results may not be entirely representative of the three-dimensional system. Perched water is one example of a feature of the site not captured in one-dimensional calculations. Lateral flow also cannot be reproduced in one dimension. The ramifications of either perching or lateral flow is that the net percolation flux below such features is over-estimated in one-dimensional models, as these models force the entire surface percolation flux through the entire column. The LBNL hydrologic studies handled this limitation by adjusting the fracture properties in three-dimensional models to obtain perching conditions. For our screening studies we assume that the one-dimensional system provides an adequate representation for the purpose of evaluating multiple parameter sets and eliminating those sets that produce behavior that duplicates the results of an equivalent parameter set. Given the limitations just discussed, conservatively retain more parameter sets than would be suggested based only on the one-dimensional results, factoring in the probable outcome if multidimensional screening analyses had been performed.

The parameter sets obtained from the LBNL studies (Bodvarsson et al., 1997) are summarized in Table 5-2. The naming convention in the present study is to use the number of the table in the LBNL report to identify the parameter set. When there is more than one parameter set

Property Sets

in a table, the letters **a** and **b** are added to the name. A brief description of the assumptions underlying each parameter set is also provided in Table 5-2. For additional details on the

Table 5-2. Summary of hydrologic parameter sets from Bodvarsson et al. (1997) (shaded rows indicate parameter set not simulated in the present study)

Parameter Set	Location in Bodvarsson et al. (1997)	Description
6412a	Chapter 6, Table 6.4.1-2, first column	DKM, 3.6 mm/y, const. reduction factor, init. Fm=0.667
6412b	Chapter 6, Table 6.4.1-2, second column	DKM, 3.6 mm/y, const. reduction factor, init. Fm=0.492
6412c	Chapter 19, Table 19.6.4-1	LBL re-calibration of CHnv units using the 3D model and matching saturations in SD-7 and SD-12
6412d	Based on 6541c	6412c with 5x increase in zeolite permeability.
6511	Appendix A-6, Table 6.5.1-1	DKM, 3.6 mm/y, const. reduction factor, init. Fm=0.492, init. red. factor=0.05
6521	Appendix A-6, Table 6.5.2-1	DKM, 18 mm/y, const. reduction factor, init. Fm=0.492
6522	Appendix A-6, Table 6.5.2-2	DKM, 0.72 mm/y, const. reduction factor, init. Fm=0.492
6531a	Appendix A-6, Table 6.5.3-1, first column	DKM, 3.6 mm/y, upstream S reduction factor, init. Fm=0.492
6531b	Appendix A-6, Table 6.5.3-1, second column	DKM, 3.6 mm/y, upstream S reduction factor, init. Fm=0.667
6541	Appendix A-6, Table 6.5.4-1	DKM, 3.6 mm/y, upstream S reduction factor (nonwelded), upstream RLP reduction factor (welded)
6541b	Based on Appendix A-6, Table 6.5.4-1	6541 with no reduction factor in PTn only. Applied in Chapter 6

assumptions employed for each parameter set and a detailed discussion of the inversion process, see Bodvarsson et al. (1997).

Of the thirteen parameter sets listed in Table 5-2, two were not examined with an SD-9 simulation in the present study. Parameter set 6542 was determined to be very similar to set 6541, the only difference being the treatment of the fifth PTn layer. This difference is not expected to influence radionuclide transport below the potential repository horizon, because the differences are restricted mainly to the uppermost TSw layers (Bodvarsson et al., 1997). Therefore, parameter set 6542 was not studied here. Parameter set 6551, an ECM inversion result, was not included in the present study because dual permeability is used for the radionuclide transport predictions.

The results of the hydrologic simulations for the eight cases are shown in Figures 5-6 through 5-13. Figure a of each figure contains the computed matrix and fracture saturations, with the field measurements of matrix saturation shown for comparison. Even though the individual simulations themselves differ somewhat from one another, the results show that the computed matrix saturations agree well with the data, as they should for calibrated data sets. Differences from one parameter set to the next result from different fits to the data and from the fact that the LBNL inversions involved several wellbores, so that the fit to any one well may be better or worse, depending on the model assumptions. This test provides a visual confirmation that the various parameter sets have been correctly implemented.

Figure b of each figure shows the simulated fracture and matrix flow fractions (which sum to unity) throughout the one-dimensional column. The general pattern of the flow fractions is similar regardless of the parameter set chosen:

- fracture-dominated flow through the TCw units;
- a rapid transition to matrix-dominated flow in the PTn;
- rapid transition back to fracture dominated (at least 90% fracture flow) in the TSw units (two of the parameter sets (6531b and 6541) have an intermediate flow fraction value in the uppermost TSw unit but higher fracture flow values in the underlying TSw units);

Figure 5-6. Case 6412a hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

Figure 5-7. Case 6412b hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

Figure 5-8. Case 6511 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

Figure 5-9. Case 6521 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

Figure 5-10.Case 6522 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

Figure 5-11.Case 6531a hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

Figure 5-12.Case 6531b hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

Figure 5-13.Case 6541 hydrologic simulation results for SD-9. a) matrix and fracture saturations, along with field data points; b) matrix and fracture fluxes; c) matrix and fracture fluxes, expanded scale including the vitric/zeolitic interface.

- matrix-dominated flow in the first vitric Calico Hills unit (shown on an expanded scale in the **c** figures); and
- differing levels of fracture flow in the zeolitic rocks comprising the remainder of the column, ranging from about 20% to 90% fracture flow, depending on the parameter set.

It is the details of flow and transport beneath the potential repository that will control radionuclide migration to the water table. Therefore, differences in flow fractions in the zeolitic rocks and the transition from matrix-dominated to an intermediate case at the vitric/zeolitic interface will exert an important influence on transport of radionuclides. We now present transport results for ^{99}Tc and ^{237}Np for an instantaneous release of one mole of radionuclide from the potential repository horizon at time 0. An important distinction for interpreting these breakthrough curves is that the actual release rate from the near-field environment (NFE) is expected to be much more drawn out over time. These curves are, in effect, generic breakthrough curves that define the distribution of residence times in the unsaturated zone. Given a steady-state flow assumption, the curves are a Green's function for the unsaturated-zone transport system, and can be converted to a combined NFE/UZ breakthrough curve by convolution of the Green's function with a NFE source term model. The convolution process and results are described in Chapters 8 and 10.

Figures 5-14 through 5-21 show the one-dimensional SD-9 transport results for the eight parameter sets (the **a** figures are ^{99}Tc , assuming no sorption, and the **b** figures are ^{237}Np , assuming sorption with a K_d value of 2.5 cc/g in the matrix of the zeolitic tuffs and 0 elsewhere). We now provide a brief discussion of each model result, using the first case to elaborate on the relevant flow and transport processes controlling the results.

5.5.1 Case 6412a

The ^{99}Tc response curve for this case is characteristic of many of the response curves for the various parameter sets. The response curve is bimodal, with early breakthrough occurring due to fracture transport through the zeolitic rocks between the vitric CHn and the water table. The minimum residence time of about 400 years represents matrix flow and transport through this vitric unit.

Figure 5-14.Case 6412a transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Figure 5-15.Case 6412b transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Figure 5-16.Case 6511 transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Figure 5-17.Case 6521 transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Figure 5-18.Case 6522 transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Figure 5-19.Case 6531a transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Figure 5-20.Case 6531b transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Figure 5-21. Case 6541 transport results for the one-dimensional SD-9 model. a) ^{99}Tc ; b) ^{237}Np . One mole of radionuclide is released at the repository horizon at time = 0.

Therefore, site-scale models must accurately include this unit to capture the early breakthrough of radionuclides properly. The spikes in the breakthrough curve are caused by several factors, which work in combination to yield the discrete pulses of arrivals at the water table. These factors include the instantaneous injection of radionuclide, the assumption of no hydrodynamic dispersion, and the one-dimensional flow field. To elaborate on the last point, all particles travel through the same locations in space, and different transit pathways arise only due to transitions from one continuum to the other at different locations. Thus the responses are spiky, but this behavior will not be present in multidimensional simulations with dispersion or reasonable NFE source-term models. The second peak in the ^{99}Tc breakthrough curve is due to matrix flow in the zeolitic rocks, which results in travel times on the order of 10,000 years. The relative areas under these two peaks are proportional to the relative fluxes through the two continua in the zeolitic rocks (shown to be roughly 60% matrix flow in Figure 5-6).

The ^{237}Np breakthrough curve for this case (Figure 5-14b) also exhibits bimodal behavior, but the later-arriving peak is delayed by more than a factor of 10, a consequence of ^{237}Np sorption on the zeolitic rocks. The first peak is virtually undelayed in time relative to the nonsorbing ^{99}Tc , despite the fact that the matrix diffusion model allows for sorption on the zeolitic rocks even for the case of fracture flow. Matrix diffusion over these time scales is relatively inefficient at delaying the arrival times of a transporting solute. Nevertheless, matrix diffusion and subsequent sorption does spread the arrival time distribution and, thus, lower the peak value for this first peak. Note that because the two radionuclides are simulated on a one-mole basis, the peak values are directly comparable. The lower peak value for ^{237}Np is a consequence of matrix diffusion and sorption, even though the effect is not sufficiently large to delay the peak time. Whether this process is important to overall performance of the unsaturated-zone transport system must await multidimensional analyses incorporating a realistic NFE source term model.

5.5.2 Case 6412b

Transport for this case is very similar to that of case 6412a, which is not surprising given that the hydrologic parameters differ only as a consequence of a different starting condition for the LBNL parameter estimation simulation. The slight differences are due to the somewhat greater matrix flow through the zeolitic rocks for 6412b, resulting in a delay in the arrival time for the first peak and an earlier arrival for the second (greater matrix flux decreases the travel time for the matrix-flow peak).

Because the LBNL 6412 parameter set is one of the primary data sets provided to the Project, we decided to retain at least one of these cases for multidimensional transport simulations. However, this analysis shows that there is no reason to use both sets, given the minor differences in the results. In Bodvarsson et al. (1997) LBNL performed an additional analysis in which they adjusted the parameters in the CHn and underlying units to obtain a better fit to the data in the three-dimensional site-scale flow model. They used 6412b as the starting point for this analysis. Therefore, to allow us to also test that adjusted parameter set along with the base-case set, we select case 6412b to carry forth in the multidimensional transport model calculations.

5.5.3 Case 6511

This parameter set is based on the same fracture/matrix interaction model as in the 6412 cases, but the initial guess of the reduction factor was different in the LBNL parameter estimation simulation. Figure 5-16 shows the transport results which exhibit similar behavior to that of case 6412b, though some differences can be seen. Examining the flow fractions in the CHn units for this case (Figure 5-8 b and c), we note that the transition to the eventual matrix flow fraction of 0.65 occurs more gradually. This effect could perhaps give rise to a qualitatively different response under different conditions, such as during transient flow or a different infiltration rate scenario. However, given the similarity of the model results and conceptualization of the fracture/matrix interaction process to that of 6412b, we decided not to use case 6511 in further simulations.

5.5.4 Case 6521

This case assumed an infiltration rate of 18 mm/y, five times that of the base case. The consequence to transport is straightforward and well known. Figure 5-17 shows an earlier arrival time for both peaks (and for both radionuclides) due to the faster flow velocities. A corollary to faster travel times is higher peak values, compared to the previous cases considered. Ordinarily, one would also expect the fraction of mass traveling through the fractures of the zeolitic rocks to increase, giving rise to a higher first peak in the bimodal breakthrough curves. This effect is true only to a limited extent in this simulation. The reason is illustrated in the flow fraction plot in Figure 5-9b, which shows a gradual decline in matrix flux with depth down to a 50% value. This parameter set apparently required a higher value for zeolite matrix permeability to obtain a good fit to the data, thus allowing half the fluid to travel through the matrix even at 18 mm/y. This effect must have been controlled by fits at wells other than SD-9, for the following reason. The model and the data both show almost complete saturation throughout the zeolitic rocks. The rationale for an inversion code raising the matrix permeability would be to achieve a better fit, but at complete saturation of both data and model, there would be no point in the code changing the permeability. Therefore, the higher permeability must be controlled by data at other wells.

Regarding the use of this parameter set in multidimensional simulations, we have chosen a somewhat different approach to studying the influence of infiltration rate sensitivity. In Section 8.7, we show that varying the infiltration rate for the same parameter set results in rather small changes in matrix saturation. Therefore, we use the parameter sets developed at the base infiltration rate as the starting point for simulations but assume that increases in the infiltration rate by a factor of 3 has an insignificant influence on the quality of the model calibration. Therefore, we choose not to carry forth this parameter set to the multidimensional simulations, instead assuming that hydrologic parameter sets developed for the base infiltration map apply to moderately higher or lower infiltration rates.

5.5.5 Case 6522

This case achieved the fit at 0.7 mm/y at SD-9, five times lower than the base infiltration value. Figure 5-18 shows the transport results. Travel times are much greater for both ^{99}Tc and ^{237}Np , and the relative mass arriving in the first peak is reduced relative to the base infiltration case, in keeping with the predicted flow split in the zeolitic rocks of about 80% matrix flow (Figure 5-10).

In contrast to Case 6521 (the 5X higher infiltration case), we decided to incorporate this parameter set into our multidimensional calculations. One result of our environmental isotope and water chemistry modeling studies is that a somewhat lower infiltration rate than the base-case infiltration map is consistent with the chemical data sets. This result implies that a parameter set developed under a lower infiltration rate scenario should be studied more thoroughly. We therefore use this parameter set in the sensitivity study of infiltration rate in multiple dimensions, assuming it to be an appropriate alternative at lower flux values.

5.5.6 Case 6531a

This case, and case 6531b, use the upstream saturation weighting scheme in all layers. For the parameter set provided for this model, matrix flow decreased to about 10% of the flux within the first several meters of the zeolitic CHn (Figure 5-11). The impact on the breakthrough curves for the radionuclides is dramatic (Figure 5-19). Most of the radionuclide mass travels through the CHn and underlying units in fractures. The late-arriving mass of ^{99}Tc that does travel through the zeolitic matrix has a travel time in excess of 50 ky due to the low flux through the matrix, but this constitutes only a small portion of the released mass. ^{237}Np has a similar breakthrough-curve shape, with somewhat lower values caused by matrix diffusion and sorption to zeolites. Nevertheless, like ^{99}Tc , ^{237}Np breaks through early (less than 1 ky) and remains at about this level for about 30 ky. This level response is caused by transition from matrix flow to mostly fracture flow in a ten-meter interval, rather than abruptly. This result means that a variety of different transition depths exist, and each of these transport pathways has a different residence time in the zeolitic matrix before transitioning to fracture flow.

Since this fracture/matrix interaction model is different than the others studied, and because the one-dimensional breakthrough curve appears to have a different character than the others, we include one of the 6531 cases in the multidimensional analyses. We chose the following case, 6531b, but in fact, there is little of importance to separate the two cases, and thus, either one could have been selected.

5.5.7 Case 6531b

The breakthrough curves for this case are shown in Figure 5-20. At first glance, they differ from that of case 6531a in terms of the high initial spike of radionuclide. However, spikes of short duration play little role in the determination of breakthrough at the water table or the accessible environment for actual release scenarios (as opposed to instantaneous releases). Therefore, the two cases actually are similar, and 6531b was selected for further study. One remaining issue related to these cases is the reason for the dramatically lower flux through the matrix of the zeolites than for the other cases studied. It is not clear why this should be so, given that for the first CHn vitric and zeolite layers, the saturation weighting scheme is the same as for case 6541 (below), yet that case yielded more matrix flow through the zeolites. In cases 6531a and b, the inversion appears to have settled on property values that yield slightly lower matrix saturations in the zeolites at the expense of more fracture flow. This point is potentially important to performance, and thus, the details of the inversion to obtain property values for this case should be examined in greater detail.

5.5.8 Case 6541

The breakthrough curves for this case (Figure 5-21) were discussed previously, as this parameter set was used for comparison to TOUGH2 simulations. Because this form of weighting scheme (upstream saturation in the nonwelded units and upstream relative permeability in the welded units) was deemed appropriate for PA analyses, we select it also for further study. However, we note that the behavior of the flow fractions and radionuclide breakthrough curves are similar to several other cases presented above.

Property Sets

The results of the screening analyses are summarized in Table 5-3, which is essentially a **Table 5-3. Parameter sets to include in multi-dimensional transport simulations. Parameter sets carried forward are unshaded in the table.**

Parameter Set	Location in Bodvarsson et al. (1997)	Description
6412b	Chapter 6, Table 6.4.1-2, second column	DKM, 3.6 mm/y, const. reduction factor, init. $F_m=0.492$
6412c	Same as 6412b except as specified in Table 5-2	DKM, 3.6 mm/y, const. reduction factor, init. $F_m=0.492$, CHn props. revised in 3D runs
6522	Appendix A-6, Table 6.5.2-2	DKM, 0.72 mm/y, const. reduction factor, init. $F_m=0.492$
6531b	Appendix A-6, Table 6.5.3-1, second column	DKM, 3.6 mm/y, upstream S reduction factor, init. $F_m=0.667$
6541	Appendix A-6, Table 6.5.4-1	DKM, 3.6 mm/y, upstream S reduction factor (nonwelded), upstream RLP reduction factor (welded)
6541b	Based on Appendix A-6, Table 6.5.4-1	Same as 6541 except no upstream S reduction factor is used in PTn only. This set is used in Chapter 6

repeat of Table 5-2, except that the shading denotes that a parameter set is not being carried forward to the multidimensional simulations. One additional parameter set, Case 6412c, is included and identified for further study even though there were no one-dimensional simulations. This case is

equivalent to case 6412b in all units except in several of the CHn and underlying units. Bodvarsson et al. (1997) performed a three-dimensional simulation using case 6412c and found that better matches in three dimensions could be obtained by changing these properties somewhat. Because the units in question are below the potential repository, this adjustment could be important for transport, and thus we include them as a separate case to examine the effects.

5.6 Conclusions

This chapter summarizes the hydrologic property sets provided by LBNL in the site unsaturated-zone flow model (Bodvarsson et al., 1997), along with modifications tested in the present study for their impact on predicted performance. Since it is not feasible to complete transport sensitivity analyses in multiple dimensions for every parameter set in the LBNL milestone, as well as test variations of these as sensitivity studies, an initial screening was performed to determine which parameter sets are yield significantly different performance predictions.

Steady state, dual permeability fluid flow fields were generated for each property set, after which particle tracking transport simulations of ^{99}Tc and ^{237}Np from the repository to the water table were computed. Because transport of radionuclides escaping the repository is the subject of the study, the emphasis is on predicted flow and transport below the repository horizon. Parameter sets may differ in their predictions of hydrologic conditions above the repository; these possible differences are addressed in Chapter 6.

Despite the potentially large spectrum of possible behaviors for different hydrologic property sets, the most striking result of this screening study is the similarity of the flow and transport results. In all cases, fracture-dominated flow in the TSw transitions to almost completely matrix flow in the vitric Calico Hills unit at SD-9. Then, at the transition from the vitric to zeolitic tuff, significant fracture flow through the zeolitic tuffs is predicted for every parameter set, even at the 3.6 mm/y infiltration rate assigned at SD-9. The permeability of the zeolitic tuffs is too low to

support the 3.6 mm/y infiltration in the matrix, thus resulting in the fracture flow, which will be even more pronounced in high flux areas within the potential repository area. There are subtle differences from one parameter set to the next which are identified and used as the criteria for selecting sets for the multi-dimensional simulations.

This study also served as a code validation check by comparing the results of FEHM and TOUGH2 for the one-dimensional column. Steady state flow fields were generated with each code, and the flow fields were used by FEHM to compute particle tracking breakthrough curves. The saturation profiles agreed closely. Transport of ^{99}Tc differed somewhat due to a slight, predicted difference in the flow split between fractures and matrix in the two cases. More importantly, the transitions from matrix flow to partial fracture flow at the vitric/zeolitic interface is predicted to occur abruptly above the interface in FEHM and more gradually within the first zeolitic grid block in TOUGH2. Matrix flow within the zeolite in TOUGH2 gave rise to significantly delayed first arrival of ^{237}Np at the water table. We attribute the TOUGH2 behavior to the combination of upstream weighting of the absolute permeability and the large size of the first zeolitic grid block; FEHM calculations using this same weighting scheme yielded results that approached those of the TOUGH2 run. We therefore recommend that in mountain-scale flow and transport simulations, the absolute permeability be harmonically weighted, and/or the grid be more finely discretized near interfaces of contrasting flow and transport properties.

Chapter 6 - Environmental Isotopes and The Hydrologic System

6.1 Abstract

In this chapter, we examine the role of studies coupling environmental isotope data and numerical modeling in identifying flow and transport processes as well as constraining the property sets used to model them. Starting with various hydrologic property sets from LBNL, which are all based on fits matching matrix fluid saturation and capillary pressure (Bodvarsson et al, 1997), we perform a series of transport studies with one-, two-, and three-dimensional transport models and examine how well the isotopic data are matched. The most significant finding in this study is that minor changes in hydrologic parameters lead to different flow distributions in the PTn matrix and fractures which, in turn, affect the predictions of whether fast paths exist between the surface and potential repository horizon. Since multiple property sets were developed by Bodvarsson et al. (1997), reducing the uncertainty in property set choice based on environmental isotope transport observations and modeling may have significant implications on repository design and performance modeling. For example, if fracture flow in the PTn is ubiquitous, then repository design may need to account for periodic high fluxes resulting from transient infiltration events that are not damped in the PTn matrix. Also, sustained PTn fracture flow leads to a greater incidence of predicted bomb-pulse $^{36}\text{Cl}/\text{Cl}$ ratios in the TSw and CHn, even away from fault zones. With current material property estimates, episodic infiltration events (precipitation-related infiltration events lasting on the order of days) are more likely to be damped in regions of thicker PTn. The direct correlation of the PTn's damping potential to the magnitude of the infiltration rate is not straightforward. Fully assessing whether episodic infiltration is a significant concern for repository design and performance assessment requires a better understanding of the hydrologic properties and fracture flow potential of the PTn.

The environmental isotope data coupled with transport modeling also serve to evaluate alternative conceptual models and the infiltration estimates to which the parameters are calibrated. One alternative conceptual model suggests that all elevated $^{36}\text{Cl}/\text{Cl}$ ratios found in the ESF represent very recent water. The current LBNL hydrologic properties tend to support this model more so than previous property sets in that simulated fracture flow in the PTn is more readily initiated and sustained. With the current infiltration map and the corresponding material properties, the simulations in this section show that more rapid transport from ground surface to the ESF should be occurring along the southern portion of the ESF than in the North. To the contrary, however, the data indicate that more early arrivals occur in the North, particularly in faulted regions. In fact there is virtually no evidence for fast travel to the ESF beyond station 50+00.

6.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al, 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 6-1. The GEOMESH/X3D grid generation tool kit, (Gable et al., 1995, Gable et al., 1996, Trease et al., 1996) is non-Q with respect to YMP and Los Alamos quality assurance requirements. However, version control software tracking procedures identical to those used for the Q code FEHM are followed. GEOMESH/X3D utilizes well established procedures to insure robust mesh generation results and to insure traceability of changes to the software. A suite of test problems has been developed and all new releases are verified against the test suite.

Table 6-1. QA Status of Data used in this Chapter			
REFERENCE	Q Status	DTN	COMMENT

[illegible]

Flint et al., 1996, Conceptual and numerical model of infiltration for the Yucca Mountain Area, Nevada", USGS WRIR MOL 19970409.0087	Q	GS960908312211.003/NA	USGS DTN 3GUI623M
Levy et al., 1997, "Investigations of Structural Controls and Mineralogic Associations of Chlorine-36 fast pathways in the ESF,. Yucca Mountain Project milestone report SP2301M4	Q	LASL831222AQ97.001	
Yang et al., 1992, "Flow and Transport Through Unsaturated Rock - Data From Two Test Holes, Yucca Mountain, Nevada, proc. Third International Conference on High-Level Radioactive Waste Management, Las Vegas, NV.	Q		
Yang, et al., 1996, Interpretations of Chemical and Isotopic Data From Boreholes in the Unsaturated Zone at Yucca Mountain, Nevada, USGS Water-Resources Investigations Report WRIR 96-4058	Q		
Yang et al., 1997, Hydrochemical Investigations and Geochemical Modeling in Characterizing the Unsaturated Zone at Yucca Mountain, Nevada, (In Preparation).	non-Q	Indeterminate	Not Available Yet

6.3 Introduction

Direct measurement of unsaturated zone percolation flux is difficult if not impossible, especially at depths greater than a few meters. To date, no such measurement has been made at Yucca Mountain. Inferring unsaturated zone flux from physical measurements of saturation and water potential is also extremely difficult, especially in multiporosity systems such as fractured volcanic tuff. Likewise, dissolved solute migration processes have not been measured in the unsaturated zone at Yucca Mountain, although the Project is now planning an unsaturated-zone transport test. In saturated-zone studies, tracer tests are often used to help quantify physical and chemical attributes of the system as well as the groundwater flux. However, performing liquid

phase tracer tests in fractured, unsaturated porous media is not a simple task, especially at the scales of interest in the site-scale characterization program of Yucca Mountain. Fortunately, there are naturally occurring environmental isotopes that, if considered properly, provide important insights into the system characteristics. Further, they capture a time scale that cannot be addressed with conventional tracer tests. An additional utility in evaluation of environmental isotopic tracer tests is the man-made signal associated predominantly with the atmospheric testing of more than 70 nuclear weapons in the Pacific Proving Grounds between 1952 and 1958 (Glasstone, 1962). The $^{36}\text{Cl}/\text{Cl}$ ratio in fallout in 1957 was about 400 times greater than the present ratio of 500×10^{-15} , thus providing the “bomb-pulse”. Significantly elevated ratios of $^{36}\text{Cl}/\text{Cl}$, ^{14}C , ^{99}Tc , and tritium associated with the atmospheric weapons testing are generally referred to as bomb-pulse signals and their presence in subsurface samples indicate travel from ground surface to the sample location in less than 50 years.

Recent efforts to characterize the hydrologic properties of the various units at Yucca Mountain have involved laboratory measurements of porosity, permeability, and characteristic curves by Flint (1996), air permeability tests (Bodvarsson et al., 1996; Le Cain and Patterson 1996), and numerical modeling to match measured saturations and gas phase pressure response (Bodvarsson et al., 1996, 1997). Although extremely beneficial in characterizing matrix material properties, these studies do not capture the “bulk response” of the system, particularly the combined influence of fracture-matrix interactions on transport processes in the liquid phase. For that reason, the environmental tracer studies described here serve to validate and enhance our conceptual and operational model of flow and transport in Yucca Mountain.

6.3.1 Implications

This study demonstrates that different hydrologic properties which yield similar simulations of saturation may lead to significantly different predictions of transport rates. Simulated transport times for solutes through the PTn range from several tens of years to tens of thousand years, with differences in the percent of solute traveling along the fastest pathways

depending on which parameter set is used. Environmental isotopes and conservative ions help identify both whether fast pathways exist and the age of sampled porewaters away from fast pathways, thus providing a useful tool for narrowing the field of possible parameter sets for hydrologic characterization, particularly when the difference in parameter sets is the method of representing fracture-matrix interactions. In conjunction with transport modeling, these data also prove useful in evaluating infiltration estimates.

6.4 Background

6.4.1 Previous Chlorine-36 Studies at Yucca Mountain and Their Implication for Flow and Transport Modeling

Investigations of the significance of ^{36}Cl data for identifying appropriate numerical model formulations for flow and transport processes in the unsaturated zone at Yucca Mountain were initially reported by Robinson et al. (1995). That report showed that only with the dual permeability model formulation could the observed rapid transport of bomb-pulse ^{36}Cl to the PTn be simulated. With the equivalent continuum model, all of the bomb-pulse stayed in the upper TCw. Also, Robinson et al. (1995) demonstrated that infiltration rates closer to those estimated by Flint et al. (1996), which average about 5 mm/year over the site-scale model domain, could be used with the dual permeability model and still simulate matrix saturations consistent with those Wittwer et al. (1995) achieved using an equivalent continuum formulation and infiltration rates on the order of 0.1 mm/yr.

The first implications of the ^{36}Cl studies coupling Exploratory Studies Facility (ESF) sample analysis with flow and transport modeling were reported by Fabryka-Martin et al. (1996a). These studies were later elaborated on by Fabryka-Martin et al. (1996b) and Robinson et al. (1996). These reports demonstrate that fracture flow must play a role in the Yucca Mountain UZ flow and transport system and that infiltration rates close to those estimated by Flint et al. (1996), at least over the ESF, are required to generate the $^{36}\text{Cl}/\text{Cl}$ signals that have been measured

in the ESF (both bomb pulse and non-bomb pulse). The most recent site-scale hydrologic model of Bodvarsson et al. (1997) primarily adopts the estimates of Flint et al. (1996) and a dual permeability model in the parameter calibration activity.

Bodvarsson et al. (1996) and Bodvarsson et al. (1997) investigate, through numerical simulation, some issues associated with bomb-pulse isotopes measured at the ESF. These simulations focus only on "fast path processes." The 1996 report examines processes in a single fault using a continuum model to specify different properties in the fault zone and host rock bounding the fault zone. The 1997 report examines transport from the surface to the ESF over the last 50 years using a two-dimensional, dual-permeability model that extends along the main drift of the ESF and which has modified PTn material properties at certain faults and hypothesized faults (undetected at the surface or at depth). Although the modification of fracture properties is not described in the 1997 report, we assume it is similar to, or based on, the approach of Robinson et al. (1996), where fracture permeabilities in fault zones are increased consistent with either more fractures or larger fracture apertures that result from tectonic activity. In transport simulations, Bodvarsson et al. (1997) use an input signal to their model which incorporates bomb-pulse $^{36}\text{Cl}/\text{Cl}$ ratios that range up to 1.6×10^{-10} . The simulated $^{36}\text{Cl}/\text{Cl}$ ratios at depth are identified as bomb-pulse in some locations, but they are less than present-day background (which is approximately 500×10^{-15}). However, these simulations show, consistent with previous simulations by Robinson et al. (1996), is that in zones with properties modified to represent faulted conditions, solutes entering the mountain at the surface can arrive at the ESF in less than 50 years. Issues regarding different numerical model formulations and constraints when simulating transport in a dual permeability framework are discussed in Robinson et al. (1997).

A final concern regarding processes affecting bomb-pulse arrivals at the ESF is how significant transients on the scale of days are. Robinson et al. (1996) used a synthetic 100 year record of daily infiltration for which the infiltration rate on any given day was either 0.0 or 0.06 mm/day with an average of 30 days per year having non zero infiltration. The annual average was

2 mm/yr and the simulations showed no significant difference from the uniform infiltration rate simulations. In the simulations of transient effects by Bodvarsson et al. (1997), a pulse of 3.5 mm/day for 7 days is applied once and then the effects are examined over the next 10 years. The large pulse apparently has an effect on the maximum penetration depth, especially in faults. In this study, we further investigate the role and significance of transients by varying the length, magnitude, and frequency of recurring transients in and away from faults.

Significant Previous Findings

Fabryka-Martin et al. (1996b) and Robinson et al. (1996) reported on a detailed study in which base-case PTn properties were modified in accordance with the increased fracturing that is expected to occur as a result of tectonic activity. In those studies, using base-case hydrologic properties from the Project's most recent analyses at the time (Bodvarsson et al., 1996; Robinson et al., 1995), we found that infiltration rates in the range of 1 to 5 mm/yr led to simulations consistent both with bomb-pulse signals found near fault zones and non-bomb pulse signals found away from fault zones in the ESF. Specifically, in regions away from faults where the PTn was not modified, simulated travel times of chloride to the ESF were on the order of 5,000 to 20,000 years and yielded $^{36}\text{Cl}/\text{Cl}$ ratios in the $600 \text{ to } 900 \times 10^{-15}$ range, just as have been measured in ESF samples. With infiltration rates considerably less than 1 mm/yr, travel times often exceeded 100,000 years and decay of ^{36}Cl resulted in predictions of ratios less than the measured values. With infiltration rates much greater than 5 mm/yr, all simulated travel times to the ESF are less than 10,000 years, and the resulting signal reflects a Holocene source with a nearly constant $^{36}\text{Cl}/\text{Cl}$ ratio of 500×10^{-15} .

When the PTn fracture permeability is increased in simulations to reflect increased fracturing in fault zones, bomb-pulse arrivals at the ESF are predicted for infiltration rates greater than 1 mm/yr. Thus, the simulations in and away from fault zones in the ESF yield consistent results for the same range of infiltration rates.

6.4.2 Infiltration Studies

Estimates of infiltration rates at Yucca Mountain are performed by invoking a variety of measurements and models associated with surface and near surface processes. Flint and Flint (1994) estimated the spatial distribution of steady state infiltration rate in matrix material based on the relative permeability of the rock underlying the alluvium. Hudson and Flint (1996) developed a model correlating precipitation, alluvial depth, and measurements of water-content changes in shallow boreholes. At the shallow boreholes, Hudson and Flint used neutron logs to estimate water content. Changes in water content are assumed to give conservative estimate (low) of the volume of water that percolated deeper over the time period between neutron logging. Also Hevesi and Flint (1996) used an independent approach to estimate the spatial distribution of steady state infiltration rates. Their model is based on the assumption that basin wide recharge can be estimated for many basins in Nevada. Through geostatistical modeling of precipitation distribution and improved average annual precipitation estimates, they redistribute the infiltration rates over the extended Yucca Mountain study area, thereby providing another map of steady-state infiltration rates.

Each of these methods tends to estimate infiltration rates that are higher by at least an order of magnitude than the potential repository horizon percolation rates which have been used in the past for flow models (e.g. Wittwer et al., 1995, Bodvarsson et al., 1996). Previous studies tended to base percolation rates on the maximum hydrologic capacity of the matrix material. In such cases, however, the role of fracture flow was generally neglected. The role of fracture flow also may complicate the estimates of Flint et al. (1996) since the rock unit below the alluvium is often of low matrix permeability but highly fractured. However, between the surface and the sampling locations described below, water must move through alternating layers of fractured and less fractured volcanic tuff, thus dampening episodic events and giving rise to a constant percolation flux in the system.

6.4.3 Present Study Summary

The environmental isotope analyses have played a significant role in guiding both the flow and transport modeling efforts. In this chapter, we review briefly the findings from previous studies and then evaluate them through analyses using our site-scale flow and transport model which integrates with ISM geologic model (Clayton et al., 1996), parameter sets from the site-scale flow model (Bodvarsson et al., 1997), an numerous field and laboratory observations.

The recent multiple hydrologic property sets of LBNL (Bodvarsson et al., 1997) all represent calibration to measured saturation in boreholes. There are at least seven different sets of properties, each having some unique difference from the other sets. One set is based on calibrating the matrix-fracture interaction term as a scaled value of the connection area. Another uses simulated fracture saturation as a scaling term and then calibrates all other parameters. Another uses fracture saturation in the nonwelded tuff and fracture relative permeability in the welded tuff as the scaling function of the fracture-matrix interaction. Variations on each of these include the results obtained by starting the saturation calibration with different initial guesses of certain model parameters.

Bodvarsson et al. (1997) place no apparent preference on any of their calibrated parameter sets. Likewise, it is not immediately clear, a priori, whether significant differences in flow and transport simulation would occur when different sets are used. As many of the sets were evaluated in Chapter 5, here we analyze only a few of the parameter sets which with respect to their implication for simulation of environmental isotope distributions in Yucca Mountain. As with our previous studies (Robinson et al., 1996) we use one-, two- and three-dimensional models in the analysis. The one-dimensional models are used for examining in detail process-level intricacies. Here, the flow distribution between matrix and fracture can be examined as well the effect on vertical flow and transport of varying material properties. The two- and three-dimensional models provide platforms for studying the role of site-scale conditions such as dipping stratigraphy of variable thickness and extent, and spatially distributed infiltration.

In this chapter, we first describe the hydrogeologic setting in which the environmental isotope studies are conducted. Then we present the data which were collected and the interpretations which can be drawn from the data alone. Finally, we present corroborating numerical simulations and demonstrate how their results help reduce uncertainty in the interpretation of the data as well as improve our understanding of the transport environment of the unsaturated zone at Yucca Mountain.

6.5 Hydrogeologic Setting

6.5.1 Stratigraphy

The stratigraphic structure of the unsaturated zone of Yucca Mountain is comprised of alternating layers of welded and nonwelded volcanic tuffs which are tilted, uplifted, fractured, and faulted. Characterization of these units and their subunits has advanced as more information from boreholes, the ESF, and other studies has become available. The classification of Scott and Bonk (1984) provided a framework around which hydrostratigraphic models were developed (e.g. Wittwer et al.; 1995; Robinson et al., 1996). Recently, Buesch et al. (1996) developed a classification scheme which enhances continuity between the various efforts in the site characterization program. Flint (1996) has provided the necessary link between the hydrologic characterization of the various units and the geologic structure of the system.

Figure 6-1 shows a schematic of the hydrostratigraphy encountered from the surface, through the potential repository horizon, and down to the water table. Below any surface alluvium, which varies in depth from 0 to 50 meters, the first major hydrologic unit encountered is the Tiva Canyon welded tuff (TCw). The saturated matrix permeability of the TCw is relatively low (on the order of 10^{-17} m^2) but the high degree of fracturing leads to a much higher bulk permeability. Below the TCw is the Paintbrush nonwelded tuff (PTn). The matrix permeability of the PTn is generally several orders of magnitude greater than the TCw, but the degree of fracturing is substantially less. The Topopah Spring welded tuff (TSw), below the PTn, is the horizon of the

potential repository and where most of the ESF samples have been collected for ^{36}Cl analysis. The TSw is much thicker than the TCw and PTn together and is broken down into subunits depending on the density of lithophysal cavities and amount of devitrification that has occurred as a secondary process. Like the TCw, the matrix permeabilities in the TSw are low but the bulk permeability is substantially greater due to a high degree of fracturing. Finally, between the TSw and the water table are the Calico Hills, Prow Pass, and Bullfrog nonwelded tuffs. These are often grouped together and referred to as the Calico Hills nonwelded unit (CHn). A unique characteristic of the CHn is the degree of alteration to zeolitic material that has occurred. The zeolites are of particular interest for radionuclide transport sensitivity studies since they have been shown to be the primary mineral onto which Neptunium and Uranium sorb significantly. The zeolite distribution and the role zeolites play in the geologic barrier system between the potential repository and the water table are discussed in Chapters 8 and 9 of this report.

6.5.2 Conceptual model of flow and transport

Base-Case

Our conceptual model between ground surface and the potential repository horizon, which has evolved from the conceptual hydrologic model of Montazer and Wilson (1984), can be described with the following components: Infiltration into the fractured TCw, sustained flow in the fractures of the TCw, matrix dominated flow in the PTn, and then fracture dominated flow again throughout most of the TSw. The flow in the CHn is complex due to the occurrence and distribution of vitric and zeolitic rocks. The degree of alteration to zeolites ranges from none to fully altered. Thus, the reduction in matrix permeability resulting from alteration must span a range from the high permeability vitric units to that almost as low as the welded units of the TCw and TSw. Where the matrix permeability is less than the percolation flux, flow must either occur in fractures or laterally bypass those low permeability zones. Due to the uncertainty in the permeability distribution, the degree of fracturing, and the actual percolation flux, the exact distribution and pathway of flow can not be determined. However, in Chapter 12 we demonstrate a geostatistical

Figure 6-1. Conceptual model of hydrologic pathways at Yucca Mountain.

approach to address this uncertainty and attempt to identify the role that these distributed properties play.

Infiltration enters the system following episodic precipitation events and, that which does not go back to the atmosphere via evapotranspiration, moves quickly through the TCw. Once the moisture enters the PTn, however, the downward velocity is slowed as the water moves almost entirely in the high porosity matrix material. Due to the transition from fracture flow to matrix flow, the episodicity of the percolation is damped and, by the bottom of the PTn, the flow rate resembles an average annual percolation rate. At the PTn/ TSw boundary, there is a permeability barrier due to the highly contrasting matrix permeabilities. However, the increased fracture permeability of the TSw can support the flow coming out of the PTn matrix. One question regarding this interface is how much flow may be diverted laterally, rather than continuing vertically from the PTn matrix to the TSw fractures (the TSw matrix by itself can not support

anywhere near the flow rate of the PTn matrix). Due to the low matrix permeability, water which continues to percolate vertically flows mostly in the fractures of the TSw and is, hence, characterized with much greater velocities than the matrix flow of the PTn.

Clearly, in our conceptual model of this complex layered system, the PTn plays an important role in arresting the velocity of downward moving water as well as dampening out the episodicity of infiltration events. If this behavior can be understood and validated, then a better understanding of the actual flow rate through the system can be inferred as well. Conversely, if episodic pulses of flow are not damped by the PTn and persist at the potential repository horizon, then dripping into the drifts is more likely and the near field performance may be reduced. $^{36}\text{Cl}/\text{Cl}$ ratios measured in the ESF provide, in conjunction with numerical modeling studies, significant insight into the relationship between flow rates, flow distribution between fractures and matrix material, and travel times through the system of welded and nonwelded tuff.

Transport of radionuclides, both naturally occurring and from a potential repository source, are considered in this system. The naturally occurring radionuclides, ^{36}Cl , tends to migrate with the water as a conservative tracer. Inventory radionuclides like ^{237}Np may move at a slower rate than a nonreactive tracer due to sorption in the zeolites of the CHn. Thus, the naturally occurring radionuclides serve to identify flow paths and travel times of water while the entire geochemical system must be considered to determine the actual rate ^{237}Np will move at.

Fault Zones

The identification of bomb-pulse $^{36}\text{Cl}/\text{Cl}$ ratios in the ESF indicates that not all vertical flow velocities are arrested in the PTn. Otherwise, we could never get any chloride from the surface to such depth in the necessary 50 years or less to show bomb pulse. Levy et al. (1997) extend on 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. One of these key issues that we have focussed on relates to the transport of chlorine-36 through fault zones that cut the PTn.

6.6 Chlorine-36 Study

6.6.1 Background

The construction of the ESF provided an opportunity to develop a spatial distribution of $^{36}\text{Cl}/\text{Cl}$ ratios within the same horizon for assessment of the hydrologic processes in the Yucca Mountain subsurface. An additional purpose for conducting the chlorine-36 sampling project in the ESF was to identify if and where any fast paths for water movement from the surface, through the PTn, and to depth could occur. If bomb-pulse $^{36}\text{Cl}/\text{Cl}$ ratios were detected, that would provide an indication of travel times of several hundred meters in less than 50 years. The ability to collect samples for $^{36}\text{Cl}/\text{Cl}$ ratio analysis in the ESF marked a unique opportunity which helped to overcome previous problems with analyses from borehole samples. First, the ESF provides the opportunity to look at and characterize the rock and geologic structure at and in the vicinity of samples. This contrasts greatly with the blind samples that were obtained previously from boreholes. Only in the ESF can the samples be classified as coming from intact rock, breccias, or fracture zones, and their precise location relative to the faults be mapped. This classification has proven extremely valuable in the correlation of structural features with $^{36}\text{Cl}/\text{Cl}$ ratios measured for these ESF samples (Levy et al, 1997). Second, borehole samples could only be collected as ground-up ream-bit cuttings. This increased the error in analysis as a result of dead chloride released during the pulverizing of the rock. The ESF samples are not pulverized and, hence, represent the ambient system more accurately.

6.6.2 Chlorine-36 input

Chlorine-36 was originally picked as a tracer for very old groundwater because it has a half life of 301,000 years. Early estimates of infiltration flux into Yucca Mountain were less than 0.1

mm/yr, which could lead to pore water ages on the order of a half a million to a million years. For such ages, carbon-14, with a half life of 5700 years, was thought to be inappropriate due to its maximum age resolution of about 50,000 years. Further, whereas ^{14}C can travel both in the liquid and vapor (CO_2) phase and participate actively in mineral precipitation and dissolution, ^{36}Cl remains in the liquid phase. Finally, the bomb-pulse signature of ^{36}Cl is more distinct and differentiable from present and past background signals than that of ^{14}C (Fabryka-Martin et al., 1996c).

Rather than indicating hundreds of thousands of years of decay as initially expected, about half of the $^{36}\text{Cl}/\text{Cl}$ ratios measured in the ESF are actually greater than present day background. These analyses became available at the same time as the revised, higher surface infiltration rate estimates (Flint et al., 1996) were being developed. One hypothesis was that the elevated ratios reflected a widespread presence of bomb-pulse in the deep subsurface. However, an alternative interpretation was that most of the elevated signals reflected past variations in background levels. Fossil packrat midden samples dated both with ^{14}C and ^{36}Cl indicate that, since the end of the Pleistocene (the last 10,000 years ago), the $^{36}\text{Cl}/\text{Cl}$ ratio has been relatively constant at the present day level. However, in the later part of the Pleistocene, a time period in which packrat midden samples can still be dated with ^{14}C , the $^{36}\text{Cl}/\text{Cl}$ ratio was as much as two and a half times greater than the present day background. Coupling the packrat midden samples with a theoretical model of tropospheric ^{36}Cl production based on Earth's geomagnetic flux variation, Fabryka-Martin et al. (1996b) and Plummer et al. (1996) developed the reconstructed $^{36}\text{Cl}/\text{Cl}$ function shown in Figure 6-2 which is used as an input to the numerical simulations in this study. Those reports give the full detail of the signal reconstruction process and the data sources.

The infiltration estimates of Flint et al. (1996) are generally on the order of 1 to 10 mm/yr above the ESF sample locations. Such infiltration rates correspond to groundwater ages on the order of 1 to 20 thousand years. These travel times are entirely consistent with the sample ratios

Figure 6-2. Reconstructed $^{36}\text{Cl}/\text{Cl}$ ratio in infiltrating water (Fabryka-Martin et al., 1996c, Plummer et al., 1996), a) last 500,000 years, b) last 50,000 years, c) last 50 years.

found in the ESF. The radioactive decay dating of ^{36}Cl loses its applicability for such short travel times, but the elevated ratio at the end of the Pleistocene provides a valuable tracer for groundwater age dating.

6.6.3 ESF Data

The ESF data collection involves sampling systematically every 200 m as well as additional feature based samples from fractures, breccias. Levy et al. (1997) report isotopic and mineralogic sample analyses up to station 68 (6.8 km into a tunnel from the entrance). Sweetkind et al. (1997) report on the additional samples that have been collected since the report of Levy et al. was published. However, the data from Levy et al. (1997) are plotted later in Figure 6-21 in conjunction with the simulated $^{36}\text{Cl}/\text{Cl}$ ratios obtained with the 3D transport model. Additional interpretation of the data can be found in Sweetkind et al. (1997).

6.7 Flow and Transport Model

6.7.1 Numerical Flow and Transport Model: FEHM

The numerical simulations are conducted using FEHM (Finite Element Heat and Mass transfer) (Zyvoloski et al., 1992). The mathematical models and numerical methods are summarized in Zyvoloski et al. (1992, 1995a,b), Robinson et al. (1996) and Viswanathan (1996). Solute transport in FEHM can either be performed with the particle tracking module or the finite-element reactive transport module. Both are coupled to the flow module so transport in steady state or transient flow conditions can be simulated. The particle tracker accommodates advection, dispersion, matrix diffusion, linear sorption, and radioactive decay. With the finite-element solver, the additional process of multi-species reactions such as complexation, competitive sorption, and precipitation/dissolution can be simulated (Viswanathan et al., 1997; Chapter 11 of this study). Here, since only aqueous chloride transport is considered, the particle tracker is adequate and substantially more efficient than the finite element solver. A large part of the efficiency associated with the particle tracker results from its ability to yield accurate solutions, even when fluid velocities are large. In fractured tuff, the velocities in fractures are substantially greater than in the rock matrix and resolution of solute movement would require very small time steps or unrealistic

grid resolution with standard finite-element or finite-difference techniques. However, the particle tracker is based entirely on the already computed flux in and out of each element and its computational limitation is only dependent on the number of particles used (see Chapter 4 of this report). With modern computer workstations, transport of millions of particles can be simulated efficiently. For example, a typical run using 1 million particles to simulate $^{36}\text{Cl}/\text{Cl}$ distributions on the 143,000 node three-dimensional grid for a time period spanning over 1 million years takes less than 10 minutes.

6.7.2 Transient Signals of Isotopic Sources

Among its many unique features, one utility in the FEHM particle tracker that proves invaluable in the simulation of environmental isotope transport and distribution is the capability to efficiently integrate a transient source function. All of the particles are introduced into the system instantaneously and the simulation proceeds until all particles leave the system. A record is kept at each node of when the particles went through that node. That time indicates the travel time to the node and hence the time before present that a packet of water would have had to enter the system to arrive at the node now. Hundreds or thousands of particles pass each node, giving a distribution of travel times (or ages of water) at each node. With each age, a unique $^{36}\text{Cl}/\text{Cl}$ ratio is obtained from the function plotted in Figure 6-2. These ages are integrated at each node to give a distribution of $^{36}\text{Cl}/\text{Cl}$ signals throughout the system. This capability enables the simulation of long-time processes (e.g. 10^4 - 10^5 years) as well as the bomb-pulse time scales (50 years) within the same simulation. With the standard finite-element or finite-difference method, the time of simulation would be substantially greater due to the small time steps required to accurately model transport in a dual permeability formulation. One limitation of the particle tracking method is that, to simulate the transient source function for environmental isotopes, steady-state flow is required due to the integration process. However, for simply simulating breakthrough curves at one location for a pulse of particles injected into the system at the top, transient flow can be invoked.

6.8 Simulated Systems

6.8.1 Overview

One-, two-, and three-dimensional numerical models are used to interpret the $^{36}\text{Cl}/\text{Cl}$ observations and the general hydrologic behavior of the system above the potential repository to aid in the prediction of migration rates of other radionuclides from the potential repository to the accessible environment. A schematic of the locations of the various models used in this chapter as well as in the radionuclide transport chapters is given in Figure 6-3. Based on the findings of Robinson et al. (1995, 1996), all simulations of environmental isotope transport and distribution are performed with the dual-permeability model formulation. Only with the dual-permeability formulation can rapid transport in fractures be simulated in conjunction with slow transport in matrix material as well as the coupling between the two media.

Two- and three-dimensional models are used to study the site-scale behavior including the effects of dipping stratigraphy of variable thickness and extent, spatially varying infiltration rates, and spatially distributed material attributes. The one-dimensional models are used to examine specific fracture-matrix interactions associated with flow and transport in and away from fault zones. Also, transport under transient and steady-state flow conditions is evaluated with the one-dimensional models to assess whether time scales associated with episodic infiltration events (days) need to be addressed in the model when simulations of tens of thousands of years in two and three dimensions are required for radionuclide migration studies.

The layers in the numerical models are derived from the three-dimensional, site-scale stratigraphic framework model (Zelinski and Clayton, 1996). The GEOMESH software package (Gable et al., 1996) is used to extract the region of interest from the geologic model and generate unstructured finite element grids. Material properties and parameters for the different layers are obtained from Bodvarsson et al (1997) and listed in Appendix 1.

Figure 6-3. Plan view of three-dimensional model domain with locations of ESF, ESF stations, two-dimensional east-west model transect, north-south transect, and one-dimensional vertical column studies.

6.9 Carbon-14 Study

6.9.1 Background

Groundwater ages in countless aquifer systems have been estimated based on the residual carbon-14 activity in the porewater. Prior to the advent of atmospheric testing of nuclear devices, ^{14}C was present in the atmosphere solely due to its production by cosmic rays bombarding nitrogen nuclei. By definition, the specific activity of ^{14}C in carbon that was in equilibrium with the atmosphere prior to the nuclear age is 100 percent modern carbon (pmc), corresponding to about 14 disintegrations per minute (dpm) per gram of carbon. Numerous lines of evidence indicate that this initial activity has been relatively constant for the past several tens of thousands of years, with fluctuations of about 10%. Copious quantities of ^{14}C were produced in the atmosphere by nuclear testing in the 1950s and 1960s, and have also been released as a by-product of the reprocessing of spent nuclear fuel. The peak activity occurred in 1963, at 200 pmc. The decrease since then has been due to exchange of ^{14}C for nonradioactive carbon in the biosphere and hydrosphere, principally the ocean. In contrast with the present-day atmospheric signals for tritium and ^{36}Cl , the modern ^{14}C value is still elevated about 20% above natural background due to the comparatively long residence time of carbon in the atmosphere.

In groundwater investigations, ^{14}C measurements are made on inorganic carbon extracted from water and gas samples. For Yucca Mountain samples, typical volumes have been 100 mL for water, and 2 L for gas samples. In the absence of a bomb-pulse signal, an apparent age can be calculated based on the radioactive decay equation, using the ^{14}C half-life of 5730 yr. However, this unadjusted age represents a maximum, and the true age can be substantially less. Because carbon-bearing species distribute among the solid, liquid and gas phases in unsaturated geologic systems, many geochemical reactions occur that dilute the initial signal. These include re-equilibration with soil-gas CO_2 , which has lower ^{14}C activity than that in the atmosphere; release from or isotopic exchange with carbonate and gypsum minerals along the flow path; oxidation of

dissolved organic matter; and re-equilibration with CO_2 in the vapor phase in the unsaturated zone. Numerous geochemical models are available to correct the measured signal, and generally require the user to specify solution chemistry and carbonate speciation, relevant mineral-water reactions that control the carbonate chemistry of the system, stable carbon isotope signals and ^{14}C activities of the various carbon sources and sinks in the system, and prevailing water temperature.

Due to the reactions that ^{14}C can be involved in, it is not a conservative liquid-phase environmental tracer as ^{36}Cl is. However, it does provide an additional constraining data set for porewater ages in the unsaturated zone at Yucca Mountain. Measurements of liquid and gas phase ^{14}C have been reported by Yang et al. (1992, 1996, 1997). These data are primarily from nonwelded tuffs and from perched water as the sample size necessary for analysis can not be readily extracted from the welded tuff. The ^{14}C analyses on the perched water samples indicate apparent ages ranging between 3,000 and 11,000 years (Yang et al., 1996) and the apparent ages in porewater of the Calico Hills range between 800 and 5500 years. Corrections to these estimates to account for caliche dissolution are discussed in Chapter 7 of this study.

6.9.2 Carbon-14 Data

Yang et al. (1997) present ^{14}C analyses for perched water samples and porewater samples. Of particular interest are the data in the units at or below the potential repository. As porewater collection has not been achieved in the welded TSw units, the ^{14}C analyses have been performed on perched water samples found at the TSw-CHn interface and in the vitric CHn units. Table 6-2 shows the apparent ages of these samples, which are discussed in greater detail in Chapter 7. The apparent ages for the porewater samples, which occur below the perched water, are younger than the apparent ages in the perched water.

Table 6-2. Apparent ^{14}C Ages from Yang et al. (1997)

Location	Well	Apparent Age Range (years)
CHn Porewater	UZ-14	400-3500
	NRG7	4100
	SD7	2700-4100
	SD9	400-1600
	SD12	2900-2500 800-2300
	UZ16	800-5350
Perched Water	UZ14	8000-12000
	NRG7	2000-6000

A possible explanation for the apparent discrepancy

One possible explanation for this seemingly contradictory indication is that under past, wetter climates, lateral flow occurred at the basal vitrophere because the entire flux could not be transmitted vertically through that low permeability unit. Under present drier climate conditions, most of the flow, even from the zones of highest infiltration, moves vertically through the basal vitrophere and does not divert laterally until it reaches the zeolites. This scenario would lead to younger water overlain by older water as the source of the perched water is primarily attributed to the previous climate. The sample locations such as UZ-14 tend to be in locations with relatively low infiltration. Thus the contribution of modern water to the perched body through the vertical pathway is small relative that from zones of higher percolation flux. However, it would serve to bring the apparent age up some do to mixing of water of different ages. This conceptual model is described more fully in Chapter 7.

6.9.3 Carbon-14 Input

Carbon-14 is input into the transport model at a uniform concentration of 100 percent

Figure 6-4. Carbon-14 decay function resulting from a 5730 year half-life.

modern carbon (pmc). As it migrates into the subsurface, decay is accounted for according to the curve shown in Figure 6-34. As with the ^{36}Cl particle tracking model, this is performed efficiently and without the time-step constraints of conventional finite-element or finite-difference methods. However, the simulations are limited to steady state flow fields.

6.10 Chlorine-36 Modeling: Two- and Three- Dimensional Studies

6.10.1 Model Description

Domain and Boundary Conditions

Migration and distribution of environmental isotopes chlorine-36 and carbon-14 are simulated on the east-west cross section two-dimensional model and in the site-scale three-dimensional mode. The two-dimensional model domain is located at Nevada State Plane Northing coordinate 232,100 m and crosses several faults between Easting coordinates 168,275 m and 174,175 m. This domain (Figure 6-5) includes the Solitario Canyon Fault, the Ghost Dance Fault, and the Bow Ridge Fault. The infiltration map of Flint et al. (1996) (the I map) and a scaled version

of that map where all rates are scaled by one third (the I/3 map) are used as the top boundary condition for the two-dimensional model (see Table 6-4). These maps are shown in Figure 6-6 for the two-dimensional cross section. The aerial domain of the three-dimensional model is shown on top of the I infiltration map of Flint et al. (1996) in Figure 6-7. The extent of faults and general structure of the three-dimensional model are described in Chapter 3 of this report. A saturation boundary condition is prescribed at the water table for all models.

Material Properties

The hydrologic properties used in this study either are taken directly from or are based on the hydrologic properties of Bodvarsson et al. (1997), which were developed through calibration to measured matrix saturation. Table 6-3 lists the property sets are used in the environmental isotope models and the source and modifications associated with each set. The input parameters to FEHM are provided in Appendix 1 of this report. The major difference in property sets is the modification of set 6541a to 6541b, in which the fracture/matrix reduction factors in the PTn were eliminated.

Table 6-3. Parameter Sets for Environmental Isotope Simulations

Property Set Name for This Report	LBNL set on which properties are based	Location in LBNL report of property set	Modification of LBNL properties
6541a	Table 6.5.4-1	A-28	None
6541b	Table 6.5.4-1	A-28	No reduction factor for fracture/matrix interaction in PTn
6541ft	Table 6.5.4-1	A-28	PTn fracture permeability increased by one order of magnitude
6541perched	Table 6.5.4-1	A-28	Basal vitrophere fracture permeability reduced and fault zone fracture permeability reduced.
6412b	Table 6.4.1-2b	6-12	None

Table 6-4. Infiltration Maps Used in Environmental Isotope Simulations

Infiltration Map Name	Source	Modification
I	Flint et al. (1996)	None
I/3	Flint et al. (1996)	Scaled everywhere by 1/3

Figure 6-5. Finite-element grid for the two-dimensional cross section at Antler Ridge. Northing location in NSP coordinates is 232100 m. Red arrows show easting location for potential repository horizon breakthrough study.

6.10.2 Two-Dimensional Chlorine-36 Simulation Results

The transport of chloride and resulting $^{36}\text{Cl}/\text{Cl}$ ratios throughout the two-dimensional cross section are simulated for a variety of different conditions. The results are evaluated both in terms of the overall resulting distributions and the temporal breakthrough curves at selected locations. Those selected locations are all at elevation 1080 m (approximately the potential repository elevation) and are generally spread out across the cross section every 500 m (see Figure 6-5 for the locations of these points).

The first simulation utilizes the 6541a material set and the resulting matrix $^{36}\text{Cl}/\text{Cl}$ ratios are plotted in Figure 6-8. In the TSw, the simulated values resemble those that are measured in the

Figure 6-6. Infiltration rates extracted from Flint et al. (1996) map along Antler Ridge cross section. I map is directly extracted, I/3 map is the I map scaled by 3. Green arrows show easting location for potential repository horizon breakthrough study.

same horizon in the ESF. That range spans from 500 to 1100×10^{-15} . What is striking about this simulation, however, is that bomb-pulse ratios are simulated ubiquitously at the TSw/CHn interface under the entire potential repository horizon and even somewhat east of the Ghost Dance fault. This means that, for these model parameters, some flow is getting through the entire PTn in less than 50 years. Recall, however, that this parameter set reduces the fracture matrix coupling area by the fracture saturations (see Chapter 5). For this scenario, any water that gets into the fractures, in a dual permeability simulation, will be less likely to transition to matrix flow than when the coupling term is not reduced.

Since these parameter sets represent calibration to saturation (Bodvarsson et al., 1997), we tested the sensitivity of that calibration only in the PTn, the unit that controls the migration rate of ^{36}Cl from the surface to the ESF and potential repository horizon. Figure 6-9 shows the resulting

Figure 6-7. Plan view of three-dimensional domain outline on top of Flint et al. (1996) infiltration map. Also shown for reference are the ESF, the east-west two-dimensional cross section and the north-south transect (used to interpret the three-dimensional model results).

$^{36}\text{Cl}/\text{Cl}$ ratios when parameter set 6541b is used. The only difference from set 6541a is that the PTn fracture/matrix coupling is not scaled by the simulated fracture saturation. The results clearly show that for the modified parameter set, bomb-pulse levels of ^{36}Cl are constrained to being below areas where there is no PTn (e.g. in the west end of the cross section) and near faults. The difference between the two simulations is shown in Figure 6-10. This figure shows that the only significant differences between the two simulations occur in the vicinity of the simulated bomb-pulse signals in either run. For the bulk of the system, the resulting ratios are about the same for these two simulations.

Figure 6-8. Simulated $^{36}\text{Cl}/\text{Cl}$ ratios in Antler Ridge 2-D cross section with 6541a material property set and Flint (1996) infiltration map.

Figure 6-9. Simulated $^{36}\text{Cl}/\text{Cl}$ ratios in Antler Ridge 2-D cross section with 6541b material property set (*no weighting of fracture-matrix connection in PTn*) and Flint (1996) infiltration map. Note the reduced amount of bomb-pulse in the top of the CHn.

Figure 6-10. Difference in simulated $^{36}\text{Cl}/\text{Cl}$ ratios for property sets 6541a and 6541b.

Figure 6-11. Simulated saturations in Antler Ridge 2-D cross section with 6541a material property set and Flint (1996) infiltration map.

Figure 6-12. Simulated saturations in Antler Ridge 2-D cross section with 6541b material property set and Flint (1996) infiltration map.

Figure 6-13. Difference in simulated saturations for property sets 6541a and 6541b.

Is one simulation better than the other? Certainly the first, 6541a, is computed with parameters calibrated to saturations. However, Figures 6-11 and 6-12 show the simulated saturations for the two parameters sets. They look virtually identical. This is confirmed in Figure 6-13 which plots the difference between the two. In the PTn, the only unit for which fracture/matrix coupling was modified, the maximum saturation difference is only 0.05.

Implications

Two parameter sets that yield virtually identical saturation fields lead to very different predictions regarding whether or not bomb-pulse ^{36}Cl penetrates the PTn above the potential repository. As a preliminary investigation into the implication regarding whether transients will be damped out in the PTn, solute breakthrough curves were computed for a set of locations in the model at the potential repository horizon. The assumption LBNL (Bodvarsson et al., 1997) and we have made in the past is that the PTn fully damps the signal and hence episodic events need not be considered in simulations spanning thousands of years. However, if travel times through the PTn are less than 50 years, then we may need to reconsider this assumption.

Breakthrough curves, which also represent the cumulative age distribution of a sample at a location in the model domain, are used to further investigate the differences that result from what appears to be a minor property change. Figure 6-15 shows the travel time breakthrough curves at the potential repository elevation throughout the cross section using the 6541a parameter set. These curves indicate that, for the 6541a set, a significant amount (7-65%) of the water going through the potential repository horizon is less than fifty years old. Moving eastward, the amount of early arrival decreases as the infiltration rate at the surface decreases. In contrast, Figure 6-15 shows that the slight modification in parameter set 6541b leads to substantially less bomb-pulse age water moving all the way through the PTn and to the potential repository elevation in the TSw constituting less than 1% of water for all samples underlying undisturbed PTn. This is far more consistent with the data reported by Levy et al. (1997). Figures 6-17 and 6-18 show the same property set comparison, but with the I/3 map. Substantial early arrivals are still predicted with the

6541a set whereas 6541b eliminates most PTn fracture flow in all locations except directly under the crest and in the Ghost Dance Fault. Note however, that the fault-zone properties in this simulation are specified with low matrix permeability and high fracture permeability.

In the next section, we demonstrate with the three-dimensional model that rapid travel through the PTn is more likely to be simulated in the southern portion of our domain (where this two-dimensional cross section falls) than in the northern portion. A final analysis with the two-dimensional model examines whether the bomb-pulse signals simulated at the top of the CHn with the 6541a parameter set actually resulted from vertical flow or whether lateral flow played a major role. Figure 6-14 shows the pathways in both the matrix and fractures for particles introduced in the system near faults and in faults. For particles injected near faults, there is a small amount of lateral diversion simulated at the top of the CHn, but most particles travel downward. For the particles injected in the fault, there is some tendency to move out of the fault into the high permeability matrix of the CHn, but most particles move downward staying in the fault.

6.10.3 Three-Dimensional Chlorine-36 Simulation Results

Simulations of chloride transport and $^{36}\text{Cl}/\text{Cl}$ ratio distributions are simulated on the 143,000 node site-scale transport model. The domain and boundary conditions for the three-dimensional simulations are described in Section 6.8 and the grids are described in Chapter 3 of this report. Figure 6-19 shows north-south and east-west slices that intersect in the proposed repository block. The bomb-pulse has reached the top of the PTn, where its downward migration velocity has been retarded as the flow partitions from the TCw fractures into the PTn matrix. Figure 6-20 shows the results from the three-dimensional simulation at the ESF horizon. The simulated values in the matrix match those that were measured on ESF samples. Comparing Figure 6-20 and Figure 6-7, one can see the influence that the spatially distributed infiltration map of Flint et al. (1996) has on the simulated matrix $^{36}\text{Cl}/\text{Cl}$ ratios. The lack of infiltration variability over most of

Figure 6-14. Particle pathways from 5 different surface locations. a) pathways in the matrix for locations near faults, b) pathways in the fractures for locations near faults, c) pathways in the matrix for locations in faults, d) pathways in the fractures for locations in faults.

the ESF relative to that over much of the rest of the domain leads to a fairly uniform simulated $^{36}\text{Cl}/\text{Cl}$ ratio.

Figure 6-21 shows the resulting simulated matrix $^{36}\text{Cl}/\text{Cl}$ ratio in the ESF from the three-dimensional model compared with the analyzed $^{36}\text{Cl}/\text{Cl}$ ratios reported by Levy et al. (1997) and Sweetkind et al. (1997). After the ESF falls below the PTn, about 1100 m into the tunnel, the

Figure 6-15. Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541a set. Infiltration rate at surface is the Flint et al. (1996) map. Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m).

Figure 6-16. Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541b set (*no weighting of fracture-matrix connection in PTn*). Infiltration rate at surface is the Flint et al. (1996) map. Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m)

Figure 6-17. Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541 set. Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m).

Figure 6-18. Breakthrough (or cumulative age distribution of a porewater sample) at elevation 1080 m at 10 locations along E-W Antler Ridge transect. Material properties are the 6541b set (*no weighting of fracture-matrix connection in PTn*). Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate Easting location in NSP coordinates (Northing coordinate is 232100 m).

simulated values tend to capture the mean behavior of the observed measurements. The bomb-pulse signals are not simulated as frequently as they occur in the data because only two of the many faults that cross the ESF (the Bow Ridge fault and the Ghost Dance fault) are included in the ISM geologic model (Clayton et al., 1996) and thus in our model. The effect of other faults such as the Sundance fault, the Drillhole Wash fault, and other unnamed faults (see Levy et al, 1997, Figure 1) must therefore be addressed with other means. These other means are discussed in the next section on process modeling in vertical columns.

Figure 6-19. Three-dimensional simulation of $^{36}\text{Cl}/\text{Cl}$ ratios using property set 6541a and infiltration map of Flint et al. (1996).

Figure 6-20. Simulated $^{36}\text{Cl}/\text{Cl}$ ratios at ESF horizon with three-dimensional transport model. Uses material property set 6541a and infiltration map from Flint et al. (1996).

Flow and Transport Pathways

With the three-dimensional model, nonvertical pathways can be identified. Here, we inject a set of particles in Yucca Crest above the potential repository and model the path they take as they move into the system. The plume does not follow one distinct pathline due to the multiporosity system in which a particle has the option of several different paths at any given location. Chapter 4, the detailed description of the particle tracking algorithm, demonstrates how a particle in the matrix may either stay in the matrix or move into the fractures. Likewise a particle in the fracture

Figure 6-21. Simulated $^{36}\text{Cl}/\text{Cl}$ ratio in ESF with 3-D transport model and measured ratios from Levy et al. (1997). Simulations performed using 6541 parameter set and Flint et al. (1996) infiltration map

may either move in the matrix or stay in the fracture. This type of behavior leads to dispersion of an instantaneously injected source in both time and space. Thus in the next section, a set of particles injected instantaneously over the surface yields breakthrough curves at the ESF spanning up to 4 orders of magnitude in time. Here the plume's injection location is (170600,234000) and Figure 6-22 shows the travel paths taken in the matrix. The less intense color in the TSw results because most simulated flow and, hence, particle transport occur in the matrix. After reaching the PTn, there is some spreading as flow partitions into the matrix. After reaching the CHn, there is increased lateral flow simulated due to the permeability contrast between the vitric and zeolitic material. Figure 6-23 shows the pathway in the Fractures for the same simulation. This figure highlights the density of flow in the TCw, TSw, and CHn zeolite fractures. Lower fracture flux densities are shown in the PTn and CHn vitric units. Even less later flow occurs in fractures than in the vitric matrix material of the CHn.

Figure 6-22.Solute travel path in the matrix from the top of Yucca Crest into the UZ system.

Figure 6-23.Solute travel path in the fractures from the top of Yucca Crest into the UZ system.

Three-Dimensional Model Breakthrough Curve Analysis

As part of the parameter set comparison that was already described for the two-dimensional model, three-dimensional simulations using the 6541b property set, which was modified to eliminate the reduction factor for fracture/matrix coupling in the PTn, is compared with three-dimensional simulations using the 6541a set. The comparison involves simulating the breakthrough of solutes at the ESF from an instantaneously injected set of particles distributed over the surface of Yucca Mountain. The points for comparison are distributed along the ESF as well as at a few other representative locations such as a point in the middle of the potential repository and a point coincident with one in the two-dimensional model.

Starting with the ESF locations, Figure 6-24 shows the breakthrough curves for the first half of the ESF, up through Station 35. Figure 6-25 shows the breakthrough curves for the second half of the ESF, Stations 39 through 76. The difference between the northern half and the southern half is quite striking. Only at Station 8, a station in the TCw with no PTn above it, is there significant young water. This is entirely expected as our models predict rapid transport through the highly fractured TCw. Once the ESF dips under the PTn, however, very little early arrival is simulated all the way up through station 35. The simulated water ages at those stations range between 1000 and 10,000 years, and hence are on the correct order to yield $^{36}\text{Cl}/\text{Cl}$ ratios similar to those measured by Fabryka-Martin et al. (1996b). The results for the southern half of the ESF differ significantly from those in the northern half. Early arrivals (up to 25% mass breakthrough) dominate this portion of the ESF, particularly once past Station 45. Simulated water ages for selected locations beyond Station 45 range between 2500 and 7000 years (Figure 6-25). However, in contrast with the breakthrough curves in the northern half of the ESF, is this faster breakthrough behavior due to the thinning of the PTn as measured in SD7 by Rautman and Engstrom (1996)? Or, do the parameters in 6541a lead to an unreasonably large proportion of fracture flow in the PTn? The occurrence of significant fracture flow in the PTn would have significant implications for repository design and performance as mentioned earlier. However, sample analyses in this

portion of the ESF show a dramatic decrease in measured $^{36}\text{Cl}/\text{Cl}$ ratios in the southern half of the ESF, with none that can be identified as containing unambiguous bomb-pulse. In fact, as Figure 6-21 shows, some of the measured signals drop off well below background values, indicating possible decay of ^{36}Cl due to long travel times from the surface to the ESF.

The poorer match of the simulated values to measured $^{36}\text{Cl}/\text{Cl}$ ratios in the southern ESF than in the northern ESF is one cause for concern. Another is the divergence of these three-dimensional results from those obtained with the property set of Bandurraga (1996). With those properties, the simulated $^{36}\text{Cl}/\text{Cl}$ ratios decreased substantially in the regions where measured sample ratios also decreased. There was a strong correlation between the data, those results, and the decreased infiltration rate above that region of low $^{36}\text{C}/\text{Cl}$ values. Thus, we must consider the reliability of the geologic model and the property sets. The geologic model is assumed to represent the PTn fairly accurately since nearby SD7 is one of the wells on which the structural model is based. Like SD7, the geologic model is thinner in the PTn in the south, but the PTn is intact over the region where bomb-pulse time-scale arrivals are simulated.

To address the effect of small changes in PTn fracture/matrix coupling, we use the same parameter modification as we used in the two-dimensional study. Namely, the reduction in fracture-matrix coupling as a function of simulated fracture saturation is removed. Figure 6-26 shows the simulated cumulative age distributions of water at the northern ESF station when the modified parameter set, 6541b, is used. Comparing Figure 6-26 with Figure 6-24, we see that the small amount of early arrival is completely eliminated as a result of reduced fracture flow in the PTn. Station 8 still is dominated by early arrival of water as there is no PTn to delay transport to the ESF at that location.

Figure 6-27 shows the cumulative age distributions at the ESF stations beyond Station 35. Although not completely eliminated, the amount of early arrival at the ESF is significantly reduced from the 6541a case shown in Figure 6-24 for the same stations. This indicates that even without

the reduced fracture-matrix coupling, some fracture flow is simulated in the PTn with these properties.

What controls whether or not significant flow occurs in the PTn fractures? Certainly, the fracture permeability plays a major role (Robinson et al., 1996) as well as the matrix potential at the simulated saturation. Both of these are a function of the flow rate. Therefore, as with the two-dimensional model runs, we examine the effect of reducing the flow rate by a factor of three. Figures 6-28 and 6-29 show the breakthrough curves for the case of modified PTn parameters, 6541b, and reduced infiltration rate. One interesting observation is that this case leads to even less fracture flow in the PTn and hence less early arrival in the ESF at the southern stations. More interesting, however, is that at all stations, the mean sample age is closer to 10,000 years than 1,000 years. This result is a closer match to our inferred ages from all of the ESF samples. Namely, to explain the $^{36}\text{Cl}/\text{Cl}$ ratios measured in the ESF, age distributions that include some portion which is greater than 10,000 is necessary.

An alternative conceptual model is that all ESF samples with $^{36}\text{Cl}/\text{Cl}$ ratios greater than present day background indicate mixing with bomb-pulse signals. This conceptual model suggests that all water flowing past the ESF is very young and that none of the elevated signals represent water that entered the system with a higher than present day background signal over 10,000 years ago. We find that model inconsistent with data and any of the hydrologic parameters. With that model, greater bomb-pulse era arrivals should be simulated in the northern part of the ESF where the elevated $^{36}\text{Cl}/\text{Cl}$ ratios are measured. To the contrary, we simulate more fast transport to the ESF in the southern ESF where most of the data are at or below present day background. This line of evidence is further explored in Chapter 7 where other chemical data sets are evaluated.

6.10.4 South to North Trends in Potential Repository Block Region

In the three-dimensional simulations, we observe a greater propensity of early arrival at the southern stations of the ESF than in the northern stations. Likewise, all of the potential repository horizon locations, as well as some east of the Ghost Dance Fault, in the two-dimensional cross

Figure 6-24. Breakthrough (or cumulative age distribution of a porewater sample) in northern ESF simulated with 3D model. Material properties are the 6541a set. Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters).

Figure 6-25. Breakthrough (or cumulative age distribution of a porewater sample) in southern ESF simulated with 3D model. Material properties are the 6541a set. Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters).

Figure 6-26. Breakthrough (or cumulative age distribution of a porewater sample) in northern ESF simulated with 3D model. Material properties are the 6541b set (*no weighting of fracture-matrix connection in PTn*). Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters).

Figure 6-27. Breakthrough (or cumulative age distribution of a porewater sample) in southern ESF simulated with 3D model. Material properties are the 6541b set (*no weighting of fracture-matrix connection in PTn*). Infiltration rate at surface is the map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters).

Figure 6-28. Breakthrough (or cumulative age distribution of a porewater sample) in northern ESF simulated with 3D model. Material properties are the 6541b set (*no weighting of fracture-matrix connection in PTn*). Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters).

Figure 6-29. Breakthrough (or cumulative age distribution of a porewater sample) in southern ESF simulated with 3D model. Material properties are the 6541b set (*no weighting of fracture-matrix connection in PTn*). Infiltration rate at surface is the scaled (I/3) map of Flint et al. (1996). Legend numbers indicate ESF Station numbers (in hundreds of meters).

section (located at NSP Northing coordinate 232100) showed some indication that fast paths through the PTn were possible, especially with the 6141a property set. Since the PTn is thinner at that northing location, we performed a series of breakthrough simulations along a north-south transect through the potential repository block under a band of high infiltration estimates. This transect is located in Figure 6-3, and Figure 6-7 shows it relative to the estimated infiltration map of Flint et al. (1996).

The north-south transect was also designed to intersect the east-west transect that was used for the two-dimensional model simulations as well a one-dimensional column study in the middle of the potential repository block. Results from the three-dimensional transport model at eight locations approximately 500 m apart are reported for this analysis. The locations range from Northing 231500 m to 235000. Figures 6-30 and 6-31 show the breakthrough curves at all 8 locations for 4 different parameter sets. Starting with the 6541a parameter set and the I infiltration map, we see clearly that moving north leads to a decrease in the amount of early arrival that is simulated at the ESF. The potential repository location at NSP Northing 232100, which corresponds with the east-west cross sections, shows the same behavior at this Easting coordinate. Comparison of the red line in Figure 6-15 and the red line in Figure 6-30a shows virtually identical behavior with the exception that the median age in the two-dimensional simulation is slightly younger than that in the three-dimensional simulation. Both simulations show about 20 percent (25 percent in the two-dimensional case) early arrival (i.e., less than 50 years).

Moving north along the north-south transect, the effects of the thickening PTn are clearly evident. The percentage of bomb-pulse era arrivals decreases until they represent less than a few percent north of NSP 233500 m. Also, as the PTn thickens, the median travel time to the repository horizon increases.

Figure 6-30 also show the simulated breakthrough curves along the north-south transect using the 6541b set (no reduction in fracture-matrix coupling in PTn). The results for this case show similar curves to the 6541a case, except there is far less early arrival because fracture flow is

Figure 6-30. North-South transect cumulative age distributions at potential repository horizon using I infiltration map. a) Property set 6541a b) Property set 6541b. Legend indicates northing coordinate. Easting coordinate is 170600 m.

Figure 6-31. North-South transect cumulative age distributions at potential repository horizon using I/3 infiltration map. a) Property set 6541a b) Property set 6541b. Legend indicates northing coordinate. Easting coordinate is 170600 m.

not sustained in the PTn, even for these high infiltration rates. Recall that the simulated saturations in the PTn differ minimally between the 6541a and 6541b case. The implications in terms of significance of episodic pulses however may be significant. We investigate such issues in the next

section with one-dimensional column models at three representative locations capturing the variation in PTn thickness.

As part of the north-south transect study, we also investigate the response for a reduced infiltration map. Here we use the I/3 map (Flint's map scaled by 1/3). Figure 6-31 shows the response for both parameters sets 6541a and 6541b. Even with the lower infiltration rate, early arrivals are simulated with the 6541a set whereas they are almost non existent, even in the southern locations, with the 6541b set.

Implications

Two important results can be derived from this study using the three-dimensional model to simulate arrival times along the north-south transect. First, a slight variation in material properties leads to significantly different results, especially in the southern section of the proposed repository block where the overlying PTn is thinnest. Reducing the uncertainty could have significant implications for repository design under that section of the PTn. Further north, the difference is less dramatic and probably less important. Second, regardless of the properties used to simulate flow and transport through the PTn, the higher percolation fluxes above the potential repository, relative to those above the ESF, lead to significantly faster transport from the surface to the potential repository. In most locations along this north-south transect, simulated ages are less than 10,000 years. Thus, we would expect to find $^{36}\text{Cl}/\text{Cl}$ ratios more representative of the present day signal of 500×10^{-15} . If data collection ever occurs in that regions, detection of ubiquitous bomb-pulse signals for ^{36}Cl and other isotopes would help determine which PTn property set is most appropriate.

In the next section, we perform more detailed analysis at three locations. One of the locations is identical to the location used in the north-south transect study at NSP coordinates (230600, 234000) which is the red dotted line in Figures 6-30 and 6-31.

6.11 Carbon-14 Modeling

Although ^{14}C can move both in the gas phase and the liquid phase, only liquid phase migration is considered in the present study. Previous studies by Viswanathan (1996) have examined gas phase migration of ^{14}C and phase partitioning with the liquid phase at Yucca Mountain. A basic conclusion of that study is that if the gas phase and liquid phase are in contact with each other, rapid gas phase migration could cause liquid phase measurement to yield apparent ages lower than the actual age of the water at the sample location. Neglecting that possibility, accounting for liquid phase only leads to a conservative estimation on the age of the porewater or perched water samples.

In the present study, infiltration variations as estimated by Flint et al. (1996) and a scaled version of that map are considered on the two-dimensional cross section. Two different property sets are also considered. One property set leads to more fracture flow in the PTn than the other. We also consider two different representations of the basal vitrophere properties. One case is the base-case property distribution. The other has a reduced fracture permeability that leads to lateral diversion to a semi-impervious fault which creates a perched water body.

Figure 6-32 shows the simulated percent modern carbon distribution in the matrix material. Due the fracture dominated flow in the welded units, there is no modern carbon in the matrix of those units. The PTn matrix shows very modern water as a result of the rapid transport through the TCw. After moving through the PTn, the water again moves quickly through the TSw to the CHn. Therefore, the ages in the CHn are representative of the travel time through the PTn. The ages simulated there are consistent with the breakthrough curves in Figure 6-15 with apparent ages ranging between 400 and 5000 years between Yucca Crest and the Ghost Dance fault and falling off to the west with the decrease in infiltration. Using the 6541b parameter set leads to no apparent difference as is shown in Figure 6-33. This is not surprising since the property change led only to small differences in the amount of flow in the PTn fractures. The majority of the flow, and hence the major contribution to an apparent age follows the same travel path which includes some flow

in the PTn matrix. Also, no bomb-pulse signature is attached to the ^{14}C input function in these simulations, hence no elevated signal at depth. Figure 6-34 shows the resulting ^{14}C for the perched water model. Percent modern carbon in the fractures is displayed as well as in the matrix. Since 6541a properties are used, young water, albeit a small amount, is shown throughout the PTn fractures under regions of adequate infiltration rate. In the saturated matrix, the apparent ages are on the order of 1000 years for the I infiltration map and 2000 years for the I/3 map. These ages certainly are not indicative of the travel times to the ESF but they occur in a region of much higher infiltration rate. Moving east, the apparent ages increase with decreasing infiltration rate. For further discussion on the perched water model, see Chapter 7.

Figure 6-32. Carbon-14 simulation with Flint et al. (1996) infiltration map and 6541a properties. Values plotted are percent modern carbon (PMC)

Figure 6-33. Carbon-14 simulation with Flint et al. (1996) infiltration map and 6541b properties. Values plotted are percent modern carbon (PMC)

Figure 6-34. Carbon-14 simulation for perched water simulation. Values plotted are percent modern carbon (PMC). a) Matrix PMC with I map, b) Fracture PMV with I map, c) Matrix PMC with I/3 map, d) Fracture PMC with I/3 map

6.12 One-Dimensional Process Level Modeling

A series of column modeling studies were performed to provide greater sensitivity analysis of significant processes and model parameters. Three columns were selected as representative of the different regimes encountered at Yucca Mountain. Two are coincident with ESF stations 35 and 57 and one falls in the middle of the proposed repository block and occurs at the same location as one of the north-south transect study points.

6.12.1 Column Descriptions

Figure 6-35 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 3D site-scale geologic model. In each of the column descriptions below, the PTn thickness is listed.

ESF Station 35

Station 35 is a location where the three-dimensional simulation did not predict significant early arrivals. It is also a location near which both bomb-pulse $^{36}\text{Cl}/\text{Cl}$ 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 we demonstrated the consistency between fault-zone and base-case model results as compared with field data and infiltration estimates (Robinson et al., 1996). The location of the Station 35 column in 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/

Figure 6-35. Schematics of three columns used in one-dimensional model studies.

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 is also near where the Ghost Dance fault crosses the ESF. However, unlike Station 35, where bomb-pulse $^{36}\text{Cl} / \text{Cl}$ 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 $^{36}\text{Cl}/\text{Cl}$ has been found. The Station 57 column is located at NSP coordinates (171160, 231200)

6.12.2 One-Dimensional Modeling Approach

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, 6541ft, that is based on the 6541a set but with increased fracture permeability in the PTn. 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.

Steady-State Flow

Flow and transport are first simulated 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

Both the UZ flow model (Bodvarsson et al., 1997) and this UZ transport model 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, we also examine the migration of solutes from the surface to depth under transient cases.

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 if 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.

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), we use a recurring episodic pulse and simulate for up to thousands of years. To compare with the transport on steady state flow fields, we 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 6-5 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.

Table 6-5. Transient Model Description

Model Name	Avg. Annual Infiltration rate (mm/yr)	Frequency of Application (yrs)	Duration (days)
10-1-10	10	1	10
10-1-90	10	1	90
10-5-10	10	5	10
10-5-90	10	5	90
20-5-10	20	5	10
50-5-90	50	5	10

6.12.3 Steady-State Results

Steady-State Flow with Parameter Set 6541a (the PA base-case)

Starting with the base-case parameter set, 6541a, and different flow rates, we examine the sensitivity of transport from the surface to the ESF or potential repository horizon. Figure 6-36 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% breakthrough (the median age), is fairly small. For example, at 5 mm/yr the difference between the high and low 50% 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% arrival ranges from 20,000 down to 800 years. With $^{36}\text{Cl}/\text{Cl}$ 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 6-38 shows the simulated distribution of flux between matrix and fractures in the Station 35 column and Figure 6-39 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% of the total flow at higher infiltration rates as is shown in the expanded view in Figure 6-39. This small component of high velocity flow leads to the small component of early arrival at Station 57 as shown in Figure 6-36.

These results are consistent with both the two- and three-dimensional simulations reported earlier in this chapter. Namely, in the southern portion of the study area, the PTn is thinner and simulations with the 6541a parameter set 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.

The $^{36}\text{Cl}/\text{Cl}$ 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 chapter we investigate the potential for episodic infiltration events to yield fast paths to the ESF at all three of the locations considered here.

Figure 6-36. Breakthrough curves at ESF/potential repository level for a) Station 35, b) Station 57, c) the repository column, and d) a comparison of the three at 5 mm/yr. Steady state infiltration varying between 0.5 and 20 mm/yr, and parameter set 6541a

Figure 6-37. Simulated saturations in the Station 35 column for parameter set 6541a.

Figure 6-38. Simulated flux distribution in the Station 35 Column for parameter set 6541a at 1 and 10 mm/yr infiltration rate.

Figure 6-39. Simulated flux distribution in the Station 57 column for parameter set 6541a at 1 and 10 mm/yr infiltration rate. Also shown is lowest 5% of total flow to highlight non-zero fracture fluxes in PTn. Layer resolution in Calico Hills not provided due to focus above ESF in these simulations.

Comparing Parameter Sets

As discussed in Chapter 5, 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, we primarily focus on parameter set 6541a and modifications to it due to the Performance Assessment interest in this parameter 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 Chapter 5 with respect to simulated saturation profiles and simulated radionuclide transport from the potential repository to the water table. Here we briefly examine the differences in simulations of ^{36}Cl from ground surface to the potential repository horizon using these two parameter sets. Figure 6-40 shows the breakthrough curve at ESF station 35 for property set 6412b. Comparing this plot with the Station 35 breakthrough in Figure 6-36 where parameter set 6541a was used, we see that there is substantially more early arrivals resulting from fracture flow in the PTn. To demonstrate this,

Figure 6-40 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. The implications with respect to perched water are addressed in Chapter 7. In the ESF, we 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.

Figure 6-40. Breakthrough curve at ESF for property set 6412b at Station 35. Also shown is flux distribution for 5 mm/yr case.

Parameter Set 6541b: A Slight Modification to the 6541a Parameter Set

The 6541b parameter set was described in the two- and three-dimensional simulations sections earlier in this chapter. It is our modification to LBNL's 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 6-41 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.

Figure 6-41. Comparison of saturations simulated in upper section of repository column with property sets 6541a and 6541b for 10 mm/yr.

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, 6541ft, 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.

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.

6.12.4 Comparisons: 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 6541ft) and for steady-state flow at 10 mm/yr and transient flow with 10-5-10 application. Figure 6-42 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 6541ft 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 6541ft properties.

Figure 6-43 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 6541ft 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, we can infer 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 6-44 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 6-45 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.

Figure 6-42. Station 35 breakthrough comparison for 10 mm/yr average annual infiltration rate. Three property sets (identified in legend) compared for steady-state (ss) and transient (trans) infiltration. Transient model, 10-5-10, is 10 mm/yr accumulation applied over 10 days every 5 years. Log plot on right illustrates component of early arrival

Figure 6-43. Station 57 breakthrough comparison for 3 property sets at steady state (ss) and in transient (trans) infiltration model, each with mean annual infiltration rate of 10 mm/yr. Property sets are 6541a, 6541b, and 6541flt. Transient mode is 10 mm/yr accumulated over 10 days every 5 years (10-5-10).

When considering the effect of transients on facilitating fracture flow in the PTn, the duration of the transient should be considered. In Figure 6-46, we examine the difference between 10 day infiltration events and 90 day infiltration events. In each case, the average infiltration rate

Figure 6-44.Repository column breakthrough comparison for 3 property sets at steady state (ss) and in transient (trans) infiltration model, each with mean annual infiltration rate of 10 mm/yr. Property sets are 6541a, 6541b, and 6541flt. Transient mode is 10 mm/yr accumulated over 10 days every 5 years (10-5-10).

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 used, 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 6-47 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. The 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 of decrease in flux of about ten percent at the potential repository after each infiltration event. The flux profile also follows the long-time

Figure 6-45. Comparison of breakthrough curves at potential repository horizon for transient simulations with a) 20 mm/yr average and b) 50 mm/yr average. All water is applied for 10 days every 5 years.

Figure 6-46. Comparison of breakthrough curves for 10 day duration transient and 90 day duration transients. Average annual infiltration for all cases is 10 mm/yr.

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

Figure 6-47. Simulated flux variation responses at potential repository for 10-5-10 transient infiltration rate at the surface. 300 years of transient followed by 700 years of uniform 10 mm/yr infiltration. The shaded portion of the 6541flt curve represents the short-term transients at the repository horizon.

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.

6.13 Conclusions

In this study, we reconfirm that modified, fault-zone PTn properties lead to simulations of bomb-pulse age solutes arriving at the potential repository horizon. Away from fault zones, the prediction of bomb-pulse age solutes arriving at depth is sensitive to the hydrologic parameter set (namely the model for fracture-matrix interaction), the PTn thickness, and the flow rate. Transport models were used to investigate relationships of these three controlling factors in order to develop a methodology for predicting fast paths at unsampled locations.

The fracture-matrix interaction model for the PTn plays a significant role in predictions of whether bomb-pulse signals penetrate the PTn completely. Varying the model has little effect on simulated saturations, but large effect on the depth to which bomb-pulse solutes can travel. Of the parameter sets investigated (see Table 8-2), set 6412b led to substantial amounts of PTn fracture flow. Sets 6541a and 6541b show similar characteristics in terms of the very small amount of PTn fracture flow that is simulated. However, the small difference between the two sets is the difference between simulations of bomb-pulse signals arriving at depth or not. Therefore, this study indicates that incorporating subsurface geochemistry into the calibration process may serve to reduce the number of possible parameter sets that characterize the flow system.

The relationship between PTn thickness and infiltration rate on bomb-pulse signal predictions at the potential repository horizon was investigated with the site-scale three-dimensional transport model. As the PTn thins to the south, the prediction of bomb-pulse arrivals at the potential repository horizon increase. With the 6541a parameter set, the bomb-pulse predictions extended further north (under thicker PTn) than for the 6541b parameter set. The 6541a parameter set also leads to predictions of more bomb-pulse travel times to the ESF than the 6541b set. However, there is a strong inconsistency between either model parameter set and the data. Whereas the incidence of bomb-pulse predictions with the model is highest in the south ramp of the ESF due to thin overlying PTn, the data indicate virtually no bomb-pulse signals. The ESF stations where bomb-pulse has been found are further north, where the model predicts less bomb-

pulse travel to the ESF. However, the locations where bomb-pulse signals have been measured are mostly well correlated with faults. The site-scale model does not resolve any of the faults associated with the ESF bomb-pulse predictions. Thus, one-dimensional models are used to investigate processes and parameters. As described above, modification of PTn properties to represent increased fracturing in faults leads to predictions of bomb-pulse in the ESF, even where the PTn is relatively thick.

The transport model was also used to investigate the role of episodic infiltration on bomb-pulse transport and flux variations at the potential repository horizon. Differences between parameter sets 6541a and 6541b were minor as the PTn damps the pulsed input almost completely. The difference in predicted bomb-pulse transport between these two sets remains due to slight differences in the small amount of sustained fracture flow in the PTn. The difference is greatest where the PTn is thinnest because fracture flow with set 6541a is sustained through the entire PTn. As the PTn thickens, fracture flow through the entire unit is not sustained with either set. In fault zones, on the other hand, fracture flow is sustained and the pulses are not damped completely. Where the PTn is thinnest, simulated pulses at the potential repository are greatest, an issue of potential significance to repository design and performance assessment.

With the new geologic model (Zelinski and Clayton, 1996), the new hydrologic parameters (Bodvarsson et al., 1997), and the spatially distributed infiltration map (Flint et al., 1996), simulated transport times through the PTn matrix are quicker than with previous models (Fabryka-Martin et al., 1996b). Whereas most of the solutes arriving at the ESF or the potential repository horizon in transport simulations had travel times on the order of 10,000 years in earlier studies, the median simulated travel times are now between 1,000 and 7,000 years. There still is a component of simulated solute arrival times (or water ages) that are greater than 10,000 years, but it represents only 5 to 20% of a sample. Thus, there may be a discrepancy in the interpretation of elevated $^{36}\text{Cl}/\text{Cl}$ signals away from fault zones. Previously, our interpretation was that these signals indicated water that entered the system in the Pleistocene with a higher $^{36}\text{Cl}/\text{Cl}$ ratio than

is found in modern water. We also interpreted these signals as being representative of the water flowing past the sample location. If, however, most of the water flowing past a sample location is really Holocene in origin (e.g. less than 10,000 years old) as the current simulations indicate, then the ESF samples are not representative of most of the water flowing through the TSw. In other words, an ESF sample may, in fact, indicate water older than 10,000 years, but it may indicate very little about the age of water flowing in the TSw fractures. The other conceptual model is that all of the elevated ESF samples contain a small bomb-pulse component. Whereas the recent PTn model of fracture-matrix interaction supports this conceptual model, the TSw fracture-matrix interaction model would need revision to prevent all bomb-pulse signals from completely bypassing the TSw matrix. Such a conceptual model also leaves questions about the apparent present day signal under thin PTn and higher signals in fault zones and away from fault zones under thicker PTn. We expect further closure on these apparent discrepancies will be reached with additional geochemical analysis.

Chapter 7 - Water Chemistry

7.1 Abstract

This chapter explores alternative explanations for the water chemistry variations in perched and pore waters at Yucca Mountain. The chemical databases are integrated with hydrologic databases to evaluate different model conceptualizations of the hydrologic behavior of the mountain at low and high infiltration rates. The premise for this exercise is that chemical data, when combined with hydrologic data, provide the most relevant information for “validating” radionuclide transport models, as the movement of naturally occurring solutes is closely related to the potential migration of radionuclides. Our philosophy for model development and testing is one in which these data are used in the building of conceptual models, rather than as data to “calibrate” these models.

Two end-member hypotheses are evaluated for the origin of perched water at Yucca Mountain: a “fast pathway” fracture flow hypothesis, and a transient-mixing hypothesis. The overall consequence of the “fast pathway” fracture flow hypothesis is that it predicts poor performance for the unsaturated zone, as the Calico Hills transport barrier is essentially bypassed by fracture flow. Since this model is based on non-calibrated fracture properties, we propose an alternative to this model in the transient-mixing hypothesis, based on the integration of all elements of the hydrologic and chemical databases at Yucca Mountain. Its main strength is that it explains simultaneously the inversion in apparent ages between pore waters and perched water in the unsaturated zone, and the low chloride content of perched waters relative to the overlying TSw and underlying CHn pore waters. The apparent disequilibrium in pore water and perched water contents is consistent with a model of transient flow, and we demonstrate that the stable isotope data and apparent ^{14}C and $^{36}\text{Cl}/\text{Cl}$ ages can be interpreted as resulting from a mixture of late Pleistocene/early Holocene water with modern waters. The existence of these mixed ancient/modern perched water bodies therefore implies that an additional retardation mechanism exists

above the Calico Hills formation. The ubiquitous young ^{14}C ages in the Calico Hills formation pore waters beneath the perched water bodies and in other areas, also imply that a lateral component of flow occurs in the vitric Calico Hills and that this formation is not bypassed. Finally, this model is consistent with the three-dimensional stochastic mineral distribution model in which zeolitic and vitric tuffs are intricately interbedded beneath the repository horizon (see Chapter 12 for a description of the stochastic model).

The implication of this model for Performance Assessment is an improved performance of the Calico Hills Transport barrier beneath the repository by 1) the additional barrier component which regulates the downward flow of water to the saturated zone, and 2) increased water residence times in the unsaturated zone due to the presence of long term perched water bodies at depth (i.e., no flow), and lateral diversion above the basal vitrophyre and/or the underlying Calico Hills vitric units. This should result in increased sorption of radionuclides at those boundaries, as actinides sorb in clays and vitric tuffs as well as in zeolites. Finally, we expect that the transient mixing model may give rise to different performance predictions than are achieved with the current hydrologic properties.

7.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al, 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 7-1 below.

Table 7-1. QA Status of Data used in this Chapter	
1	2
3	4
5	6
7	8
9	10
11	12
13	14
15	16
17	18
19	20
21	22
23	24
25	26
27	28
29	30
31	32
33	34
35	36
37	38
39	40
41	42
43	44
45	46
47	48
49	50
51	52
53	54
55	56
57	58
59	60
61	62
63	64
65	66
67	68
69	70
71	72
73	74
75	76
77	78
79	80
81	82
83	84
85	86
87	88
89	90
91	92
93	94
95	96
97	98
99	100

[illegible]

Fabryka-Martin et al., 1996c "Summary Report of Chlorine-36 Studies, YMP Milestone Report 3782M"	non-Q	LAJF831222AN96.001	Accession Nos. MOL.19961015.0094 MOL.19961015.0097
	Q	LAJF831222AQ96.002	
	Q	LAJF831222AQ96.003	
	Q	LAJF831222AQ96.004	
	Q	LAJF831222AQ96.005	
	Q	LAJF831222AQ96.006	
	Q	LAJF831222AQ96.007	
	Q	LAJF831222AQ96.008	
	Q	LAJF831222AN96.009	
	Q	LAJF831222AQ96.010	
	Q	LAJF831222AQ96.011	
	Q	LAJF831222AQ96.012	
	Q	LAJF831222AQ96.013	
	Q	LAJF831222AQ96.014	
	Q	LAJF831222AQ96.015	
	Q	LAJF831222AQ96.016	
	Q	LAJF831222AQ95.003	
	non-Q	LAJF831222AN95.004	
Flint et al., 1996 Conceptual and numerical model of infiltration for the Yucca Mountain Area, Nevada", USGS WRIR MOL 19970409.0087	Q	GS960908312211.003/ MOL.19970409.0087	USGS DTN 3GUI623M
Kwicklis, E.M. and J.P. Rousseau, 1996 Implications for waste isolation, <i>Hydrogeology of the unsaturated zone, North Ramp area of the Exploratory Studies Facility, Yucca Mountain, Nevada</i> , J.P. Rousseau, E.M. Kwicklis and D.C. Gillies (Eds.), U.S. Geological Survey Water Resources Investigations Report, (<i>in review</i>).	non-Q	Indeterminate	
Levy et al., 1997 "Investigations of Structural Controls and Mineralogic Associations of Chlorine-36 fast pathways in the ESF,. Yucca Mountain Project milestone report SP2301M4	Q	LASL831222AQ97.001	LANL DTN
Phillips, F.M., 1994 Environmental tracers for water movement in desert soils of the American southwest, Soil Sci.Soc. Am. J., 58, 15-24.	non-Q		

Rousseau, J.P. et al., 1996 Hydrogeology of the unsaturated zone, north ramp area of the exploratory studies facility, Yucca Mountain, Nevada, U.S. Geological Survey Water Resources Investigation Report 96-####, (in review).	non-Q	GS960308312232.001	USGS DTN
Yang et al., 1996, Interpretations of Chemical and Isotopic Data From Boreholes in the Unsaturated Zone at Yucca Mountain, Nevada, USGS Water-Resources Investigations Report WRIR 96-4058.	Q	GS951108312271.006	USGS DTN
Yang et al., 1997, Hydrochemical Investigations and Geochemical Modeling in Characterizing the Unsaturated Zone at Yucca Mountain, Nevada, (In Preparation).	non-Q		

7.3 Introduction

In other chapters, we have used geochemical databases to constrain and evaluate the validity of our flow and transport models in order to provide a strong scientific basis for the abstractions of these products used by Performance Assessment. In this chapter, we will focus on the water chemistry databases for pore waters and perched waters, in order to identify and discuss alternate models for flow and transport in Yucca Mountain.

7.4 Hydrochemical setting

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 databases (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 databases collected over the past 14 or so years (see Table 15-1). This model is

represented briefly by the following components involving two end member behaviors as a function of low and high infiltration rates, representing modern and ancient climates respectively.

7.4.1 TCw: Surface Infiltration

Under all climatic conditions the downward migration of meteoric waters through the Tiva Canyon welded tuffs (i.e., 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 Paintbrush non-welded Tuffs (i.e., PTn).

7.4.2 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% of the total water flux (see Chapter 6, this report). 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”. 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.

7.4.3 TSw: Transition from matrix dominated flow to fracture dominated flow

Under all infiltration rates, the flow of water in the non-lithophysal welded Topopah Spring hydrologic unit (i.e., 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 very short. 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, lateral diversion, down-dip along the top of the basal TSw vitrophyre is possible in most areas in the vicinity of the repository area, leading to the formation of perched water bodies on the down dropped side (i.e., west) of large normal faults (e.g., Figure 7-1; boreholes USW UZ-14/UZ-1, USW SD-9, USW SD-12). Exceptions include areas where the vitrophyre is missing (e.g., borehole USW G-2), areas possibly associated with complex tectonic disruptions in the vicinity of intersecting fault zones (e.g., boreholes NRG 7/7a, Drill Hole Wash/Ghost Dance fault intersections; USW SD-7, south-west splay of Ghost Dance fault/ Dune Wash / Abandoned Wash fault intersections; c.f., Figure 7-1). 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 non-welded tuffs of the Calico Hills formation (i.e., CHn).

At low infiltrations 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 Calico Hills formation.

7.4.4 CHn: composite vertical flow and lateral diversion

The Calico Hills non-welded tuffs are comprised of interlayered zeolitized and non-zeolitized units underlying the repository horizon. The flow in the CHn is probably complex due to the intricate spatial distribution of vitric and zeolitic rocks (c.f., Chapters 6 and 12). Slow percolation through matrix and fractures through intermingled 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 (c.f., Chapters 6 and 12). The perched water chemistries should reflect mixing as in the case of the basal TSw vitrophyre.

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 the underlying zeolitic Calico Hills

Figure 7-1. Map of Yucca Mountain site area with boreholes (i.e., G-2, UZ-14, NRG-7/7a, SD-9, SD-12, and SD-7) where perched water has been observed. Green line: site-scale model boundary. Blue: ESF. Red line: outline of potential repository.

formation, when present. This mechanism is possible for any infiltration rate and can result in the formation of perched water and lateral flow within the vitric Calico Hills. The pore water chemistry is expected to be strongly influenced by both the lateral flow component in the matrix (i.e., high chloride concentrations) and the vertical flow beneath the perched water bodies (i.e., low chloride concentrations).

7.4.5 Water Table: Seepage via fractures and matrix flow from the base of the CHn into the saturated zone

This conceptual model allows for some fast pathways through tectonic disruptions of 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.

7.5 Perched Water Chemistry

7.5.1 The Origin and Significance of Perched Water

In complex geologic settings, where the intrinsic hydrologic properties of rocks can be heterogeneous, the state of saturation above the static water table can vary so that saturated zones may be nested in vadose zones. These saturated conditions can be discontinuous in time as well as space, and form when contrasting hydrologic properties between juxtaposed formations give rise to a barrier to flow (Freeze and Cherry, 1979). The formation of perched water bodies also described as inverted water tables, typically involves a discrete boundary with a large permeability contrast and/or the transition from predominantly fracture flow to predominantly matrix flow and vis-versa. Heavy rainfall or climatic changes can lead to the formation of a temporary perched water body via transient effects in ground water flow systems. These are the result of time-dependent changes in the inflows and outflows at the ground surface (e.g., precipitation, evapotranspiration and infiltration). Therefore transient perched water bodies can exist at different time scales; from hours to months, associated with heavy rainfalls and snow melt infiltration, to

hundreds or several thousands of years, associated with climatic changes, and depending on the nature of the flow barrier, its depth from the surface, and the source and sink terms controlling the flux of water into and out of the perched system.

Several lines of evidence point to the existence of perched water bodies at depth in the vicinity of the potential repository site at Yucca Mountain. Their existence can be explained in part by the heterogeneous hydrologic properties exhibited by the various volcanic layers, enhanced locally by the juxtaposition and alteration of rocks along major fault systems identified in the mountain. Our knowledge of the existence of perched water bodies, their geometry and the processes that lead to their formation, is key to understanding the response behavior of Yucca Mountain to changes in local precipitation and global climate.

Unfortunately, the hydrologic characterization of these bodies when detected is scarce, the geochemistry complex and the hydrologic and chemical databases are often incomplete and/or unreliable. These factors allow for the existence of a wide range of conceptual models for the formation of perched water at Yucca Mountain, with the potential for drastically differing consequences for the performance of the unsaturated zone barrier and its ability to retard the migration of radionuclides from the repository to the water table and the accessible environment.

With some variations, most conceptual models advanced thus far, to explain the presence of perched water, involve a hydrologic system dominated by fracture flow (Bodvarsson et al., 1996, 1997), in which the movement of surface waters through fast vertical pathways in the mountain, rapidly reach the water table or saturated zone. In these models, perched waters represent local perturbations to that system due to the presence of anomalous properties associated with faults and some localized hydrologic heterogeneities in the tuffs of Yucca Mountain (i.e., Paintbrush Group, Topopah Spring and Calico Hills formations). These models imply that strong lateral and vertical heterogeneities exist in the Yucca Mountain hydrologic system. For example the basal vitrophyre must be relatively impermeable to vertical flow in areas where perched water exists (i.e., negligible fracture and matrix permeabilities), but be highly permeable elsewhere (i.e.,

high fracture permeability) so as to allow water to reach the saturated zone via fracture flow. Such a system also implies lateral flow is minimal to non-existent, and that perched waters represent modern meteoric waters. Furthermore these models imply that the fault systems that act as fast path channels for surface waters into the perched water zones also serve as lateral barriers to flow, in those same zones.

In this section, we will attempt to demonstrate that a “piston model” or “fast pathway” fracture flow model as presented above for the unsaturated zone at Yucca Mountain is not consistent with all observations. The alternative model involves a complex heterogeneous system of fracture/matrix interactions and takes into account the past climatic history of the Yucca Mountain area. The consequences for the performance of the unsaturated zone barrier at Yucca Mountain are diametrically opposite to those of the previous conceptual models. We will demonstrate that in the vicinity of the repository environment, the hydrologic and hydrochemical databases can be best explained by a basal vitrophyre of mostly homogeneous intrinsic hydrologic characteristics, which impede vertical flow during high infiltration conditions, and accommodates vertical flow during low infiltration conditions. This model allows for a lateral component of flow at high infiltration conditions, essential to the formation of perched water bodies. Perched water bodies in the repository footprint (i.e., boreholes USW UZ14/UZ1, USW SD-9 and USW SD12) are all composed of waters ranging from late Pleistocene to early Holocene in age implying that an inverted water table may exist at depth that retards the vertical movement of surface waters for up to 10,000 years prior to release through seepage into the underlying Calico Hills formation. They also exhibit lower chloride concentrations than from areas directly above or below them. These two conditions require that a lateral component of flow be present in the formation of perched water. The existence of these mixed/ancient perched water bodies therefore imply that an additional retardation mechanism exists above the Calico Hills formation.

7.5.2 Hydrologic evidence for perched waters

Perched water occurrences in the unsaturated zone at Yucca Mountain have been summarized by Striffler et al., 1996, and their detailed hydrologic and chemical signatures

described elsewhere (e.g., Kwicklis et al., 1996; Yang et al., 1996; Bovardsson et al., 1997). Typically peroxide waters at Yucca Mountain have been reported in the vicinity of a zone of fractured rock overlying a layer of lower fracture permeability (e.g., boreholes USW UZ-14/1, NRG-7/7a, USW SD-7, USW SD-9, USW SD-12, and G-2). However the data on most perched water bodies is incomplete as only USW UZ-14 and USW SD-7 produced sufficient perched water for pumping tests. Consequently, the hydrologic and geochemical evidence available for understanding the origin of perched-waters at Yucca Mountain does not allow for definitive answers.

The following represent the conditions necessary for perched waters to exist at Yucca Mountain.

Based on the observation of free water in some boreholes, it implies that:

- In perched water bodies where free water is observed, the water saturation within must have been high enough to initiate fracture flow. The water pressure within that body of water must have also exceeded static atmospheric gas pressures that would be expected for that elevation.
- A large contrast in hydraulic conductivity between adjacent geologic formations must be present.
- A permeable layer or pathway system must overlie a relatively impermeable layer and/or a well-connected fractured formation must overlie a locally unfractured or poorly connected fractured formation.
- Perched water in a dipping horizontal plane suggests a lateral barrier to down dip flow, such as a fault, may be present.
- The flow pathway is unlikely to pass vertically through the unsaturated zone to the water table, without being retarded by the perched water body, as water must be diverted laterally to a higher permeability channel to reach the water table.

Perched water at depth was located, from North to South, in boreholes G-2, USW UZ-14/ UZ-1, NRG-7/7a, USW SD-9, USW SD-12, USW SD-7 (Figure 7-1).

G-2: Located at the northern end of Yucca Mountain, near the site model boundary (Figure 7-1), a composite water level (c.f., Czarnecki et al., 1994), was detected at an elevation of 1,032m in 1981. Since then, the water level has declined by approximately 12m to an elevation of 1,020m (Czarnecki, 1997, personal comm.). The water level was originally thought to represent the saturated zone potentiometric surface, implying a large hydraulic gradient to the north of Yucca Mountain. The persistent decline since in water level since 1981, and the observation of water flow in fractures above the apparent water table point to the possibility that in fact this is a perched water body. Several pump tests were conducted recently (O'Brien, 1996), and perched water chemistry can be found in Yang et al. (1996).

USW UZ-14/UZ-1: As can be seen in Figure 7-1, both are situated in Drill Hole Wash. The USW UZ-14 hole is located N-NW of the potential repository site at Yucca Mountain, and 26.2 m from USW UZ-1. A perched water zone was detected initially as wet core samples just above the top of the Topopah Spring basal vitrophyre, at a depth of 383m. Several pumping tests were conducted and complete drawdown recovery occurred after 5.6 days indicating a large body of perched water (Czarnecki, 1997, personal comm.). USW UZ-14 comprises the most complete hydrologic and geochemical database of any of the perched water bodies at Yucca Mountain. Drilling of the hole was started on April 15, 1993, cored to a depth of 672.6m by April 29, 1994, and completed to a total depth of 677.8m on May 1994. Predominant rock types are similar to those of USW UZ-1 reported by Whitfield et al. (1990). Methods of sample collection and analysis are given in Yang et al. (1996).

NRG-7/7a: As for USW UZ-14/UZ-1, this borehole is also located in Drill Hole Wash (Figure 7-1). Drilled to a depth of 461 m, through the basal TSw vitrophyre and into the top of the Calico Hills non-welded tuffs, perched water was present between 458m and 461 m depth, near the contact of a series of highly fractured welded tuffs overlying relatively unfractured, nonwelded tuffs.

No pump tests were conducted for this borehole. Perched water chemistry was collected by Yang et al. (1996).

USW SD-9: This borehole is located to the south of Drill Hole Wash and west of the Ghost Dance fault. Perched water was detected at 413 m depth, above the basal vitrophyre of the TSw, and 157 m above the water table. Insufficient free water yield prevented the use of hydraulic pump tests. Perched water chemistry was collected by Yang et al. (1996).

USW SD-12: This borehole is located in Antler wash, west of the Ghost Dance fault (Rautman and Engstrom, 1996). Perched water was detected by the attenuation of the surface barometric signal within and below the crystal poor vitric zone of the TSw. No pump tests or water chemistry data are available to confirm the existence of a perched water body at this location.

USW SD-7: This hole is located near the southwest corner of the ESF, west of the Ghost Dance fault and east of the Abandoned Wash fault trace. Perched water in this hole was located at the bottom of the Calico Hills zeolitic unit, 150m above the water table. Although pump test were conducted, a small volume of water is indicated as the water level did not recover to the pre-pumping levels after testing (Bodvarsson et al., 1997). Perched water chemistry was collected by Yang et al. (1996).

7.5.3 Chemical Composition of Perched Waters

Perched waters are generally dilute sodium bicarbonate waters, chemically distinct from pore waters in the unsaturated zone, and somewhat compositionally similar to saturated zone waters. Below are some of the key characteristics of perched waters from Yucca Mountain.

- Lower chloride concentrations with respect to unsaturated zone pore waters situated above and below. Pore waters have chloride concentrations that range from 10-250 mg/l whereas perched waters have concentrations of 4-8 mg/l.
- Cl/Br ratios in perched waters have a range of 25-120 with an average of approximately 100. This distinguishes them from saturated zone waters which range from 120-150 with an average of approximately 130. Unsaturated zone pore waters ratios range from 162-428, whereas the

ratio in precipitation at Yucca Mountain averages about 74.

- Older apparent ages, with respect to pore water ages from the PTn and the CHn, located above and below respectively (Yang et al., 1997). Apparent ages range from 3,000 to 12,000 years using ^{14}C radioisotope techniques (Yang et al., 1996, 1997). At USW UZ-14 the ^{14}C apparent ages range from 8,000 to 12,000 years (Fabryka-Martin et al., 1996). Although younger ages have been suggested based on age correction schemes (Yang et al., 1996; Kwicklis et al., 1996) ^{14}C and ^{36}Cl isotope data indicate that Yucca Mountain perched waters (i.e., USW UZ-14, USW SD-7 and NRG 7/7a) fall along a characteristic “meteoric water curve” suggesting that the perched water primarily consists of meteoric water, unaffected by any geochemical processes affecting carbon or chloride isotopic compositions (Fabryka-Martin et al., 1996a-c).
- Stable isotope values measured in perched waters at Yucca Mountain fall in the range of -12.1 to -13.8 per mil $\delta^{18}\text{O}$, and -87.4 to -100 per mil δD (Yang et al., 1997). These values are generally between modern and ancient water compositions. Pore waters show a greater range and slightly higher average values compared to perched waters (Yang et al., 1996). However, perched waters $\delta^{18}\text{O}$ and δD values are slightly higher than those measured in most of the saturated zone waters analyzed to date except UE 29a#2 and J-13, dated at 10,000 years B.P. (Benson and McKinley, 1985).

7.6 Conceptual Models for Perched Water

7.6.1 Fast-Pathway Fracture Flow Hypothesis

Geochemical Models: The US Geological Survey has used hydrological and hydrochemical data to develop a model for the origin of perched water found in USW UZ-14 and several other drill holes (Rousseau et al., 1996). This model builds on an earlier USGS model (Montazer and Wilson, 1984), and attributes the perched zones primarily to vertically percolating fracture waters that were intercepted and accumulated at the boundary of a low permeability barrier (i.e., the top of the To-

popah Springs vitrophyre, or the top of the Calico Hills non-welded tuff). This model is essentially a “steady-state” model in that the volume of water in the perched zones represents a balance of inflow from fracture flow and outflow by leakage to the underlying units.

Several types of data have been cited in support of the “steady-state” model. One is the finding of “bomb-pulse” tritium (^3H), and ^{36}Cl in the Calico Hills tuff and in the ESF. This finding is taken to mean that there are preferential “fast pathways” (i.e., <50 years B.P.) for the percolation of water in Yucca Mountain. These pathways are assumed to be intercepted by the low permeability layers below the Topopah Springs welded unit to create the perched layers.

Another set of data used to support the “steady-state” model involves ^{14}C ages. Data presented by Yang et al (1996), suggests that pore waters in the Calico Hills tuff are younger than pore waters higher in the system. According to Rousseau et al. (1996), such an arrangement of ages cannot be explained by vertical percolation through the matrix of the units overlying the Calico Hills tuff.

The observation that chloride concentrations in pore waters in the Calico Hills tuff exceed chloride concentrations in perched waters by factors of 3-4 when combined with the ^{14}C “age inversion” was interpreted by Rousseau et al. (1996) to indicate that the Calico Hills pore waters did not originate by slow seepage of perched water through the low permeability layers. The observed chloride concentrations in pore waters from the nonwelded unit at the base of the Topopah Springs tuff are significantly higher than those found in perched water or Calico Hills pore water. This is also taken as evidence that vertical percolation of pore water from the lower Topopah Springs welded unit into the upper non-welded Calico Hills tuff is not consistent with the data.

Hydrologic Models: The formation of perched water was recently investigated by Bodvarsson et al. (1996, 1997), using a 3-D steady-state hydrologic model. The simulations involved the use of hydrologic data from up to 6 boreholes with perched water bodies (i.e., USW UZ-14/UZ-1, G-2, USW SD-7, USW SD-9, USW SD-12 and NRG-7/7a), and combined with the surface infiltration

map of Flint et al. (1996). Steady-state simulations were compared to measured water saturation and water potential profiles. Perched water ages were also used to compare with the simulated groundwater travel times.

In the latest conceptual model, it is assumed that perched water occurs where the percolation flux exceeds the capacity of the geologic media in the unsaturated zone to transmit flux. Based on an interpretation of the geochemical data base of Yang et al. (1996), they conclude that perched waters at Yucca Mountain are unlikely to represent past climate conditions. This is based on the interpretation that:

- Corrected ^{14}C perched water ages are relatively modern.
- Perched water chloride concentrations are low relative to the overlying PTn values.
- Perched water and pore water chemistries at depth, are in apparent disequilibrium.
- ^3H and ^{36}Cl bomb pulse signals in the ESF and the Calico Hills respectively (Yang et al., 1996; Fabryka-Martin et al., 1996a-c; Levy et al., 1997), require fast pathways at depth.

The “fast-pathway” fracture flow model hypothesis therefore requires circumventing the PTn matrix and the Calico Hills zeolitic tuffs, and is characterized by the following:

- With the exception of local occurrences of perched water, water fluxes travel predominantly through fractures from the surface to the water table with little interaction with matrix pore waters in the unsaturated zone.
- Surface waters undergo very little evaporation prior to infiltration.
- Rapid transport through the PTn, circumventing matrix flow, represents the largest flux component of water.
- With the exception of zones of local perching, a large volume water fraction circumvents the Calico Hills through fracture flow in the zeolites.
- Travel times to the water table vary generally from 50 to 3,000 years.

7.6.2 Transient Mixing Hypothesis

Hydro-geochemical Models: Beginning with the observation that there are zones of perched water present at depth today, it follows that under earlier, higher infiltration conditions (i.e., 10,000-20,000 years ago), larger perched water bodies must have formed in the same locations and possibly other locations as well. If the rate of seepage from these perched water bodies is sufficiently slow, one would expect that with some modifications, remnants of the Late Pleistocene/Early Holocene perched water bodies still exist today.

7.6.2.1 Climatic Evidence

Over the past 10,000 years, the global climate has changed from a glacial cycle to a post- or interglacial cycle (Imbrie and Imbrie, 1980; Imbrie, 1985). In the southwestern United States, the glacial periods are typically associated with wet climates, whereas the interglacials are dry (Gauthier and Wilson, 1994). Studies of Yucca Mountain demonstrate that the present climate belongs to a dry, interglacial period and that the recent past climate (i.e., 10,000 years ago.) was pluvial (Spaulding, 1985; Benson and Klieforth, 1989; Paces et al., 1993; Marshall et al., 1993; Forester, 1994; Forester and Smith, 1994). Although glacial cycles recur every 100,000 years or so, higher frequency global climate perturbations are also indicated (Imbrie et al., 1984; GRIP, 1993; Gauthier and Wilson, 1994). Although there is no direct evidence for smaller recurrence interval climate fluctuations (i.e., 1,000 to 2,000 year intervals) in the Yucca Mountain area (Winograd et al., 1992), there is evidence for an important climate change over the past 10,000 years, which is expected to have had a significant influence on net infiltration at Yucca Mountain (Gauthier and Wilson, 1994).

The evaluation of past and present climates at Yucca Mountain has recently been used to relate changes in near-surface processes (e.g., infiltration) to climate, and understand the hydrologic response above and below the potential repository site at Yucca Mountain (Flint et al., 1996). Using global ocean $\delta^{18}\text{O}$ time data from Imbrie et al. (1984), Flint et al. (1996) have simulated hydrologic conditions at Yucca Mountain over the past 600,000 years. Based on this

analysis, they predict a mean recharge rate of almost 5mm/yr, consistent with average infiltration estimates from environmental isotope studies (Fabryka-Martin et al., 1996a-c), and with a maximum of 10mm/yr 18,000 years ago and a minimum of 1mm/yr at present. They also predict that over the past 10,000 years the average precipitation at Yucca Mountain has decreased from approximately 260mm/yr to a present day average precipitation rate of 170mm/yr (Hevesi et al., 1991). Based on model predictions, Flint et al. (1996) estimate that this results in a decrease in infiltration rate of approximately a factor of 5 (Flint et al., 1996).

Given that climate changes of the same order of magnitude are possible over the next 10,000 years (Gauthier and Wilson, 1994; Flint et al., 1996), understanding the effects of this recent past climate on the hydrologic system at Yucca mountain is a key to understanding the behavior of the system in the future. In order to accomplish this, it is necessary to comprehend the coupled hydrochemical system at Yucca Mountain. In this model, the measured environmental isotope abundances, radioactive and stable isotopic compositions and water chemistry of perched- and pore waters, are believed to capture both the recent and ancient hydrologic histories of the mountain, integrated temporally and spatially. Kinetically driven equilibrium and disequilibrium hydrochemical processes, coupled with an understanding of the complex dynamics of the hydrochemical system at Yucca Mountain, can provide an insight into the behavior of the system over time. This concept represents the foundation for the conceptual model presented in this work. By integrating hydrologic and geochemical databases into model simulations, we will attempt to demonstrate that the pluvial pleistocene period and its transition to a dry interglacial present climate are both captured by the hydrochemical signature of Yucca Mountain at depth.

7.6.2.2 Structural and Mineralogical Setting

As discussed previously, the formation of perched water bodies typically requires that a flow barrier exist. The most commonly invoked flow barriers involve a discrete boundary with a large permeability contrast (Bodvarsson et al., 1997) and/or the transition from predominantly fracture flow to predominantly matrix flow and visa-versa (Robinson et al., 1996, present study; Fabryka-Martin et al., 1996a-c).

In the case of Yucca Mountain several such hydrologic settings might exist, the simplest and most obvious ones being the boundary between the base of the PTn and the upper TSw vitrophyre, and the boundary between the top of the basal Tsw vitrophyre and the overlying devitrified tuffs of the Topopah Spring formation. The first of these boundaries, the PTn, represents a matrix permeability contrast of approximately 2 orders of magnitude (i.e., 0.2×10^{-12} to $0.67 \times 10^{-14} \text{ m}^2$; Bodvarsson et al., 1997), and results in a transition from matrix flow to fracture flow with increasing depth, at low to intermediate infiltration rates (Fabryka-Martin et al., 1996a-c; Robinson et al., 1996, present study). Conversely, the permeability contrast between the top of the basal TSw vitrophyre and the overlying devitrified TSw tuffs is only a factor of 2 (Bodvarsson et al., 1997). The absolute matrix permeabilities however, are much lower (i.e., 0.13×10^{-15} to $0.65 \times 10^{-16} \text{ m}^2$), so that both lithologies exhibit dominantly fracture flow properties.

Vitrophyre Mineralogy: With the exception of data from boreholes USW SD-7 and NRG 7/7a, most perched water bodies in the vicinity of the potential repository footprint, are located just above the vitrophyre (e.g., USW UZ-14, USW SD-9, and USW SD-12). As both the basal vitrophyre and the overlying TSw devitrified tuffs have low matrix permeabilities and are predominantly characterized by fracture flow, perching at the vitrophyre is consistent with a model of lower fracture permeabilities than in the overlying devitrified TSw. Field investigations support this contention as fractures in the basal TSw vitrophyre can be classified into three categories:

- Perlitic cracks which tend to be concentric, non-penetrative curvilinear features of very small aperture and with no secondary alteration (Vaniman et al., 1993). The absence of clays or alteration products suggest that these do not contribute to the bulk fracture permeability of the vitrophyre.
- Penetrative fractures filled with devitrification alteration products (e.g., clays, clinoptilolite, opal CT). Although these features penetrate into the top of the vitrophyre they are not throughgoing (Levy and O'Neil, 1989). They have not been observed at the base of the vitrophyre and are interpreted as representing an alteration front associated with the

devitrification of the unit.

- Discrete fracture systems, commonly throughgoing and which may be associated with lower order fault systems. Generally poorly documented, these are believed to be tectonic in origin and are commonly associated with alteration minerals (e.g., clinoptilolite and smectite; Broxton et al., 1993).

Therefore, whereas fractures are commonly observed throughout the basal TSw vitrophyre in outcrop, most of them though penetrative, are not throughgoing. Though of variable aperture (i.e., 1-10mm), the throughgoing discrete fractures identified in the vitrophyre, are commonly coated or filled with secondary mineral alteration products (Vaniman, 1997, personal comm.). Based on these observations, fracture permeabilities of the basal vitrophyre are expected to be substantially lower than those of the overlying devitrified tuffs of the Topopah Springs formation. The basal vitrophyre is therefore likely to produce a barrier to flow under moderate to high infiltration rates, consistent with the occurrence of perched water above the basal vitrophyre (e.g., USW UZ-14, USW SD-9, USW SD-12).

Fault Mineralogy and Hydrologic Consequences: Given the nearly continuous eastward dip of the depositional bedding planes at Yucca Mountain (up to 10 degrees eastward dip), and the apparent ability of the basal vitrophyre in the vicinity of the potential repository area to impede vertical flow, it is likely that lateral diversion also occurs. As we will see in other sections perched water chemistries and isotopic compositions support a lateral flow hypothesis. However, in addition to a vertical barrier to flow, in this scenario a lateral barrier is also required for perched water to exist at Yucca Mountain. Based on these and other observations, several authors have suggested that faults may be responsible for impeding lateral flow at Yucca Mountain (Winograd, 1981; Montazer and Wilson, 1984; Bodvarsson et al., 1997).

Whereas fractures associated with larger scale fault structures have been shown to result in preferential pathways through the PTn (Fabryka-Martin et al., 1996a-c; Paces et al., 1996; Robinson et al., 1996, present study), it has been suggested that faults may also act as barriers to

flow at depth (Winograd, 1981; Montazer and Wilson, 1984). Because the Calico Hills nonwelded unit is more ductile than the overlying Topopah Spring welded unit, and because it is prone to gouge formation along shear zones and faults, Montazer and Wilson, (1984) suggest that fracture/fault conductivities may be smaller in the vicinity of faults. Further, we suspect that this is most likely at depth. Under most geologic settings and in the presence of humidity, gouge material typically increases mineral surface areas and consequently enhances reaction rates, leading to recrystallization along fault planes. This idea is supported by the extensive alteration observed in the vicinity of major fault zones affecting the Calico Hills formation (e.g., Fran Ridge, Busted Butte and Paintbrush faults; Broxton et al., 1993). At Busted Butte where such an exposure exists, the nonwelded tuffs of the Calico Hills formation which are largely unaltered, exhibit a localized zone of intense zeolitization next to a major normal fault that displaces the vitrophyre by several tens of meters vertically. Next to the fault, the non-welded base of the Topopah Spring member is thoroughly altered to clinoptilolite (i.e., 98wt.% zeolite). Though not as extensive, this zeolitization extends down the fault plane to the underlying vitric tuffs of the Calico Hills, and extends laterally several tens of meters on either side of the fault. Such a system is expected to act as a lateral barrier to flow because of the decreased fracture and matrix permeabilities associated with alteration, or in the absence of alteration, because of matrix/fracture permeability contrasts on either side of the fault. Even in the absence of alteration, the juxtaposition of low permeability devitrified Tsw tuffs on the downthrown side of the fault against high permeability vitric Calico Hills tuffs may form a capillary barrier. The effectiveness of the barrier will depend on the flux of water, and in a dual permeability system, the relative fracture and matrix permeabilities. A fault may act as a barrier at high infiltration rates if the fracture permeability contrast is high enough so as to impede flow. The same system however, may drain at lower infiltration rates. Conversely a fault may act as a barrier at low infiltration rates if the matrix permeability contrast and effective permeability are high enough so as to impede fracture flow. Again, this same configuration may drain via fracture flow at higher infiltration rates. At Yucca Mountain, in the probable case where geologic structures such as faults cause hydraulic discontinuities, water may either become

perched or drain depending on both extrinsic (e.g., infiltration rates and water flux) and intrinsic variables (e.g., effective fracture/matrix hydrologic properties). For perched water to be present either at the upper contact of the basal TSw vitrophyre (e.g., USW UZ-14, USW SD-12, USW-SD9) or the upper contact of the Calico Hills nonwelded unit (e.g., USW SD-7, NRG7/7a, USW G-2), such favorable conditions to flow barrier formation must exist. The proximity of USW UZ-14 to the down dropped side of the Drill Hole Wash fault (SW side) and of USW SD-12, USW SD-9, NRG7/7a, and USW SD-7 to the down dropped side of the Ghost Dance fault may be evidence that this perching mechanism is viable for Yucca Mountain in the area of the potential repository horizon.

From the evidence presented above, lateral flow along barriers to vertical flow is likely at Yucca Mountain. The existence of perched water bodies today, in the vicinity of known fault systems suggests that faults may impede continued horizontal water movement at depth by acting as barriers to lateral flow. Because the emplacement and tectonic histories responsible for the formation of the major geologic structures at Yucca Mountain (i.e., layering and fault systems), are believed to be over 5 million years old, and because associated fractures and mineral alterations in the basal vitrophyre and along faults are equally ancient (Levy and O'Neil, 1989), it follows that given the correct intrinsic hydrologic properties, the lithologic and structural boundaries that perform as barriers to flow today (i.e., low to moderate infiltration conditions), should have performed as such during the last ice age 10,000 to 20,000 years (i.e., moderate to high infiltration conditions).

7.6.2.3 Hydro-geochemical Model

A model which includes the existence of modified Late Pleistocene/Early Holocene perched water bodies at depth in Yucca Mountain would be distinguishable from a model that does not, using the following simple and testable geochemical and hydrologic criteria:

Hydrologic Criteria: There is a paucity of hydrologic criteria that can help distinguish between the various conceptual models. Calibrations to saturation data are non-unique and can therefore not be

used reliably to distinguish between the required hydrologic properties required for the models (Robinson et al., 1995, 1996, present study; Bodvarsson et al., 1996). The important difference between the proposed transient mixed water model, and fast pathway fracture models for the existence of perched water at Yucca Mountain, is that of a transient versus a steady-state model respectively. If one is to recognize the importance of the drastic change in climatic conditions over the past 10,000 years to the development of the present day hydrochemistry of the Yucca Mountain system, then only a transient model is appropriate. But, because this involves a past history, present day hydrologic measurements cannot hope to explain any differences between these models. Finally the alternative steady-state models, calibrated to matrix properties, invoke transient fracture flow as a dominant mechanism to provide a bypass mechanism of the PTn matrix (c.f., Bodvarsson et al., 1997), even though fracture properties are not well constrained by this approach.

Geochemical Criteria: Pleistocene perched water would be mixed with modern (i.e., post-pleistocene) waters so that the perched water bodies today would be identified as having a range of ages (assuming linear mixing) less than or equal to approximately 10,000 years old and always greater than present age. A model that does not allow for the presence of Pleistocene water implies all modern ages for the perched water body as travel times to the vitrophyre via fracture flow (Ptn bypass) are between several tens of years (e.g., ^3H ; Yang et al., 1996) to less than 1,000 years (present study).

At Yucca Mountain, the apparent ages for perched waters range from 3,000 to 12,000 years using ^{14}C radioisotope techniques (Yang et al., 1996, 1997). However, Yang et al. (1996, 1997) has suggested that these apparent ^{14}C ages be corrected for caliche contamination and soil-zone carbon. When this correction is applied, residence times of perched waters drop to values between 2,000 to 8,000 years.

However, samples of perched water from USW UZ-14 measured for $^{36}\text{Cl}/\text{Cl}$ range from 392×10^{-15} to 675×10^{-15} , are consistent with late Pleistocene to Holocene ages (Fabryka-Martin et

al., 1996a-c), and are in agreement with the apparent ^{14}C age determinations above. Fabryka-Martin et al. (1996a-c) have shown that the independent behavior through time between ^{14}C and ^{36}Cl isotope decay rates (Figure 7-2, allows us to predict that “ancient water” will fall along a

Figure 7-2. Reconstructed ^{14}C and ^{36}Cl activities for the last 50 ka. From Fabryka-Martin et al. (1996c).

characteristic “meteoric water curve” when plotted on a graph of ^{14}C versus $^{36}\text{Cl}/\text{Cl}$ (Figure 7-3). When perched water data are plotted together for various boreholes (i.e., USW UZ-14, USW SD-7 and NRG 7/7a), the data is shown to fall along “the meteoric water curve.” This result strongly suggests that the ^{14}C and ^{36}Cl reconstructions are reasonable, and also that the perched water primarily consists of meteoric water, unaffected by geochemical processes such as caliche contamination, affecting carbon or chloride isotopic compositions. At a minimum, this would indicate that the apparent ^{14}C age determinations for perched water are more reliable than their

Figure 7-3. Reconstructed ^{14}C and ^{36}Cl activities in the atmosphere for the last 20 ka, from Fabryka-Martin et al. (1996c). USW UZ-14A and UZ-14C were shown to be contaminated with modern water. Perched water data from UZ-14, SD-7 and NRG-7/7a.

ages corrected for caliche contamination (e.g., Yang et al., 1996, 1997), and that the perched water at USW UZ-14 for example, is between 8,000 and 12,000 years B.P. corresponding to 25 to 40 pmc and consistent with the very low ^3H analyses from USW UZ-14 (Yang et al., 1996). This data also implies that a simple piston-flow (i.e., no mixing) model is not correct for the origin of perched water (Fabryka-Martin et al., 1996a-c).

In a Pleistocene perched water model, the isotopic compositions of perched waters measured today (i.e., $\delta^{18}\text{O}$ and δD) should fall between values from the last ice age (e.g., -15 to -20 per mil $\delta^{18}\text{O}$; -105 to -120 per mil δD ; Yang et al., 1996) and modern compositions. Modern stable isotopic compositions vary a great deal both spatially and temporally but reported values

from 1984 rain storms at Yucca Mountain fall in the range -10.0 to -15.0 per mil for $\delta^{18}\text{O}$ and -70 to -90 per mil for δD (Yang et al., 1997).

In a mixed Pleistocene to modern perched water model, stable isotopic compositions are therefore expected to range between -13 and -15 per mil for $\delta^{18}\text{O}$, and -90 and -105 per mil for δD . In alternate models not involving a mixture with Pleistocene waters, the stable isotope values for perched waters should be modern.

The range of stable isotope values measured in perched waters today at Yucca Mountain fall in the range of -12.1 to -13.8 per mil $\delta^{18}\text{O}$, and -87.4 to -100 per mil δD (Yang et al., 1997), consistent with a mixing trend between modern and pleistocene waters, consistent with infiltration under cooler late-Pleistocene/early-Holocene climatic conditions, thus supporting the proposed model for perched water formation.

In order to be consistent with a Pleistocene perched water model, the chloride concentration (i.e., $[\text{Cl}]_{\text{pw}} = 4$ to 8 mg/l) of present day perched waters $[\text{Cl}]_{\text{pw}}$, should be consistent with higher infiltration rates (i.e., pluvial conditions) relative to the overlying unsaturated zone which should reflect the influence of a more recent, drier interglacial climate. Models that do not invoke Pleistocene water are required to bypass the PTn matrix, as the chloride concentration is high (i.e., $[\text{Cl}] = 45$ to 100mg/l, USW-14; Yang et al., 1996). Two bypass scenarios have been proposed:

- water travels via fracture flow through the PTn thereby bypassing the Ptn matrix and retaining low chloride concentrations (Bodvarsson et al., 1997; Kwicklis et al., 1996).
- water travels via fast matrix/fracture flow through the PTn in areas of high infiltration (i.e., lower chloride concentration), and then moves laterally underneath the low infiltration areas until a lateral barrier is encountered.

The chloride concentrations measured in the perched water bodies today are lower than those measured in PTn pore waters (e.g., 4 to 8 mg/l vs. 45 to 100mg/l, USW UZ-14; Yang et al., 1996). Simulations using calibrated hydrologic properties of Yucca Mountain rocks demonstrate

that this condition can be met *without bypassing the PTn matrix*, using a transient integrated flow model consistent with the past 10,000 year climatic history where a transition from pluvial (i.e., high infiltration) conditions to drier interglacial conditions existed.

Conversely, the other models postulate minimal matrix flow through the PTn and require fracture-dominated flow above the perched water bodies. This scenario is inconsistent with $^{36}\text{Cl}/\text{Cl}$ data from the ESF which indicate that a dominant proportion of the flow involves matrix flow through the PTn (Fabryka-Martin et al., 1996a-c; see also Chapter 6, present study). Whereas fast flow paths have been identified in the PTn using environmental isotope data (Fabryka-Martin et al., 1996a-c), these pathways have been shown to be discrete features, strongly correlated to large structural discontinuities (i.e., major fault zones; Levy et al., 1997) and are therefore not capable of handling large volumes fractions of water under present climatic conditions. Fast water velocities are therefore not synonymous with large water volumes or fluxes, and present day measured matrix saturations are well calibrated for conditions of matrix dominated flow in the PTn (see Chapter 6, present study). Without completely bypassing the PTn matrix, these conceptual models fail to produce low enough chloride concentrations in perched water bodies under present day infiltration conditions.

7.6.3 Evaluation of Alternate Conceptual Models

The alternate conceptual models for the origin of perched water bodies in the unsaturated zone at Yucca Mountain can be grouped into “fast pathway” fracture flow models and transient mixing models. The first are “steady-state, piston flow models that predict that perched waters primarily reflect current climatic conditions. The latter are transient flow models that put a greater emphasis on paleoclimatic conditions in addition to present-day conditions. Hydrochemical data can be used to evaluate these various models. The intent in this section is to review the available hydrochemical data and to discuss the extent to which the data are consistent with or contradict the existing models.

7.6.3.1 The Radioactive Isotopes of Hydrogen, Chlorine and Carbon

Hydrogen and Chlorine: Radioactive isotopes of hydrogen (^3H) and chlorine (^{36}Cl) have been particularly useful in investigations of potential flowpaths of waters infiltrating Yucca Mountain under current conditions. This results from the fact that substantial quantities of these isotopes were added to the atmosphere during nuclear weapons testing between 1952 and 1958 and the early 1960's (Fabryka-Martin et al., 1996a-c). Elevated concentrations of ^3H and ^{36}Cl (i.e., bomb-pulse) have been detected in pore waters in the PTn, and occasionally in the Calico Hills tuffs (Yang et al., 1996; Fabryka-Martin, 1996a-c). Elevated concentrations of ^{36}Cl have also been found at several locations in the Exploratory Science Facility (ESF). These observations are difficult to explain with any model that does not include fast flow paths within the unsaturated zone at Yucca Mountain at least to the depth of the ESF.

What the available data do not constrain very well are the amounts of water that moved through the fast-pathways to transport the observed ^3H and ^{36}Cl . Tritium in particular, travels readily in the gas phase which obscures its interpretation with respect to water pathways and travel times. The current thinking is that these fast flow paths are associated primarily with highly fractured or faulted portions of Yucca Mountain in discrete zones and that they involve relatively small water fluxes due to small catchment areas associated with their surface expressions.

Carbon: The radioactive isotope of carbon (^{14}C) can be used to trace pre-bomb waters in the unsaturated zone at Yucca Mountain. Analyses of ^{14}C activities in pore waters from samples representing the PTn, the Topopah Springs tuffs and the Calico Hills tuffs have been reported by Yang et al. (1996; 1997). Analyses of perched waters were also reported. Surprisingly, the measured range of activities of ^{14}C in the Calico Hills pore waters are similar to the range of activities measured in the PTn pore waters. Both are relatively young (i.e., 300-5,000 years B.P.). The activities measured in perched waters are generally less than those measured in the pore waters and imply

that the perched waters are older than the pore waters, even though the pore waters are found above and below the perched waters.

According to the USGS model, the inverted age distribution below perched water bodies precludes explanations that require vertical percolation of water from the TSw matrix above the perched water, to the CHn matrix below the perched water bodies. This not necessarily the case, however, because the pore waters in the Calico Hills tuffs could represent mixtures of perched water and pore water from above the perched water bodies via lateral flow mechanisms identified in the simulations. The proportions of perched water and pore water required cannot reasonably be quantified because the chemical parameters involved are rather poorly constrained. Importantly, the young “ages” derived for Calico Hills *pore waters* have significant implications for transport that are discussed further below.

The apparent ^{14}C “ages” calculated for perched waters are in the range of 3,500-11,000 years B.P. (Yang et al., 1996). As noted above this implies the perched waters are older than pore waters above and below them. It could be argued that the apparent “ages” for perched waters must be corrected for dead carbon dissolved along the path these waters took to their current location (Yang et al., 1997). However, this correction is controversial in that there is not agreement on how much dead carbon the perched waters may have dissolved. The assumption by Yang et al. (1997) that the concentrations of the major chemical constituents in perched and pore waters are entirely the result of water rock interaction, and not the result of evapotranspiration of precipitation incident on Yucca Mountain is difficult to defend as discussed further below. In any case, a dead carbon correction would also need to be applied to the pore waters. In the end, perched waters would still be older than the pore waters.

7.6.3.2 The Conservative Environmental Tracers Chloride and Cl/Br

Chloride: Pore waters are chemically distinct from perched waters in the unsaturated zone. This is clearly evident in a comparison of chloride concentrations. Pore waters have chloride concentra-

tions that range from 10-250 mg/l whereas perched waters have concentrations of 5-8 mg/l. Pore waters in the PTn tend to have higher chloride concentrations than pore waters in the Calico Hills Tuff. Further, pore waters in the PTn tend to be calcium and magnesium chloride/sulfate waters while pore waters in the Calico Hills tuffs tend to be sodium bicarbonate/carbonate types. Perched waters tend to be similar to saturated zone waters in composition in most constituents and are generally dilute sodium bicarbonate waters.

The concentration of chloride in pore waters obtained from the three samples of the Topopah Springs tuff that overlap the perched zone in UZ-14 have chloride concentrations that are a factor of 10 higher than the chloride concentrations in perched waters (Yang et al., 1996). The implication is that perched waters have not chemically equilibrated with pore waters in this horizon over time frames of thousands of years. The reasons for this apparent disequilibrium are unclear, but likely involve processes unique to the devitrified-vitrophyre transition at the base of the Tpt. The fractures in this zone are highly mineralized (Levy, 1984), and various unusual mineral species have found in this zone. For example, a ferrous chloride phase (Lawrencite) was identified in at least one sample from this zone (Vaniman, 1997, personal comm.). Further, the amounts of pore water recovered from samples in this zone was small compared to samples from the PTn and the Calico Hills Tuff (Yang et al., 1997). The pore waters recovered from this zone have unusually high chloride to sulfate ratios compared to pore waters from the PTn (Meijer, 1996). These observations taken together suggest that the pore waters from the lower Topopah Springs Tuff are unusual and that the chemical data for them must be interpreted with caution.

Yang et al. (1997) have suggested that the concentrations of chloride in pore waters and perched waters reflects primarily chemical reaction of dilute infiltrating waters with the host rock. This suggestion ignores the fact that the chloride mass balance method for calculating infiltration rates produces values that are in the range of values calculated on the basis of physical measurements (Flint et al., 1996). To the contrary, a detailed analysis of pore water chemistry data (Meijer, 1996), has lead to the opposite conclusion that pore waters in the unsaturated zone at Yucca Mountain show very little evidence of reaction with the host rock.

Cl/Br: One other aspect of water chemistry that is of interest in this discussion is the ratio of chloride to bromide in waters within the Yucca Mountain flow system. Because both of these constituents show conservative behavior in ground water systems, their ratio can be used to differentiate waters of different origins. This ratio has been measured in many different waters by Fabryka-Martin et al. (1996, and personal comm.) and Yang et al. (1996). The available data suggest Cl/Br ratios in saturated zone waters at Yucca Mountain are in the range of 120 to 150 with an average of approximately 130. Perched waters have a range of 25 to 120 with an average of approximately 100. Pore water ratios range from 162 to 428. The ratio in precipitation at Yucca Mountain averages about 74. At low infiltration rates, the concentrations are assumed to differ from the chloride values by a factor similar to that in the PTn pore water samples (200:1, chloride:bromide). In high infiltration zones, the fluids are assumed to enter the mountain at ratios typical of precipitation, namely 100:1, chloride:bromide.

These data suggest that Cl/Br ratios could be quite useful as a tracer in the Yucca Mountain flow system. For example, the Cl/Br ratio is one of the few chemical parameters that distinguishes perched waters from saturated zone waters.

7.6.3.3 The Stable Isotopes

In terms of stable isotopes $\delta^{18}\text{O}$ and δD , pore waters show a greater range and generally higher average values compared to perched waters (Yang et al., 1996). However, perched waters $\delta^{18}\text{O}$ and δD values are slightly higher than those measured in most of the saturated zone waters analyzed to date except J-13 and UE 29a#2. This is consistent with the fact that J-13 and UE 29a#2 waters and perched waters have similar apparent ages (i.e., 9-10 ka) and were therefore infiltrated under similar climatic conditions. The somewhat higher average $\delta^{18}\text{O}$ and δD values in perched waters are also consistent with the younger apparent ^{14}C ages for these waters (Yang et al., 1996) and a post-glacial climate.

Values for $\delta^{13}\text{C}$ have been measured in many water samples by Yang et al. (1996). This parameter is of importance to the interpretation of ^{14}C data because it provides an approximate means of assessing the amount of dead carbon that may have been incorporated into a given water sample. Yang et al. (1996) have used this parameter to “correct” apparent ^{14}C ages for inherited dead carbon. Unfortunately, the values obtained for this parameter in pore waters show rather large variability. Further, the pore water $\delta^{13}\text{C}$ values are commonly out of equilibrium with the $\delta^{13}\text{C}$ measured in the gas phase (Yang et al., 1996), which is problematic. Finally, there could be effects from drilling and the pore water extraction techniques that further complicate the analysis. The conclusion reached here is that “correction” of $\delta^{14}\text{C}$ “ages” using the $\delta^{13}\text{C}$ data is fraught with difficulties and the results should be interpreted with caution.

7.7 Simulation Results

In this section we present results of simulations of major ions present in unsaturated zone fluids, including chloride, sulfate, and bromide. To recap the conceptual model used to simulate these species, each are assumed to be conservative ions once they transport into the bedrock below the soil zone. The distinction between chloride, sulfate, and bromide is in the source term for fluid entering the bedrock. Chloride is assumed to be conservative even in the soil zone, whereas sulfate and bromide undergo reactions within the soil zone that lower their concentrations. For sulfate and bromide, these reactions could be biological in origin, whereas sulfate concentrations could also be affected by precipitation under conditions of evapotranspiration (Freeze and Cherry, 1979).

The concentration of dissolved species such as chloride are presumed in this modeling to be inversely proportional to the deep percolation rate, that is, the flow escaping evapotranspiration and entering the bedrock. This model assumes that the deposition rate of chloride is constant and not spatially variable, and that the controlling factor for concentration in the percolating fluid is evapotranspiration. Therefore, fluid entering at high infiltration rate has low chloride

concentration, and low infiltration correlates to high concentration. The concentration values themselves are based on the midpoint of the range of estimated values of chloride deposition rate of 75 to 150 mg/m²/y reported by Phillips (1994).

For reasons discussed above, the ratios of sulfate and bromide to chloride appear to be lower than in precipitation for many pore water samples in the unsaturated zone. An exception is in the perched waters, which appear to have ratios more reflective of precipitation. To simulate this behavior, we postulate that in regions of high enough infiltration, evapotranspirative effects and reactivity of bromide and sulfate are minimal due to short residence times in the soil zone. This results not only in lower concentrations of major ions, but also a shift to ratios closer to that of precipitation in fluids that enter at high infiltration. Therefore, the concentrations in the infiltrating fluids are assumed to fall into two categories. At low infiltration rates, the concentrations are assumed to differ from the chloride values by a factor similar to that in the PTn pore water samples (1:1 for chloride:sulfate, and 200:1 chloride:bromide). In high infiltration zones, the fluids are assumed to enter the mountain at ratios typical of precipitation, namely 1:3 sulfate:chloride and 100:1 chloride:bromide. The cut-off infiltration rate is assumed to be 10 mm/y, above which the latter ratios apply. In essence, the ion ratios are used as an additional natural tracer in the simulations.

When this chloride concentration model is input at the top of the three-dimensional site model, the concentration entering the mountain is a strong function of the spatially varying chloride map as controlled by infiltration rate. Figure 7-4 shows the concentration along the top surface of the model (top figure) and in a horizontal slice at an elevation of 900 m (bottom figure). The concentration entering the system is a direct result of the correlation to infiltration rate described earlier. At depth, the concentration patterns are somewhat more uniform due to mixing of fluids with different concentrations. The values, in general, appear lower because the fluid that supplies the subsurface is heavily weighted toward the high infiltration (low concentration) values.

Figure 7-4. Chloride concentration predicted in the three-dimensional model. Top figure: map of chloride concentrations at the surface. Bottom figure: concentrations at and elevation of 900 m. Top of page is North.

In the surface chloride map, only a small fraction of the fluid actually entering the mountain has high concentrations.

Figure 7-5 shows the predicted chloride concentrations for a steady state dual permeability

Figure 7-5. EW cross-section of transport site-scale model looking North. Chloride concentrations for the base infiltration map (upper figure) and the 1/3 infiltration scenario (bottom figure).

model for the base case infiltration map of Flint et al. (1996), and a scaled map where all values are reduced by a factor of three. Within a given simulation, the concentration values in the subsurface are a reflection of the concentration entering at the surface, which in turn is inversely

proportional to infiltration rate. The pore water compositions measured in the PTn fluids are therefore strongly controlled by the infiltration rate at the surface.

An infiltration rate equal to one third (i.e., $I/3$) of the Flint et al. (1996) values, distributed over the entire site surface, was used to investigate the effects of infiltration rate on the behavior of the hydrologic system. The $I/3$ values were used as they provided the best fit to the $^{36}\text{Cl}/\text{Cl}$ values from the ESF (Chapter 6, present study). The values themselves are higher at all locations for the $I/3$ case, since the chloride deposition rate is held constant throughout this simulation exercise. At greater depths, there is some tendency for lateral diversion to result in lower concentrations beneath higher concentrations (see close-up in Figure), but in general, for the property values used in this calculation, the flow and transport is predominantly vertical.7-6)

Figure 7-6. Detail of EW cross-section of transport site-scale model in area of relatively low infiltration. Close-up of chloride concentrations to the East of the Ghost Dance fault for the $I/3$ scenario.

The simulations for bromide and sulfate are closely related to those for chloride, except for the presumed difference in ion ratio for fluids of infiltration rate greater than 10 mm/y. Figure 7-7

Figure 7-7. EW cross-section of full site scale transport model, lookin North through Yucca Crest. Chloride to bromide ratio (top figure) and sulfate to chloride ratio (bottom figure) simulated using the base case property set and infiltration map.

plots the chloride to bromide ratio (top figure) and sulfate to chloride ratio (bottom figure) predicted by the model for the base infiltration case. Beneath high infiltration zones the ratio is that of precipitation, whereas elsewhere in the model the ratios reflect the presumed values in lower infiltration areas. Although there is a hint of lateral diversion at the TSw basal vitrophyre leading

to water with “precipitation ratios” underlying fluid with the low infiltration ratio, the effect is minor.

Although they explain some of the features observed in the data, the calculations presented so far, predict predominantly vertical downward flow, consistent with model parameters of the unsaturated zone flow model (Bodvarsson et al., 1997). However, such model results are seemingly at odds with data suggesting lateral diversion, including the perched water data (see Section 7.6). We now develop a model that includes perching due to lateral diversion at the TSw basal vitrophyre. We accomplish this by uniformly reducing the fracture permeability in this unit to $4 \times 10^{-17} \text{ m}^2$ while leaving the matrix permeability unchanged from the base-case value. This permeability is too low to support flow issuing from high infiltration regions, so that lateral diversion downdip from the source region occurs. To obtain perched water, we also reduce the permeability of the fault zones to values sufficiently low that they cannot drain the perched water. Figure 7-8 shows the steady state saturation profiles in the fracture domain for the base infiltration and the scaled I/3 scenario. The perched water bodies can exist under steady state conditions for these hydrologic parameters. Higher infiltration rates lead to a more extensive perched water body adjacent to the Ghost Dance fault.

Chemical transport predictions for this type of perched water scenario are now presented. Figure 7-9 shows the simulated chloride concentration (top figure), sulfate concentration (middle figure), and sulfate to chloride ratio (bottom figure) in the fracture domain for the base infiltration case. Dilute fluids enter the mountain in high infiltration regions, travel laterally at the basal vitrophyre, and result in a perched water body that is low in concentration of sulfate and chloride (about 11 ppm chloride), and higher in sulfate to chloride ratio than fluids entering at lower infiltration regions. Thus, several aspects of the perched and pore water chemical composition data are explained well with this lateral diversion model.

formation, when present. This mechanism is possible for any infiltration rate and can result in the formation of perched water and lateral flow within the vitric Calico Hills. The pore water chemistry is expected to be strongly influenced by both the lateral flow component in the matrix (i.e., high chloride concentrations) and the vertical flow beneath the perched water bodies (i.e., low chloride concentrations).

7.4.5 Water Table: Seepage via fractures and matrix flow from the base of the CHn into the saturated zone

This conceptual model allows for some fast pathways through tectonic disruptions of 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.

7.5 Perched Water Chemistry

7.5.1 The Origin and Significance of Perched Water

In complex geologic settings, where the intrinsic hydrologic properties of rocks can be heterogeneous, the state of saturation above the static water table can vary so that saturated zones may be nested in vadose zones. These saturated conditions can be discontinuous in time as well as space, and form when contrasting hydrologic properties between juxtaposed formations give rise to a barrier to flow (Freeze and Cherry, 1979). The formation of perched water bodies also described as inverted water tables, typically involves a discrete boundary with a large permeability contrast and/or the transition from predominantly fracture flow to predominantly matrix flow and vis-versa. Heavy rainfall or climatic changes can lead to the formation of a temporary perched water body via transient effects in ground water flow systems. These are the result of time-dependent changes in the inflows and outflows at the ground surface (e.g., precipitation, evapotranspiration and infiltration). Therefore transient perched water bodies can exist at different time scales; from hours to months, associated with heavy rainfalls and snow melt infiltration, to

hundreds or several thousands of years, associated with climatic changes, and depending on the nature of the flow barrier, its depth from the surface, and the source and sink terms controlling the flux of water into and out of the perched system.

Several lines of evidence point to the existence of perched water bodies at depth in the vicinity of the potential repository site at Yucca Mountain. Their existence can be explained in part by the heterogeneous hydrologic properties exhibited by the various volcanic layers, enhanced locally by the juxtaposition and alteration of rocks along major fault systems identified in the mountain. Our knowledge of the existence of perched water bodies, their geometry and the processes that lead to their formation, is key to understanding the response behavior of Yucca Mountain to changes in local precipitation and global climate.

Unfortunately, the hydrologic characterization of these bodies when detected is scarce, the geochemistry complex and the hydrologic and chemical databases are often incomplete and/or unreliable. These factors allow for the existence of a wide range of conceptual models for the formation of perched water at Yucca Mountain, with the potential for drastically differing consequences for the performance of the unsaturated zone barrier and its ability to retard the migration of radionuclides from the repository to the water table and the accessible environment.

With some variations, most conceptual models advanced thus far, to explain the presence of perched water, involve a hydrologic system dominated by fracture flow (Bodvarsson et al., 1996, 1997), in which the movement of surface waters through fast vertical pathways in the mountain, rapidly reach the water table or saturated zone. In these models, perched waters represent local perturbations to that system due to the presence of anomalous properties associated with faults and some localized hydrologic heterogeneities in the tuffs of Yucca Mountain (i.e., Paintbrush Group, Topopah Spring and Calico Hills formations). These models imply that strong lateral and vertical heterogeneities exist in the Yucca Mountain hydrologic system. For example the basal vitrophyre must be relatively impermeable to vertical flow in areas where perched water exists (i.e., negligible fracture and matrix permeabilities), but be highly permeable elsewhere (i.e.,

high fracture permeability) so as to allow water to reach the saturated zone via fracture flow. Such a system also implies lateral flow is minimal to non-existent, and that perched waters represent modern meteoric waters. Furthermore these models imply that the fault systems that act as fast path channels for surface waters into the perched water zones also serve as lateral barriers to flow, in those same zones.

In this section, we will attempt to demonstrate that a "piston model" or "fast pathway" fracture flow model as presented above for the unsaturated zone at Yucca Mountain is not consistent with all observations. The alternative model involves a complex heterogeneous system of fracture/matrix interactions and takes into account the past climatic history of the Yucca Mountain area. The consequences for the performance of the unsaturated zone barrier at Yucca Mountain are diametrically opposite to those of the previous conceptual models. We will demonstrate that in the vicinity of the repository environment, the hydrologic and hydrochemical databases can be best explained by a basal vitrophyre of mostly homogeneous intrinsic hydrologic characteristics, which impede vertical flow during high infiltration conditions, and accommodates vertical flow during low infiltration conditions. This model allows for a lateral component of flow at high infiltration conditions, essential to the formation of perched water bodies. Perched water bodies in the repository footprint (i.e., boreholes USW UZ14/UZ1, USW SD-9 and USW SD12) are all composed of waters ranging from late Pleistocene to early Holocene in age implying that an inverted water table may exist at depth that retards the vertical movement of surface waters for up to 10,000 years prior to release through seepage into the underlying Calico Hills formation. They also exhibit lower chloride concentrations than from areas directly above or below them. These two conditions require that a lateral component of flow be present in the formation of perched water. The existence of these mixed/ancient perched water bodies therefore imply that an additional retardation mechanism exists above the Calico Hills formation.

7.5.2 Hydrologic evidence for perched waters

Perched water occurrences in the unsaturated zone at Yucca Mountain have been summarized by Striffler et al., 1996, and their detailed hydrologic and chemical signatures

described elsewhere (e.g., Kwicklis et al., 1996; Yang et al., 1996; Bovardsson et al., 1997). Typically peroxide waters at Yucca Mountain have been reported in the vicinity of a zone of fractured rock overlying a layer of lower fracture permeability (e.g., boreholes USW UZ-14/1, NRG-7/7a, USW SD-7, USW SD-9, USW SD-12, and G-2). However the data on most perched water bodies is incomplete as only USW UZ-14 and USW SD-7 produced sufficient perched water for pumping tests. Consequently, the hydrologic and geochemical evidence available for understanding the origin of perched-waters at Yucca Mountain does not allow for definitive answers.

The following represent the conditions necessary for perched waters to exist at Yucca Mountain.

Based on the observation of free water in some boreholes, it implies that:

- In perched water bodies where free water is observed, the water saturation within must have been high enough to initiate fracture flow. The water pressure within that body of water must have also exceeded static atmospheric gas pressures that would be expected for that elevation.
- A large contrast in hydraulic conductivity between adjacent geologic formations must be present.
- A permeable layer or pathway system must overly a relatively impermeable layer and/or a well-connected fractured formation must overly a locally unfractured or poorly connected fractured formation.
- Perched water in a dipping horizontal plane suggests a lateral barrier to down dip flow, such as a fault, may be present.
- The flow pathway is unlikely to pass vertically through the unsaturated zone to the water table, without being retarded by the perched water body, as water must be diverted laterally to a higher permeability channel to reach the water table.

Perched water at depth was located, from North to South, in boreholes G-2, USW UZ-14/ UZ-1, NRG-7/7a, USW SD-9, USW SD-12, USW SD-7 (Figure 7-1).

G-2: Located at the northern end of Yucca Mountain, near the site model boundary (Figure 7-1), a composite water level (c.f., Czarnecki et al., 1994), was detected at an elevation of 1,032m in 1981. Since then, the water level has declined by approximately 12m to an elevation of 1,020m (Czarnecki, 1997, personal comm.). The water level was originally thought to represent the saturated zone potentiometric surface, implying a large hydraulic gradient to the north of Yucca Mountain. The persistent decline since in water level since 1981, and the observation of water flow in fractures above the apparent water table point to the possibility that in fact this is a perched water body. Several pump tests were conducted recently (O'Brien, 1996), and perched water chemistry can be found in Yang et al. (1996).

USW UZ-14/UZ-1: As can be seen in Figure 7-1, both are situated in Drill Hole Wash. The USW UZ-14 hole is located N-NW of the potential repository site at Yucca Mountain, and 26.2 m from USW UZ-1. A perched water zone was detected initially as wet core samples just above the top of the Topopah Spring basal vitrophyre, at a depth of 383m. Several pumping tests were conducted and complete drawdown recovery occurred after 5.6 days indicating a large body of perched water (Czarnecki, 1997, personal comm.). USW UZ-14 comprises the most complete hydrologic and geochemical database of any of the perched water bodies at Yucca Mountain. Drilling of the hole was started on April 15, 1993, cored to a depth of 672.6m by April 29, 1994, and completed to a total depth of 677.8m on May 1994. Predominant rock types are similar to those of USW UZ-1 reported by Whitfield et al. (1990). Methods of sample collection and analysis are given in Yang et al. (1996).

NRG-7/7a: As for USW UZ-14/UZ-1, this borehole is also located in Drill Hole Wash (Figure 7-1). Drilled to a depth of 461 m, through the basal TSw vitrophyre and into the top of the Calico Hills non-welded tuffs, perched water was present between 458m and 461 m depth, near the contact of a series of highly fractured welded tuffs overlying relatively unfractured, nonwelded tuffs.

No pump tests were conducted for this borehole. Perched water chemistry was collected by Yang et al. (1996).

USW SD-9: This borehole is located to the south of Drill Hole Wash and west of the Ghost Dance fault. Perched water was detected at 413 m depth, above the basal vitrophyre of the TSw, and 157 m above the water table. Insufficient free water yield prevented the use of hydraulic pump tests. Perched water chemistry was collected by Yang et al. (1996).

USW SD-12: This borehole is located in Antler wash, west of the Ghost Dance fault (Rautman and Engstrom, 1996). Perched water was detected by the attenuation of the surface barometric signal within and below the crystal poor vitric zone of the TSw. No pump tests or water chemistry data are available to confirm the existence of a perched water body at this location.

USW SD-7: This hole is located near the southwest corner of the ESF, west of the Ghost Dance fault and east of the Abandoned Wash fault trace. Perched water in this hole was located at the bottom of the Calico Hills zeolitic unit, 150m above the water table. Although pump test were conducted, a small volume of water is indicated as the water level did not recover to the pre-pumping levels after testing (Bodvarsson et al., 1997). Perched water chemistry was collected by Yang et al. (1996).

7.5.3 Chemical Composition of Perched Waters

Perched waters are generally dilute sodium bicarbonate waters, chemically distinct from pore waters in the unsaturated zone, and somewhat compositionally similar to saturated zone waters. Below are some of the key characteristics of perched waters from Yucca Mountain.

- Lower chloride concentrations with respect to unsaturated zone pore waters situated above and below. Pore waters have chloride concentrations that range from 10-250 mg/l whereas perched waters have concentrations of 4-8 mg/l.
- Cl/Br ratios in perched waters have a range of 25-120 with an average of approximately 100. This distinguishes them from saturated zone waters which range from 120-150 with an average of approximately 130. Unsaturated zone pore waters ratios range from 162-428, whereas the

ratio in precipitation at Yucca Mountain averages about 74.

- Older apparent ages, with respect to pore water ages from the PTn and the CHn, located above and below respectively (Yang et al., 1997). Apparent ages range from 3,000 to 12,000 years using ^{14}C radioisotope techniques (Yang et al., 1996, 1997). At USW UZ-14 the ^{14}C apparent ages range from 8,000 to 12,000 years (Fabryka-Martin et al., 1996). Although younger ages have been suggested based on age correction schemes (Yang et al., 1996; Kwicklis et al., 1996) ^{14}C and ^{36}Cl isotope data indicate that Yucca Mountain perched waters (i.e., USW UZ-14, USW SD-7 and NRG 7/7a) fall along a characteristic "meteoric water curve" suggesting that the perched water primarily consists of meteoric water, unaffected by any geochemical processes affecting carbon or chloride isotopic compositions (Fabryka-Martin et al., 1996a-c).
- Stable isotope values measured in perched waters at Yucca Mountain fall in the range of -12.1 to -13.8 per mil $\delta^{18}\text{O}$, and -87.4 to -100 per mil δD (Yang et al., 1997). These values are generally between modern and ancient water compositions. Pore waters show a greater range and slightly higher average values compared to perched waters (Yang et al., 1996). However, perched waters $\delta^{18}\text{O}$ and δD values are slightly higher than those measured in most of the saturated zone waters analyzed to date except UE 29a#2 and J-13, dated at 10,000 years B.P. (Benson and McKinley, 1985).

7.6 Conceptual Models for Perched Water

7.6.1 Fast-Pathway Fracture Flow Hypothesis

Geochemical Models: The US Geological Survey has used hydrological and hydrochemical data to develop a model for the origin of perched water found in USW UZ-14 and several other drill holes (Rousseau et al., 1996). This model builds on an earlier USGS model (Montazer and Wilson, 1984), and attributes the perched zones primarily to vertically percolating fracture waters that were intercepted and accumulated at the boundary of a low permeability barrier (i.e., the top of the To-

popah Springs vitrophyre, or the top of the Calico Hills non-welded tuff). This model is essentially a “steady-state” model in that the volume of water in the perched zones represents a balance of inflow from fracture flow and outflow by leakage to the underlying units.

Several types of data have been cited in support of the “steady-state” model. One is the finding of “bomb-pulse” tritium (^3H), and ^{36}Cl in the Calico Hills tuff and in the ESF. This finding is taken to mean that there are preferential “fast pathways” (i.e., <50 years B.P.) for the percolation of water in Yucca Mountain. These pathways are assumed to be intercepted by the low permeability layers below the Topopah Springs welded unit to create the perched layers.

Another set of data used to support the “steady-state” model involves ^{14}C ages. Data presented by Yang et al (1996), suggests that pore waters in the Calico Hills tuff are younger than pore waters higher in the system. According to Rousseau et al. (1996), such an arrangement of ages cannot be explained by vertical percolation through the matrix of the units overlying the Calico Hills tuff.

The observation that chloride concentrations in pore waters in the Calico Hills tuff exceed chloride concentrations in perched waters by factors of 3-4 when combined with the ^{14}C “age inversion” was interpreted by Rousseau et al. (1996) to indicate that the Calico Hills pore waters did not originate by slow seepage of perched water through the low permeability layers. The observed chloride concentrations in pore waters from the nonwelded unit at the base of the Topopah Springs tuff are significantly higher than those found in perched water or Calico Hills pore water. This is also taken as evidence that vertical percolation of pore water from the lower Topopah Springs welded unit into the upper non-welded Calico Hills tuff is not consistent with the data.

Hydrologic Models: The formation of perched water was recently investigated by Bodvarsson et al. (1996, 1997), using a 3-D steady-state hydrologic model. The simulations involved the use of hydrologic data from up to 6 boreholes with perched water bodies (i.e., USW UZ-14/UZ-1, G-2, USW SD-7, USW SD-9, USW SD-12 and NRG-7/7a), and combined with the surface infiltration

map of Flint et al. (1996). Steady-state simulations were compared to measured water saturation and water potential profiles. Perched water ages were also used to compare with the simulated groundwater travel times.

In the latest conceptual model, it is assumed that perched water occurs where the percolation flux exceeds the capacity of the geologic media in the unsaturated zone to transmit flux. Based on an interpretation of the geochemical data base of Yang et al. (1996), they conclude that perched waters at Yucca Mountain are unlikely to represent past climate conditions. This is based on the interpretation that:

- Corrected ^{14}C perched water ages are relatively modern.
- Perched water chloride concentrations are low relative to the overlying PTn values.
- Perched water and pore water chemistries at depth, are in apparent disequilibrium.
- ^3H and ^{36}Cl bomb pulse signals in the ESF and the Calico Hills respectively (Yang et al., 1996; Fabryka-Martin et al., 1996a-c; Levy et al., 1997), require fast pathways at depth.

The “fast-pathway” fracture flow model hypothesis therefore requires circumventing the PTn matrix and the Calico Hills zeolitic tuffs, and is characterized by the following:

- With the exception of local occurrences of perched water, water fluxes travel predominantly through fractures from the surface to the water table with little interaction with matrix pore waters in the unsaturated zone.
- Surface waters undergo very little evaporation prior to infiltration.
- Rapid transport through the PTn, circumventing matrix flow, represents the largest flux component of water.
- With the exception of zones of local perching, a large volume water fraction circumvents the Calico Hills through fracture flow in the zeolites.
- Travel times to the water table vary generally from 50 to 3,000 years.

7.6.2 Transient Mixing Hypothesis

Hydro-geochemical Models: Beginning with the observation that there are zones of perched water present at depth today, it follows that under earlier, higher infiltration conditions (i.e., 10,000-20,000 years ago), larger perched water bodies must have formed in the same locations and possibly other locations as well. If the rate of seepage from these perched water bodies is sufficiently slow, one would expect that with some modifications, remnants of the Late Pleistocene/Early Holocene perched water bodies still exist today.

7.6.2.1 Climatic Evidence

Over the past 10,000 years, the global climate has changed from a glacial cycle to a post- or interglacial cycle (Imbrie and Imbrie, 1980; Imbrie, 1985). In the southwestern United States, the glacial periods are typically associated with wet climates, whereas the interglacials are dry (Gauthier and Wilson, 1994). Studies of Yucca Mountain demonstrate that the present climate belongs to a dry, interglacial period and that the recent past climate (i.e., 10,000 years ago.) was pluvial (Spaulding, 1985; Benson and Klieforth, 1989; Paces et al., 1993; Marshall et al., 1993; Forester, 1994; Forester and Smith, 1994). Although glacial cycles recur every 100,000 years or so, higher frequency global climate perturbations are also indicated (Imbrie et al., 1984; GRIP, 1993; Gauthier and Wilson, 1994). Although there is no direct evidence for smaller recurrence interval climate fluctuations (i.e., 1,000 to 2,000 year intervals) in the Yucca Mountain area (Winograd et al., 1992), there is evidence for an important climate change over the past 10,000 years, which is expected to have had a significant influence on net infiltration at Yucca Mountain (Gauthier and Wilson, 1994).

The evaluation of past and present climates at Yucca Mountain has recently been used to relate changes in near-surface processes (e.g., infiltration) to climate, and understand the hydrologic response above and below the potential repository site at Yucca Mountain (Flint et al., 1996). Using global ocean $\delta^{18}\text{O}$ time data from Imbrie et al. (1984), Flint et al. (1996) have simulated hydrologic conditions at Yucca Mountain over the past 600,000 years. Based on this

analysis, they predict a mean recharge rate of almost 5mm/yr, consistent with average infiltration estimates from environmental isotope studies (Fabryka-Martin et al., 1996a-c), and with a maximum of 10mm/yr 18,000 years ago and a minimum of 1mm/yr at present. They also predict that over the past 10,000 years the average precipitation at Yucca Mountain has decreased from approximately 260mm/yr to a present day average precipitation rate of 170mm/yr (Hevesi et al., 1991). Based on model predictions, Flint et al. (1996) estimate that this results in a decrease in infiltration rate of approximately a factor of 5 (Flint et al., 1996).

Given that climate changes of the same order of magnitude are possible over the next 10,000 years (Gauthier and Wilson, 1994; Flint et al., 1996), understanding the effects of this recent past climate on the hydrologic system at Yucca mountain is a key to understanding the behavior of the system in the future. In order to accomplish this, it is necessary to comprehend the coupled hydrochemical system at Yucca Mountain. In this model, the measured environmental isotope abundances, radioactive and stable isotopic compositions and water chemistry of perched- and pore waters, are believed to capture both the recent and ancient hydrologic histories of the mountain, integrated temporally and spatially. Kinetically driven equilibrium and disequilibrium hydrochemical processes, coupled with an understanding of the complex dynamics of the hydrochemical system at Yucca Mountain, can provide an insight into the behavior of the system over time. This concept represents the foundation for the conceptual model presented in this work. By integrating hydrologic and geochemical databases into model simulations, we will attempt to demonstrate that the pluvial pleistocene period and its transition to a dry interglacial present climate are both captured by the hydrochemical signature of Yucca Mountain at depth.

7.6.2.2 Structural and Mineralogical Setting

As discussed previously, the formation of perched water bodies typically requires that a flow barrier exist. The most commonly invoked flow barriers involve a discrete boundary with a large permeability contrast (Bodvarsson et al., 1997) and/or the transition from predominantly fracture flow to predominantly matrix flow and visa-versa (Robinson et al., 1996, present study; Fabryka-Martin et al., 1996a-c).

In the case of Yucca Mountain several such hydrologic settings might exist, the simplest and most obvious ones being the boundary between the base of the PTn and the upper TSw vitrophyre, and the boundary between the top of the basal Tsw vitrophyre and the overlying devitrified tuffs of the Topopah Spring formation. The first of these boundaries, the PTn, represents a matrix permeability contrast of approximately 2 orders of magnitude (i.e., 0.2×10^{-12} to $0.67 \times 10^{-14} \text{ m}^2$; Bodvarsson et al., 1997), and results in a transition from matrix flow to fracture flow with increasing depth, at low to intermediate infiltration rates (Fabryka-Martin et al., 1996a-c; Robinson et al., 1996, present study). Conversely, the permeability contrast between the top of the basal TSw vitrophyre and the overlying devitrified TSw tuffs is only a factor of 2 (Bodvarsson et al., 1997). The absolute matrix permeabilities however, are much lower (i.e., 0.13×10^{-15} to $0.65 \times 10^{-16} \text{ m}^2$), so that both lithologies exhibit dominantly fracture flow properties.

Vitrophyre Mineralogy: With the exception of data from boreholes USW SD-7 and NRG 7/7a, most perched water bodies in the vicinity of the potential repository footprint, are located just above the vitrophyre (e.g., USW UZ-14, USW SD-9, and USW SD-12). As both the basal vitrophyre and the overlying TSw devitrified tuffs have low matrix permeabilities and are predominantly characterized by fracture flow, perching at the vitrophyre is consistent with a model of lower fracture permeabilities than in the overlying devitrified TSw. Field investigations support this contention as fractures in the basal TSw vitrophyre can be classified into three categories:

- Perlite cracks which tend to be concentric, non-penetrative curvilinear features of very small aperture and with no secondary alteration (Vaniman et al., 1993). The absence of clays or alteration products suggest that these do not contribute to the bulk fracture permeability of the vitrophyre.
- Penetrative fractures filled with devitrification alteration products (e.g., clays, clinoptilolite, opal CT). Although these features penetrate into the top of the vitrophyre they are not throughgoing (Levy and O'Neil, 1989). They have not been observed at the base of the vitrophyre and are interpreted as representing an alteration front associated with the

devitrification of the unit.

- Discrete fracture systems, commonly throughgoing and which may be associated with lower order fault systems. Generally poorly documented, these are believed to be tectonic in origin and are commonly associated with alteration minerals (e.g., clinoptilolite and smectite; Broxton et al., 1993).

Therefore, whereas fractures are commonly observed throughout the basal TS_w vitrophyre in outcrop, most of them though penetrative, are not throughgoing. Though of variable aperture (i.e., 1-10mm), the throughgoing discrete fractures identified in the vitrophyre, are commonly coated or filled with secondary mineral alteration products (Vaniman, 1997, personal comm.). Based on these observations, fracture permeabilities of the basal vitrophyre are expected to be substantially lower than those of the overlying devitrified tuffs of the Topopah Springs formation. The basal vitrophyre is therefore likely to produce a barrier to flow under moderate to high infiltration rates, consistent with the occurrence of perched water above the basal vitrophyre (e.g., USW UZ-14, USW SD-9, USW SD-12).

Fault Mineralogy and Hydrologic Consequences: Given the nearly continuous eastward dip of the depositional bedding planes at Yucca Mountain (up to 10 degrees eastward dip), and the apparent ability of the basal vitrophyre in the vicinity of the potential repository area to impede vertical flow, it is likely that lateral diversion also occurs. As we will see in other sections perched water chemistries and isotopic compositions support a lateral flow hypothesis. However, in addition to a vertical barrier to flow, in this scenario a lateral barrier is also required for perched water to exist at Yucca Mountain. Based on these and other observations, several authors have suggested that faults may be responsible for impeding lateral flow at Yucca Mountain (Winograd, 1981; Montazer and Wilson, 1984; Bodvarsson et al., 1997).

Whereas fractures associated with larger scale fault structures have been shown to result in preferential pathways through the PT_n (Fabryka-Martin et al., 1996a-c; Paces et al., 1996; Robinson et al., 1996, present study), it has been suggested that faults may also act as barriers to

flow at depth (Winograd, 1981; Montazer and Wilson, 1984). Because the Calico Hills nonwelded unit is more ductile than the overlying Topopah Spring welded unit, and because it is prone to gouge formation along shear zones and faults, Montazer and Wilson, (1984) suggest that fracture/fault conductivities may be smaller in the vicinity of faults. Further, we suspect that this is most likely at depth. Under most geologic settings and in the presence of humidity, gouge material typically increases mineral surface areas and consequently enhances reaction rates, leading to recrystallization along fault planes. This idea is supported by the extensive alteration observed in the vicinity of major fault zones affecting the Calico Hills formation (e.g., Fran Ridge, Busted Butte and Paintbrush faults; Broxton et al., 1993). At Busted Butte where such an exposure exists, the nonwelded tuffs of the Calico Hills formation which are largely unaltered, exhibit a localized zone of intense zeolitization next to a major normal fault that displaces the vitrophyre by several tens of meters vertically. Next to the fault, the non-welded base of the Topopah Spring member is thoroughly altered to clinoptilolite (i.e., 98wt.% zeolite). Though not as extensive, this zeolitization extends down the fault plane to the underlying vitric tuffs of the Calico Hills, and extends laterally several tens of meters on either side of the fault. Such a system is expected to act as a lateral barrier to flow because of the decreased fracture and matrix permeabilities associated with alteration, or in the absence of alteration, because of matrix/fracture permeability contrasts on either side of the fault. Even in the absence of alteration, the juxtaposition of low permeability devitrified Tsw tuffs on the downthrown side of the fault against high permeability vitric Calico Hills tuffs may form a capillary barrier. The effectiveness of the barrier will depend on the flux of water, and in a dual permeability system, the relative fracture and matrix permeabilities. A fault may act as a barrier at high infiltration rates if the fracture permeability contrast is high enough so as to impede flow. The same system however, may drain at lower infiltration rates. Conversely a fault may act as a barrier at low infiltration rates if the matrix permeability contrast and effective permeability are high enough so as to impede fracture flow. Again, this same configuration may drain via fracture flow at higher infiltration rates. At Yucca Mountain, in the probable case where geologic structures such as faults cause hydraulic discontinuities, water may either become

perched or drain depending on both extrinsic (e.g., infiltration rates and water flux) and intrinsic variables (e.g., effective fracture/matrix hydrologic properties). For perched water to be present either at the upper contact of the basal TSw vitrophyre (e.g., USW UZ-14, USW SD-12, USW SD9) or the upper contact of the Calico Hills nonwelded unit (e.g., USW SD-7, NRG7/7a, USW G-2), such favorable conditions to flow barrier formation must exist. The proximity of USW UZ-14 to the down dropped side of the Drill Hole Wash fault (SW side) and of USW SD-12, USW SD-9, NRG7/7a, and USW SD-7 to the down dropped side of the Ghost Dance fault may be evidence that this perching mechanism is viable for Yucca Mountain in the area of the potential repository horizon.

From the evidence presented above, lateral flow along barriers to vertical flow is likely at Yucca Mountain. The existence of perched water bodies today, in the vicinity of known fault systems suggests that faults may impede continued horizontal water movement at depth by acting as barriers to lateral flow. Because the emplacement and tectonic histories responsible for the formation of the major geologic structures at Yucca Mountain (i.e., layering and fault systems), are believed to be over 5 million years old, and because associated fractures and mineral alterations in the basal vitrophyre and along faults are equally ancient (Levy and O'Neil, 1989), it follows that given the correct intrinsic hydrologic properties, the lithologic and structural boundaries that perform as barriers to flow today (i.e., low to moderate infiltration conditions), should have performed as such during the last ice age 10,000 to 20,000 years (i.e., moderate to high infiltration conditions).

7.6.2.3 Hydro-geochemical Model

A model which includes the existence of modified Late Pleistocene/Early Holocene perched water bodies at depth in Yucca Mountain would be distinguishable from a model that does not, using the following simple and testable geochemical and hydrologic criteria:

Hydrologic Criteria: There is a paucity of hydrologic criteria that can help distinguish between the various conceptual models. Calibrations to saturation data are non-unique and can therefore not be

used reliably to distinguish between the required hydrologic properties required for the models (Robinson et al., 1995, 1996, present study; Bodvarsson et al., 1996). The important difference between the proposed transient mixed water model, and fast pathway fracture models for the existence of perched water at Yucca Mountain, is that of a transient versus a steady-state model respectively. If one is to recognize the importance of the drastic change in climatic conditions over the past 10,000 years to the development of the present day hydrochemistry of the Yucca Mountain system, then only a transient model is appropriate. But, because this involves a past history, present day hydrologic measurements cannot hope to explain any differences between these models. Finally the alternative steady-state models, calibrated to matrix properties, invoke transient fracture flow as a dominant mechanism to provide a bypass mechanism of the PTn matrix (c.f., Bodvarsson et al., 1997), even though fracture properties are not well constrained by this approach.

Geochemical Criteria: Pleistocene perched water would be mixed with modern (i.e., post-pleistocene) waters so that the perched water bodies today would be identified as having a range of ages (assuming linear mixing) less than or equal to approximately 10,000 years old and always greater than present age. A model that does not allow for the presence of Pleistocene water implies all modern ages for the perched water body as travel times to the vitrophyre via fracture flow (PTn bypass) are between several tens of years (e.g., ^3H ; Yang et al., 1996) to less than 1,000 years (present study).

At Yucca Mountain, the apparent ages for perched waters range from 3,000 to 12,000 years using ^{14}C radioisotope techniques (Yang et al., 1996, 1997). However, Yang et al. (1996, 1997) has suggested that these apparent ^{14}C ages be corrected for caliche contamination and soil-zone carbon. When this correction is applied, residence times of perched waters drop to values between 2,000 to 8,000 years.

However, samples of perched water from USW UZ-14 measured for $^{36}\text{Cl}/\text{Cl}$ range from 392×10^{-15} to 675×10^{-15} , are consistent with late Pleistocene to Holocene ages (Fabryka-Martin et

al., 1996a-c), and are in agreement with the apparent ^{14}C age determinations above. Fabryka-Martin et al. (1996a-c) have shown that the independent behavior through time between ^{14}C and ^{36}Cl isotope decay rates (Figure 7-2, allows us to predict that “ancient water” will fall along a

Figure 7-2. Reconstructed ^{14}C and ^{36}Cl activities for the last 50 ka. From Fabryka-Martin et al. (1996c).

characteristic “meteoric water curve” when plotted on a graph of ^{14}C versus $^{36}\text{Cl}/\text{Cl}$ (Figure 7-3). When perched water data are plotted together for various boreholes (i.e., USW UZ-14, USW SD-7 and NRG 7/7a), the data is shown to fall along “the meteoric water curve.” This result strongly suggests that the ^{14}C and ^{36}Cl reconstructions are reasonable, and also that the perched water primarily consists of meteoric water, unaffected by geochemical processes such as caliche contamination, affecting carbon or chloride isotopic compositions. At a minimum, this would indicate that the apparent ^{14}C age determinations for perched water are more reliable than their

Figure 7-3. Reconstructed ^{14}C and ^{36}Cl activities in the atmosphere for the last 20 ka, from Fabryka-Martin et al. (1996c). USW UZ-14A and UZ-14C were shown to be contaminated with modern water. Perched water data from UZ-14, SD-7 and NRG-7/7a.

ages corrected for caliche contamination (e.g., Yang et al., 1996, 1997), and that the perched water at USW UZ-14 for example, is between 8,000 and 12,000 years B.P. corresponding to 25 to 40 pmc and consistent with the very low ^3H analyses from USW UZ-14 (Yang et al., 1996). This data also implies that a simple piston-flow (i.e., no mixing) model is not correct for the origin of perched water (Fabryka-Martin et al., 1996a-c).

In a Pleistocene perched water model, the isotopic compositions of perched waters measured today (i.e., $\delta^{18}\text{O}$ and δD) should fall between values from the last ice age (e.g., -15 to -20 per mil $\delta^{18}\text{O}$; -105 to -120 per mil δD ; Yang et al., 1996) and modern compositions. Modern stable isotopic compositions vary a great deal both spatially and temporally but reported values

from 1984 rain storms at Yucca Mountain fall in the range -10.0 to -15.0 per mil for $\delta^{18}\text{O}$ and -70 to -90 per mil for δD (Yang et al., 1997).

In a mixed Pleistocene to modern perched water model, stable isotopic compositions are therefore expected to range between -13 and -15 per mil for $\delta^{18}\text{O}$, and -90 and -105 per mil for δD . In alternate models not involving a mixture with Pleistocene waters, the stable isotope values for perched waters should be modern.

The range of stable isotope values measured in perched waters today at Yucca Mountain fall in the range of -12.1 to -13.8 per mil $\delta^{18}\text{O}$, and -87.4 to -100 per mil δD (Yang et al., 1997), consistent with a mixing trend between modern and pleistocene waters, consistent with infiltration under cooler late-Pleistocene/early-Holocene climatic conditions, thus supporting the proposed model for perched water formation.

In order to be consistent with a Pleistocene perched water model, the chloride concentration (i.e., $[\text{Cl}]_{\text{pw}} = 4$ to 8 mg/l) of present day perched waters $[\text{Cl}]_{\text{pw}}$, should be consistent with higher infiltration rates (i.e., pluvial conditions) relative to the overlying unsaturated zone which should reflect the influence of a more recent, drier interglacial climate. Models that do not invoke Pleistocene water are required to bypass the PTn matrix, as the chloride concentration is high (i.e., $[\text{Cl}] = 45$ to 100mg/l, USW-14; Yang et al., 1996). Two bypass scenarios have been proposed:

- water travels via fracture flow through the PTn thereby bypassing the Ptn matrix and retaining low chloride concentrations (Bodvarsson et al., 1997; Kwicklis et al., 1996).
- water travels via fast matrix/fracture flow through the PTn in areas of high infiltration (i.e., lower chloride concentration), and then moves laterally underneath the low infiltration areas until a lateral barrier is encountered.

The chloride concentrations measured in the perched water bodies today are lower than those measured in PTn pore waters (e.g., 4 to 8 mg/l vs. 45 to 100mg/l, USW UZ-14; Yang et al., 1996). Simulations using calibrated hydrologic properties of Yucca Mountain rocks demonstrate

that this condition can be met *without bypassing the PTn matrix*, using a transient integrated flow model consistent with the past 10,000 year climatic history where a transition from pluvial (i.e., high infiltration) conditions to drier interglacial conditions existed.

Conversely, the other models postulate minimal matrix flow through the PTn and require fracture-dominated flow above the perched water bodies. This scenario is inconsistent with $^{36}\text{Cl}/\text{Cl}$ data from the ESF which indicate that a dominant proportion of the flow involves matrix flow through the PTn (Fabryka-Martin et al., 1996a-c; see also Chapter 6, present study). Whereas fast flow paths have been identified in the PTn using environmental isotope data (Fabryka-Martin et al., 1996a-c), these pathways have been shown to be discrete features, strongly correlated to large structural discontinuities (i.e., major fault zones; Levy et al., 1997) and are therefore not capable of handling large volumes fractions of water under present climatic conditions. Fast water velocities are therefore not synonymous with large water volumes or fluxes, and present day measured matrix saturations are well calibrated for conditions of matrix dominated flow in the PTn (see Chapter 6, present study). Without completely bypassing the PTn matrix, these conceptual models fail to produce low enough chloride concentrations in perched water bodies under present day infiltration conditions.

7.6.3 Evaluation of Alternate Conceptual Models

The alternate conceptual models for the origin of perched water bodies in the unsaturated zone at Yucca Mountain can be grouped into “fast pathway” fracture flow models and transient mixing models. The first are “steady-state, piston flow models that predict that perched waters primarily reflect current climatic conditions. The latter are transient flow models that put a greater emphasis on paleoclimatic conditions in addition to present-day conditions. Hydrochemical data can be used to evaluate these various models. The intent in this section is to review the available hydrochemical data and to discuss the extent to which the data are consistent with or contradict the existing models.

7.6.3.1 The Radioactive Isotopes of Hydrogen, Chlorine and Carbon

Hydrogen and Chlorine: Radioactive isotopes of hydrogen (^3H) and chlorine (^{36}Cl) have been particularly useful in investigations of potential flowpaths of waters infiltrating Yucca Mountain under current conditions. This results from the fact that substantial quantities of these isotopes were added to the atmosphere during nuclear weapons testing between 1952 and 1958 and the early 1960's (Fabryka-Martin et al., 1996a-c). Elevated concentrations of ^3H and ^{36}Cl (i.e., bomb-pulse) have been detected in pore waters in the PTn, and occasionally in the Calico Hills tuffs (Yang et al., 1996; Fabryka-Martin, 1996a-c). Elevated concentrations of ^{36}Cl have also been found at several locations in the Exploratory Science Facility (ESF). These observations are difficult to explain with any model that does not include fast flow paths within the unsaturated zone at Yucca Mountain at least to the depth of the ESF.

What the available data do not constrain very well are the amounts of water that moved through the fast-pathways to transport the observed ^3H and ^{36}Cl . Tritium in particular, travels readily in the gas phase which obscures its interpretation with respect to water pathways and travel times. The current thinking is that these fast flow paths are associated primarily with highly fractured or faulted portions of Yucca Mountain in discrete zones and that they involve relatively small water fluxes due to small catchment areas associated with their surface expressions.

Carbon: The radioactive isotope of carbon (^{14}C) can be used to trace pre-bomb waters in the unsaturated zone at Yucca Mountain. Analyses of ^{14}C activities in pore waters from samples representing the PTn, the Topopah Springs tuffs and the Calico Hills tuffs have been reported by Yang et al. (1996; 1997). Analyses of perched waters were also reported. Surprisingly, the measured range of activities of ^{14}C in the Calico Hills pore waters are similar to the range of activities measured in the PTn pore waters. Both are relatively young (i.e., 300-5,000 years B.P.). The activities measured in perched waters are generally less than those measured in the pore waters and imply

that the perched waters are older than the pore waters, even though the pore waters are found above and below the perched waters.

According to the USGS model, the inverted age distribution below perched water bodies precludes explanations that require vertical percolation of water from the TSw matrix above the perched water, to the CHn matrix below the perched water bodies. This not necessarily the case, however, because the pore waters in the Calico Hills tuffs could represent mixtures of perched water and pore water from above the perched water bodies via lateral flow mechanisms identified in the simulations. The proportions of perched water and pore water required cannot reasonably be quantified because the chemical parameters involved are rather poorly constrained. Importantly, the young “ages” derived for Calico Hills *pore waters* have significant implications for transport that are discussed further below.

The apparent ^{14}C “ages” calculated for perched waters are in the range of 3,500-11,000 years B.P. (Yang et al., 1996). As noted above this implies the perched waters are older than pore waters above and below them. It could be argued that the apparent “ages” for perched waters must be corrected for dead carbon dissolved along the path these waters took to their current location (Yang et al., 1997). However, this correction is controversial in that there is not agreement on how much dead carbon the perched waters may have dissolved. The assumption by Yang et al. (1997) that the concentrations of the major chemical constituents in perched and pore waters are entirely the result of water rock interaction, and not the result of evapotranspiration of precipitation incident on Yucca Mountain is difficult to defend as discussed further below. In any case, a dead carbon correction would also need to be applied to the pore waters. In the end, perched waters would still be older than the pore waters.

7.6.3.2 The Conservative Environmental Tracers Chloride and Cl/Br

Chloride: Pore waters are chemically distinct from perched waters in the unsaturated zone. This is clearly evident in a comparison of chloride concentrations. Pore waters have chloride concentra-

tions that range from 10-250 mg/l whereas perched waters have concentrations of 5-8 mg/l. Pore waters in the PTn tend to have higher chloride concentrations than pore waters in the Calico Hills Tuff. Further, pore waters in the PTn tend to be calcium and magnesium chloride/sulfate waters while pore waters in the Calico Hills tuffs tend to be sodium bicarbonate/carbonate types. Perched waters tend to be similar to saturated zone waters in composition in most constituents and are generally dilute sodium bicarbonate waters.

The concentration of chloride in pore waters obtained from the three samples of the Topopah Springs tuff that overlap the perched zone in UZ-14 have chloride concentrations that are a factor of 10 higher than the chloride concentrations in perched waters (Yang et al., 1996). The implication is that perched waters have not chemically equilibrated with pore waters in this horizon over time frames of thousands of years. The reasons for this apparent disequilibrium are unclear, but likely involve processes unique to the devitrified-vitrophyre transition at the base of the Tpt. The fractures in this zone are highly mineralized (Levy, 1984), and various unusual mineral species have found in this zone. For example, a ferrous chloride phase (Lawrencite) was identified in at least one sample from this zone (Vaniman, 1997, personal comm.). Further, the amounts of pore water recovered from samples in this zone was small compared to samples from the PTn and the Calico Hills Tuff (Yang et al., 1997). The pore waters recovered from this zone have unusually high chloride to sulfate ratios compared to pore waters from the PTn (Meijer, 1996). These observations taken together suggest that the pore waters from the lower Topopah Springs Tuff are unusual and that the chemical data for them must be interpreted with caution.

Yang et al. (1997) have suggested that the concentrations of chloride in pore waters and perched waters reflects primarily chemical reaction of dilute infiltrating waters with the host rock. This suggestion ignores the fact that the chloride mass balance method for calculating infiltration rates produces values that are in the range of values calculated on the basis of physical measurements (Flint et al., 1996). To the contrary, a detailed analysis of pore water chemistry data (Meijer, 1996), has lead to the opposite conclusion that pore waters in the unsaturated zone at Yucca Mountain show very little evidence of reaction with the host rock.

Cl/Br: One other aspect of water chemistry that is of interest in this discussion is the ratio of chloride to bromide in waters within the Yucca Mountain flow system. Because both of these constituents show conservative behavior in ground water systems, their ratio can be used to differentiate waters of different origins. This ratio has been measured in many different waters by Fabryka-Martin et al. (1996, and personal comm.) and Yang et al. (1996). The available data suggest Cl/Br ratios in saturated zone waters at Yucca Mountain are in the range of 120 to 150 with an average of approximately 130. Perched waters have a range of 25 to 120 with an average of approximately 100. Pore water ratios range from 162 to 428. The ratio in precipitation at Yucca Mountain averages about 74. At low infiltration rates, the concentrations are assumed to differ from the chloride values by a factor similar to that in the PTn pore water samples (200:1, chloride:bromide). In high infiltration zones, the fluids are assumed to enter the mountain at ratios typical of precipitation, namely 100:1, chloride:bromide.

These data suggest that Cl/Br ratios could be quite useful as a tracer in the Yucca Mountain flow system. For example, the Cl/Br ratio is one of the few chemical parameters that distinguishes perched waters from saturated zone waters.

7.6.3.3 The Stable Isotopes

In terms of stable isotopes $\delta^{18}\text{O}$ and δD , pore waters show a greater range and generally higher average values compared to perched waters (Yang et al., 1996). However, perched waters $\delta^{18}\text{O}$ and δD values are slightly higher than those measured in most of the saturated zone waters analyzed to date except J-13 and UE 29a#2. This is consistent with the fact that J-13 and UE 29a#2 waters and perched waters have similar apparent ages (i.e., 9-10 ka) and were therefore infiltrated under similar climatic conditions. The somewhat higher average $\delta^{18}\text{O}$ and δD values in perched waters are also consistent with the younger apparent ^{14}C ages for these waters (Yang et al., 1996) and a post-glacial climate.

Values for $\delta^{13}\text{C}$ have been measured in many water samples by Yang et al. (1996). This parameter is of importance to the interpretation of ^{14}C data because it provides an approximate means of assessing the amount of dead carbon that may have been incorporated into a given water sample. Yang et al. (1996) have used this parameter to “correct” apparent ^{14}C ages for inherited dead carbon. Unfortunately, the values obtained for this parameter in pore waters show rather large variability. Further, the pore water $\delta^{13}\text{C}$ values are commonly out of equilibrium with the $\delta^{13}\text{C}$ measured in the gas phase (Yang et al., 1996), which is problematic. Finally, there could be effects from drilling and the pore water extraction techniques that further complicate the analysis. The conclusion reached here is that “correction” of $\delta^{14}\text{C}$ “ages” using the $\delta^{13}\text{C}$ data is fraught with difficulties and the results should be interpreted with caution.

7.7 Simulation Results

In this section we present results of simulations of major ions present in unsaturated zone fluids, including chloride, sulfate, and bromide. To recap the conceptual model used to simulate these species, each are assumed to be conservative ions once they transport into the bedrock below the soil zone. The distinction between chloride, sulfate, and bromide is in the source term for fluid entering the bedrock. Chloride is assumed to be conservative even in the soil zone, whereas sulfate and bromide undergo reactions within the soil zone that lower their concentrations. For sulfate and bromide, these reactions could be biological in origin, whereas sulfate concentrations could also be affected by precipitation under conditions of evapotranspiration (Freeze and Cherry, 1979).

The concentration of dissolved species such as chloride are presumed in this modeling to be inversely proportional to the deep percolation rate, that is, the flow escaping evapotranspiration and entering the bedrock. This model assumes that the deposition rate of chloride is constant and not spatially variable, and that the controlling factor for concentration in the percolating fluid is evapotranspiration. Therefore, fluid entering at high infiltration rate has low chloride

concentration, and low infiltration correlates to high concentration. The concentration values themselves are based on the midpoint of the range of estimated values of chloride deposition rate of 75 to 150 mg/m²/y reported by Phillips (1994).

For reasons discussed above, the ratios of sulfate and bromide to chloride appear to be lower than in precipitation for many pore water samples in the unsaturated zone. An exception is in the perched waters, which appear to have ratios more reflective of precipitation. To simulate this behavior, we postulate that in regions of high enough infiltration, evapotranspirative effects and reactivity of bromide and sulfate are minimal due to short residence times in the soil zone. This results not only in lower concentrations of major ions, but also a shift to ratios closer to that of precipitation in fluids that enter at high infiltration. Therefore, the concentrations in the infiltrating fluids are assumed to fall into two categories. At low infiltration rates, the concentrations are assumed to differ from the chloride values by a factor similar to that in the PTn pore water samples (1:1 for chloride:sulfate, and 200:1 chloride:bromide). In high infiltration zones, the fluids are assumed to enter the mountain at ratios typical of precipitation, namely 1:3 sulfate:chloride and 100:1 chloride:bromide. The cut-off infiltration rate is assumed to be 10 mm/y, above which the latter ratios apply. In essence, the ion ratios are used as an additional natural tracer in the simulations.

When this chloride concentration model is input at the top of the three-dimensional site model, the concentration entering the mountain is a strong function of the spatially varying chloride map as controlled by infiltration rate. Figure 7-4 shows the concentration along the top surface of the model (top figure) and in a horizontal slice at an elevation of 900 m (bottom figure). The concentration entering the system is a direct result of the correlation to infiltration rate described earlier. At depth, the concentration patterns are somewhat more uniform due to mixing of fluids with different concentrations. The values, in general, appear lower because the fluid that supplies the subsurface is heavily weighted toward the high infiltration (low concentration) values.

Figure 7-4. Chloride concentration predicted in the three-dimensional model. Top figure: map of chloride concentrations at the surface. Bottom figure: concentrations at and elevation of 900 m. Top of page is North.

In the surface chloride map, only a small fraction of the fluid actually entering the mountain has high concentrations.

Figure 7-5 shows the predicted chloride concentrations for a steady state dual permeability

Figure 7-5. EW cross-section of transport site-scale model looking North. Chloride concentrations for the base infiltration map (upper figure) and the 1/3 infiltration scenario (bottom figure).

model for the base case infiltration map of Flint et al. (1996), and a scaled map where all values are reduced by a factor of three. Within a given simulation, the concentration values in the subsurface are a reflection of the concentration entering at the surface, which in turn is inversely

proportional to infiltration rate. The pore water compositions measured in the PTn fluids are therefore strongly controlled by the infiltration rate at the surface.

An infiltration rate equal to one third (i.e., $I/3$) of the Flint et al. (1996) values, distributed over the entire site surface, was used to investigate the effects of infiltration rate on the behavior of the hydrologic system. The $I/3$ values were used as they provided the best fit to the $^{36}\text{Cl}/\text{Cl}$ values from the ESF (Chapter 6, present study). The values themselves are higher at all locations for the $I/3$ case, since the chloride deposition rate is held constant throughout this simulation exercise. At greater depths, there is some tendency for lateral diversion to result in lower concentrations beneath higher concentrations (see close-up in Figure), but in general, for the property values used in this calculation, the flow and transport is predominantly vertical.7-6)

Figure 7-6. Detail of EW cross-section of transport site-scale model in area of relatively low infiltration. Close-up of chloride concentrations to the East of the Ghost Dance fault for the $I/3$ scenario.

The simulations for bromide and sulfate are closely related to those for chloride, except for the presumed difference in ion ratio for fluids of infiltration rate greater than 10 mm/y. Figure 7-7

Figure 7-7. EW cross-section of full site scale transport model, looking North through Yucca Crest. Chloride to bromide ratio (top figure) and sulfate to chloride ratio (bottom figure) simulated using the base case property set and infiltration map.

plots the chloride to bromide ratio (top figure) and sulfate to chloride ratio (bottom figure) predicted by the model for the base infiltration case. Beneath high infiltration zones the ratio is that of precipitation, whereas elsewhere in the model the ratios reflect the presumed values in lower infiltration areas. Although there is a hint of lateral diversion at the TSw basal vitrophyre leading

to water with “precipitation ratios” underlying fluid with the low infiltration ratio, the effect is minor.

Although they explain some of the features observed in the data, the calculations presented so far, predict predominantly vertical downward flow, consistent with model parameters of the unsaturated zone flow model (Bodvarsson et al., 1997). However, such model results are seemingly at odds with data suggesting lateral diversion, including the perched water data (see Section 7.6). We now develop a model that includes perching due to lateral diversion at the TSw basal vitrophyre. We accomplish this by uniformly reducing the fracture permeability in this unit to $4 \times 10^{-17} \text{ m}^2$ while leaving the matrix permeability unchanged from the base-case value. This permeability is too low to support flow issuing from high infiltration regions, so that lateral diversion downdip from the source region occurs. To obtain perched water, we also reduce the permeability of the fault zones to values sufficiently low that they cannot drain the perched water. Figure 7-8 shows the steady state saturation profiles in the fracture domain for the base infiltration and the scaled 1/3 scenario. The perched water bodies can exist under steady state conditions for these hydrologic parameters. Higher infiltration rates lead to a more extensive perched water body adjacent to the Ghost Dance fault.

Chemical transport predictions for this type of perched water scenario are now presented. Figure 7-9 shows the simulated chloride concentration (top figure), sulfate concentration (middle figure), and sulfate to chloride ratio (bottom figure) in the fracture domain for the base infiltration case. Dilute fluids enter the mountain in high infiltration regions, travel laterally at the basal vitrophyre, and result in a perched water body that is low in concentration of sulfate and chloride (about 11 ppm chloride), and higher in sulfate to chloride ratio than fluids entering at lower infiltration regions. Thus, several aspects of the perched and pore water chemical composition data are explained well with this lateral diversion model.

Figure 7-8. Detail of EW cross-sections from the site scale transport model through Yucca Crest, looking North. Fracture saturations predicted using the basal vitrophyre lateral diversion model. Top figure: base infiltration map; bottom figure: 1/3 scaled map. Blue areas represent zones of perching with lateral flow along the top of the basal TSw vitrophyre. Principal source of water is from Yucca Crest in both cases.

7.8 Summary and Discussion

The LBNL and USGS models for flow in the unsaturated zone at Yucca Mountain emphasize fracture flow in which chemical constituents in the fracture waters have minimal interaction with the rock matrix (Rousseau et al., 1996; Bodvarsson et al., 1997). In addition,

Figure 7-9. Detail of EW cross-section (looking North) of transport site-scale model through Yucca Crest. Chloride concentration (top), sulfate concentration (middle), and sulfate to chloride ratio (bottom) for the basal vitrophyre lateral diversion hydrologic scenario.

Bodvarsson et al. (1997) believe that the matrix of the Calico Hills tuff will also be bypassed as downward percolating waters follow fracture pathways to the saturated zone. This has significant implications for transport modeling because the Calico Hills tuff has been thought of as, potentially, a significant barrier to radionuclide transport. The previous evaluations of retardation in the Calico Hills tuff are not consistent with available hydrologic and hydrochemical data. This section discusses an alternative position on the hydrologic behavior of the unsaturated zone at Yucca Mountain.

7.8.1 The Hydrologic Data

Whereas a substantial amount of hydrologic data exists for the Yucca Mountain area, the integrated flow and transport models that rely on these data cannot be calibrated on hydrologic data alone (Robinson et al., 1995, 1996, present study). Indeed, water saturation data can be fit to a wide range of parameter values. This is particularly true when advancing disequilibrium fracture flow as a dominant mechanism for water infiltration as the water saturation data is based on matrix saturations. Water potentials could be more useful but the data range greatly and are difficult to match (Bodvarsson et al., 1996, 1997). Hence, models calibrated only to hydrologic data are non-unique, and need additional constraints from hydrochemical data.

7.8.2 The Hydrochemical Data

As noted above, relatively high ^{14}C activities have been measured in pore waters extracted from samples of the Calico Hills tuff (Yang et al., 1996; 1997). These activities imply apparent ages of 300-3,000 years. Many of these pore waters were extracted from samples that were strongly zeolitized (e.g., USW UZ-14). These observations imply that relatively young waters are percolating into and presumably through zeolitic zones in the Calico Hills Tuff everywhere that measurements have been made. This observation points to the fact that the Calico Hills matrix is not bypassed by water migrating to the water table, in contradiction with some conceptual models. It also points to the fact that the pathways, volumes of water and their residence times can only be

constrained using environmental chemical tracers; synthesis and integration of hydrochemical data provides more robust conceptual models.

Chloride concentrations and environmental isotope, stable isotope and radioisotope measurements in other regions of high infiltration at Yucca Mountain are required to evaluate the conceptual models presented to date for Yucca Mountain. At present, no such measurements exist and our ability to distinguish further between the various conceptual models for flow is impaired by this lack of data.

7.8.3 The Model Results

Several aspects of the perched and pore water chemical composition data are explained well with the numerical model presented above. Despite this encouraging start, several aspects of the model need improvement. First, the anion compositions in the pore water above the perched water body adjacent to the Ghost Dance fault are lower in the model than in wellbores such as USW UZ-14. However, since this cross section is to the south of USW UZ-14, the model is really a tool for exploring various conceptualizations of the processes giving rise to the chemical measurements, rather than a calibrated model capturing the data at any location. More importantly, this simulation predicts fluids with low concentrations beneath the perched water zones, whereas the data at USW UZ-14 suggests higher concentrations, and, perhaps more problematic, younger ages than the perched water. Since the fluid underlying the perched water in these simulations clearly results from slow percolation through the basal vitrophyre (a leaking perched water body), its age will be similar to that of the perched water itself, and thus inconsistent with the data. Finally, the age of the perched water in this steady state simulation is roughly 1,000 years, much younger than the measured values.

Several corrections to the hydrologic properties would be required to improve the model. First, a transient model is clearly in order, since the chemical signature in the pore and perched waters is suggestive of a residual imprint from the Pleistocene Epoch. A transient model would also explain the generally higher chloride compositions in PTn pore water, which, based on water

ages, clearly reflect infiltration from the past 1000 years or so. This model would require a significantly lower infiltration rate under present day conditions to prevent relatively modern water from completely flushing the system in the past 10 ka. Finally, younger waters beneath the perched water zones may indicate additional lateral diversion of fluid in the vitric Calico Hills matrix on top of zones of extensive zeolitic alteration. This relatively modern water could be passing laterally under the perched water zones, thereby resulting in waters of distinctly different origin than the perched water. In fact, the current three-dimensional site scale transport model predicts such lateral diversion, as shown for a particle pathway calculation in Figure 7-10. This lateral diversion

Figure 7-10. Three-dimensional simulation of transport for particles injected at the surface above the potential repository. Image represents a cut-away fence diagram from the three-dimensional grid. Represented are the Solitario (S.C.) and Ghost Dance (G.D.) faults and the ESF. Particles undergoing matrix flow are represented with the red colors corresponding to a high particle concentration. Significant lateral diversion is predicted in the vitric Calico Hills matrix (beneath the TSw basal vitrophyre) and to the Ghost Dance fault.

predicted in the vitric Calico Hills, beneath the TSw basal vitrophyre, is consistent with the observation that chloride concentrations and younger pore waters are observed from core samples taken beneath perched water bodies (Yang et al., 1996). This scenario is also consistent with the tendency for perching to occur to the west of the Ghost Dance fault. Therefore, an adjustment to the current model that will be explored is a combined model with the following features:

- partial lateral diversion at the TSw basal vitrophyre at higher infiltration;
- minimal lateral diversion at lower infiltration, such as present-day conditions;
- partial lateral diversion of fluid within the vitric Calico Hills.

A model with these features will be consistent with the transient mixing model for the hydro-geochemical system in the unsaturated zone.

7.9 Conclusions

In terms of extrapolation for Performance Assessment, the relevance of a conceptual model may be overlooked if the predictions overlap with other models. We have attempted in this chapter, to tie the different conceptual models to the fundamental physically-based differences among them so that extrapolations of the results can be better assessed. However, it is important to note that the weakest link in the analysis and evaluation of the different conceptual models, resides in the paucity of data for zones of high infiltration at Yucca Mountain (e.g., Yucca Ridge), and in some cases the reliability of both the hydrologic and geochemical databases on which the models are based.

The exploration of these alternative explanations for the perched and pore water chemical data are important in that different model conceptualizations may give rise to different performance predictions. We believe that when combined with the hydrologic database, the chemical data provide the most relevant information for “validating” radionuclide transport models, because the movement of naturally occurring solutes is closely related to the potential

migration of radionuclides. Our philosophy for model development and testing is one in which these data are used in the building of conceptual models, rather than as data to calibrate models against. The sparseness of the data sets and the ambiguities regarding the processes giving rise to the chemical composition preclude a more formal calibration exercise at this time. Therefore, we will examine these and other conceptual models for explaining the chemical data in future studies, attempting to pin down the conceptual model for flow and transport that is most consistent with all observations.

The overall consequence of the "fast pathway" fracture flow hypothesis is that it predicts poor performance for the unsaturated zone, as the Calico Hills transport barrier is essentially bypassed by fracture flow. It is important to note however, that the flow model which encompasses this hypothesis (Bodvarsson et al., 1997), and the transport simulations using the hydrologic parameters derived from that model (present study), are not uniquely calibrated as fracture and fault properties are still unknowns. Indeed the flow model calibrations were accomplished using measured matrix properties and fracture and fault properties derived from air permeability measurements. Bounding fracture and fault properties therefore relies on additional sources of data, and in particular the use of hydrochemistry. This is because the modern and ancient flow pathways have left behind a chemical imprint in the mountain, that if deciphered will help bound the effective properties and behavior of the unsaturated zone at Yucca Mountain. The transient-mixing hypothesis was designed to capture all elements of the hydrologic and chemical databases at Yucca Mountain. Its main strength is that it explains simultaneously the inversion in apparent ages between pore waters and perched water in the unsaturated zone, and the low chloride content of perched waters relative to the overlying TSw and underlying CHn pore waters. The apparent disequilibrium in pore water and perched water contents is consistent with a model of transient flow, and it has been demonstrated that the stable isotope data and apparent ^{14}C and $^{36}\text{Cl}/\text{Cl}$ age determinations can be interpreted as being caused by a mixture of late Pleistocene/early Holocene to modern perched waters. The existence of these mixed ancient/modern perched water bodies therefore implies that an additional retardation mechanism exists above the Calico Hills formation.

The ubiquitous young ^{14}C ages in the Calico Hills formation pore waters beneath the perched water bodies and in other areas, also imply that a lateral component of flow occurs in the vitric Calico Hills and that this formation is not bypassed. Finally, this model is consistent with the 3-D stochastic mineral distribution model (Robinson et al., 1996; Chipera et al., 1997a; see also Chapter 12, present study) in which zeolitic and vitric tuffs are intricately interbedded beneath the repository horizon.

The implication of this model for performance assessment is improved performance of the Calico Hills transport barrier beneath the repository by 1) the addition of an additional barrier component which regulates the downward flow of water to the saturated zone, and 2) increased water residence times in the unsaturated zone due to the presence of long term perched water bodies at depth (i.e., no flow), and lateral diversion above the basal vitrophyre and/or the underlying Calico Hills vitric units. This should result in increased sorption of radionuclides at those boundaries as actinides sorb in clays and vitric tuffs as well as in zeolites (Triay et al., 1996a).

In conclusion, it appears that although the fracture and fault properties of Yucca Mountain tuffs require further refinement, the hydrochemical database is consistent with a model in which the flow system can be represented by an interlayered sequence of units characterized by matrix or fracture dominated flow, depending on infiltration rate. The current integration of the databases at Yucca Mountain continue to support a conceptual model for waste isolation, based on the concept of multiple and redundant engineered and natural barriers to radionuclide migration. It is suggested that the unsaturated zone remains one of the principal natural barriers to radionuclide migration to the accessible environment and that the Calico Hills formation and the overlying basal vitrophyre of the Topopah Springs unit play an important role in that process.

Chapter 8 - Two-Dimensional Radionuclide Transport Simulations

8.1 Abstract

Multiple parameter sets from the LBNL site-scale flow model and variations on the USGS infiltration map are used in the simulation of radionuclides from the potential repository to the water table. For most of these parameter sets, radionuclides tend to bypass the zeolites of the Calico Hills via fracture transport when base-case infiltration conditions are simulated. When the infiltration rate is scaled by $1/3$, significant fracture flow in the zeolites is still simulated with most parameter sets. A modification increasing CHn zeolite matrix permeabilities by a factor of 5 leads to substantially greater matrix flow and, hence, retardation of sorbing radionuclides. This process leads to dramatic improvement in the performance of the unsaturated zone. These studies demonstrate the overriding importance of proper characterization of the zeolitic units for performance assessment. Colloid-facilitated transport of otherwise immobile radionuclides like Plutonium is also addressed in this chapter. In the absence of sorption onto the mobile colloids, Plutonium migration is limited due to large sorption coefficients. When partitioning onto colloids approaches 99% or more, Plutonium mobility becomes potentially important to performance. Long-time transients capturing climate change related phenomena are also simulated in this chapter. The results show that the flux of radionuclide at the water table under transient conditions is closely approximated by a quasi-steady state model. Finally, for wetter future climates, the combined effect of a shorter travel distance to the water table and high fluxes under a wetter climate scenario yield prediction of poorer unsaturated zone performance than for the present climate conditions.

8.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al., 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 8-1.

Table 8-1. QA Status of Data used in this Chapter			
REFERENCE	Q Status	DTN	COMMENT
Bodvarsson et. al., 1997 "The Site-Scale Unsaturated Zone model of Yucca Mountain, Nevada, For the Viability Assessment, LBL Technical Report LBNL-40378	Q	LB970601233129.001	No DTN assigned. Hydrologic property sets computed in this report are used throughout the present study.
Chipera et. al., 1997b "Mineralogic Variation in Drill Holes USW NRG-6, NRG-7-7a, SD-7, SD-9, SD-12, and UZ#14: New Data from 1996-1997 Analysis Los alamos National Laboratory YMP Deliverable SP321BM4"	Q	LADV831321AQ97.001	
Triay et. al., 1996a , "Summary and Synthesis Report on Radionuclide Retardation for the Yucca Mountain Site Characterization Project, Los Alamos National Laboratory YMP Milestone Report 3784	Q	LAIT831341AQ96.001	
Triay et. al., 1996b , "Radionuclide Sorption in Yucca Mountain Tuffs with J-13 Well Water,: neptunium, Uranium, and Plutonium, Los alamos National Laboratory Yucca Mountain Site Characterization Project" Milestone 3338, Technical Report LA-12956-MS	Q	LAIT831341AQ96.001 *	* DTN supersedes DTN LA000000000104.001
Triay et. al., 1996c , "Batch Sorption Results for neptunium transport through Yucca Mountain tuffs, Los alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3349, Technical Report LA-12961-MS	Q	LAIT831341AQ96.001 *	*DTN supersedes DTN LA000000000090.001

Triay et. al., 1996d, "Comparison of Neptunium Sorption Results using batch and column techniques, Los Alamos National laboratory Yucca Mountain Site Characterization Project Milestone 3041, Technical Report LA-12958-MS	Q	LA000000000106.001	DTN Accession No. MOL.19950630.0300
---	---	--------------------	--

8.3 Introduction

In this chapter we use the two-dimensional, East-West cross section model to examine the predicted behavior of radionuclides migrating through the unsaturated zone from the potential repository to the water table. In this report we use a combination of one-, two-, and three-dimensional simulations to evaluate the sensitivity of transport to many uncertain aspects of the unsaturated zone. One-dimensional simulations were used mainly for mechanistic and screening studies, where the number of parameters or the time scale of the process being studied precludes performing the simulations in two or three dimensions. The two-dimensional simulations presented in this chapter examine behavior that does not necessarily require full three-dimensional simulations, including the following uncertainties and processes:

- hydrostratigraphy and its effect on radionuclide transport;
- percolation flux;
- different hydrologic parameter sets;
- transport parameters;
- long-term transients due to climate change;
- heterogeneities and the distribution of zeolites;
- repository heat effects; and
- near-field environment (NFE) radionuclide release rate.

8.4 Methodology for Radionuclide Migration Calculations

In the present study the particle tracking module in FEHM is used to simulate radionuclide migration from the potential repository to the water table. The reasons for using this module for transport relate to the extensive use of the dual-permeability model formulation in the present study. In Chapter 4, we discuss in detail the computational advantage of the cell-based particle tracking technique for unsaturated zone dual-permeability transport modeling is discussed.

Green's Function Analysis Method

The primary type of simulation presented in this chapter is a steady state flow field simulation, followed by particle tracking on this flow field. Some individual analyses are performed to test the steady state flow assumption by examining the possibility of climate-induced, long-term transients (Section 8.1.1), heat-induced transients (Chapter 11), and episodic transients in Chapter 6. However, for steady state flow, the flow and radionuclide transport system can be decoupled from the NFE (near-field environment) source term through the use of convolution. Instead of determining the transport of a radionuclide for each NFE release scenario, we compute the response of the system to an instantaneous release of a unit mass of radionuclide at time 0. Generally, the response is the resulting radionuclide mass flux at the water table resulting from the instantaneous injection of particles at the potential repository. This response curve, $f_{UZ}(t)$, is a Green's function (e.g. Wylie, 1975), which can be used as the kernel of a convolution calculation in which the time-varying source term mass flux, m_{NFE} , is applied to the following equation:

$$m_{UZ}(\tau) = \int_0^{\tau} m_{NFE}(t) f_{UZ}(\tau - t) dt \quad (\text{Eq. 8-1})$$

In words, Equation 8-1 states that the radionuclide mass flux at the water table is the sum of the contributions of all paths of different residence times of radionuclides that entered the system that long ago. The primary assumptions underlying the convolution approach are steady state flow and a release rate that is independent of position in the repository. The more complex cases are consid-

ered separately and do not use this convolution approach.

The instantaneous injection breakthrough curve f_{UZ} will be presented first for a number of hydrologic cases, infiltration rates, and transport parameters, but in the presentation of the two-dimensional base case, we also show the nature of the convolution results for an actual release scenario. In this way, an example is provided that will allow the reader to interpret the instantaneous injection breakthrough curve results more readily. The integral of the f_{UZ} curve, is also presented which is a cumulative residence time distribution function. This curve demonstrates the extent to which radionuclides take rapid pathways to the water table, compared to slower pathways.

8.5 The Base Case

The Ten Percent Zeolite Map

Because zeolites are formed through secondary alteration, the distribution of zeolites in the Calico Hills do not follow the primary geologic stratigraphy (Chipera et al., 1997b). Therefore, we map the zeolite distribution of Chipera et al. (1997b) on top of the geologic model as described in detail in Chapter 2. In Chapter 12 we investigate the relationship between percent alteration, permeability changes due to alteration, and the effect on radionuclide transport. In this chapter, we choose a threshold percent alteration and assign zeolitic parameters according to the unit in which the alteration occurs. For example, the first geologic layer in the CHn below the basal vitrophyre contains both altered and unaltered minerals. If a location in this unit is identified as having zeolitic alteration greater than the threshold value, 10% in this case, then cells in the grid are assigned CHn1z (zeolitic) properties while all other cells in that layer are assigned CHn1v (vitric rather than zeolitic hydrologic properties). Thus, each cell in each geologic unit in the CHn can be either zeolitic and non-zeolitic depending on where it falls. This approach is a major improvement to the previous approach of representing entire layers in the CHn as either zeolitic or nonzeolitic.

Property Sets

In this chapter, the hydrologic property set 6541 is regarded as the base case for the purposes of comparison and the execution of variations from the base-case conditions. Recall that this property set was derived by LBNL (Bodvarsson et al., 1997) using a calibration to matrix fluid saturations assuming the base-case infiltration map of Flint et al. (1996), with upstream saturation weighting for the reduction factor for fracture/matrix flux in nonwelded units, and upstream relative permeability weighting for the reduction factor for fracture/matrix flux in the welded units. This parameter set is used for this base-case analysis because both PA and site characterization scientists identified it as most appropriate. This set, and all other sets from LBNL, contain hydrologic properties for both zeolitic and non-zeolitic material in each CHn geologic unit. Therefore, with the threshold approach described above, the properties of each cell in each layer are obtained from the LBNL property set being used for the particular simulation. All property sets referred to in this chapter are described in Table 5-2.

Simulations

The steady-state fluid flow field is generated after mapping the spatially varying infiltration values (see Figure 6-6, this report) onto the two-dimensional domain. The steady-state matrix and fracture saturations for this case are shown in Figure 8-1 along with the infiltration maps. Estimated infiltration rates above the potential repository are highest at, and just east of, the crest of Yucca Mountain (Flint et al., 1996). These higher infiltration rates have ramifications for radionuclide migration, as shown below. The basic pattern of fluid saturations in the matrix is typical of the models presented in Bodvarsson et al. (1997), and, in general, follow the results from the one-dimensional results in Chapter 5. Fracture saturations are controlled by the matrix permeabilities (what cannot pass through the matrix must percolate through the fracture domain), the local percolation rate, and the fracture properties. In addition, the fracture/matrix interaction term controls the ability of fluid to transfer from fracture to matrix and back at the interface

Two-Dimensional Radionuclide Transport Simulations

Figure 8-1. Matrix (top) and fracture (bottom) saturations for the two-dimensional base-case simulation. Parameter set 6541, base infiltration map (I). Also shown are the I and I/3 infiltration map values across the model domain.

between units of contrasting properties. The implications of this effect for flow and transport in nonwelded tuff were addressed in Chapters 5 and 6 of this report.

Radionuclide transport from the repository to the accessible environment is simulated using the steady-state flow field. The repository is identified as a series of point sources located 22 m apart at the repository horizon. An equal number of particles is released at each node at time 0. The simulation is carried out to one-million years, and the exit time of each particle is written to an FEHM output file where the particle times are sorted and binned to produce the residence time distributions (the Green's function). Figure 8-2 shows the results for the base case for different

Figure 8-2. Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map.

assumed sorption coefficients in the zeolitic tuff (no sorption occurs in the other units for this case). The assumption of sorption only in the zeolites is appropriate for ^{237}Np and uranium (Triay et al., 1996a), with values of 2 cc/g typical of ^{237}Np , and values ranging from 5 to 20 cc/g for uranium. The no-sorption case is indicative of the migration of non-reactive radionuclides such as ^{99}Tc , ^{127}I , and perhaps ^{14}C . Sorption occurs in the matrix only, either as a retardation of the portion of the radionuclide travelling through the matrix by advective transport, or via sorption in the matrix diffusion model of fracture transport. For the matrix diffusion model, the matrix diffusion coefficient and fracture aperture are required: for this case they are $10^{-11} \text{ m}^2/\text{s}$ and 0.1 mm, respectively. Recall from the one-dimensional screening analysis in Chapter 5 that for the property sets supplied by LBNL, the curves exhibited a distinct bimodal character, with rapid transport through fractures and long-time pathways through the matrix. This behavior is also exhibited in the two-dimensional simulations. With no sorption (the $K_d = 0$ curve), a small fraction of the inventory travels quite rapidly to the water table (less than 100 years), but the main portion of the first peak takes roughly 1000 years to reach the water table. This is roughly the travel time of the radionuclide through the matrix of the vitric nonwelded tuffs. Sorption onto the zeolites has little impact on this peak, since the early-arriving radionuclide travels through fractures, where it has only limited contact with the zeolitic tuffs in the matrix diffusion model.

The second peak is associated with matrix flow and transport. Without sorption, this peak arrives at roughly 10 ky, and for sorption onto zeolitic tuffs, the peak is pushed out to much larger residence times. The relative proportions of radionuclides arriving at the water table in each peak is more easily seen in Figure 8-3, the cumulative breakthrough curve. The first plateau in each curve represents the fraction of the radionuclide inventory that travels rapidly through to the water table. For the base-case parameters and infiltration rate, roughly 70% of the mass travels through these paths. Sorption onto zeolites affects this value very little, since there is not much contact of radionuclides with the zeolitic matrix for these flow paths.

Figure 8-3. Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map.

Before presenting the sensitivity analyses for the uncertain parameters discussed, the water table breakthrough curve for a more typical release scenario is presented. For this calculation particles at a uniform rate across the repository starting at 1,000 years for a period of 30 ky are released, after which the inventory is depleted and the releases stop. This case simulates a 1,000 year canister lifetime and solubility-limited release thereafter. This release scenario and a zeolite K_d of 2 cc/g is a typical scenario for ^{237}Np . Figure 8-4 shows the results for this simulation for the base case. Breakthroughs at the water table commence at slightly greater than 1,000 years, and plateau to a constant value 5 to 10 ky. This plateau persists until the releases stop at 31 ky. During this period, all fast pathways associated with the first peak in the Green's function are supplying

Figure 8-4. Water table breakthrough of ^{237}Np for the base case. Parameter set 6541, base infiltration map, $k_d = 2$ cc/g, constant release rate from 1 ky to 31 ky.

radionuclides at the water table, and the breakthrough curve, thus, resembles the first plateau of the cumulative breakthrough curve shown in Figure 8-3. Once the inventory is depleted, these pathways no longer are supplied with radionuclides, and are in essence flushed out with fresh water. The remaining portion of the breakthrough curve comes from contributions to the slower moving pathways. Since the duration of the source term (30 ky) is much shorter than the spread in travel times associated with these pathways, the breakthrough of this portion of the inventory resembles the Green's function response (Figure 8-2, red curve), in contrast to the early breakthroughs.

Since this curve is the source term for the saturated zone transport system, we can foresee the ramifications of the current unsaturated zone flow model predictions on total system performance. Namely, if waste packages degrade on the order of 1,000 years after waste emplacement and the system's flow partitioning between fractures and matrix is as predicted with the current model parameters (6541), then the peak mass flux at the water table for radionuclides, such as ^{237}Np , will be reached within 10 ky. For this scenario, any parameter or process in the unsaturated zone (NFE, waste package, unsaturated zone system) that reduces release rates will directly influence total system performance. In addition, saturated zone flow or transport processes will be critical to characterize for their ability to dilute this initial breakthrough. The breakthrough of the second peak would appear to not be important for determination of peak dose for radionuclides, such as ^{237}Np , unless the first peak becomes significantly diluted and the compliance period is extended to 100,000 y or greater. This conclusion is based on radionuclide sorption onto zeolites (and no sorption elsewhere), and does not apply to other more sorptive radionuclides.

For non-sorbing radionuclides such as ^{99}Tc , the breakthrough of the second peak occurs within 10,000 y (Figure 8-2, $K_d = 0$ case). Because this residence time is of the same order of magnitude as the time period over which radionuclides are released from the potential repository, the breakthrough curve will not exhibit the distinct bimodal character predicted for ^{237}Np . Rather, the entire inventory breaks through to the water table within about 50 ky, and the distinction between the fracture peak and the matrix peak becomes blurred. Thus, the influence of sorption for ^{237}Np is to delay the arrival of the matrix transport portion of the inventory. This is in effect a dispersion mechanism that lowers the peak dose to levels below what would be obtained in the absence of sorption.

Finally, we note that small portions of the inventory travelling through very fast pathways (less than 100 year travel times) have an insignificant impact on the water table breakthrough curves. If indeed the observations of bomb-pulse ^{36}Cl at the ESF represent only a small fraction of

the total flow, this analysis shows that these flow paths are mitigated by the small proportion of radionuclide mass travelling through them, as well as by slow release rates. In addition, the saturated zone will tend to dilute any rapid-moving radionuclide in the unsaturated zone system, as long as the proportion of mass travelling through these paths is small.

8.6 The Influence of Hydrologic and Geochemical Parameter Sets

The LBNL site-scale model report provided a variety of parameter sets, which were screened in Chapter 5 to eliminate redundant sets not likely to give rise to different behavior than the base case. In this section we examine two-dimensional transport for other parameter sets developed by LBNL. The goal is to determine if the method of model calibration or the form of the fracture/matrix interaction conceptual model influences radionuclide transport significantly. If it does, then this conceptual model uncertainty should be reflected in total system performance analyses since it would represent an important uncertainty.

Case 6412b: The Green's function curves for different sorption coefficients and the cumulative residence time distribution curves for this case are shown in Figures 8-5 and 8-6, respectively. The response curves are not significantly different than the base case. Recall that case 6412b employs a constant reduction factor in each unit as the multiplier in the fracture/matrix interaction model. At steady state, it is expected that the constant reduction factors obtained by parameter estimation should be similar to the weighting factor obtained with the alternative interaction models used for the base case, since both data sets were made to fit the available saturation data. Under transient conditions, the models could perhaps yield different results, but for steady state flow fields, it is not surprising that the breakthrough curves are quite similar.

Case 6531b: The Green's function curves for different sorption coefficients and the cumulative residence time distribution curves for this case are shown in Figures 8-7 and 8-8, respectively. Breakthrough occurs earlier and at higher levels in this case, and the bulk of the inventory travels rapidly to the water table within 10 ky. The only significant difference in the breakthrough curves

Figure 8-5. Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6412b, base infiltration map.

Figure 8-6. Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6412b, base infiltration map.

Two-Dimensional Radionuclide Transport Simulations

Figure 8-7. Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6531b, base infiltration map.

Figure 8-8. Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6531b, base infiltration map.

as a function of sorption coefficient is at early times, where the no-sorption case has a higher mass flux than the sorbing radionuclide cases. This is a characteristic of a system in which matrix transport is limited to the high permeability vitric units, and flow through the zeolites is almost exclusively fracture flow and transport. Therefore, sorption on zeolites has a relatively minor impact on the results for this case. In contrast to case 6412b, case 6531b exhibits significantly different behavior than the base case. It represents the extreme of fracture-dominated flow through the zeolites, with little influence of sorption. Of course, radionuclides that sorb onto the vitric units would be delayed relative to nonsorbing species.

These three cases represent how, under base-case infiltration conditions, the range of uncertainty in hydrologic parameter values results in different performance for the unsaturated zone. In general, zeolite permeabilities in these parameter sets are too low to transmit a large fraction of the flow. This minimizes the degree to which sorption of radionuclides such as ^{237}Np and uranium are retarded by sorption onto the zeolitic rocks. It must be recognized that the permeability of the zeolites are model-derived quantity, albeit constrained by measured data. If the upscaled permeability of a zeolitized region of rock exhibits a higher permeability than the averaged values obtained by parameter estimation techniques, then significantly more retardation is to be expected. The study of matrix hydrologic and transport properties in a heterogeneous region of varying zeolitic abundance is presented in Chapter 12. In that chapter, we demonstrate that if permeability is correlated with degree of alteration, greater matrix flow and, hence, retardation onto zeolites is predicted than occurs in this chapter's simulations.

8.7 The Effect of Percolation Flux

Percolation flux is one of the key uncertainty parameters that will influence the mass flux values and arrival time distributions at the water table. Strictly speaking, to simulate the flow field assuming a different percolation flux, a new set of hydrologic parameters should first be developed. However, given that matrix saturations are relatively insensitive to percolation flux

over the range of uncertainty at Yucca Mountain (about a factor of three higher or lower than the base case map), we assume that the hydrologic parameter sets developed using the base-case map can be applied directly for fluxes a factor of three higher or lower than the base case. This concept is demonstrated in Figure 8-9, a plot of the difference in predicted fluid saturations in the matrix

Figure 8-9. Difference in predicted fluid saturations from an increase in the base-case percolation flux map by a factor of three at all locations. Top figure: matrix saturation difference; bottom figure: fracture saturation difference.

(top figure) and fractures (bottom figure) for an increase in percolation flux of a factor of three at all locations on the base infiltration map. The matrix saturations, the only ones for which data are available, exhibit changes of less than 0.05 at almost all locations, with differences of 0.1 only in

a few locations. At the higher percolation rate, the fracture saturations increase more dramatically, a characteristic of increased fracture flow. Nevertheless, we conclude from this comparison that as a first approximation, the use of the parameter values determined with the base map is warranted for this range of percolation flux values.

This analysis begins with a set of calculations for the base case parameter set (case 6541) assuming an increase by a factor of three (called the “3 x I” case) and a decrease by a factor of three (1/3). Figures 8-10 and 8-11 show the Green’s function and cumulative breakthrough curve for the

Figure 8-10. Green’s function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, 3 x I infiltration map.

3 x I case, and Figures 8-12 and 8-13 are the same curves for the 1/3 case. As expected, the percolation flux is a key parameter controlling the transport response. At the higher percolation flux, the first peak of the breakthrough curve occurs much earlier (less than 1000 years), and perhaps more importantly, a greater fraction of the inventory travels through the fast pathways, thereby bypassing the zeolites. This observation is most easily seen in Figure 8-11 for the 3 x I case

Figure 8-11. Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, 3 x I infiltration map.

Figure 8-12. Green's function residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, I/3 infiltration map.

Figure 8-13. Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, I/3 infiltration map.

(the converse is true for the I/3 case in Figure 8-13), the cumulative breakthrough curve. For this parameter set, even the I/3 case predicts a significant fraction of radionuclides traveling through the faster pathways. This is caused by the low zeolite permeabilities derived in the parameter estimation simulations combined with the still high percolation flux values predicted above the potential repository (up to 6 mm/yr).

Since the possibility exists for lower percolation flux values than predicted by the base case map, we continue to explore the ramifications of lower percolation for the other parameter sets. It is important to realize that higher infiltration is also possible, as in the 3 x I case, but given the behavior of each parameter set for the base case map, there is little point in further testing at the higher values. At higher flux values, the unsaturated zone transport is dominated by fractures, and travel times are short. We conclude that unless these parameter sets underestimate significantly the permeability of the zeolitic rocks, fracture flow through these horizons dominates transport at the

base infiltration rate or higher rates. If bulk fracture permeability is also low in the zeolites, then lateral flow would be induced.

For comparison, we present transport results for the $I/3$ infiltration map for the remaining parameter sets in Figures 8-14 and 8-15. The results presented in this figure, when compared to the base-case results, illustrate the degree of uncertainty in transport caused by the various parameter sets and conceptual models. Comparing cases 6412b and 6412c, we note that the adjustment of parameter values in the vitric units below the potential repository has minor effect on the transport behavior. This effect is in spite of a difference in the simulated matrix saturations of the two models, shown in Figure 8-16, in plots of the matrix saturations, and the difference in saturations for the two models. Although the property values resulted in lower saturations in these units (apparently more in keeping with the measured data), the distribution of flux in fracture and matrix in these units and the zeolitic rocks apparently did not change significantly.

Case 6522 parameters were developed by Bodvarsson et al. (1997) assuming that the infiltration rate is one-fifth that of the base-case map. It is being used here for the $I/3$ case, assuming that the somewhat different infiltration rates have little influence on the calibration. The Green's function is also similar to the previous cases, except for a somewhat greater fraction of the inventory travelling through the faster pathways. The only case to exhibit significantly different behavior for the $I/3$ infiltration scenario is case 6531b, the upstream saturation weighting parameter set. This set exhibited significantly more rapid transport than the others in one dimension and at the base infiltration rate. This trend continued to hold for this infiltration rate as well.

8.8 Grid Refinement Study

An accurate transport solution requires that the numerical grid be discretized finely enough to capture accurately the details of the flow and transport system. Two types of grid errors are possible. First, a coarse grid can smear the details of a complex, heterogeneous flow and transport

Two-Dimensional Radionuclide Transport Simulations

Figure 8-14. Green's function (left column of figures) and cumulative residence time distributions for the I/3 infiltration map for various parameter sets. a, b) 6412b; c, d) 6412c.

Two-Dimensional Radionuclide Transport Simulations

Figure 8-15. Green's function (left column of figures) and cumulative residence time distributions for the I/3 infiltration map for various parameter sets. a, b) 6522; c, d) 6531b.

parameter field, requiring spatial averaging to capture with a single set of parameters a complex system with widely contrasting parameter values within each computational grid element. Even for

Figure 8-16. Comparison of matrix saturations for cases 6412b and 6412c. $I/3$ infiltration map. Top figure: 6412b; middle figure: 6412c; bottom figure: difference in saturations (6412c - 6412b).

Two-Dimensional Radionuclide Transport Simulations

a layered system with homogeneous properties within each unit, several computational grid points may be required to capture the details of a complex flow and transport system. This grid resolution numerical accuracy issue is examined here, for the ten percent zeolite map on the same refined grid used in Chapter 12 for the heterogeneity study where all zeolite alteration percentages are considered.

In this calculation we map the zeolite distribution defined as the 10% zeolite case onto each grid, and compute steady state fluid flow transport to the water table for both cases. Therefore the only difference is the numerical grid in the region at and below the potential repository. Figures 8-17 and 8-18 show the results for the two models for K_d values of 0, 2, and 10 cc/g. The responses

Figure 8-17. Comparison of Green's functions (base-case grid and refined grid) for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map.

are quite similar for the two cases in terms of the peak arrival times and general shapes. The refined grid exhibits a slightly higher peak value with less dispersion. However, recall that when an actual NFE source term is applied in these models, the area under this peak (or the plateau value in the

Figure 8-18. Comparison of cumulative residence time distributions (base-case grid and refined grid) for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map.

cumulative breakthrough curve) is more important than details regarding the shape. Figure 8-18 shows that, based on this criterion, the responses are quite similar. Therefore, we conclude that the coarser grid is adequately resolved for the purposes of this study. The refined grid is still necessary for the study of the impact of heterogeneities presented in Chapter 12.

8.9 Uncertainties in Zeolite Distributions and Property Variations

No parameter in the site-scale flow model is known to a high degree of precision. Therefore, it is important to understand the impact of uncertainty in key parameters on the performance of the unsaturated zone as a barrier to radionuclide migration. One aspect of the system that has presented itself most noticeably in this version of the site-scale radionuclide transport model is the prediction that a significant portion of the inventory may bypass the zeolites through fractures. This result has occurred due to the low matrix permeabilities and the weak coupling between fractures and matrix brought on by the new fracture/matrix coupling models.

The implications are important for both nonsorbing and sorbing radionuclides. Nonsorbing species, such as ^{99}Tc , travel more rapidly to the water table because of the increased velocities in the fractures of the zeolitic units, rather than slow matrix percolation. Radionuclides, such as ^{237}Np and uranium, also sorb onto zeolites, so the impact of fracture flow is even more pronounced. Therefore, one of the critical uncertainties is the distribution and hydrologic property values for zeolitic tuffs.

To study the influence of property value uncertainty, we simulated a case in which the parameter set 6412c was revised to increase the matrix permeability of each zeolitic unit by a factor of five. We call this simulation case 6412d. Both property sets are described in Table 5-2. The justification for this level of increase is that the measured values easily spans a range that encompasses this variation, and that the LBNL parameter estimation results imply that a rather wide range of zeolite matrix permeabilities could fit the data almost equally as well. Of course, this logic would imply that either an increase or decrease is warranted. We choose to study only the increase because even for the base case values, the zeolites are being bypassed to a large extent. Lowering the zeolite matrix permeability would not fundamentally change this result.

Figures 8-19 and 8-20 show the Green's function and cumulative residence time distributions for this case assuming the I/3 infiltration map, and Figures 8-21 and 8-22 are the results for the base infiltration case. The performance of the unsaturated zone improves dramatically for this variation in property values, especially when combined with the I/3 infiltration map. The nonsorbing radionuclide has first arrivals greater than 1,000 years and the peak occurs at about 10 ky. In essence, due to matrix flow through the zeolites, the first peak in the breakthrough curve vanishes, and virtually all radionuclide travels slowly through the matrix below the basal vitrophyre. This dramatically improves the performance with respect to radionuclides that sorb to the zeolites, as shown in Figures 8-19 to 8-22. For example, peak breakthrough for the $K_d = 2 \text{ cc/g}$ case (a typical value for ^{237}Np) occurs at about 100 ky for this case. For the base infiltration map, performance is not as good (Figures 8-21 and 8-22), but the

Figure 8-19.Green's function for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), I/3 infiltration map.

Figure 8-20.Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), I/3 infiltration map.

Figure 8-21.Green's function for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), base infiltration map.

Figure 8-22.Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6412d (high-K zeolites), base infiltration map.

fraction of radionuclide travelling through the rapid pathways is still greatly reduced from the base case parameter values. Clearly, the properties of the zeolites is of overriding importance to the performance of the unsaturated zone.

8.10 Plutonium Colloid Transport Analysis

In this section we expand upon a topic in which significant advances have been made recently in our ability to capture the potentially important process of colloid-facilitated transport. The Unsaturated Zone (UZ) Flow and Transport Workshop conducted by TSPA personnel identified the issue of colloidal transport as a key uncertainty in transport of radionuclides in the unsaturated zone, reiterating statements made for many years by scientists at Los Alamos and elsewhere. Specifically, the workshop participants concluded that in order to develop a credible model for plutonium movement in the UZ, 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 PA 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 must be included in future analyses. Even if the “bottom line” remains the same (i.e. that plutonium does not contribute significantly to the predicted dose at the accessible environment), including colloidal transport will add an additional degree of credibility to the analyses.

At the PA Workshop, a model of plutonium migration was proposed that included aqueous transport, kinetically controlled sorption of plutonium onto natural and man-made colloids, as well as transport as radiocolloids. However, simpler models are needed to fulfill several goals:

- scoping the problem to identifying key processes and parameters;
- providing simple analytical solutions to check against the more sophisticated models when

they are developed;

- perhaps to provide a simplified model to PA that could be used to bound the problem for TSPA-VA (this goal would be realized only if the simple model was indeed a defensible abstraction of the more complex model).

In this section we derive an analytical solution, 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 for which colloid-facilitated transport is possible. This model is then incorporated into the site-scale transport model to predict plutonium migration. More sophisticated models and analyses that include estimates of specific parameters for radionuclides such as plutonium will follow at a later date.

8.10.1 Model Development

The model developed herein is a one-dimensional transport model of a contaminant travelling in a fractured medium. 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 unsaturated zone 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, a scenario not ruled out in present site scale models.

Aqueous transport: We first consider a model available in the literature for the one-dimensional 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 8-23). The transport equation to be solved in the fracture assumes the one-dimensional advective-

Figure 8-23. Schematic of the fracture transport model (after Tang et al., 1981)

dispersion equation, diffusion into the porous rock matrix, equilibrium, and linear sorption on the fracture surface:

$$R_f \frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - \frac{q}{b} \quad (\text{Eq. 8-2})$$

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 $2b$ is the fracture aperture. Assuming no advective flow of fluid in the matrix, the transport equation for the contaminant is:

$$R_m \frac{\partial c}{\partial t} = D_m \frac{\partial^2 c}{\partial y^2} \quad (\text{Eq. 8-3})$$

where c (lower case) is the concentration of contaminant in the matrix, y is the direction perpen-

pendicular to the flow direction, R_m is the retardation factor for the matrix, and D_m is the 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:

$$q = -\theta D_m \frac{\partial c}{\partial y} \Big|_{y=b} \quad (\text{Eq. 8-4})$$

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. In this paper, we give the result for a position $z = L$ in the fracture, presumed to be at the water table:

$$\frac{C}{C_{in}} = \frac{2 \cdot \exp(Pe/2)}{\pi^{1/2}} \int_l^\infty \exp\left[-\xi^2 - \frac{Pe^2}{16\xi^2}\right] \text{erfc}\left(\frac{PeA}{4\xi^2 \sqrt{\frac{t}{\tau} - \frac{PeR_f}{2\xi^2}}}\right) d\xi \quad (\text{Eq. 8-5})$$

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 ,

$$l = \left(\frac{\tau Pe R_f}{4t}\right)^{1/2} \quad (\text{Eq. 8-6})$$

and

$$A = \frac{\theta(\tau R_m D_{mol})^{1/2}}{2b} \quad (\text{Eq. 8-7})$$

Equation 8-6 also makes use of the relation between D_z and dispersivity α , namely that

$D_z = \alpha v$, 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

$$\frac{C}{C_{in}} = \operatorname{erfc}\left(\frac{A}{\sqrt{t/\tau - R_f}}\right) \quad t > R_f \tau \quad (\text{Eq. 8-8})$$

with $C/C_{in} = 0$ for $t \leq R_f \tau$. Although the finite dispersion model Equation 8-5 will be used for the simulations presented below, Equation 8-8 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.

Colloid-facilitated transport: In this section we include a simplified colloidal transport model equation to derive a combined aqueous/colloidal transport model. The key assumptions of the colloidal transport sub-model are:

- 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;
- the colloids interact reversibly with the fracture surface.
- Given these assumptions, a transport equation for contaminant attached to colloids can be written as

$$R_c \frac{\partial C_c}{\partial t} = D_z \frac{\partial^2 C_c}{\partial z^2} - v \frac{\partial C_c}{\partial z} \quad (\text{Eq. 8-9})$$

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 8-9) to the aqueous transport equation (Equation 8-2), reversible, linear sorption of contaminant onto colloids is assumed:

$$C_c = K_c C \quad (\text{Eq. 8-10})$$

where K_c is the distribution parameter relating the concentrations. Since both C and C_c are expressed as moles 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 8-2 and 8-9, and making use of Equation 8-10, we obtain for the concentration of the contaminant in the aqueous phase for the fracture:

$$\left(\frac{R_f + K_c R_c}{1 + K_c} \right) \frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} - \frac{q}{b(1 + K_c)} \quad (\text{Eq. 8-11})$$

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 8-11 and 8-2 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 8-5 to 8-8) need only be modified slightly to include colloid contaminant transport. To do this, we define revised constants $\hat{R}_f$ and $\hat{A}$ as follows:

$$\hat{R}_f = \frac{R_f + K_c R_c}{1 + K_c} \quad (\text{Eq. 8-12})$$

$$\hat{A} = \frac{\theta(\tau R_m D_{mol})^{1/2}}{2b(1 + K_c)} \quad (\text{Eq. 8-13})$$

Using these expressions instead of their counterparts R_f and A allow us to consider colloidal

Two-Dimensional Radionuclide Transport Simulations

transport under dispersive conditions (Equation 8-5) or plug-flow fracture conditions (Equation 8-8). 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.

8.10.2 Results of Colloid Transport Simulations

One-Dimensional Simulation Results: This discussion begins with a general description of the features of the model for an aqueous contaminant traveling through a fracture, and then examine the differences between aqueous-only and colloid-facilitated contaminant transport. We use parameter values that we regard as possible for unsaturated zone transport through a fracture system directly connecting the repository to the water table. The range of values of each parameter are listed in Table 8-2 below:

Table 8-2. Potential ranges of hydrologic and transport parameter values

Parameter	Range	Note
τ	0.1-10 y	Example: 350 m flow distance, 3.5 mm/y through fractures, fracture volumetric water content of 10^{-5} : $\tau = 1$ year.
Dispersivity	1 - 100 m	0 is no dispersion (Equation (Eq. 8-8))
R_f	1-100	1 is no fracture sorption, 100 is possible for a strongly sorbing radionuclide
R_m	1-1000	1 is no fracture sorption, 1000 would result from $K_d = 100$ cc/g, rock density of 2 cc/g, and $\theta = 0.2$
D_{mol}	10^{-12} - 10^{-10} m ² /s	Low values are indicative of diffusion into low-water content matrix. High values are typical of measured values under saturated conditions.
b	0.1-10 mm	Poorly known parameter
θ	0.1-0.3	0.1 is the matrix water content for a welded, nearly saturated rock or nonwelded, partially saturated rock. 0.3 would be typical for a saturated, zeolitic rock. 0.2 used for all simulations in this report.
K_c	0-1000	0 no colloids or minimal sorption, 1000 is the equivalent of 999 parts out of 1000 residing on colloids

Two-Dimensional Radionuclide Transport Simulations

Table 8-2. Potential ranges of hydrologic and transport parameter values(Continued)

Parameter	Range	Note
R_c	1-100	Little known about colloid filtration or sorption in fractures

Some of these parameters should be considered to be correlated, even though the sensitivity analysis treats them as separate. Examples include the fact that fracture fluid travel time should be related to the aperture, and that diffusion coefficient is a strong function of matrix volumetric water content. Because the parameters are here assumed to be uncorrelated, the calculations presented below are to be considered a first step toward an understanding of the important parameters, rather than a sensitivity analysis that could be used directly in Performance Assessment activities.

Aqueous transport without colloids: To place the colloid transport calculations in perspective, we first present results of the transport model without colloids present. All transport results are presented as dimensionless breakthrough curves at the water table to a constant-concentration release at the repository. Figure 8-24 shows the results of the model for different

Figure 8-24. Influence of fluid residence time and dispersivity on breakthrough at the water table. No matrix diffusion or sorption.

fluid residence times in the absence of matrix diffusion. As expected, if retardation effects are neglected, the model reduces to conventional convective-dispersive behavior with a mean travel time equal to the fluid travel time. The dispersivity is varied from 1 m to 100 m for the 0.1 y fluid travel time case, and is shown to have very little impact on the breakthrough curve. Therefore, 10 m dispersivity is assumed for all subsequent simulations. If travel times are indeed this short, no amount of dispersion in the fractures will spread the transporting radionuclide significantly. Figure 8-25 illustrates the influence of sorption on fracture surfaces, which of course delays the breakthrough even in the absence of matrix diffusion.

Figure 8-25. Influence of fracture sorption on breakthrough at the water table. No matrix diffusion or sorption. Fluid travel time = 0.1 y.

When matrix diffusion is included in the model, increased travel times and greater apparent dispersion of the breakthrough curves are predicted. Figure 8-26 illustrates the influence of matrix diffusion coefficient on the breakthrough curves over the range selected to represent uncertainty in the unsaturated zone. Significant increases in travel times are predicted, even for the case of 1 y

fluid travel time. The influence of the matrix sorption retardation factor is identical to the effect of

Figure 8-26. Influence of matrix diffusion on breakthrough at the water table. Fluid travel time = 1 y. No matrix or fracture sorption.

diffusion coefficient, as seen by comparing the results of Figure 8-27 with those of Figure 8-26. This similarity is due to the equivalent roles played by R_m and D_{mol} in the mathematical development in the model (they appear as a product in the expression for A in Equation 8-5).

Colloid-Facilitated Transport: Colloid-facilitated transport due to sorption on existing colloid particles is suspected to be most important for strongly sorbing radionuclides such as plutonium. To illustrate the extent to which colloids could increase the mobility of plutonium, we examine the case in which the predicted large retardation of plutonium (the $R_m = 1000$ case in Figure 8-27) are influenced by colloids. In the absence of sorption of aqueous plutonium or retardation of colloids in the fracture itself, the fracture retardation factor $\hat{R}_f$ of Equation 8-12 is 1, regardless of the colloid distribution parameter K_c . However, the diffusion parameter $\hat{A}$

Figure 8-27. Influence of matrix sorption on breakthrough at the water table. Fluid travel time = 1 y. $D_{mol} = 10^{-12} \text{ m}^2/\text{s}$. No fracture sorption.

decreases with increasing K_c , so that sorption on colloids reduces significantly the travel times and apparent dispersion of the breakthrough curves (Figure 8-28). One way to assess the relative impact of colloid and matrix diffusion/sorption qualitatively is to compare the matrix retardation factor R_m to the term $1 + K_c$ in the denominator of Equation 8-13. If diffusion and strong sorption to the rock matrix is predicted to be important in the absence of colloid-facilitated transport, as is the case with plutonium when modeled as an aqueous radionuclide, then Equation 8-13 implies that colloid-facilitated transport will negate the benefits of matrix sorption as $1 + K_c$ approaches or exceeds $\sqrt{R_m}$. For plutonium, a K_d of 100 cc/g roughly corresponds to $R_m = 1000$, or $\sqrt{R_m} \approx 30$. For K_c values less than 1 (equal amounts of radionuclide on colloids and as an aqueous solute), the role of colloids in reducing matrix diffusion and sorption will be slight. Conversely, if 90% or more of the plutonium resides on the colloids ($K_c = 10$), the performance benefit of

Figure 8-28. Influence of colloid-facilitated transport on breakthrough at the water table. Fluid travel time = 1 y. $D_{mol} = 10^{-12} \text{ m}^2/\text{s}$. $R_m = 1000$. No fracture sorption.

matrix diffusion and sorption will be largely negated, and fracture transport will resemble that of a solute with little or no access to the rock matrix.

This analysis has so far neglected any retardation of aqueous or colloidal contaminant in the fracture itself. Equation 8-12 shows the interplay between sorption of aqueous plutonium on fractures and sorption on colloids. When fracture retardation factors are large ($R_f \approx 100$ or more), the earliest arriving radionuclide is delayed somewhat relative to the case of no fracture sorption (Figure 8-29). Also, Figure 8-29 also shows the performance benefit of assuming retardation of the colloids during transport. The effect of increased R_c appears small for this simulation, but the nature of the model suggests that the benefit of delay in colloids is present no matter how high the value of K_c . In other words, filtration or attachment/detachment processes is the last transport “barrier” to mitigate the deleterious performance brought on by colloid-facilitated transport.

Figure 8-29. Influence of colloid-facilitated transport on breakthrough at the water table.

Fluid travel time = 1 y. $D_{mol} = 10^{-12} \text{ m}^2/\text{s}$. $R_m = 1000$. Varying fracture sorption, colloid attachment, colloid sorption.

8.10.3 Discussion of Colloid-Facilitated Transport Results

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, we 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 radionuclides that form stable radiocolloids, the influence of introduced colloids must consider the interaction between the colloids and aqueous radionuclide.

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 the "base case" of aqueous transport. Colloid-facilitated transport is shown 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. The next step will be to update this study with estimated values of K_c based on field measurements of colloid concentrations and estimated or measured values of the sorption coefficient.

There are additional processes and proposed scenarios that are not directly handled with this model. One important scenario not discussed is the migration of plutonium radiocolloids, which are thought to be extremely stable in Yucca Mountain groundwater. This scenario can be handled by separating plutonium radiocolloids into their own category, one in which, in the absence of filtration, a radionuclide travels essentially at the fluid velocity in the fractures. The key parameter will then be the source concentration of radiocolloid. For both radiocolloids and pseudocolloids, a key uncertainty is the nature of the interaction of colloids with the rock, captured in the present model in the R_c term. It is perhaps too early to devote much effort to constrain R_c , given that the chemical interactions of radionuclides with colloids and the radiocolloid source term may be too small to degrade performance significantly. If future analyses uncover scenarios in

which performance is significantly impacted by colloids, then colloid filtration studies would be warranted.

Although the model is idealized, it can be used quite simply as a building block of a more sophisticated hydrologic model. We now present results of plutonium migration in the unsaturated zone that includes the influence of colloid-facilitated transport. 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 unsaturated zone.

The form of the mathematical model is identical to the fracture transport sub-model employed by the particle tracking module of FEHM. Therefore, with literally no changes to the code, colloid-facilitated transport can be included. Of course, the underlying conceptual model assumptions must be acceptable (valid, or at least bounding) in order for this approach to be used. The main assumption that must be justified is reversible, linear interaction between radionuclide and colloids. Irreversible or kinetically limited sorption onto colloids (or the rock) would limit the applicability of this model, although this could perhaps be handled approximately through adjustment of K_c . The benefits of using the same code, model hydrostratigraphy, and hydrologic flow fields for colloid-facilitated and aqueous transport of radionuclides are great enough that further work to justify the model assumptions or to determine that the assumptions are bounding is advisable.

When 2- or 3-dimensional site-scale transport simulations are performed with the dual permeability model formulation, the predicted partitioning of flow between fractures and matrix is treated, rather than the overly conservative assumption of pervasive fracture flow directly to the water table. Since in some units matrix flow predominates, model parameters characterizing colloid transport through the rock matrix are required. Sensitivity analyses are likely to show that

if colloid-facilitated transport is important, filtration during transport through the matrix of the CHn would be a key barrier to the movement of colloids. In future studies we will investigate this issue using the UZ flow and transport model. If colloids are deemed to degrade performance under assumptions of minimal filtration, then additional work will be required. Ultimately, an unsaturated zone field transport test using particulate tracers, such as was done in the saturated zone at the C-Wells, could provide critical information on filtration. For TSPA-VA, we will be at the stage of being able to address these uncertainties in models; these uncertainties could be reduced with a field-scale transport test, with results available in time for the Licence Application.

Figure 8-30 shows the Green's function for plutonium transport to the water table for case

Figure 8-30. Green's function for Plutonium transport through the unsaturated zone for different values of K_c . Case 6541, base infiltration map, aqueous plutonium $K_d = 100$ cc/g, all units.

6541, base infiltration map. The impact of the colloidal plutonium partitioning coefficient K_c is dramatic, with very little radionuclide even reaching the water table for $K_c = 0$ (no colloid-

facilitation). For progressively higher values of K_c , the breakthroughs occur at earlier times, and at much higher mass flux values. The effect is equivalent to lowering the sorption coefficient from its assumed aqueous value of 100 cc/g to values of 1 or less. Clearly, if most of the plutonium present travels in colloidal form, the impact on performance is substantial. When more than half of the plutonium is aqueous ($K_c < 1$), the predicted migration potential of plutonium is small, and the impact on overall system performance will be minimal. The cumulative breakthrough curves for this case are shown in Figure 8-31. Although there is some tendency toward very short travel times

Figure 8-31. Cumulative breakthrough curves for plutonium transport through the unsaturated zone for different values of K_c . Case 6541, base infiltration map, aqueous plutonium $K_d = 100$ cc/g, all units. Dotted curves are simulations with no matrix diffusion.

in the Green's function (on the order of 1 year to the water table), the cumulative curves indicate that this is only a very small portion of the plutonium inventory. Also plotted in this figure are simulations in which the effective sorption coefficient for matrix flow and transport is reduced due to colloids, and the fractures are assumed to exhibit no matrix diffusion. The close agreement

between these curves and their equivalent curves with matrix diffusion illustrates that the primary effect of colloid-facilitated transport is in reducing the effective sorption coefficient in units exhibiting matrix flow and transport, such as the vitric Calico Hills units. It is these units that provides the migration barrier to plutonium migration, rather than the units exhibiting fracture-dominated flow. Therefore, a conservative assumption of no matrix diffusion in fractures can be made, since the key uncertainties in the transport results reside in matrix-flow units.

Studies are underway to evaluate the possibility of colloid-facilitated transport of plutonium, with respect to the physical and chemical parameters that are likely to influence the parameter K_c (Triay et al., 1997). The stability and mobility of colloids in groundwater depend most strongly on the chemical conditions and less so on the fluid temperature. Higher ionic strength fluid tends to reduce the stability of colloids, flocculation being the principle mechanism reducing the concentration of colloids in solution. As quantitative results from these studies become available, they will be incorporated into the modeling effort through reduction in the uncertainty in K_c .

8.11 Transient Analyses

Considerations

Most flow and transport calculations of the unsaturated zone have assumed steady state 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 Chapter 6 where sensitivity of this assumption to parameter sets and PTn thickness is evaluated. 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 state 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 state model does a good job at explaining the data at Yucca Mountain, it should be favored over a more complex, transient flow model.

Even if steady state 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 unsaturated zone 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. Ultimately, the method to be used in TSPA-VA analyses to capture long-term transients is likely to be some form of a quasi-steady state model in which the flow is assumed to jump instantaneously from one steady state 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 this section we present an analysis of flow and transport in a transient system is presented that should provide the basis for making the simplifying assumptions just discussed.

Transport Results

We start with an analysis of the magnitude and time scale of changes in the flow field in response to 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 x I case;
- Case 2: decrease in percolation rate from the base map to the I/3 case;
- Case 3: increase in percolation rate from the I/3 case to the base map.

Hydrologic parameter set 6412b was chosen to perform this analysis.

Figure 8-32 shows the water flow rate exiting the two-dimensional model domain versus

Figure 8-32. Water mass flow rate versus time exiting the two-dimensional model domain for the three transient cases considered. Changes in percolation rate occur at $t = 0$.

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 5000 years. There is a very gradual rise to the plateau values at long times, probably due to the filling of matrix pores to their new, higher saturation values. The time scale for that process is governed by the fracture/matrix interaction parameter. 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 8-33 (Paintbrush tuff non-welded unit ptn3 and vitric Prow Pass tuff ch4v) and 8-34

Figure 8-33. Matrix saturations in the ptn3 and ch4v units in response to the long-term transients in percolation flux. Black curves: ptn3; Blue curves: ch4v.

(Topopah Springs lower lithophysal tuff tsw5 and zeolitic Bullfrog tuff bf2z). 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

Figure 8-34. Matrix saturations in the tsw5 and bf2z units in response to the long-term transients in percolation flux. Black curves: bf2z; Red curves: tsw5.

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.

The nature of radionuclide transport during a long-term transient is examined with the following scenario. The radionuclide ^{237}Np is simulated with $K_d = 2 \text{ cc/g}$ in the zeolitic tuff, with a constant release rate from the NFE from 1 ky to 31 ky, after which it was assumed that the entire inventory was depleted. At 5 ky after waste emplacement, we assume a climate-change-driven increase in the infiltration rate from the base map to the 3 x 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 8-35, along with other simulation results used for comparison. The radionuclide mass

Figure 8-35. Breakthrough curve of ^{237}Np at the water table for the long-term transient scenario. Also shown are the steady state curves for the two infiltration rates, and the quasi-steady state method.

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 3 x 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 3 x I steady state curve. Once the concentrated fluid is flushed from the unsaturated zone, 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 (base case curve before 5 ky, 3 x I thereafter).

The final comparison in this transient analysis is to examine the quasi-steady state assumption, comparing it to the full transient simulation. As shown previously in Figure 8-35, the agreement is extremely good, even during the transient period. The differences are shown on an expanded scale in Figure 8-36. The quasi-steady state assumption was run assuming that the new,

Figure 8-36. Breakthrough curve of ^{237}Np at the water table for the long-term transient scenario, shown on an expanded scale near the time of the change in percolation flux.

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, we showed earlier that the flow field achieves a steady state at a higher infiltration rate within 100 to 1000 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 NFE release rates should be carried out, it appears that the quasi-steady state assumption is an excellent approximation to the full transient solution.

One other climate-related effect that must be considered is the possibility of a rise in the water table due to a future wetter climate. For the unsaturated zone model, the elevation of the water table is set as an imposed boundary condition, and thus is not a computed result. Estimates based on mineralogic studies and saturated zone modeling efforts have placed the water table as roughly 100 m above the present-day elevation in the past. 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. Figures 8-37 and

Figure 8-37. Green's function for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map, elevated water table.

8-38 are the resulting Green's function and cumulative residence time distribution curves for this system. Arrival times at the water table are somewhat higher for the elevated water table compared

Figure 8-38. Cumulative residence time distributions for transport of radionuclides in the two-dimensional site scale model. Property set 6541, base infiltration map, elevated water table.

to the base case (Figures 8-2 and 8-3), as expected. 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. This effect, combined with the negative impact of higher percolation rate on unsaturated zone transport, makes the unsaturated zone perform considerably more poorly as a barrier under a future, wetter climate scenario.

8.12 Conclusions

In most of the two- and three-dimensional simulations presented in this study, steady state flow fields are generated using FEHM, and then radionuclide transport is computed for various transport parameters. The steady state assumption allows us to compute generic breakthrough curves in response to a pulse of radionuclide injected at time 0. This simulation provides the basis for computing, external to the FEHM simulation, the actual response to a realistic near field release

scenario. Numerical convolution is employed for this purpose. In this section we present the results of the generic response curves for a variety of flow and transport parameter sets and scenarios.

Multiple parameter sets from the LBNL site-scale flow model and variations on the USGS infiltration map are used in the simulation of radionuclides from the potential repository to the water table. For most of these parameter sets, radionuclides tend to bypass the zeolites of the Calico Hills via fracture transport when base-case infiltration conditions are simulated. When the infiltration rate is scaled by 1/3, significant fracture flow in the zeolites is still simulated with most parameter sets. A modification increasing CHn zeolite matrix permeabilities by a factor of 5 leads to substantially greater matrix flow and, hence, retardation of sorbing radionuclides. This process leads to dramatic improvement in the performance of the unsaturated zone. These studies demonstrate the overriding importance of proper characterization of the zeolitic units for performance assessment. Presently, there is significant uncertainty in the characteristics of these units due to data limitations. Enhanced characterization efforts, including the planned field test at Busted Butte, would reduce the uncertainty in these calculations and lead to a better understanding of the true nature of the hydrologic and transport behavior of the CHn.

Colloid-facilitated transport of otherwise immobile radionuclides like Plutonium is also addressed in this chapter. As very little data are currently available for Plutonium sorption onto colloids, we present a sensitivity analysis of the significant processes to mobility. In the absence of sorption onto the mobile colloids, Plutonium migration is limited due to large sorption coefficients. Assuming reversible sorption, when partitioning onto colloids approaches 99% or more, Plutonium mobility becomes potentially important to performance. If sorption of Plutonium onto colloids is irreversible, any colloid transport would directly affect performance assessment.

Long-time transients capturing climate change related phenomena are also simulated in this chapter. The results show that the flux of radionuclide at the water table under transient conditions is closely approximated by a quasi-steady state model. Thus multiple steady-state simulations can be used to predict the effects of wetter or drier future climates on radionuclide flux at the water

table. The effect of potential water table rise due to wetter, future climates is also considered. The combined effect of a shorter travel distance to the water table and high fluxes under a wetter climate scenario yield prediction of poorer unsaturated zone performance than for the present climate conditions.

Chapter 9 - Three-Dimensional Radionuclide Transport

9.1 Abstract

The two-dimensional approach presented in the last chapter is extended to the three-dimensional site-scale model of radionuclide transport. With the full three-dimensional transport model, implications of lateral flow, spatial variability of flux, and underlying strata thickness are examined. Compared to the two-dimensional model, greater spreading in the travel time distribution curve is simulated due to the additional flow paths sampling a greater range of property variability that occur in three dimensions. This variability leads to cumulative breakthrough curves at the water table spanning the range from 10 years to 10,000 years. Regarding spatial variability in releases, radionuclides released from the northern end of the repository are more likely to experience rapid transport to the water table than radionuclides released in the southern portion of the potential repository. As in the two-dimensional simulations, slightly higher matrix permeability in the zeolites leads to substantially greater retardation of radionuclides. The diffusion coefficient influences the breakthrough curves simulated with the three-dimensional model, especially for the portion of the inventory with the shortest travel times. By contrast, the dispersion coefficient is shown to have very little impact on the unsaturated zone transport.

9.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al., 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 9-1.

Table 9-1. QA Status of Data used in this Chapter

REFERENCE	Q Status	DTN	COMMENT
Bodvarsson et. al., 1997 "The Site-Scale Unsaturated Zone model of Yucca Mountain, Nevada, For the Viability Assessment, LBL Technical Report LBNL-40378	Q	LB970601233129.001	No DTN assigned. Hydrologic property sets computed in this report are used throughout the present study.
Flint et. al., 1996, Conceptual and numerical model of infiltration for the Yucca Mountain Area, Nevada", USGS WRIR MOL 19970409.0087	Q	GS960908312211.003/ NA	USGS DTN 3GUI623M
Triay et. al., 1996a, "Summary and Synthesis Report on Radionuclide Retardation for the Yucca Mountain Site Characterization Project, Los Alamos National Laboratory YMP Milestone Report 3784	Q	LAIT831341AQ96.001	

9.3 Introduction

In the previous chapter, we presented transport results on a representative two-dimensional, East-West cross section model. Many of the uncertainties of the flow and transport model were investigated, including different hydrologic parameter sets, infiltration rates, and transport parameters. A great number of processes and parameters can be examined quickly in two dimensions without compromising the validity of the result through dimensionality reduction. However, there are some issues that can only be examined thoroughly in three dimensions. These include any heterogeneities that vary in the direction not modeled in the two-dimensional simulation (the North-South direction in the present study), variations in hydrologic layer thicknesses or mineralogic thicknesses, and spatial variability in infiltration. In addition, lateral flow effects can only really be studied thoroughly in three dimensions.

In this chapter we use the three-dimensional model to investigate these and other issues. In addition, we present results of variations in transport properties not examined explicitly in the two-

dimensional results, such as diffusion and dispersion parameters. The process for incorporating the three-dimensional geologic framework model into a numerical grid was described in Chapter 3. As with the two-dimensional simulations, zeolite abundance from the min-pet model was mapped onto the three-dimensional numerical grid using a cut-off value approach (described in Chapter 2). Locations with zeolite abundance greater than 10% are given zeolite properties. At other locations in the model, the unaltered properties of the geologic units are used to populate the model with property values that are homogeneous within a layer.

9.4 Radionuclide Simulations, Base Infiltration Rate

In this set of simulations, we compute the radionuclide transport functions for the base case hydrologic property set, Case 6541 (see Table 5-3), with the base-case infiltration map from Flint et al. (1996) which is shown in Figure 6-7. For sorption, we examine the behavior assuming sorption only in zeolitic rocks, where the K_d value is varied from 0 (no sorption anywhere in the model) to 20 cc/g. As discussed earlier, this series of simulations covers the nonsorbing radionuclides (^{99}Tc , ^{129}I , ^{14}C , and ^{36}Cl) and sorbing radionuclides ^{237}Np and the various isotopes of uranium. The Green's function, or response of the system to a pulse of radionuclide released instantaneously at time 0, is used to characterize transport through the three-dimensional model to the water table. Figure 9-1 shows the results of the simulations for this case. With the current hydrologic property set, there is a tendency for some fraction of the inventory to travel rapidly through fractures to the water table. This fracture transport is through the zeolitic rocks where, for the current property sets, the matrix permeability is insufficient to transmit the flow. There is a hint of a bimodal distribution of transit times in the $K_d = 0$ curve, but, in general, the overall response exhibits a wide range of residence times without much tendency of clustering into distinct arrival times. This behavior is due to the wide range of hydrogeologic conditions present across the repository. The three-dimensional model samples the entire range of this variability, rather than

Figure 9-1. Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map.

biasing the results through the selection of a particular cross section. The spatial variability issue is addressed further in a moment.

As sorption coefficients in the zeolitic tuffs are increased, retardation reduces the amount of early arriving radionuclide through the matrix diffusion/sorption process. In addition, radionuclide percolating through the zeolite matrix is delayed. The result is a reduction in peak concentration in the Green's function, and a shift to longer residence times of a larger portion of the inventory. This effect is seen more readily in the integrated cumulative breakthrough curves in Figure 9-2. The portion of the inventory with transit times less than 100 years is small, and the distribution of times for different fractions of the radionuclide inventory to reach the water table spans a range of 10 years to greater than 10 ky. This variability is greater than for the two-dimensional simulations presented previously, and is an essential feature that must be captured in

Figure 9-2. Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map.

any total system performance assessment. Sorption onto zeolites shifts the curves to longer residence times, which means that for a given time such as a hypothetical 10 ky compliance period, less of the radionuclide is predicted to reach the water table. Thus the benefit of sorption onto zeolites is present in the three-dimensional model to perhaps a greater degree than for the two-dimensional simulations.

9.5 Radionuclide Simulations, 1/3 Infiltration Map

In this section we examine the influence of infiltration rate on the performance of the unsaturated zone transport system. Property set 6541 is used as in the previous section, but with an infiltration map in which the flux is reduced by a factor of three from the base map at all locations. The resulting Green's function is shown in Figure 9-3, and the cumulative breakthrough curves are

Figure 9-3. Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.

presented in Figure 9-4. Relative to the base infiltration map, transport times and peak values for the Green's function curves differ by more than the factor of three that would be expected in this comparison, all else being equal. Lower infiltration rates allow a greater fraction of the radionuclide inventory to travel through the zeolitic tuffs, thereby increasing the travel times. This is the case even for nonsorbing radionuclides such as ^{99}Tc . The cumulative breakthrough curves exhibit a wide range of transit times as before, but the values are from several hundred to tens of thousands of years for nonsorbing radionuclides, and from one thousand to one hundred thousand years for radionuclides that sorb to the zeolitic tuffs. Clearly, the uncertainty in infiltration rate must be captured in total-system performance analyses or else the full range of possible behaviors will not be captured in the unsaturated zone model.

Figure 9-4. Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.

9.6 Large Scale Spatial Variability in Radionuclide Transport

Variability across the repository horizon in the stratigraphic thicknesses, mineralogic abundances, and infiltration rates, as well as possible lateral diversion effects, could all play a role in the overall behavior of the unsaturated zone. One way to further understand the integrated transport results just presented is to examine the difference in travel time distributions as a function of position in the repository. To do this, we break the nodes representing the repository into five patches of nodes, each section representing approximately a rectangle comprising one fifth of the repository. The divisions were made from north to south to examine variability with respect the northing location of the release. One fifth of the total radionuclide inventory is released from each

of these patches, and the individual response curve for each patch is recorded. For all simulations presented in this analysis, the $K_d = 2$ cc/g case is used for the analysis.

Figure 9-5 shows the Green's function for each of the five individual sections of the

Figure 9-5. Green's function for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, base infiltration map.

repository along with the total integrated curve. Figure 9-6 shows the cumulative breakthrough curves for these same simulations. Radionuclide released in the northern end of the repository tends to have a greater tendency for rapid transport to the water table. The southern release points, by contrast, exhibit greater matrix flow and transport and a greater tendency toward longer residence times. We attribute this behavior to a more sporadic distribution of zeolites below the southern section of the repository. With the current property values, zeolites are effectively barriers to flow and transport, so that radionuclides travel through fractures in regions of high zeolitic alteration, such as areas in the north sections below the repository. We must point out, however,

Figure 9-6. Cumulative breakthrough curves for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, base infiltration map.

that if the hydrologic property values of zeolites had been set somewhat higher, and matrix flow and transport through zeolites had occurred to a greater degree, this situation would have been reversed, i.e., zeolites would be a benefit to performance rather than a detriment, and releases from the northern end of the repository would have the longer residence times. This analysis points out the critical importance in characterizing the hydrologic properties of units with zeolitic alteration. A similar pattern emerges for the I/3 case, as shown in Figures 9-7 and 9-8. The tendency for rapid transport to the water table is decreased at the lower infiltration rates, but the general pattern of more rapid transport times for northern release points is still present.

Figure 9-7. Green's function for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.

9.7 Transport parameter sensitivity studies

Earlier in this chapter, the influence of hydrologic parameter value uncertainty and the influence of sorption coefficient in the zeolitic tuffs was discussed. For each case, we assumed a diffusion coefficient in the matrix diffusion model of $1 \times 10^{-11} \text{ m}^2/\text{s}$, and no dispersivity. In this section we examine the importance of transport properties not yet examined, namely the matrix diffusion coefficient and dispersivity. We perform these calculations using the three-dimensional model because these processes, in a sense, "compete with" large-scale dispersion processes to influence the overall breakthrough curves. Therefore, the most accurate information of the importance of these parameters on system performance is gained by running the three-dimensional model, where large scale heterogeneities such as those discussed earlier are captured most directly. In all cases, we use the base infiltration map to perform this sensitivity study.

Figure 9-8. Cumulative breakthrough curves for radionuclide transport from five subsections of the repository in the three-dimensional site scale model. Property set 6541, I/3 infiltration map.

To examine the influence of the matrix diffusion coefficient, we performed simulations both for conservative radionuclides and the zeolite $K_d = 2$ cc/g case, a typical value for ^{237}Np . Figure 9-9 shows the results for conservative radionuclide breakthrough (Green's function) as a function of diffusion coefficient, and Figure 9-10 shows the cumulative breakthrough curves for the same simulations. The impact of the diffusion coefficient on the early arriving radionuclides is dramatic. This is due to the fact that this portion of the inventory is predicted to travel rapidly to the water table through fractures throughout the entire unsaturated zone. Under these circumstances, matrix diffusion is the only retardation process that delays the arrival times significantly. The range of values from 10^{-12} to 10^{-10} m^2/s spans the range of measured values for solutes diffusing through the saturated matrix. This is a close approximation in the zeolitic tuffs, which typically exhibit saturations very close to one. The cumulative breakthrough curves indicate

Figure 9-9. Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, no sorption, different diffusion coefficients.

that, without matrix diffusion, roughly 70% of the transport pathways have residence times of less than 10 years in the absence of matrix diffusion effects. This percentage seems large, and may be due to the current parameter sets, which simply do not allow significant fluid flow through the zeolite matrix.

Although the importance of the matrix diffusion concept in the unsaturated zone seems obvious from these results, recall that these results are for the instantaneous release scenario, and must be convoluted with an actual NFE source term to compute the arrival of radionuclides in an actual transport prediction. In Chapter 10, we show that radionuclide mass with extremely early arrivals at the water table, though counter to the notion of the unsaturated zone as a barrier to radionuclide migration, do not necessarily result in poorer overall system performance than a case with travel times on the order of 1000 years. For example, the $10^{-11} \text{ m}^2/\text{s}$ diffusion rate curve in

Figure 9-10. Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, no sorption, different diffusion coefficients.

Figure 9-10 still transmits about half the radionuclide inventory to the water table within a few thousand years. Under a 10 ky performance criterion, this system would probably not perform significantly more poorly than the case with no diffusion, the difference being primarily in the arrival times at the saturated zone, rather than in the peak concentration at the accessible environment. Nonetheless, at high enough values of diffusion coefficient, significant retardation occurs, and should be captured in models of unsaturated zone performance.

When matrix diffusion is coupled to sorption in the matrix, the impact of the diffusion coefficient is greater. Figure 9-11 shows the Green's function calculation for different diffusion coefficients for the $K_d = 2$ cc/g case, and Figure 9-12 plots the simulations for the cumulative breakthrough curves. Without matrix diffusion, the early arriving portion of the inventory cannot

Figure 9-11. Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, zeolite $K_d = 2$ cc/g, different diffusion coefficients.

contact the zeolite matrix, and the breakthrough is virtually identical to that of the conservative radionuclide case presented earlier. For moderate amounts of matrix diffusion, the combined effect of matrix diffusion and sorption delays the arrivals even more than for the $K_d = 0$ case. Thus one of the most important aspects of matrix diffusion is that the process allows contact of radionuclides with the sorptive minerals that would otherwise be bypassed through fractures.

As mentioned above, all calculations presented so far have assumed no hydrodynamic dispersion of radionuclides. In Figure 9-13, we plot the Green's function curves for a conservative radionuclide for different values of the dispersivity. This dispersivity is a local hydrodynamic parameter that captures all dispersive processes occurring at scales smaller than the grid cell size. As indicated in Figure 9-13, small scale dispersion has very little effect on the overall response of

Figure 9-12. Cumulative breakthrough curves for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, zeolite $K_d = 2$ cc/g, different diffusion coefficients.

the unsaturated zone. The only significant differences occur at early times, where dispersion results in somewhat earlier (and later) arrivals due to spreading of radionuclide through dispersion in the fracture domain. The differences are small, and thus not likely to have much impact on the overall performance of the unsaturated zone. Large scale processes and variability in thicknesses of units and spatial variability of infiltration rate have much more significance as processes that spread the arrival time distribution at the water table.

9.8 Conclusions

In this chapter, the two-dimensional approach presented in the last chapter is extended to the three-dimensional site-scale model of radionuclide transport. Variations in additional transport

Figure 9-13. Green's function for radionuclide transport in the three-dimensional site scale model. Property set 6541, base infiltration map, no sorption, different dispersivity values.

properties such as the diffusion coefficient and the dispersion coefficient are also examined in this chapter. With the full three-dimensional transport model, implications of lateral flow, spatial variability of flux, and underlying strata thickness are examined. Compared to the two-dimensional model, greater spreading in the travel time distribution curve is simulated due to the additional flow paths sampling a greater range of property variability that occur in three dimensions. This variability leads to cumulative breakthrough curves at the water table spanning the range from 10 years to 10,000 years. The three-dimensional model predicts some sorption on the zeolites as did the two-dimensional model. However, pathways bypassing the zeolites still exist and may lead to peaks in the mass flux at the water table which are not affected by retardation in the zeolites.

The three-dimensional transport model was used to examine the difference in travel time distribution as a function of the release position in the potential repository. Radionuclides released

Three-Dimensional Radionuclide Transport

from the northern end of the repository are more likely to experience rapid transport to the water table than radionuclides released in the southern portion of the potential repository. The behavior is attributed to less uniform distribution of zeolites in the southern portion of the domain. This results in slower matrix flow in the nonzeolitic units in the south compared to fracture dominated flow in the zeolitic units in the north. As with the two-dimensional simulations, this finding highlights the critical need to develop greater certainty on the material properties assigned to the zeolitic units. A slightly higher matrix permeability in the zeolites leads to substantially greater retardation of radionuclides.

The diffusion coefficient influences the breakthrough curves simulated with the three-dimensional model, especially for the portion of the inventory with the shortest travel times. Diffusion into the rock matrix and sorption onto zeolites retard the transport of ^{237}Np traveling in fractures. The dispersion coefficient is shown to have very little impact on the unsaturated zone transport.

Chapter 10 - Unsaturated Zone Performance Predictions

10.1 Abstract

This chapter describes the process of convolving NFE models with far-field transport simulations. With this approach, the differences in response due varying hydrologic conditions near waste emplacement drifts, different engineered barrier designs, time varying thermodynamic effects on canister corrosion, differing models of release due to dissolution of waste, and uncertainty in transport processes in backfill and invert material can all be evaluated. For nonsorbing radionuclides like ^{99}Tc , there is no retardation in any material between the potential repository and the water table. For all of the different property sets developed by LBNL, fracture flow through the zeolitic Calico Hills and underlying units results in bypassing of most of the unsaturated zone matrix material for the USGS infiltration estimates. NFE release rates control the maximum concentration flux at the water table. Releases that occur over longer periods of time lead to lower peak mass fluxes at the water table. Reducing the infiltration rate in these predictions also leads to lower peak mass flux rates at the water table, and they occur significantly later in time. As with nonsorbing radionuclides, those that do sorb to zeolites like ^{237}Np still show early arrivals at the water table after release begins for these parameter sets due to the simulated fracture bypass of the zeolites. However, relatively small changes from the LBNL parameters lead to vastly improved performance with respect to sorbing radionuclides. Reduction in uncertainty of the actual hydrologic properties of the Calico Hills (both fracture and matrix) would clarify whether these predictions of poor far-field barrier performance is indeed warranted or whether they are a by-product of a calibration procedure that was not well constrained by data indicating the degree of fracture flow in the units below the basal vitrophyre.

10.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al., 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 10-1.

Table 10-1. QA Status of Data used in this Chapter			
REFERENCE	Q Status	DTN	COMMENT
Bodvarsson et. al., 1997 "The Site-Scale Unsaturated Zone model of Yucca Mountain, Nevada, For the Viability Assessment, LBL Technical Report LBNL-40378	Q	LB970601233129.001	No DTN assigned. Hydrologic property sets computed in this report are used throughout the present study.
Flint et. al., 1996 , Conceptual and numerical model of infiltration for the Yucca Mountain Area, Nevada", USGS WRIR MOL 19970409.0087	Q	GS960908312211.003/ NA	USGS DTN 3GUI623M
Triay et. al., 1996a , "Summary and Synthesis Report on Radionuclide Retardation for the Yucca Mountain Site Characterization Project, Los Alamos National Laboratory YMP Milestone Report 3784	Q	LAIT831341AQ96.001	
Triay et. al., 1996b , "Radionuclide Sorption in Yucca Mountain Tuffs with J-13 Well Water, neptunium, Uranium, and Plutonium, Los alamos National Laboratory Yucca Mountain Site Characterization Project" Milestone 3338, Technical Report LA-12956-MS	Q	LAIT831341AQ96.001 *	* DTN supersedes DTN LA000000000104.001
Triay et. al., 1996c , "Batch Sorption Results for neptunium transport through Yucca Mountain tuffs, Los alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3349, Technical Report LA-12961-MS	Q	LAIT831341AQ96.001 *	*DTN supersedes DTN LA000000000090.001

Unsaturated Zone Performance Predictions

Triay et. al., 1996d , "Comparison of Neptunium Sorption Results using batch and column techniques, Los Alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3041, Technical Report LA-12958-MS	Q	LA000000000106.001	DTN Accession No. MOL.19950630.0300
---	---	--------------------	--

10.3 Introduction

In the previous two chapters, we simulated the transport of radionuclides from the repository to the water table for a variety of different hydrologic parameter sets and infiltration rates, determining the behavior for both steady state and transient flow fields. In those analyses we made extensive use of the Green's function, or response of the system to a pulse of radionuclide released from the repository at time 0. For steady state flow fields, we showed how this curve can be converted by numerical convolution into a breakthrough curve for an arbitrary input function of radionuclide from the near field environment (NFE). In this chapter a series of sensitivity analyses of radionuclide migration for various NFE release scenarios is performed. The convolution approach allows us to rapidly assess the interplay between release scenarios and unsaturated zone transport, so that an individual unsaturated zone simulation need not be performed for each scenario. The purpose of these analyses is to examine the interplay between expected performance of the unsaturated zone transport system with different NFE performance scenarios. This allows us to examine the importance of various aspects of the unsaturated zone system, including:

- the existence of very rapid transport paths (less than 100 year travel time);
- the flow split between fractures and matrix in the CHn units as a function of infiltration scenario;
- the extent to which the unsaturated zone provides travel time delays approaching the proposed 10,000 year time frame proposed as a regulatory compliance period, as well as longer periods.

This section is not a total-system performance analysis, as it omits key elements that must be included in such studies. These factors include:

- Detailed models of the NFE release rate of radionuclides. The release rate is approximated with very simple functions designed to capture the correct trends.
- Spatial variability in release rate within the repository region due to differences in repository heat effects and variability in local percolation rate. These factors are lumped into a single source term that applies to the repository as a whole. The convolution approach could have been applied to various sections of the repository and the results combined by superposition, but this approach was not carried out in the present study.
- The lack of coupling to a saturated zone model to compute concentrations at the accessible environment that can then be converted to a dose. The computation of concentrations in the saturated zone is treated in Zyvoloski et al. (1997).

Thus, this chapter is designed to elaborate on the combined behavior of the NFE source term/unsaturated zone transport system. The output from this combined model is in the form of mass flux at the water table versus time, which is the feed to a saturated zone model. The influence of various parameters and model uncertainties on the breakthrough curves will be demonstrated and explained. As such, this section provides the underlying explanations and understanding of how the unsaturated zone transport system, in combination with typical NFE source term models, is likely to transmit radionuclides to the water table.

10.4 Radionuclide Transport Properties

The radionuclides examined in this chapter include the actinides ^{237}Np , ^{233}U , ^{242}Pu , as well as ^{99}Tc . The nonsorbing ^{99}Tc can be thought of as a surrogate for other conservative radionuclides such as ^{36}Cl , ^{14}C , and ^{129}I , so the others are not modeled explicitly in the present study even though they are potentially important to total system performance. Transport properties for these radionuclides and others have been discussed in detail in other studies (Triay et al., 1996a, Robinson et al., 1995, 1996) and will be summarized briefly here. Sorption coefficients for these

radionuclides have been measured on a variety of tuff types at different chemical conditions. The radionuclides ^{99}Tc , ^{14}C , and ^{129}I are all assumed to be conservative, nonsorbing radionuclides in the unsaturated zone, based on measurements and their presumed oxidation states in the Yucca Mountain fluids. ^{14}C is perhaps better described by a multi-phase model that includes partitioning between the liquid and gas phases, and precipitation as calcite on the rock. Viswanathan (1996) modeled the gas-liquid partitioning as a function of fluid chemistry to describe the behavior of natural gas-phase ^{14}C in the unsaturated zone. However, the most conservative approach is to assume liquid-phase only transport, as gas-phase partitioning would disperse radionuclide to the atmosphere, thereby reducing the dose associated with groundwater transport.

The actinides exhibit small to moderate amounts of sorption onto the Yucca Mountain tuffs, especially to zeolites such as clinoptilolite and mordenite, which are present beneath the potential repository. ^{237}Np and ^{233}U sorption appear to be minimal onto vitric and devitrified tuffs that have not been altered by secondary mineralization. The sorption coefficient of ^{237}Np onto zeolitic tuff is discussed in detail in Chapter 8. An appropriate value for this radionuclide is about 2 cc/g on zeolitic tuff; we conservatively assume a value of 0 elsewhere, although measurable sorption has been observed on other tuff samples. Uranium exhibits somewhat higher sorption coefficients on zeolites (3-20 cc/g, depending on concentration), but similarly small values onto unaltered tuff samples. Plutonium sorbs to all types of tuff at Yucca Mountain; for this radionuclide we use a nominal value of 50 cc/g on all tuffs. Ordinarily, this degree of sorption would imply very slow migration. However, colloid-facilitated transport of plutonium is considered in this version of the model: details of this process are described in a subsequent section as well as in Chapter 8.

The matrix diffusion model for fracture transport requires the input of a diffusion coefficient for each radionuclide into the tuff. Values in the range of 10^{-10} to 10^{-11} m^2/s have been measured under saturated conditions (Triay et al., 1997). In addition, fracture coatings do not appear to inhibit the diffusion into the rock matrix. Given the rather small sensitivity to diffusion coefficient of the Green's functions computed previously, we choose a fixed diffusion coefficient

of 10^{-11} m²/s for all simulations in this section. Dispersivity also had a relatively small impact on the results presented earlier. Therefore, to simplify the interpretation, we conservatively assume no hydrodynamic dispersion. When this assumption is made, all spreading of radionuclide is caused by large-scale heterogeneities captured explicitly in the model, such as flow partitioning between the fractures and matrix and variations in vitric and zeolite thicknesses with position beneath the repository.

10.5 Abstracted Models for NFE Source Term

A comprehensive representation of the release of radionuclides from the near-field environment (NFE) is beyond the scope of the present study, which focuses on unsaturated zone flow and radionuclide transport processes. The factors that enter into the rate of radionuclide release from the NFE include:

- hydrologic conditions near waste-emplacement drifts, including the probability of seepage into the drift and dripping onto the waste canisters;
- the presence and effectiveness of engineered barriers built into the near-field system, such as backfill and drip shields;
- the time-varying thermodynamic conditions at waste packages (temperature, pressure, relative humidity), which control the corrosion rate of the canisters;
- the dissolution rate of the waste form, depending in turn on the ability of liquid water to contact the surface of the spent fuel rods;
- the transport of radionuclides in the aqueous phase through the failed waste package, backfill (if present), and invert material;
- the solubility of radionuclides in the near-field environment.

The incorporation of these factors into NFE models has typically been the purview of researchers studying the near-field system, and the models developed have been analyzed and abstracted in

past TSPA analyses (e.g. Wilson et al., 1994, Andrews et al., 1995). The complexity of these models and the unsaturated zone flow and transport model requires that each be dissected to determine which factors are the most important ones affecting system performance. Otherwise, the overall analysis is not "transparent," i.e. there is not a clear, understandable picture of the key factors and parameters that control the performance of the overall system. Therefore, there is value in adopting simplifications in some aspects of the system while retaining the complex behavior in other parts, so that the role of the uncertainties in the complex model can be highlighted.

In the present study, we accomplish this goal for the unsaturated-zone transport barrier by abstracting the NFE source term model. By using simple parameterizations for the NFE system, we can examine the role of critical assumptions and uncertainties of the unsaturated zone transport system, discovering which processes and parameters are important under different regimes of NFE behavior. The NFE source terms, though simple, are designed to capture the range of possible release scenarios. The first function we use to describe the NFE is a constant release of finite duration, beginning at a minimum time, t_{can} , and lasting until all radionuclide has been leached from the repository region (Figure 10-1a). The duration before any releases captures whatever minimum lifetime is assumed for the waste packages before corrosive degradation allows contact of water onto the waste form. Once releases start, the constant release rate occurs over a time period that will depend on near-field conditions and the type of radionuclide. For example, for solubility-limited radionuclides, such as ^{237}Np , the solubility is thought to control the release from the near-field environment, so that even if waste form dissolution supplies radionuclide to the fluid at rates that imply concentrations greater than the solubility, precipitation in the near-field will occur, thereby, placing a maximum on the release rate. By contrast, the rate for a dissolution-rate-limited radionuclide such as ^{99}Tc will depend on the kinetics of dissolution. Release rates for either radionuclide will also be a function of the water flow rate through the waste package, since liquid water is the carrier fluid. Typically, t_{rel} , the time over which releases occur, will be shorter (faster rates) for dissolution-rate-limited radionuclides, but the value is a function of the many factors

Figure 10-1. Schematic of abstracted rate models for radionuclide release from the NFE. a) constant-release-rate model; b) exponential-release-rate model.

listed above, so in this study we examine the performance of the unsaturated zone for a range of t_{rel} values.

The second model in Figure 10-1 assumes that after the initial period of no release, the rate starts at an initial maximum value and decays exponentially in time, such that at long times, the entire inventory is leached from the near field. The time dependence is intended to capture the variability in lifetime of individual waste packages, which in turn is likely to depend strongly on variability in fluid flux associated with dripping on individual canisters. For example, an order of magnitude difference in fluid flux translates to a similar magnitude difference in the local radionuclide release rate. Some waste packages will be unfavorably located within regions of high flux; other packages will reside in zones of lower risk where little or no dripping occurs. Still others

may be damaged by rockfall or other failure scenarios in the engineered system. To capture this variability, an exponential decay in total release rate is assumed. In previous TSPA analyses such as Andrews et al. (1995), the published release rates, which did attempt to capture these effects, appear to tail off over long times in this fashion, thus justifying the use of this sort of simplified release model to examine the unsaturated zone system. Like the constant rate model, the exponential model has a parameter k_{rel} that scales the time frame over which the majority of releases occur, i.e. $1/k_{rel}$ is a characteristic time over which a radionuclide is released from the NFE. After about three times this characteristic time, most of the inventory is depleted.

The subsections that follow examine the behavior of radionuclides ^{99}Tc and ^{237}Np in greatest detail because of their importance in previous TSPA analyses. ^{242}Pu is described in its own section because of the possibility of colloid-facilitated transport. Finally, more limited modeling studies are presented for ^{233}U , ^{14}C , and ^{129}I .

10.6 Transport Predictions for ^{99}Tc

We first examine the predicted transport of ^{99}Tc under an assumed release scenario of 1000 years of no release, followed by constant release rate for 3000 years. In all cases that follow we hold t_{can} at 1000 years, since the influence is merely as a delay time, with no other impact on the breakthrough curve. The 3000 year time for complete release represents a relatively rapid dissolution of the waste form and mobilization of the radionuclide. Figure 10-2 shows the resulting breakthrough at the water table (radionuclide mass flux versus time) for four of the hydrologic parameter sets assuming the base infiltration map. After the 1000 year minimum lifetime of the waste package/near-field system, breakthroughs at the water table occur well within 10 ky, regardless of which parameter set is chosen. The peak values of about 100 moles/y are controlled mostly by the release rate, rather than by details of the unsaturated zone transport system. This occurs because for these parameter sets, the fracture flow predicted in the CHn units provides

Figure 10-2. Breakthrough of ^{99}Tc at the water table for different hydrologic parameter sets. Base infiltration map, $t_{\text{can}}=1000$ y, $t_{\text{rel}}=3000$ y.

minimal barriers to radionuclide migration. This conclusion can be stated in a general way as follows: *for all of the property sets developed by LBNL using parameter estimation techniques, fracture flow through the zeolitic Calico Hills and underlying units results in bypassing of most of the unsaturated zone through fractures at the assumed infiltration rates used here. Even without considering the retardation due to sorption, this results in only minimal effectiveness of the unsaturated zone transport barrier.* The exception in Figure 10-2 is case 6412d, which assumed higher zeolite permeability. For this case, the peak is extended to about 5000 years, and its value is lower.

Regarding rapid transport through fractures in time periods of less than 100 years, recall that parameter set 6531b exhibited the greatest amount of fast-pathway flow. However, notice that this effect is not predicted to be important for predicting mass fluxes at the water table, since the quantity traveling through such paths is predicted to be small, and finite release durations (as

Unsaturated Zone Performance Predictions

opposed to the instantaneous release used for the Green's function calculation) tend to negate the influence of small quantities of fast-path transport.

To illustrate the influence of NFE release rate on breakthrough curves, we plot breakthrough curves for a 10 ky release duration in Figure 10-3. All curves plateau at about the

Figure 10-3. Breakthrough of ^{99}Tc at the water table for different hydrologic parameter sets. Base infiltration map, $t_{\text{can}}=1000$ y, $t_{\text{rel}}=10000$ y.

same level, namely the release rate at the repository. Only for one case, parameter set 6412d, does this maximum occur over times approaching 10 ky. The others reach this level within 5,000 years. A more complete examination of the influence of release rate on the base case (6541) parameter set is shown in Figure 10-4, in which the duration of release is varied from 3 to 50 ky. The plateau in each case is at the release rate from the NFE.

For unsaturated zone performance predicted from the base case, the NFE release rate will control the maximum release rate; under these conditions the unsaturated zone is a relatively poor barrier to transport for nonsorbing radionuclides. This conclusion applies only to parameter sets

Figure 10-4. Breakthrough of ^{99}Tc at the water table for different durations of release. Base infiltration map, $t_{\text{can}}=1000$ y, parameter set 6541.

and infiltration rates similar to the base case. The unsaturated zone transport system provides a more robust barrier under somewhat different assumptions, as shown below.

Infiltration rate plays a key role in the predicted mass flux at the water table. Figure 10-5 shows the influence of infiltration rate on the ^{99}Tc breakthrough curve (3 ky duration of release) for case 6541, and Figure 10-6 is a similar plot for the high-k zeolite case, 6412d. For case 6541, the peak values are reached before 10 ky, but the peak mass flux is a function of the infiltration rate, even though the duration of release, and hence the NFE release rate, was constant in each case. This difference reflects the lower flow fraction flowing through fractures at the lower infiltration rates. A similar effect is seen for case 6412d (Figure 10-6), but in addition, travel times approaching 10 ky are predicted at the lower infiltration rate. Note also that these simulations probably underestimate the importance of infiltration rate, since the NFE release rate was held fixed despite changes in flux. A more realistic scenario would be a lower release rate (longer

Figure 10-5. Breakthrough of ^{99}Tc at the water table as a function of infiltration rate. $t_{\text{can}}=1000$ y, $t_{\text{rel}} = 3000$ y, parameter set 6541.

Figure 10-6. Breakthrough of ^{99}Tc at the water table as a function of infiltration rate. $t_{\text{can}}=1000$ y, $t_{\text{rel}} = 3000$ y, parameter set 6412d.

release duration) for the low infiltration rate case. Building these interrelationships into model predictions is the subject of TSPA analyses, and is beyond the scope of the present study.

It is unlikely that the NFE release rate will be uniform until the inventory is depleted, but rather should be a function of the time-history of waste package failures, the spatial distribution of percolation flux, and other factors. Therefore, the exponential release scenario discussed above was developed. This function appears to roughly capture the complexities built into previous TSPA analyses of release rate from the near field. For ^{99}Tc , we assume that the exponential decay time ($1/k_{\text{rel}}$) spans a similar range as the times chosen for the constant release case above, namely 3 ky to 50 ky. Figure 10-7 shows the results of the simulations. Qualitatively, the conclusions are similar

Figure 10-7. Breakthrough of ^{99}Tc at the water table for different values of k_{rel} for the exponential release scenario. Base infiltration map, $t_{\text{can}}=1000$ y, parameter set 6541.

to the constant release scenario in that the peak mass flux is controlled by the maximum value issuing from the near field. In other words, the unsaturated zone provides a meager barrier to radionuclide migration for this property set. However, the peak flux value and the shape of the

Unsaturated Zone Performance Predictions

response curve at the water table are different than those obtained with the constant source model. In particular, the peak mass flux values tend to be somewhat lower for the exponential release scenario with the same time scale as the constant release case because the exponential model spreads some releases to much longer times, presumably because of a distribution of failure times and release rates for individual canisters. Therefore, the details of the failure scenarios for the NFE model will be important for predicting total system performance.

Finally, we compare the two-dimensional and three-dimensional integrated transport simulations for ^{99}Tc in Figure 10-8. The transport predictions in three dimensions appear to be

Figure 10-8. Breakthrough of ^{99}Tc at the water table comparing the two-dimensional and three-dimensional models. Base infiltration map and I/3 map, $t_{\text{can}}=1000$ y, parameter set 6541

adequately approximated by the two-dimensional prediction for conservative radionuclides. The fundamental reason for this is the predominantly downward flow and transport through fractures simulated in each model. Subtle differences in flow appear to be relatively unimportant for ^{99}Tc .

10.7 Transport Predictions for ^{237}Np

The key differences in the behavior of ^{237}Np relative to ^{99}Tc arise due to sorption of ^{237}Np (taken as 2 cc/g in this section) and the longer duration of release (lower release rate) of this solubility-limited radionuclide. The water table breakthrough curves for the various parameter sets are shown for various durations of release in Figures 10-9 (10 ky duration of release), 10-10 (30

Figure 10-9. Breakthrough of ^{237}Np at the water table for different hydrologic parameter sets. Base infiltration map, $t_{\text{can}}=1000$ y, $t_{\text{rel}} = 10$ ky, parameter set 6541.

ky duration of release), and 10-11 (100 ky duration of release). As before, a 1000 year minimum canister life is assumed. A distinct bi-modal response is predicted for ^{237}Np , due to fracture flow bypassing the zeolites. The fraction that travels through the zeolite matrix sorbs and is delayed to times on the order of 100 ky. The level of the plateau at earlier times is controlled by the flow fraction that bypasses the zeolites through fractures. The one parameter set that yields significantly better performance is case 6412d, the high-k zeolite case (Table 5-3). This result highlights the importance of characterizing the nature of flow and transport in zeolitic tuff. As with ^{99}Tc , the

Figure 10-10. Breakthrough of ^{237}Np at the water table for different hydrologic parameter sets. Base infiltration map, $t_{\text{can}}=1000$ y, $t_{\text{rel}}=30$ ky, parameter set 6541.

Figure 10-11. Breakthrough of ^{237}Np at the water table for different hydrologic parameter sets. Base infiltration map, $t_{\text{can}}=1000$ y, $t_{\text{rel}}=100$ ky, parameter set 6541.

Unsaturated Zone Performance Predictions

early arriving portion of the inventory results in the peak mass flux at the water table for all cases except 6412d, where, for the shortest release duration (10 ky), the mass fluxes of the two sets of pathways are almost equal, and for longer release periods, the long-residence-time peak at 100 ky is the larger one.

For the base case, the effect of duration of release is shown in Figure 10-12. The curves

Figure 10-12. Breakthrough of ^{237}Np at the water table as a function of duration of release. $t_{\text{can}}=1000$ y, base infiltration map, parameter set 6541.

plateau at a value that depends on the NFE release rate and the fraction of the inventory that bypasses the zeolitic tuffs through fractures. Infiltration rate also influences the results markedly, as shown for parameter set 6541 for the 30 ky release duration in Figure 10-13. Progressively lower percentages of fluid travel through the matrix at higher infiltration rates, resulting in higher peak mass flux values associated with the early-arriving portion of the inventory. The extreme case of this situation is at the low infiltration for parameter set 6412d (Figure 10-14), where the I/3 map results in no bypassing of zeolites. For this case, no ^{237}Np reaches the water table before 40 ky,

Figure 10-13. Breakthrough of ^{237}Np at the water table as a function of infiltration rate. $t_{\text{can}}=1000$ y, $t_{\text{rel}} = 30$ ky, parameter set 6541.

Figure 10-14. Breakthrough of ^{237}Np at the water table as a function of infiltration rate. $t_{\text{can}}=1000$ y, $t_{\text{rel}} = 30$ ky, parameter set 6412d.

Unsaturated Zone Performance Predictions

and the peak occurs between 100 and 150 ky. Clearly, relatively small changes from base case conditions can lead to vastly improved performance with respect to radionuclides such as ^{237}Np . Nevertheless, under base case conditions with the parameter sets supplied by LBNL, the unsaturated zone does not provide a significant barrier to radionuclide migration. For ^{237}Np , the NFE source term, which will depend strongly on solubility and near field hydrologic conditions, is likely to control the mass flux of this critical radionuclide.

Breakthrough curve results for the exponential release scenario for case 6541, base infiltration map are shown in Figure 10-15. As was the case with ^{99}Tc , the general conclusions are

Figure 10-15. Breakthrough of ^{237}Np at the water table for different values of k_{rel} for the exponential release scenario. Base infiltration map, $t_{\text{can}}=1000$ y, parameter set 6541.

unchanged by the form of the release scenario, but the exact predictions of mass flux versus time are different for the exponential release scenario. Specifically, the peak mass flux values are somewhat lower for the exponential release scenario with the same time scale as the constant release case, for reasons explained above with regard to ^{99}Tc .

Unsaturated Zone Performance Predictions

A comparison of the two-dimensional and three-dimensional integrated transport prediction is shown in Figure 10-16. As with ^{99}Tc , the three-dimensional simulation is adequately

Figure 10-16. Breakthrough of ^{237}Np at the water table comparing the two-dimensional and three-dimensional models. Base infiltration map and I/3 map, $t_{\text{can}}=1000$ y, parameter set 6541.

approximated by the two-dimensional model, except perhaps for a significant deviation for the I/3 infiltration scenario. The two-dimensional model predicts somewhat more matrix flow through the zeolites, which shifts more of the inventory to longer residence times, when compared to the three-dimensional simulation. Overall, however, the differences are fairly small compared to other uncertainties in the models.

10.8 Transport Predictions for Uranium Isotopes

As mentioned in the previous chapter, Triay et al. (1996a) determined that uranium exhibits significant sorption only onto the zeolitic tuffs at Yucca Mountain for the geochemical conditions

Unsaturated Zone Performance Predictions

likely to be present. A nonlinear sorption isotherm behavior was determined, with K_d values of about 20 cc/g at 8×10^{-8} mol/l, compared to 3 cc/g at 10^{-4} mol/l. Robinson et al. (1996) used the nonlinear Langmuir and Freundlich isotherm models to simulate uranium transport. However, in the present study, we use the linear sorption model embodied in the particle tracking model to bound the behavior. This requires that we examine a broad range of sorption coefficients to assess the likely effects of nonlinear isotherm behavior.

The appropriate source term for uranium was also discussed in Robinson et al. (1996) and will be briefly reviewed here. The isotopes of uranium present in the inventory consist of several long-lasting isotopes, the most plentiful of which is ^{238}U . Figure 10-17 shows the predicted

Figure 10-17. Uranium isotope inventory as a function of time for a 70,000 MTU repository and -- add description of inventory assumptions (initial inventory data from Wilson et al., 1994).

proportions of the different uranium isotopes with time generated by Robinson et al. (1996) using the initial inventory figures of Wilson et al. (1994) and decay-chain models to track the ingrowth and decay of all isotopes of uranium and other radionuclides. The initial inventory data, half-lives

of radionuclides, and relevant decay chains are presented in Appendix A, along with a brief description of the calculation method for determining the isotope distribution with time.

This proportion of uranium isotopes represents the relative amounts of each isotope, regardless of where these isotopes are located in space. If all isotopes were present at the repository and available for mobilization, then a reasonable assumption would be that the release rate of each isotope would be proportional to the fraction of each present at any given time. However, production of ^{233}U occurs due to decay of ^{237}Np , most of which will probably have escaped the near-field environment by the time significant quantities of ^{233}U are generated. Therefore, special provisions are required for ^{233}U ; this special case is discussed below. For all isotopes except ^{233}U , we assume that the release rate is proportional to the fraction of each present at time 0 (e.g. congruent dissolution), and that the total NFE source term for uranium is governed by the solubility of uranium, assumed to be about one order of magnitude larger than ^{237}Np (Triay et al., 1997). A key difference with uranium relative to ^{237}Np is the much larger inventory of uranium in the waste form, resulting in much longer times for complete dissolution and mobilization of the waste.

For ^{233}U , we assume that the majority of this isotope is mobilized as the parent ^{237}Np , and that decay produces ^{233}U , which then transports with properties similar to that of ^{237}Np . In the unsaturated zone, we showed that for the current property values, the influence of zeolite K_d , the only significant difference between uranium and ^{237}Np , was relatively minor since fracture transport is the controlling feature of transport in this system. Under these assumptions, ^{233}U breakthrough curves can be generated simply from the ^{237}Np results with a post-processing calculation to account for ^{237}Np decay to ^{233}U .

Transport predictions for uranium isotopes are shown in Figure 10-18. This calculation assumed solubility-limited release at a rate ten times faster than the 30 ky release scenario for ^{237}Np , though for uranium this implies only 6.7% of the uranium inventory is released after 100 ky. The constant release rate presumed using this model is reflected at the water table after about

Figure 10-18. Water table breakthrough curves for Uranium isotopes. Property set 6541, base infiltration map, release rate similar the ^{237}Np case with 30 ky release scenario (for Uranium only --% is released after one million years).

5000 years, and the relative proportion of each isotope is controlled by the fraction of each present in the inventory, as discussed above. The exception is ^{233}U , which tracks the behavior of ^{237}Np , but at a level that increases with time due to the decay of the parent. Note also that radionuclides that reach the saturated zone as ^{237}Np will still decay to ^{233}U in the saturated zone, thereby adding to the amount of the daughter product reaching the accessible environment.

10.9 Transport Predictions for Plutonium Isotopes

In Chapter 8 we presented a model for colloid-facilitated transport of plutonium. We showed that even for the current hydrologic property values, with fracture-dominated flow through a large portion of the unsaturated zone, that retardation in nonwelded vitric tuff provides a very robust barrier to migration of this strongly sorbing radionuclide, as long as the plutonium remains primarily in the aqueous phase. When plutonium is assumed to sorb to mobile colloids, the

Unsaturated Zone Performance Predictions

situation is akin to a more weakly sorbing radionuclide, and significant releases to the water table are predicted. In this section we convolute these results with a range of NFE release rates for plutonium to predict the mass flux at the water table for some specific scenarios.

As was the case for uranium, several isotopes of plutonium are present in the waste form. Figure 10-19 shows the quantity of each isotope present versus time. In contrast to ^{233}U , there is

Figure 10-19. Plutonium isotope inventory as a function of time for a 70,000 MTU repository and --- add description of inventory assumptions (initial inventory data from Wilson et al., 1994).

no ingrowth from a mobile radionuclide to complicate the prediction of dissolution in the near field. After a few hundred years, the remaining isotopes are ^{239}Pu , ^{240}Pu , and ^{242}Pu , which are, as was the case with uranium, assumed to be released in proportion to their relative proportions in the inventory.

Plutonium is a very insoluble radionuclide under the geochemical conditions present in the unsaturated zone. Triay et al. (1997) report values at 25°C ranging from 5×10^{-8} mol/l to 1×10^{-8}

mol/l in the pH range of 5.9 to 8.5. These solubilities are roughly three orders of magnitude lower than those of ^{237}Np . Therefore, to a first approximation, NFE release rates would be predicted to be three orders of magnitude lower under similar hydrologic conditions if the limiting factor for release rate is the solubility in the aqueous phase. This is one end-member that we consider in this set of calculations. The other extreme is to consider the total fluid phase ^{242}Pu as governed by radionuclide present either in the aqueous phase or sorbed to colloids. For this extreme, we assume that the aqueous phase plutonium is at the solubility limit, but that an additional portion of the inventory is also mobilized on colloids, so that the effective "solubility" (moles plutonium present in either the aqueous or pseudo-colloidal form) equals $C_S K_C$, where C_S is the aqueous solubility and K_C is the partitioning coefficient for sorbed plutonium defined in the previous chapter. For example, the $K_C = 999$ case raises the effective solubility, and presumably the release rate from the NFE, by three orders of magnitude from the value in the absence of colloids. Under these extreme conditions, release rates similar to those predicted for ^{237}Np would occur.

Figure 10-20 shows the predicted water table breakthrough curves obtained by convoluting the results of the Green's function calculation of total plutonium for the hydrologic base case (property set 6541, base infiltration map) with different durations of release corresponding to different K_C values. The parameter K_C has a multiplicative effect on the predicted mass flux at the water table. With little or no sorption to colloids, transport is slow, as shown earlier, and furthermore the NFE releases are limited by the low aqueous solubility of plutonium. At high K_C values, transport is enhanced and the NFE release rate is increased, resulting in much earlier and much higher mass fluxes at the water table. Because of this combined effect, the partitioning parameter K_C essentially controls the predicted breakthrough flux at the water table for this radionuclide. Further work to assess the importance of ^{242}Pu in total system performance should therefore focus primarily on estimating this key parameter. The influence of dimensionality is also fairly unimportant, as shown by the three-dimensional model result for $K_C = 999$ shown as the dotted curve on Figure 10-18.

Figure 10-20. Water table breakthrough curves for total plutonium. Property set 6541, base infiltration map, different durations of release. Duration of release is directly proportional to effective solubility, which in turn is proportional to K_c .

Figure 10-21 shows the individual breakthrough curves for the various isotopes of plutonium for the $K_c = 99$ case. Because the release is solubility-limited, and congruent dissolution of the various isotopes is assumed, the quantity of long-lived isotopes such as ^{242}Pu increases as radioactive decay of ^{240}Pu and ^{239}Pu reduce the mass of each present in the near field.

10.10 Conclusions

Predicting the response at the water table to the large variety of near field environment (NFE) release scenarios is done most efficiently by decoupling the far-field and near-field models. This chapter describes the process of convolving NFE models with far-field transport simulations. With this approach, the differences in response due varying hydrologic conditions near waste emplacement drifts, different engineered barrier designs, time varying thermodynamic effects on canister corrosion, differing models of release due to dissolution of waste, and uncertainty in

Figure 10-21. Water table breakthrough curves for individual isotopes of Plutonium. Property set 6541, base infiltration map, durations of release corresponding to $K_c = 99$.

transport processes in backfill and invert material can all be evaluated. The present study focuses on the unsaturated zone transport barrier; simplified NFE source term functions are used.

For nonsorbing radionuclides like ^{99}Tc , there is no retardation in any material between the potential repository and the water table. For all of the different property sets developed by LBNL, fracture flow through the zeolitic Calico Hills and underlying units results in bypassing of most of the unsaturated zone matrix material for the USGS infiltration estimates. NFE release rates control the maximum concentration flux at the water table. Releases that occur over longer periods of time lead to lower peak mass fluxes at the water table. Reducing the infiltration rate in these predictions also leads to lower peak mass flux rates at the water table, and they occur significantly later in time.

As with nonsorbing radionuclides, those that do sorb to zeolites like ^{237}Np still show early arrivals at the water table after release begins for these parameter sets due to the simulated fracture bypass of the zeolites. This early arrival appears to control the peak mass flux as well. Under these

Unsaturated Zone Performance Predictions

conditions, two factors control the magnitude of the peak mass flux at the water table. First, the NFE release scenario directly influences the actual peak mass flux; rapid releases over short time periods yield higher peaks. Second, any parameter that influences the relative flow fraction through fractures has a direct impact on peak mass flux.

Relatively small changes from the LBNL parameters lead to vastly improved performance with respect to sorbing radionuclides. However, without modification, those parameter sets lead to predictions of very little mitigation of releases from the near field by the unsaturated zone. Reduction in uncertainty of the actual hydrologic properties of the Calico Hills (both fracture and matrix) would clarify whether these predictions of poor far-field barrier performance is indeed warranted or whether they are a by-product of a calibration procedure that was not well constrained by data indicating the degree of fracture flow in the units below the basal vitrophyre.

Chapter 11 - Reactive Transport of ^{237}Np with Repository Heat

11.1 Abstract

Characterization and performance assessment studies for the potential high-level nuclear waste repository at Yucca Mountain have identified ^{237}Np as a radionuclide of concern for the proposed repository. To predict the migration of neptunium after a repository breach, an understanding of the relevant hydrologic and geochemical processes is required. The hydrologic flow in the unsaturated zone at Yucca Mountain is dependent on the infiltration rate, stratigraphy of the vadose zone, and the heat generated by the decaying radioactive waste. The geochemical processes that strongly affect ^{237}Np migration include: solubility-limited release of ^{237}Np from the near-field environment, aqueous speciation of neptunium into non-sorbing carbonate/hydroxy complexes and the sorbing NpO_2^+ cation, sorption of neptunium onto the zeolitic tuffs via an ion exchange mechanism, and radioactive decay. We investigate the coupled effects of chemical interactions and heat on neptunium transport from the potential repository to the water table. The selective coupling method is introduced to solve these reactive transport problems. The simulations indicate that in the absence of irreversible changes in the hydrologic and transport properties, the heat pulse does not significantly affect the migration of neptunium, as the time scale of heat pulse propagation is shorter than the time scales associated with neptunium release and migration. Water chemistry, particularly pH and calcium and sodium concentrations, significantly affect the retardation of neptunium by the zeolitic rocks between the repository and the water table.

11.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is

Reactive Transport of ^{237}Np with Repository Heat

documented in (Zyvoloski et al., 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 11-1.

Table 11-1. QA Status of Data used in this Chapter			
REFERENCE	Q Status	DTN	COMMENT
Bertetti et al., 1996 "Neptunium (V) Sorption Behavior on clinoptilolite, Quartz, and Montmorillonite, Mat. Res. Soc. Symp., Proc., Vol. 412, 631-638, 1996	non-Q		
Flint et. al., 1996, Conceptual and numerical model of infiltration for the Yucca Mountain Area, Nevada", USGS WRIR MOL 19970409.0087	Q	GS960908312211.003/ NA	USGS DTN 3GUI623M
Nitsche et. al., 1993, "Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in a Typical Groundwater (J-13) from the Yucca Mountain Region", Los Alamos National Laboratory Report Number LA-12562-MS, July 1993	non-Q non-Q	LA000000000012.001 LA000000000012.002	Accession Nos. MOL.19950705.0241 NNA.19930507.0136, and MOL.19950621.0265, and MOL.19950711.0329
Ogard and Kerrisk, 1984, "Groundwater Chemistry along flow paths between a proposed repository site and the accessible environment, Los Alamos National Laboratory Report LA-10188-MS, 1984	non-Q	LA000000000038.001	Accession No. NNA.19870406.0021
Tait et. al., 1996, "Sorption as a function of groundwater chemistry to elucidate sorption mechanisms, Los Alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3219, 1996	Q Q	LAIT831341AQ95.001 LAIT831341DQ95.002	
Triay et. al., 1996a, "Summary and Synthesis Report on Radionuclide Retardation for the Yucca Mountain Site Characterization Project, Los Alamos National Laboratory YMP Milestone Report 3784	Q	LAIT831341AQ96.001	

Triay et. al, 1996b , "Radionuclide Sorption in Yucca Mountain Tuffs with J-13 Well Water,: neptunium, Uranium, and Plutonium, Los alamos National Laboratory Yucca Mountain Site Characterization Project" Milestone 3338, Technical Report LA-12956-MS	Q	LAIT831341AQ96.001 *	* DTN supersedes DTN LA000000000104.001
Triay et. al, 1996c , "Batch Sorption Results for neptunium transport through Yucca Mountain tuffs, Los alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3349, Technical Report LA-12961-MS	Q	LAIT831341AQ96.001 *	*DTN supersedes DTN LA000000000090.001
Triay et. al., 1996d , "Comparison of Neptunium Sorption Results using batch and column techniques, Los Alamos National laboratory Yucca Mountain Site Characterization Project Milestone 3041, Technical Report LA-12958-MS	Q	LA000000000106.001	DTN Accession No. MOL.19950630.0300

11.3 Introduction

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.

In this chapter, we investigate the migration of ^{237}Np from the potential repository to the water table (Viswanathan et al., 1997). 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; Andrews et al., 1995). We utilize FEHM (Zyvoloski et al., 1992) to study ^{237}Np transport. 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 utilize 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 which 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. We then apply the model to the two-dimensional 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 unsaturated zone. 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 thermohydrologic 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

thermohydrologic 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 chapter 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 will allow us to assess the validity of assumptions embodied in previous studies such as isothermal conditions and linear equilibrium sorption. Furthermore, such simplifications, if valid, will greatly facilitate future performance assessment calculations, which typically involve hundreds of Monte Carlo simulations in which numerous uncertain parameters are varied in an attempt to capture the overall uncertainty of the system.

11.4 Model Description

11.4.1 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 FEHM code is a two- or three-dimensional finite element code suitable for simulating systems with complex geometries that arise when modeling subsurface flow and transport. In the unsaturated zone, the system of governing equations arises from the principles of conservation of water and air, as well as conservation of energy. Darcy's law is assumed to be valid for the movement of each phase. In most simulations in this chapter, the relative permeability curves for the fractured rock are assumed to be adequately represented by an equivalent continuum model (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 as described in Chapters 4 and 5. Therefore, we also include a series of one-dimensional scoping calculations using a dual permeability model (DKM). In this model formulation, fractures and matrix are treated as two overlapping continua, with appropriate properties assigned for each continuum. Additionally, there is a transfer term describing fluid flow between the two continua at each location in space. An additional parameter, a characteristic length scale for transport of heat, mass, and solutes between the two continua, is also required. When this length is small, the model reverts to ECM behavior. For larger lengths, the coupling between the fracture and matrix domains is weak, and flow in the two media becomes more independent. Because fracture-dominated flow and transport is expected to be present in some stratigraphic units, it is essential to consider the relative flow fractions in the two media as demonstrated in Chapter 5. Current limitations in the model preclude simulations of the full reactive transport system with heat effects in dual permeability. Therefore, we use these dual permeability scoping calculations as a guide to adjusting an ECM model to capture the influence of fracture flow and radionuclide transport. Provisions for correcting the model for transport are developed in section 11.8.1.

11.4.2 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 not 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:

$$C_j = c_j + \sum_{i=1}^{N_x} a_{ij}x_i \quad j = 1, \dots, N_c \quad (\text{Eq. 11-1})$$

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.

11.4.2.1 Governing Equations for the PDVs

For transient fluid flow, the generalized version of the reactive transport equation for an aqueous component is

$$\frac{\partial A_j}{\partial t} = \nabla \bullet (\phi S_l D_j \nabla \rho_l C_j) - \nabla \bullet f_c + R_j \quad j = 1, \dots, N_c \quad (\text{Eq. 11-2})$$

where $A_j = \phi C_j S_l \rho_l$ is the solute mass storage per unit total volume for aqueous component liquid concentration C_j (units of concentration are moles/kg fluid); $f_c = \rho_l C_j u_l$ is the advective mass flux of solute; D is the hydrodynamic dispersion tensor; S_l is the saturation; u_l is the Darcy velocity vector; ϕ is the porosity; S_l is the liquid saturation; and R_j is the kinetic reaction source-sink term. The porosity is an effective porosity for transport, which in the code can be assigned different values than the values used for fluid flow. This provision allows us to simulate the case in which fracture flow results in a low transport porosity due to the small pore volume available for transport within the fractures. To simplify the notation for the remainder of the chapter, we define the advection-dispersion operator:

$$L_l(C) = \nabla \bullet f_c - \nabla \bullet (\phi S_l D_j \nabla \rho_l C_j) \quad (\text{Eq. 11-3})$$

Equation (Eq. 11-2), the reactive advection dispersion equation, can then be rewritten as:

$$\frac{\partial A_j}{\partial t} + L_l(C_j) = R_j \quad j = 1, \dots, N_c \quad (\text{Eq. 11-4})$$

The reaction transport equation for a vapor component takes on a similar form to Equation 11-4 and is given by

$$\frac{\partial B_k}{\partial t} + L_v(G_k) = R_k \quad k = 1, \dots, N_v \quad (\text{Eq. 11-5})$$

where B_k is the solute mass per total volume of component k , G_k is the vapor concentration of component k , u_v is the vapor Darcy velocity vector and N_v is the number of vapor components. The hydrodynamic dispersion tensor is assumed to reduce to longitudinal and transverse components (e.g. Lichtner, 1996).

Immobile components are not transported and are therefore treated using a simple mass balance given by

$$\frac{\partial M_m}{\partial t} = R_m \quad m = 1, \dots, N_{im} \quad (\text{Eq. 11-6})$$

where M_m is the immobile concentration (mass solute per unit mass rock matrix) of component m and N_{im} is the number of immobile components.

The reaction rate terms in Equations 11-4, 11-5, and 11-6 originate from the kinetic reactions in the system and may be nonlinear functions of the concentrations of the total aqueous components, uncomplexed aqueous components, aqueous complexes, immobile components and vapor components. FEHM is capable of modeling the following kinetic processes: linear adsorption, nonlinear Langmuir adsorption, ion/surface exchange, precipitation-dissolution and liquid-vapor interchange of solute. The specific kinetic models used to model ^{237}Np adsorption and dissolution will be provided in the application section. Equations 11-4, 11-5, and 11-6 result in a system of $(N_c + N_m + N_v)N_n$ nonlinear coupled partial differential equations (PDEs) where N_n is the number of spatial grid points. FEHM's method for solving this system of coupled PDEs will be discussed in the solution procedure section.

11.4.2.2 Governing Equations for the SDVs

Given all of the aqueous component concentrations, the uncomplexed aqueous component concentrations and aqueous complex concentrations can be calculated using chemical equilibrium theory. The chemical equilibrium calculations performed by FEHM are similar to the techniques

used in batch geochemical software such as EQ3/6 (Wolery, 1992). A chemical reaction describing aqueous speciation can be written in the following general form

where $\hat{C}_j$ is the chemical formula for the aqueous component j , and $\hat{X}_i$ is the chemical formula for the aqueous complex i . FEHM assumes that all aqueous speciation reactions are at local equilibrium. The mass action expression for an aqueous component is given by

$$K_i = x_i \prod_{j=1}^{N_c} (\gamma_j c_j)^{-a_{ij}} \quad (\text{Eq. 11-8})$$

where K_i is the equilibrium formation constant for complex i and γ_j is the activity coefficient for aqueous component j . In these calculations, we neglect ionic strength corrections. For example, for Yucca Mountain well UZ-14, the chemical analyses (Figure 2) yield a range of ionic strengths from 0.004 to 0.013 mol/l, for which the Davies equation for activity coefficients yields γ_j values that are very close to 1. Equations 11-1 and 11-8 can be combined to express the total aqueous concentration of component j as a function of the uncomplexed component concentrations:

$$C_j = c_j + \sum_{i=1}^{N_x} a_{ij} K_i \prod_{z=1}^{N_c} c_z^{a_{iz}} \quad j = 1, \dots, N_c \quad (\text{Eq. 11-9})$$

Equation 11-9 results in a set of N_c nonlinear algebraic equations to be solved given all of the total aqueous concentrations.

11.5 Solution Procedure

The solution of the flow and energy equations will not be discussed in this chapter. This description can be found in Zyvoloski et al. (1992).

11.5.1 Solution Method for the PDVs:

The reactive transport equations given by Equations 11-4, 11-5 and 11-6 result in a set of nonlinear coupled PDEs. Many reactive transport codes, including FEHM, utilize a two-step approach to decouple the PDEs (e.g. Cederberg et al., 1985, Narasimhan et al., 1986, Yeh and Tripathi, 1989). Two-step methods divide the solution into a transport step, in which the aqueous and vapor components are transported by advection and dispersion, and a reaction step, in which aqueous, vapor and immobile components react with one another. The transport and reaction steps are either coupled iteratively or sequentially (Walter et al., 1994). In FEHM, the transport and reaction steps are coupled iteratively. First, a trial solution for the reaction terms is computed. These reaction terms are then used in the transport step. Iteration between these two steps is performed until convergence is achieved.

The numerical implementation of the transport step can be derived by rewriting Equations 11-4, 11-5 and 11-6 in fully implicit time-discretized form:

$$\frac{C_j^{n+1} - C_j^n}{\Delta t} + L(C_j)^{n+1} = R_j^{n+1} \quad (\text{Eq. 11-10})$$

$$\frac{G_k^{n+1} - G_k^n}{\Delta t} + L(G_k)^{n+1} = R_k^{n+1} \quad (\text{Eq. 11-11})$$

$$\frac{M_m^{n+1} - M_m^n}{\Delta t} = R_m^{n+1} \quad (\text{Eq. 11-12})$$

where n indicates the time step level. The reaction rate terms in these equations can be estimated using a Taylor series expansion to linearize the reaction rate term

$$\begin{aligned} R_i^{n+1, p+1} = & R_i^{n+1, p} + \sum_{j=1}^{N_c} \left(\frac{\partial R_i}{\partial C_j} \right)^{n+1, p} (C_j^{n+1, p+1} - C_j^{n+1, p}) + \\ & \sum_{k=1}^{N_v} \left(\frac{\partial R_i}{\partial G_k} \right)^{n+1, p} (G_k^{n+1, p+1} - G_k^{n+1, p}) + \\ & \sum_{m=1}^{N_{im}} \left(\frac{\partial R_i}{\partial M_m} \right)^{n+1, p} (M_m^{n+1, p+1} - M_m^{n+1, p}) + \dots \end{aligned} \quad (\text{Eq. 11-13})$$

where p is the iteration level and i represents either an aqueous, vapor or immobile component. The iterative methods in the literature differ in the number of terms evaluated in this Taylor series expansion. Expanding each term on the left hand side of equations 11-10, 11-11 and 11-12 in a Taylor series and substituting the expanded reaction rate given by Equation 11-13 results in a set of linear algebraic equations which constitute a single iteration of the Newton-Raphson method. However, the Newton-Raphson method results in a large system of coupled linear equations stemming from the derivative terms of R_i with respect to other aqueous, vapor, or immobile components. The approximate iterative approaches in the reactive transport literature often drop terms in the Taylor series expansion to decouple the linear equations into smaller equations sets that are solved sequentially. The sequential iterative approach described by Engesgaard and Kipp (1992) and Kinzelbach et al. (1991) use only the first term in the Taylor series ($R_i^{n+1,p}$). We will refer to this method as the SIA-0 method. We have found that this method works efficiently for relatively slow kinetic reactions but becomes inefficient for faster kinetic reactions. It is possible to improve the SIA-0 method by including additional terms in order to improve the convergence of SIA. The SIA-1 method estimates the reaction term as:

$$R_i^{n+1,p+1} = R_i^{n+1,p} + \left(\frac{\partial R_i}{\partial C_j} \right)^{n+1,p} (C_j^{n+1,p+1} - C_j^{n+1,p}) \quad (\text{Eq. 11-14})$$

The SIA-1 approach seeks to improve the estimate of $R_i^{n+1,p+1}$ by considering the relationship between R_i and C_i only. That is, the relationships with other aqueous, vapor and immobile components are neglected, and the linear equations arising from the individual components are solved separately. We have found that the SIA-1 greatly improves convergence for large Damkohler number systems, when kinetics are fast compared to the transport time scale (Tebes-Stevens et al., 1997). However, for certain reactions, the SIA-1 approach becomes quite inefficient because some of the cross derivative terms ($\partial R_i / \partial C_j, i \neq j$) that are neglected by SIA-

1 are significant. Physically, this can occur when aqueous, vapor, or immobile components that are, in fact, coupled to one another are treated as though they were independent. In an extension of the SIA methods, we selectively include additional derivative terms that couple a subset of the components to one another to improve convergence. Since cross derivative terms, $\partial R_i / \partial C_j$, are now included in the calculation, sets of components must be solved simultaneously. We note in passing that in the extreme case in which only $\partial R_i / \partial C_i$ are calculated, the method reduces to the SIA-1 method. At the other extreme, in which all components are coupled, a global implicit solution scheme (Steefel and Yabusaki, 1996) is effectively obtained. We say effectively because in this implementation, a reaction iteration is still required to include the aqueous speciation reactions. Nonetheless, only a few iterations (less than 5) are typically required when the reactive transport equations for components that are strongly coupled through reaction terms are solved simultaneously.

The linear equation solver in FEHM was developed primarily for the solution of coupled fluid flow and heat transport. The solver makes use of well-tested numerical techniques that take advantage of the block structure of the coupled equations for pressure, temperature, and fluid saturation (e.g. Zyvoloski, 1983; Behie et al., 1985). Here, we use the same solver technologies for the transport solution step. In essence, for N_{dof} (where N_{dof} is the number of “degrees of freedom,” in this context equal to the number of components solved simultaneously) unknowns per grid point, the same operations on the overall matrix of a single-unknown solution are carried out, but multiplications of individual matrix elements now become matrix multiplications involving the $N_{dof} \times N_{dof}$ submatrices, and divisions are carried out as multiplications by the inverse of the submatrix. Since such operations become memory and cpu intensive for large problems, it is important to employ efficient numerical techniques. FEHM uses numerical methods suitable for the nonsymmetric matrices that arise from the finite element solution of reactive transport equations on unstructured numerical grids. The solver software uses incomplete factorization with variable fill-in level (Zyvoloski, 1986) as a preconditioner, and a generalized minimum residual

(GMRES) acceleration technique (Saad and Schultz, 1986) for the iterative solution. Details of this method as applied to heat and mass transport problems may be found in Zyvoloski et al. (1992). In a typical reactive transport solution with FEHM, the heat and mass transfer solution is also being performed, so the initial bookkeeping associated with the method is already being carried out, and the memory allocated for the solution is shared between the heat and mass solution and the transport solution.

There are definite trade-offs in computational efficiency and memory utilization between the SIA techniques and methods involving coupling of the transport equations of individual components. Coupling requires more time per iteration and more memory than typical SIA methods. However, components strongly coupled by reaction may not converge using SIA methods without using small time steps. Since flexibility is required in a general purpose code, the transport iteration in FEHM was developed with a provision we call Selective Coupling to solve the component concentrations in groups of one or more at a time, so that only those components that need to be coupled are solved simultaneously. It is necessary to solve a set of equations for each component present in the system, but the order of solution and the nature of the coupling are set by the user at run-time. This allows the user, on the basis of his knowledge of the reactive transport system, to couple only those components that are required for efficient solution of the system of equations. Selective Coupling of components linked to each other through kinetic chemical reactions allows a given problem to be solved in the fastest, yet most memory efficient manner possible (Viswanathan, 1996). The use of this provision in the solution of the neptunium transport chemical system is demonstrated in the application section of this chapter, however, the methods used should have wider applicability in the field of reactive chemical transport. Details of the computational performance of this method will be explored in a future publication.

11.5.2 Solution Method for the SDVs

Every time a trial solution for the reaction terms is calculated in a transport iteration, the SDV's must be solved for. The Newton-Raphson method is used to perform these calculations at each grid point. The solution procedure is similar to batch geochemical codes such as EQ3/6 (e.g.

Wolery, 1992). The techniques used to handle pH calculations are presented in Tebes-Stevens et al. (1997).

11.6 Yucca Mountain Unsaturated Zone Hydrology and Geochemistry

11.6.1 Hydrogeologic Setting

Characterization of the hydrostratigraphy of the unsaturated zone at Yucca Mountain generally follows from the delineation of major stratigraphic units and structural features such as faults. The stratigraphic structure of the unsaturated zone of Yucca Mountain is comprised of alternating layers of welded and nonwelded volcanic tuff that are tilted, uplifted, fractured, and faulted. Information from boreholes, the Exploratory Studies Facility (ESF), a shaft currently under construction, and surface studies has led to various classification schemes for the stratigraphy at the site (e.g. Scott and Bonk, 1984; Buesch et al., 1996). The repository, if constructed, will be located approximately 350 m above the water table and roughly 200 to 300 m below ground surface, depending on the local topography, within the nonlithophysal portion of the Topopah Springs welded (TSw) unit (Figure 11-1d). From the surface, the highly fractured Tiva Canyon welded (TCw) unit (not present at well UZ-14 in Figure 11-1d) is the first major stratigraphic unit encountered under a largely unconsolidated alluvial cover that varies in thickness from zero to more than 50 m. Below the TCw is the Paintbrush tuff nonwelded (PTn) unit, with a higher permeability in the rock matrix, but lower bulk permeability due to lower fracture density. The TSw unit consists mainly of highly welded and fractured subunits of devitrified tuff, some of which contain a high density of lithophysal cavities. The TSw extends about 100 to 150 m below the potential repository before the Calico Hills nonwelded (CHn) unit is reached. The CHn is a less fractured tuff that contains both vitric sections and a horizon that exhibits extensive zeolitic alteration. In the cross section chosen for calculations, the underlying Prow Pass and Bullfrog units also lie above the water table, and both have zones of zeolitic alteration. In general, the major hydrostratigraphic units dip to the East at angles of 5 to 10° (Figure 11-2a), creating the possibility

of lateral flow at the interfaces of units of contrasting hydrologic properties. 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. In this set of calculations, the major stratigraphic units were further subdivided to capture important differences within the units such as the low porosity vitrophyre at the base of the TSw. Hydrologic property values for each unit (listed for all units in Robinson et al., 1996) were derived from Wittwer et al. (1995) for the units from the surface to the TSw and Loeven (1993) for the CHn units.

11.6.1.1 *Unsaturated Zone Geochemistry*

The major ion chemistry of fluids in the unsaturated zone will impact many aspects of repository behavior, including the corrosion of waste packages, dissolution rate of the waste form, and solubilities, speciation, and sorption of radionuclides. In addition, the chemical data provides important insights on the hydrologic system of the unsaturated zone. The use of chloride and other ions to characterize the unsaturated zone hydrology is the subject of ongoing studies, and will be treated only very briefly here. Figure 11-1 (data published in Yang et al., 1996) shows the important anions (11-1a), cations (11-1b), and pH (11-1c) of the unsaturated zone fluids in samples extracted from non- to partially welded tuffs in wellbore UZ-14.

The chloride mass balance method (Allison et al., 1985; Scanlon, 1991) can be used to estimate the infiltration rate. The method assumes that the deposition of chloride onto the surface occurs at a constant rate both in precipitation and as dry fallout. Evapotranspiration serves to concentrate the infiltrating fluid in chloride by a factor that is proportional to the amount of evapotranspirative loss of water in the soil zone. Given this conceptual model, we obtain I using $I = C_p P / C_s$, where C_p is the concentration in precipitation (estimated to be 0.4 ppm by Triay

Figure 11-1. Geochemistry and stratigraphy of well UZ-14 (aqueous chemical data from Yang et al., 1996). (a) Chloride and bicarbonate concentration, (b) sodium, calcium, and magnesium concentration, (c) pH and (d) stratigraphy and zeolite abundance

et al., 1996a), C_s is the concentration in the sample (for example, 87 ppm for the PTn samples in UZ-14, Figure 11-1a), and P is the precipitation rate (taken to be 170 mm/y, from Hevesi and Flint, 1996). This method yields an estimated percolation rate of 0.8 mm/y in the vicinity of UZ-14, with

Figure 11-2. Stratigraphy and finite element grid used for the two-dimensional model cross section. (a) stratigraphy, including locations of zeolitic rocks, (b) finite element grid, (c) close-up of a small portion of the grid near the repository.

estimates based on the individual samples ranging from 0.3 to 1.4 mm/y. This range is lower than the current average infiltration rate in the vicinity of UZ-14 of 3 mm/y cited by Flint et al. (1996), but the agreement is adequate when considered in the context of a spatially varying infiltration model with a range of values from 0 to over 20 mm/y (see the next section).

The chloride concentration at greater depths is significantly lower than the values in the PTn. This result is consistent with either lateral diversion of fluid originating at a higher infiltration region at the surface, or a climate change scenario, in which a wetter climate during the Pleistocene Epoch (about 10 ky ago) resulted in higher infiltration rates and fluids of lower chloride concentration that have taken roughly 10 ky to percolate to this depth. In these simulations, we use

the latter explanation to justify the examination of the neptunium transport system assuming a future wetter climate scenario, in which infiltration rates increase as dissolved ion concentrations decrease.

The cation chemistry in UZ-14 (Figure 11-1b) and other wells at Yucca Mountain are used in the present study to obtain ranges of concentrations to use in simulations and to justify the use of an ion exchange model of neptunium sorption. In the section titled Neptunium Chemistry below, we show that the available sorption data are consistent with ion exchange on zeolitic tuffs. Zeolites are located above the water table in the Calico Hills unit (Figure 11-1d) and the underlying Crater Flats tuffs (located below the water table in UZ-14). With a conceptual model of local one-dimensional downward percolation of fluid, the cation concentrations in Figure 11-1b suggest a dynamic system of ion exchange within the zeolitic horizons, with Na^+ competing with K^+ (data not available for UZ-14), Ca^{2+} , and Mg^{2+} . As with chloride, the profiles are consistent with a transient hydrogeochemical system, in which the relative cation concentrations entering the zeolites are different than in the past. It appears that at the present time, divalent cations are sorbing at the top of the zeolitic horizon at the expense of Na^+ , which is transported to greater depths at higher concentrations than the fluid entering the horizon. Current modeling efforts are directed toward using the ion exchange system to understand the transient hydrologic and chemical system. For now, we use data from wellbores UZ-14 (Figure 11-1), UZ-16, and NRG-6 to bound the range of concentrations of cations in the neptunium ion exchange model. The ranges for the unsaturated zone fluids are 50-200 ppm for Na^+ , 1-50 ppm for Ca^{2+} , 0.1-20 ppm for Mg^{2+} , and 7 to 9 for pH (Figure 11-1c).

11.6.2 Fluid Percolation Rate

The percolation rate of water through the unsaturated zone will control the migration rate of radionuclides to the water table, and thus is of critical importance in these simulations. Of the estimated 170 mm/y average precipitation rate at the site, only a small fraction is expected to escape evapotranspiration and run-off and percolate deep into the subsurface rock layers. The

nature of the infiltration at the surface is likely to be episodic, associated with individual storms and seasonal differences in average precipitation. Infiltration is also almost certainly spatially variable, a result of differing near-surface hydrogeologic conditions. Estimated maps of temporally averaged infiltration rates have been constructed based on models of surface soil hydrologic processes, calibrated with measurements of moisture content in shallow boreholes (Flint et al., 1996). The deep percolation rates derived from these measurements are uncertain due to difficulties in developing a comprehensive model for the evapotranspirative processes that control the results. Nonetheless, estimates vary spatially from zero net infiltration (typically in zones of extensive alluvial cover) to 20 mm/y infiltration. The highest estimates tend to occur at greater elevations and in regions of thin soil layers. The average infiltration rate in the modeled area for this 0 to 20 mm/y range is about 5 mm/y.

Geochemical methods to estimate the rate of deep fluid percolation have recently been compared to the near-surface moisture profile monitoring studies. Estimates using the chloride mass balance method discussed above, yield site-wide estimates of infiltration rate ranging from 0.4 to 22 mm/y (infiltration estimates reported by Triay et al., 1996a). Fabryka-Martin et al. (1996b) illustrate that measurements of ^{36}Cl in samples collected in the ESF are consistent with an average infiltration rate in excess of about 1 mm/y, and perhaps as high as 10 mm/y. Robinson et al. (1996) present results of modeling of the ^{14}C liquid age measurements of Yang et al. (1996), concluding that the data are consistent with percolation rates between 1 and 5 mm/y. Finally, Paces et al. (1996) use a conceptual model of continuous deposition of calcite to estimate the infiltration rate that is consistent with the total estimated quantity of calcite present in the vadose zone. A 2 mm/y infiltration rate is predicted using this method.

The results of these studies illustrate the difficulty of obtaining precise estimates of average infiltration rate through the unsaturated zone. Some of the variability of the estimates is the result of the approximations and lack of sensitivity of the estimation techniques, but the wide range no doubt also reflects true variability in space and time of the infiltration process. In this chapter, we

deal with this variability and uncertainty through sensitivity analyses, in which different infiltration rate scenarios are selected for study. Spatially uniform infiltration rates of 1 and 4 mm/y were selected as representative of the average ranges discussed above, and a spatially variable infiltration rate based on the work of Flint et al. (1996) is used to investigate the impact of spatial variability. For this case, the average infiltration rate at the cross section (Figure 11-2a) selected is 4.8 mm/y, with values ranging from 0 to 18 mm/y. The variable infiltration scenario which is discussed in forthcoming sections refers to this work by Flint et al. (1996).

11.6.3 Computational Domain

The domain selected for the transport calculations is a two-dimensional, East-West cross section through Antler Ridge. Figure 11-2a shows the East-dipping stratigraphy at this cross section, including the zeolitic horizons important to the transport of neptunium, the location of the potential repository and major fault zones that intersect the cross section. Although there are variations in thicknesses of stratigraphic units in the North-South direction, this cross section is a representative slice through the three-dimensional system that allows the issues relevant to transport of neptunium to be studied. The finite element grid for the entire cross section is shown in Figure 3b. The X3D grid generator (Trease et al., 1996) was used to develop this grid. The grid generator is capable of honoring the stratigraphic boundaries, while providing regions with fine discretization, such as the repository region (Figure 11-2c). This allows heat to be applied to a node representing an individual drift (a tunnel containing the waste canisters) in the repository with several nodes in between to capture the heat transfer and fluid flow behavior between drifts in the near-field region. Furthermore, the neptunium inventory can be placed only at nodes representing the drifts. For this grid, the nominal node spacing in the near-field region is 3 m.

11.7 Neptunium Chemistry

The sorption coefficient K_d for ^{237}Np measured on core samples in the laboratory varies from effectively 0 to values of order 5 cc/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). We have combined these experimental results 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 saturated zone in wells J-13 and UE-25 p#1 (Table 11-2) can be used to bracket the composition of the Yucca Mountain fluids in the unsaturated zone (UZ) (Meijer, 1992). J-13 water is a dilute sodium bicarbonate water from the shallow saturated zone. Calcium, potassium, 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 saturated zone from the deeper Paleozoic carbonate aquifer, and has higher concentrations of most of these chemical species.

Table 11-2. Groundwater Compositions (ppm) of Unsaturated Zone Fluids and Fluids Used in Sorption Studies

Species(mg/L)	J-13 Ogard and Kerrisk (1984)	UE25 p#1 Ogard and Kerrisk (1984)	UZ Fluids Yang et al. (1996)
Ca	11.5	87.8(10)*	1-50
Mg	1.8	31.9	0.1-20
Na	45	171	50-200
HCO ₃	143	698	50-300
pH	6.9	6.7	7-9

*Ca concentration after degassing and precipitation of CaCO_3 is shown in parenthesis. The lower value is typical of values in sorption studies (Triay et al. 1996a).

11.7.1 Solubility and ^{237}Np Inventory

Solubility establishes an upper limit for the concentration of aqueous neptunium 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 11-3.

Table 11-3. Solubility of ^{237}Np (Efurd et al. (1996))

Temperature, °C	pH 5.9	pH 7	pH 8.5
25	6.5×10^{-4}	3.1×10^{-5}	1.5×10^{-5}
60	9.4×10^{-4}	1.6×10^{-5}	1.7×10^{-5}
90	9.0×10^{-4}	7.9×10^{-6}	5.5×10^{-6}

We model the precipitation-dissolution of neptunium 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 neptunium released into the system. The neptunium 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 neptunium at each repository node in the two-dimensional

cross section, we divide the total inventory equally into each drift node and divide by the thickness of the repository in the unmodeled direction to place the calculations on a one-meter-thick basis. In the transport simulations, we assume that once the drift node rewets, the canisters immediately are breached and radionuclides escape the near-field environment at the solubility limit of ²³⁷Np. Therefore, for simplicity and in the interest of conservatism, we assume 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.

11.7.2 Speciation

Neptunium released into the aqueous phase participates in speciation reactions. These speciation reactions are given in Table 11-4. The three total aqueous components chosen to model these reactions are NpO_2^+ , HCO_3^- , and H^+ .

Table 11-4. Reactions and Log Equilibrium Coefficients for Np Speciation

Reaction	0°C	25°C	60°C	100°C	150°C	200°C
$H_2CO_3(aq) \rightleftharpoons HCO_3^- + H^+$	-6.58	-6.34	-6.27	-6.39	-6.72	-7.24
$HCO_3^- \rightleftharpoons CO_3^{2-} + H^+$	10.62	10.33	10.13	10.08	10.20	N/A
$H_2O \rightleftharpoons OH^- + H^+$	-14.93	-14.17	-13.04	-12.26	-11.64	-11.26
$NpO_2^+ + H_2O \rightleftharpoons NpO_2(OH)^0(aq) + H^+$	N/A	8.9	8.2	7.6	7.2	N/A
$NpO_2^+ + 2H_2O \rightleftharpoons NpO_2(OH)_2^- + 2H^+$	N/A	20.2	N/A	18.0	17.0	N/A
$NpO_2^+ + HCO_3^- \rightleftharpoons NpO_2(CO_3)^- + H^+$	6.85	5.73	4.48	3.40	2.39	N/A
$NpO_2^+ + 2(HCO_3^-) \rightleftharpoons NpO_2(CO_3)_2^{3-} + 2H^+$	14.59	13.66	12.57	11.53	10.36	N/A
$NpO_2^+ + 3(HCO_3^-) \rightleftharpoons NpO_2(CO_3)_3^{5-} + 3H^+$	23.58	22.49	21.11	19.59	17.69	N/A

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 neptunium, 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 11-3 shows the speciation results for neptunium 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 neptunium present as the NpO_2^+ species 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 11-4 shows the model calculations of fraction NpO_2^+ 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 fifth reaction in Table 11-4). 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 neptunium is travelling through the zeolites, then the temperature dependence will have a significant impact on performance predictions.

Figure 11-3. ^{237}Np speciation in J-13 water at 25°C with a solubility of 5×10^{-5} mol/l.

Figure 11-4. Predicted fraction of ^{237}Np as NpO_2^+ cation as a function of temperature and fluid composition (J-13 fluid versus UE-25 p#1 fluid).

11.7.3 Sorption Data

A comprehensive set of laboratory studies have been performed to characterize the sorption of neptunium 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 ($K_d < 0.4 \text{ cc/g}$) on the vitric and devitrified tuffs in the pathways from the repository to the water table. Small but appreciable sorption (K_d values of $2.5 \pm 1.4 \text{ cc/g}$ 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 neptunium (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 neptunium 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.

11.7.4 Sorption Models

To develop a quantitative geochemical model of neptunium sorption on zeolitic tuffs as a function of groundwater composition, we rely on the batch sorption data cited above (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 11-4. Recall that Figure 11-4 illustrated the effects of the speciation calculations in a plot of the fraction of neptunium present as NpO_2^+ 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 $K_{d,\text{eff}}$ in the zeolitic Calico hills units is given by $K_{d,\text{eff}} = f_{\text{NpO}_2} \cdot K_{d,\text{max}}$, where $K_{d,\text{max}}$ is the maximum distribution coefficient in a system consisting of 100% neptunyl cation, and f_{NpO_2} is the fraction of total neptunium present as neptunyl cation. For the J-13 fluid at 25°C, the speciation results will predict a reduction in f_{NpO_2} and $K_{d,\text{eff}}$ 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 cc/g to 1.5 cc/g in this pH range (Figure 4 of that report).

The transition to neptunium 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#1 fluid. 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, we feel that the reactions of Table 11-4 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, we couple the speciation results presented above 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 ion

exchange model developed in the present study is patterned after a model first proposed by Triay et al. (1996a). Figure 11-5 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:

where *tAl* represents a tetrahedral aluminum sorption site. The first reaction alone is used to model the data of Figure 11-5. The waters at Yucca Mountain contain divalent cations like Ca²⁺ that compete for the available ion exchange sites. The equilibrium constants listed in Eqns. 11-15 and 11-16, along with the cation exchange capacity (CEC) for neptunyl cation of 5 x 10⁻⁴ equiv/L, produce a good match to the data of Figure 11-5. The resulting K_d is 3 cc/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 NpO₂⁺ in 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. We assume 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 neptunium-carbonate complexes due to the higher carbonate

Figure 11-5. ^{237}Np sorption coefficient versus Na^+ concentration in pure sodium bicarbonate solutions (data from Tait et al., 1996). The X's with accompanying curve is the predicted ion exchange model result.

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 cc/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 sorption 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 cc/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.

11.7.5 Column Studies

FEHM simulations using data from columns studies of Triay et al. (1996d) to examine whether ^{237}Np sorption may be kinetically limited under flowing conditions. The suite of column experiments analyzed were conducted using crushed zeolitic tuff with J-13 well water at a pH of 8.3. Crushed tuff was used to overcome mass transfer limitations in the column experiments. In the numerical analyses, the data for tritium, a conservative tracer, was used to fit the dispersivity of the column. The tritium breakthrough emerges from the column at one pore volume, as expected. Using the dispersivity obtained from the tritium simulation, ^{237}Np transport experiments were analyzed using a linear kinetic sorption model. The linear kinetic model was chosen over the competitive ion exchange model, since there were constant chemical conditions in the column. The K_d and the kinetic parameter were fit to the experimental data, yielding a good fit with a K_d of 1.25 cc/g and a kinetic parameter of 8 hr^{-1} . The K_d of 1.25 cc/g is consistent with the batch sorption coefficients for J13 water at a pH of 8.3 (Figure 11-4). Next, to check for kinetic control, the kinetic parameter was set to a large number (greater than 100 hr^{-1} in this system) so that the linear kinetic model would simplify down to an equilibrium model. Figure 11-6 shows that in the column studies, the sorption process is captured more accurately with a kinetic model, rather than an equilibrium formulation. Another ^{237}Np sorption experiment was analyzed using the same column at a different flow rate (2.99 ml/hr). This additional experiment confirmed that the K_d and kinetic parameters are independent of flow rate. The result agrees with the findings Pabalan (1994), who reported that the ion exchange reactions controlling ^{237}Np sorption onto Clinoptilolite equilibrate after one or two days. Despite the slight kinetic control of sorption in the laboratory, no kinetic limitations are expected in the field due to the much longer time scales of transport in the natural system.

Figure 11-6. Tritium and ^{237}Np sorption column study using J-13 well water at a pH of 8.3. The curves represent fits to the data for equilibrium and kinetic sorption models

11.8 Model Results

11.8.1 Preliminary One-Dimensional Flow and Transport Simulations

The unsaturated zone at Yucca Mountain consists of alternating layers of welded and non-welded tuffs with wide contrasts in the matrix permeability and fracture density. This hydrogeologic complexity gives rise to variations in the predicted nature of flow through the unsaturated zone, ranging from fracture-dominated to matrix-dominated flow, depending on the unit. In this section, we develop a one-dimensional isothermal model domain to investigate the nature of flow through the various rock units. The same hydrologic and transport properties are used in these calculations as the forthcoming site-scale simulations. In particular, we address the question of whether it is possible to approximate the radionuclide transport behavior using a single continuum model. In this section we demonstrate that as a first approximation, the relative flow fractions in the fractures and matrix as predicted by a DKM model allow us to assign appropriate effective porosities in the units below the potential repository for transport of radionuclides.

Therefore, an ECM model can be used for transport calculations of heat, fluid, and solute. Specifically, if the flow is predicted to be within the fracture domain, the fracture porosity is used in transport calculations, whereas if matrix flow predominates, the matrix porosity is used. Since the fracture porosity is typically several orders of magnitude smaller than the matrix porosity, this approximation will result in very rapid transport velocities through units controlled by fracture flow, and the overall performance of the unsaturated zone will then be dominated by the behavior of those units predicted to be controlled by matrix flow.

The one-dimensional model used for this analysis extends from the surface to the water table using the stratigraphy of the unsaturated zone at wellbore SD-9, located near the northeast corner of the potential repository. Figures 11-7a and b show the fracture and matrix flow fractions at 1 and 4 mm/y, respectively using the DKM model. Above the potential repository, the flow is fracture-dominated in the TCw, transitions to matrix-dominated flow in the PTn, and back to fracture-dominated (>50% flow) in the TSw. Of greatest importance for radionuclide transport is the flow below the repository, where at 1 mm/y the flow is predicted to convert to matrix flow in the vitric CHn unit and remain as matrix flow to the water table, including in the zeolitic CHn where sorption of ^{237}Np is predicted. Therefore, at relatively low infiltration rates, transport is expected to be controlled by the much larger matrix porosity of the CHn. In contrast, the TSw can be approximated as fracture-dominated flow and transport, obtained by setting the porosity in a single continuum transport model to the fracture porosity.

The situation at 4 mm/y is more complex. For the parameter values selected as the base case, the permeability of the zeolitic CHn matrix is too low to support the entire 4 mm/y flux. Therefore, the rock in the CHnz is predicted to exhibit significant fracture flow (about 60% in Figure 11-7b). There is considerable variability in the measurements of matrix permeability of all of the tuffs, including the CHnz, and this variability will have a significant impact on the prediction of the flow split between fractures and matrix. Figure 11-7c shows that a zeolite permeability a factor of 3 greater (well within the range of measured values) results in a prediction of matrix-

Figure 11-7. The fraction of flow residing in the matrix and fracture for the DKM model.

dominated flow throughout the CHn, as was the case at 1 mm/y. This analysis points out the uncertainty in the nature of flow through the zeolitic CHn unit. This uncertainty is treated by establishing bounds on the type of transport behavior, as discussed below.

Before developing the two-dimensional models of heat and mass transport, we present one-dimensional transport model results using the DKM results just presented, along with ECM results for comparison. For this analysis we use the particle tracking transport module of FEHM because of its efficiency in handling DKM systems with widely contrasting flow velocities in the two continua. For details of the particle tracking method, see Chapter 4. Figure 11-8 shows the breakthrough of ^{237}Np at the water table versus time in the one-dimensional model. At 1 mm/y,

the ECM and DKM models result in virtually equivalent breakthrough curves, a consequence of the matrix-dominated flow through the CHn predicted by the DKM model (Figure 11-8a). A very small fraction of the mass arrives early in the DKM model, but the majority of the radionuclide travels through the matrix, and the two models give very similar results. At 4 mm/y, the situation is similar for the high-permeability zeolite case. However, for the base-case properties, the DKM model predicts a significant portion of the ^{237}Np inventory traveling through fractures. The resulting breakthrough curve at the water table is bimodal, with a large flux arriving within the first 30,000 years of the simulation. Clearly, an ECM model is not capable of reproducing this behavior, except in a bounding sense. In most of the two-dimensional ^{237}Np transport simulations, we assume that flow is matrix-dominated throughout the zeolitic units since this assumption facilitates the investigation of the effect of chemical and thermal conditions on sorption and transport. However, to assess the role of the uncertainty of the flow field on predicted transport of ^{237}Np , we also present a simulation in Section 11.8.3 in which the fracture porosity is assumed for transport through the zeolites, thereby bounding the behavior.

11.8.2 Thermohydrologic Simulations

In this section, we describe the fluid flow and heat transport behavior of the model to set the stage for presenting the neptunium transport results. Current reference design conditions for the heat load issuing from the repository are assumed: the inventory of waste totalling 70,000 Metric Tons Uranium (MTU) with an 83 MTU/acre initial heat load, with waste emplaced in drifts that are 22 m apart. At the repository portion of the model, heat is supplied to individual nodes of the grid along a series of nodes spaced 22 m apart, so that in these calculations, there are roughly four unheated nodes between each node representing an emplacement drift. In the unmodeled direction, the heat load is assumed to be uniform, so that the appropriate heat load in the two-dimensional cross section (taken by convention to be 1 m thick) can be applied. The time-varying heat load exhibits a sharp drop from its initial value within the first 100-1000 years. The heat load declines to 30% of its initial heat load after 100 years and 7% after 1000 years. Infiltration rate boundary

Figure 11-8. Breakthrough of ^{237}Np at the water table in the one-dimensional model. Comparison of the ECM and DKM models for different flow scenarios.

conditions were described earlier, and the fluid flow boundary condition at the bottom of the model domain is a fluid saturation of one to represent the water table.

Thermal and air pressure boundary conditions are also required as input to the model. At the surface, temperature and pressure are assigned according to the surface elevation; time-invariant values are based on a method developed by Ross et al. (1992) which captures the elevation dependence of air properties. The bottom and side boundaries are no-flow air boundaries. The thermal boundary condition at the water table is a flux-based boundary condition for the heat flux Q_b which takes the form $Q_b = \Omega(T - T_b)$ where T_b is the temperature outside of the model boundary at which the heat exchange is taking place and Ω is a scaling parameter. Adjustment of Ω allows us to examine a range of thermal conditions at the water table ranging from isothermal to the approximate behavior assuming conduction-limited heat transfer in the saturated zone.

Figure 11-9a shows the predicted fluid saturation response of the system after 500 years for a

uniform 4 mm/y infiltration rate. Over a period of roughly 10,000 years, the model predicts dry-out, followed by rewetting, near the emplacement drifts. A two-phase, countercurrent flow zone develops above the potential repository, with buoyant and pressure-gradient-driven vapor movement in the upward direction counterbalanced by downward fluid percolation, the primary source of which is condensed steam. The resulting heat transfer mechanism, called a heat pipe, is much more efficient than thermal conduction alone, and results in an extended two-phase region of enhanced heat transfer at about 1 atm and 100°C (Figure 11-9b). This heat pipe sets up regions of net boiling and condensation that in turn impact the concentrations of dissolved ions. Figure 11-9c shows the predicted concentration pattern of Cl^- ion (assumed to be conservative), along with the general movement of vapor at a time of 2000 years. Net vaporization at the edges and below the repository concentrates the fluid in those zones, and net condensation directly above the repository dilutes the fluid. This pattern evolves through time, changing the chemical composition from its initial state. When the heat from the repository region is insufficient to maintain the two-phase zone, the zone collapses, and fluid reenters the repository region. The rewetting of the repository and return of the system to its ambient conditions occurs within 10,000 years of waste emplacement.

The temperature and fluid saturation near the emplacement drifts control the hydrologic and gas-phase moisture conditions that in turn control the degradation of the protective waste package containers, the dissolution of the solid radioactive waste, and the solubility of neptunium. At all infiltration rates selected, the model predicts complete dry-out of the rock near each drift, with the duration of complete dry-out conditions depending on position within the repository. Closer to the middle of the repository, dry-out regions associated with the individual drifts are predicted to merge, forming a continuous region of dry rock through which fluid cannot percolate. In contrast, an “edge effect” is predicted for drifts at either end of the repository, where enhanced downward percolation and lateral cooling effects result in a lack of coalescence of dry-out zones. Figure 11-10 shows the predicted temperature versus time in the rock near two emplacement drifts

Figure 11-9. Influence of repository heat on thermohydrologic and chemical conditions in the unsaturated zone. (a) Fluid saturation profile (500 years), (b) temperature profile (500 years), (c) chloride concentration (2000 years).

(one closer to the middle of the repository, called the “center” drift, and one near the edge of the repository, called the “edge” drift) for the 4 mm/y infiltration rate scenario. In addition, the figure shows the predicted influence of infiltration rate on the temperature at the center drift. The period during which a drift is elevated to temperatures greater than 100°C corresponds to dry-out of the rock to low saturation. The period of complete dryout at a given infiltration rate depends most critically on the position of the drift within the repository. For example, for the 4 mm/y case, the onset of rewetting is predicted to occur within the first 100 years near the repository edge and between 500 and 1000 years for an inner drift. These results are in agreement with one-dimensional dual permeability simulations by Ho (1997), who predicted thermally induced increases in flux lasting for about 1000 years. Thus the choice of ECM for the model calculations appears to be acceptable from the standpoint of predicting the duration of the thermal perturbation to the flow field.

Figure 11-10. Temperature versus time in the rock near an emplacement drift in the repository. “Center”-drift near the center of the repository, “Edge” -drift near the edge of the repository. The family of curves for the “Center” drift show the influence of infiltration rate on the results.

The infiltration rate also has some influence on the model results. Figure 11-11 shows the predicted temperature-time histories at two positions in the Calico Hills unit corresponding to the top of the Calico Hills zeolitic horizon. Two comparisons are presented in the figure. First, examining the solid curves labelled “Isothermal”, we note that the greater temperature rise occurs for the position that is closer to the middle of the repository heat source, despite the fact that the top of the zeolites is actually 25 m farther from the heat source than for the position below the edge of the repository. The other comparison illustrates the impact of the heat transfer boundary condition at the water table. The conduction-limited model is more insulating than the isothermal condition, resulting in a predicted rise in rock temperature at the water table (Figure 11-11) of roughly 25°C that subsequently returns to the ambient value. This effect in turn results in a higher predicted temperature rise and a longer duration of elevated temperature in all units between the water table and the repository horizon, including the zeolitic rocks.

Figure 11-11. Temperature versus time at the top of the Calico Hills zeolitic horizon. “Center”-position located beneath the center of the repository; “Edge”-drift near the edge of the repository. The influence of the assumptions regarding the heat transfer at the water table boundary are also shown.

11.8.3 Neptunium Transport Simulations

In the site scale neptunium transport simulations, we study the effects of infiltration rate, temperature, pH, and bicarbonate, sodium and calcium concentrations on neptunium migration from the repository to the water table. 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 saturated zone transport models that predict the concentration and dose associated with ^{237}Np in the underlying groundwater system. All simulations utilize the competitive ion exchange reaction model with speciation discussed above.

Figure 11-12 shows the position of the ^{237}Np plume at various times during one of the site scale simulations. The first three profiles are total aqueous ^{237}Np concentrations (mol/kg water), and the last profile is the sorbed ^{237}Np concentration (mol/kg rock). In this simulation, the infiltration rate is 4 mm/y, pH is 8, bicarbonate concentration is 150 mg/l, sodium concentration is 125 mg/l and calcium concentration is 25 mg/l. We will refer to this simulation as the base case.

Figure 11-12a shows the repository after the waste canisters begin to cool and water begins to rewet the edges of the repository. At early times, ^{237}Np begins to dissolve and quickly flows through the Topopah Springs welded unit via fracture flow. The migration of ^{237}Np is slowed significantly by the Calico Hills nonwelded units, where matrix flow dominates. The ^{237}Np then begins a slow migration through the nonwelded tuff matrix towards the water table. Figure 11-12b shows the transport plume after all drifts have rewet and releases occur throughout the repository. In Figure 11-12c, the main ^{237}Np plume has reached the water table and is more dilute, mainly due to dilution with the percolating fluid and sorption to the zeolitic tuffs. Radioactive decay of ^{237}Np is considered but at the infiltration rates chosen for study, transport times are short enough that the impact of decay on concentrations is negligible. Figure 11-12d shows the sorbed ^{237}Np site concentration. The sorbed concentration profile illustrates the combined effects of the ion exchange and speciation model. For the base case, the approximate retardation factor for ^{237}Np

Figure 11-12. ^{237}Np transport through the unsaturated zone. (a) ^{237}Np total aqueous concentration profile after rewetting of the repository region and mobilization of ^{237}Np at all locations (10,000 years), (c) ^{237}Np total aqueous concentration profile as ^{237}Np reaches the water table (50,000 years), and (d) ^{237}Np immobile site concentration profile (50,000 years).

sorption onto the zeolitic layers is about 20. However, certain scenarios discussed below can greatly diminish the retardation of the zeolitic layers.

Figure 11-13 shows the predicted fate of ^{237}Np for infiltration rates of 1 mm/y, 4 mm/y and the variable infiltration scenario. Two sets of results are presented in the figure. First, the quantity of solid ^{237}Np remaining at the repository horizon is plotted as moles versus time (the curves that decline from their initial values to 0) during the simulation. Also, the ^{237}Np breakthrough curves at the water table are presented. Recall that the variable infiltration rate averages 4.8 mm/y at the cross section, with values ranging from 0 to 18 mm/y. As expected, the higher the infiltration rate, the faster the release of ^{237}Np from the repository resulting in a shorter travel times and higher peak concentration at water table. For the variable infiltration scenario, the inventory is completely mobilized in about 20,000 years, in contrast to 70,000 years for the 1 mm/y case. Regarding the breakthrough curves at the water table, infiltration rate has the expected result: higher fluid flow rates decrease breakthrough times and increase the peak radionuclide mass flux at the water table.

Figure 11-13. Mobilization of ^{237}Np from the near field, and breakthrough curves at the water table for different infiltration scenarios. The declining curves starting at early times represent the ^{237}Np mass remaining as a solid in the repository region (the left y-axis). The breakthrough curves exhibiting peaks are ^{237}Np flux at the water table (the right y-axis).

The matrix permeability of the nonwelded zeolitic Calico Hills units is a major uncertainty in the transport calculations. Section 6.1 showed that for a infiltration rate of 4 mm/y, the base-case zeolite permeability results in primarily fracture flow through the zeolitic Calico Hills, whereas, a zeolite permeability three times the base-case value results in matrix-driven flow. Using the DKM model for the site scale simulations is beyond the scope of this reactive transport study and is dealt with in detail elsewhere in this report. To provide a lower bound on the travel time to the water table for ^{237}Np , we simulate transport through the nonwelded zeolitic units by setting the porosity of the Calico Hills zeolitic units equal to the *fracture* porosity. In addition, we assume that the fractures do not sorb ^{237}Np . This simulation serves as a worst case scenario for the site scale model. Figure 11-14 shows that the fracture bypass case leads to ^{237}Np travel times that are significantly shorter than the base case scenario. The influence of sorption onto zeolites, shown above to result in a retardation factor of about 20 in the zeolites, is negated when fracture transport is assumed. If this is the case, significant breakthrough occurs at the water table before 10,000 years. Further experimental work and modeling will be necessary to better constrain the permeability of the zeolites to resolve the flow conditions in the Calico Hills units. To examine the influence of chemistry on ^{237}Np sorption and transport, we assume hereafter that matrix transport predominates in the zeolites, while recognizing that this uncertainty remains.

We next examine the influence of fluid pH on ^{237}Np sorption and transport. 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 $\text{CO}_2(\text{aq})$ throughout the model while allowing for all other components to dilute and concentrate due to heating effects. In effect, we are

Figure 11-14. Fracture bypass of the zeolitic Calico Hills units.

assuming that the partial pressure of $\text{CO}_2(\text{g})$ 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 16). This result qualitatively agrees with the one-dimensional model results of Lichtner and Seth (1996). The extent of the pH rise is perhaps overpredicted in our model, since we do not include buffering by dissolved silicic acid, which has a dissociation constant (pK_a) of 9.8. However, we show below that this simplification results in conservative model predictions since the higher the pH at the zeolites, the less sorption of ^{237}Np . Figure 11-15 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 indicates 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, because the pH influences both the solubility and sorption behavior. As pH rises, solubility

Figure 11-15. The effect of CO_2 degassing on pH at the center of the repository (heat-pipe front) and at the top of the zeolitic layers.

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 11-16 shows the ^{237}Np breakthrough for initial pHs of 7, 8, and 9 (we will discuss the “carrier plume” model results 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, we fix the pH of the solution 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

Figure 11-16. Influence of pH on the ^{237}Np breakthrough curves at the water table. The “carrier plume” result assumes an elevated pH of 10 at the repository for a period of either 500 or 5,000 years due to cement-water reactions.

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.

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 unsaturated zone 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 11-2 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 we showed that the higher infiltration rate scenarios would result in a more rapid release of ^{237}Np , but in those simulations we assumed that sodium and calcium concentrations were independent of infiltration rate. We now present a transient simulation in which we assume 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, we assume that sodium and calcium concentrations more dilute by a factor of four during the higher infiltration period. Figure 11-17 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 cc/g to 5.5 cc/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.

The influence of repository heat on the flow field, transport properties, and ^{237}Np migration is now considered. Recall that Figure 11-4 showed that speciation and sorption could be strongly affected by temperature effects. Furthermore, the flow field itself is significantly different than that

Figure 11-17. Influence of transient infiltration and chemical conditions on the ^{237}Np breakthrough curves at the water table. The “changing climate” result assumes an increase from 1 mm/y to 4 mm/y, accompanied by a four-fold decrease in cation concentrations. The 1 mm/y and 4 mm/y steady state results without geochemical transients are shown for comparison.

of the ambient system. Despite these potential complications, Figure 11-18 shows that the details of the heat transfer process have little effect on the transport of ^{237}Np . The primary reason for this result is that the temperature of the zeolites is predicted to have returned to near ambient conditions by the time the centroid of the ^{237}Np plume reaches the zeolites. Solubility is low enough to extend the period over which releases occur to greater than 10,000 years, long enough for the relatively short-term thermohydrologic transients to have abated. The only observable heat effect impacting ^{237}Np migration in the simulations is for the conduction-limited heat boundary condition. However, even this effect is insignificant. Thus, the results indicate the current model is relatively insensitive to heat effects.

Figure 11-18. Influence of thermal boundary condition at the water table on the ^{237}Np break-through curves at the water table.

11.9 Discussion and Conclusions

The need to develop a reactive transport model for ^{237}Np migration in the unsaturated zone is motivated by its relatively weak (but significant) sorption, its high solubility, and long half-life. Considerable effort has been expended into characterizing the chemical properties of ^{237}Np in Yucca Mountain fluids, including solubility, speciation, and sorption. This chapter places these measurements into perspective for the Yucca Mountain site by combining the chemical information with the latest site characterization information on the hydrology, geochemistry, and projected thermal behavior of the unsaturated zone.

The computational intensity of performing multiple simulations of the full thermohydrologic and reactive transport behavior of ^{237}Np has led to various simplifications in previous analyses, including isothermal, steady state flow, an equivalent K_d model for sorption, and simplified source term expressions for the release of ^{237}Np from the near-field environment. Some of these simplifying assumptions will be necessary in future performance assessment

exercises due to the large number of simulations required to treat uncertainty in system parameters. As one example, the release of radionuclides from the near-field was assumed to occur as soon as rewetting of the drift takes place. In reality, time lags due to limited corrosion of the waste packages and slow transport from the packages are more realistic. To quantify the uncertainty of these and many other processes, Monte Carlo analyses are used, and thus simplifications of the full reactive transport model are required for expediency. Thus, one of the key outcomes of this work is to assess the validity of these simplifications and to propose abstractions of the complex behavior that can be used in performance assessment analyses.

The chemistry of the unsaturated zone 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 cc/g translates into an order of magnitude difference in travel time through the zeolitic rocks. These travel times within the unsaturated zone 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 unsaturated zone 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 unsaturated zone 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 cc/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 cc/g. Finally, the bicarbonate concentrations in unsaturated zone 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, the reactive transport model results can be used to justify model simplifications through a sensitivity analysis. Without the reactive transport model capability, justifying the simpler K_d approach would be much more difficult.

The impacts of waste heat and thermohydrology on the large-scale migration of ^{237}Np through the unsaturated zone appear to be relatively minor in comparison to, for example, the percolation flux through the unsaturated zone or sorption onto the zeolites for ^{237}Np . The perturbation from ambient temperature conditions has, in most simulations, subsided before the bulk of the ^{237}Np plume has reached the water table. This result is caused by several effects. First, the near-field thermohydrology (dry-out accompanied by minimal liquid flow into the drifts) is

predicted to limit releases of ^{237}Np during the period of the most vigorous thermal effects. Second, once waste packages become wet and ^{237}Np is leached, the solubility is low enough to stretch the period over which ^{237}Np escapes the near field to tens of thousands of years. This is a long enough period for the thermal wave to have returned to close to its ambient state. Although the ECM model assumption can be questioned, one-dimensional dual permeability calculations performed by C. Ho (C. Ho, pers. comm., 1997) agree with this general time scale of about 1000 years. Third, the redistribution of water in the form of vapor creates zones of concentration and dilution of dissolved species, however, the magnitude of these effects and their persistence in time are not great enough to significantly affect ^{237}Np transport. Finally, for ^{237}Np , the impact of temperature on speciation and sorption is only relevant in the zeolitic rocks, which are far enough from the heat source to minimize the potential impacts on transport. Nonetheless, under conditions of thermal-conduction-limited heat transfer in the saturated zone persists, the thermal plume persists for long enough to potentially influence the transport behavior of ^{237}Np in the zeolites. Thus, further work to bound the nature of the heat transfer coupling between the unsaturated zone and saturated zone is warranted.

Given the relative lack of significance of thermohydrology on site-scale transport, we tentatively conclude that isothermal calculations can be used in performance assessment modeling studies. However, future studies under non-isothermal conditions should address key uncertainties not treated here. The constancy of hydrologic and transport properties in the presence of wide changes in temperature is a key assumption that should be tested. Full thermohydrologic and transport simulations will also be necessary to further study the chemistry of the fluid that passes through the repository region. Nonetheless, isothermal fluid flow calculations are an appropriate platform on which to design Monte Carlo simulations in performance assessment analyses. An important aspect of the nonisothermal behavior that needs to be retained even in isothermal transport simulations is the temperature-dependence of ^{237}Np solubility, which in turn affects the duration of release. This effect can easily be included by using the rate of release from the

repository region from nonisothermal simulations as input to an isothermal simulation. Finally, the issue of fracture bypassing of the zeolites is an additional uncertainty in assessing the migration of ^{237}Np through the unsaturated zone. Using dual permeability simulations, we demonstrated that significant fracture flow through zeolites may occur, resulting in much more rapid transport to the water table and higher peak mass flux values. This uncertainty is being addressed through numerical simulations using the dual permeability model. A variety of fracture-matrix coupling models, hydrologic property data sets, and infiltration rates are being examined in two- and three-dimensional simulations, and will be the focus of a future paper. Until more definitive conclusions can be reached, our results on the influence of heat and chemical conditions must be viewed as provisional, given that, except for the zeolite bypass case, the modeling was performed under conditions in which matrix flow occurred in the zeolitic rocks.

Regarding the efficiency of the reactive transport code, the computational intensity of the simulations presented here proved to be manageable in two dimensions, allowing timely results to be obtained. The Selective Coupling method implemented in FEHM was crucial to the success of the ion exchange simulations. Ion exchange is an example of a chemical system in which chemical component concentrations are strongly coupled. In such instances, if the reaction kinetics are rapid compared to the transport time scale, convergence is difficult to attain unless the transport step is performed with coupling of the components in the ion exchange reaction. In the future, we will examine issues of computational efficiency in greater detail, determining the relative merits of Selective Coupling versus traditional SIA methods. The results of the present study suggest that the Selective Coupling method is a promising solution technique for multi-dimensional, reactive transport problems. Further development of the Selective Coupling technique will be pursued to attain additional computational and memory savings.

11.10 Conclusions

In this chapter we investigate the coupled effects of chemical interactions and heat on neptunium transport from the potential repository to the water table. The simulations indicate that in the absence of irreversible changes in the hydrologic and transport properties, the heat pulse does not significantly affect the migration of neptunium, as the time scale of heat pulse propagation is shorter than the time scales associated with neptunium release and migration. The major uncertainty in this conclusion is the possibility of rock-water interactions that permanently alter the porosity or permeability. Future investigations should focus on this possibility. Water chemistry, particularly pH and calcium and sodium concentrations, significantly affect the retardation of neptunium by the zeolitic rocks between the repository and the water table. The chemical compositions derived from field measurements appear to constrain the fluid chemistry sufficiently to allow for the estimation of sorption coefficient for ^{237}Np . Finally, the selective coupling method introduced to solve these reactive transport problems is a robust numerical technique applicable to a wide range of reactive transport problems.

Chapter 12 - Incorporation of Geostatistics into Flow and Transport Simulations

12.1 Abstract

The zeolitic threshold method used in previous chapters is replaced with more rigorous geostatistical modeling. With this more sophisticated modeling approach, we examine whether using a threshold cutoff for zeolitic abundance is an appropriate upscaling method for characterizing small scale heterogeneity. Geostatistical techniques are used to develop variograms, kriging maps, and conditional simulations of zeolitic abundance. The kriging maps are used to estimate the uncertainty in the mineralogical model. The conditional simulations result in equally probable maps of zeolitic abundance. Using the algorithms discussed in Chapters 2 and 3, a conditional simulation of zeolitic abundance was mapped on top of the geologic model. We investigate the relationship between percent alteration, permeability changes due to alteration, sorption due to alteration, and their overall effect on radionuclide transport. A key conclusion to the study is that the retardation due to sorption predicted by a conditional simulation is much larger than the retardation predicted by the zeolite threshold method. The reason for larger retardation of ^{237}Np for the conditional simulation is a small but significant K_d at locations with zeolite abundance less than 10%. For the purposes of abstracting these results, setting nonzero K_d 's in the vitric tuffs may be an effective way to mimic the more rigorous conditional simulations. Rocks with high zeolitic abundance may not be as effective in retarding radionuclides such as ^{237}Np and Uranium since these rocks also in general have low permeability and contaminants can only 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. In future studies, multiple conditional simulations will be performed using linear coregionalization models to strengthen the conclusions drawn in this study.

12.2 Software and Data QA Status

The FEHM code is used to perform all flow and transport calculations in this report. The code is qualified in accordance with Los Alamos quality assurance requirements and is documented in (Zyvoloski et al., 1992, 1995a, 1995b). The QA status of the data used in this report is shown in Table 12-1. The petroleum industry standard software packages STRATAMODEL (Stratigraphic Geocellular Modeling Reference Guide, 1995) and RC² (ResMod User's Manual, 1994) are used in the geostatistical analyses of heterogeneous materials. These are off-the-shelf software packages and are applied only to sensitivity studies in the present study. They are non-Q with respect to YMP and Los Alamos quality assurance requirements but they follow well documented methods for geostatistical representation of spatially varying attributes.

Table 12-1. QA Status of Data used in this Chapter			
REFERENCE	Q Status	DTN	COMMENT
Altman et al., 1996 , "Flow Calculations for Yucca Mountain Ground Water Travel Time (GWTT-95). Technical Report SAND96-0819, Sandia National Laboratories, 1996	non-Q	No DTN Assigned	No DTN Assigned
Bodvarsson et al., 1997 "The Site-Scale Unsaturated Zone model of Yucca Mountain, Nevada, For the Viability Assessment, LBL Technical Report LBNL-40378	Q	LB970601233129.001	No DTN assigned. Hydrologic property sets computed in this report are used throughout the present study.
Chipera et al., 1997a , "Preliminary Three Dimensional Mineralogical Model of Yucca Mountain, Nevada, YMP Milestone SP321AM4"	non-Q	Not Assigned	No DTN assigned
Chipera et al., 1997b "Mineralogic Variation in Drill Holes USW NRG-6, NRG-7-7a, SD-7, SD-9, SD-12, and UZ#14: New Data from 1996-1997 Analysis Los Alamos National Laboratory YMP Deliverable SP321BM4"	Q	LADV831321AQ97.001	

Flint et al., 1996 , Conceptual and numerical model of infiltration for the Yucca Mountain Area, Nevada", USGS WRIR MOL 19970409.0087	Q	GS960908312211.003/ NA	USGS DTN 3GUI623M
Flint, L. E., 1996 , "Matrix properties of Hydrogeologic Units at Yucca Mountain Nevada", USGS Water-Resources Investigation Report, WRIR-96	Q	GS9609083122231.004 /NA	USGS DTN Report in Preparation.
Triay et al., 1996a , "Summary and Synthesis Report on Radionuclide Retardation for the Yucca Mountain Site Characterization Project, Los Alamos National Laboratory YMP Milestone Report 3784	Q	LAIT831341AQ96.001	
Triay et al., 1996b , "Radionuclide Sorption in Yucca Mountain Tuffs with J-13 Well Water,,: neptunium, Uranium, and Plutonium, Los alamos National Laboratory Yucca Mountain Site Characterization Project" Milestone 3338, Technical Report LA-12956-MS	Q	LAIT831341AQ96.001 *	* DTN supersedes DTN LA000000000104.001
Triay et al., 1996c , "Batch Sorption Results for neptunium transport through Yucca Mountain tuffs, Los alamos National Laboratory Yucca Mountain Site Characterization Project Milestone 3349, Technical Report LA-12961-MS	Q	LAIT831341AQ96.001 *	*DTN supersedes DTN LA000000000090.001
Triay et al., 1996d , "Comparison of Neptunium Sorption Results using batch and column techniques, Los Alamos National laboratory Yucca Mountain Site Characterization Project Milestone 3041, Technical Report LA-12958-MS	Q	LA000000000106.001	DTN Accession No. MOL.19950630.0300

12.3 Motivation

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 we are concerned with zeolite heterogeneity in the unsaturated zone, 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 chapter, we study the impact of small-scale variability in the zeolite distribution on the transport of ^{237}Np through the unsaturated zone. 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 unsaturated zone.

Another assumption made in previous chapters was to use a threshold cutoff for zeolitic abundance. 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 chapter 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 12-1a shows the zeolitic distribution when a 10% threshold is used as the basis for considering a node to be zeolitically altered. The 10% threshold results in a zeolitic distribution that roughly follows the layers in Figure 2-1b of Chapter 2. A potential problem with using a simple threshold is that the zeolitic distributions change significantly as a function of the threshold. Figure 12-1b shows the zeolitic distribution for a 20% threshold: the layers lose 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.

a

b

Figure 12-1. Antler Ridge cross section (a) hydrologic zones including rock with greater than 10% zeolitic abundance, (b) hydrologic zones including rock with greater than 20% zeolitic abundance.

This new 3-D framework model of fully distributed mineralogic abundances and corresponding hydrologic properties has been further improved through the application of geostatistical techniques that help characterize the uncertainties in spatial distribution of the hydrologic and chemical properties. These improvements now make it possible to refine the contaminant transport simulations. This chapter will summarize the ongoing efforts to integrate the mineralogic model and geostatistical techniques into the flow and transport simulations to better capture the effects of chemical and physical heterogeneity.

The following topics are covered in the sections of this chapter:

- Methodology for integrating geostatistical methods into the mineralogical model using a commercially available geostatistical software package (RC²);
- A simple example to illustrate the implications of the choice of the threshold value for zeolitic cutoff;
- Field data that support a correlation between the physical and chemical properties at Yucca Mountain indicating that these correlations can have a large impact on the travel times for ²³⁷Np. These correlations are used to study the relationship between zeolitic abundance, van Genuchten parameters, permeability and reactivity;
- Geostatistical techniques are used in conjunction with the mineralogical model to develop variograms, kriging maps, and conditional simulations of zeolitic abundance;
- Transport simulations are conducted with zeolitic abundance correlated to permeability, the van Genuchten model parameter α , and the distribution coefficient K_d . This simulation is used to investigate whether correlated heterogeneities can influence the travel times of ²³⁷Np through the unsaturated zone, and if so, to devise abstracted models to capture the behavior.

12.4 Geostatistical Integration

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 we 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 (see Chapter 2) interfaces to a compatible commercially available geostatistical software package (RC²). The regularly-spaced mineralogic data and stratigraphic surfaces output from SGM are imported by RC² 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 12.7.

12.5 Implications of Zeolite Threshold Cutoff

A simple example is developed in this section to illustrate the effect of using a threshold cutoff for zeolitic abundance on the transport of ²³⁷Np. Transport through a 10 m by 10 m block of unfractured vitric tuff with a zeolitic layer will be compared to a block of vitric tuff containing

a rectangular zeolitic lens (Figure 12-2). Each block is representative of a typical element in the site-scale model for the Calico Hills unit. The layered case is analogous to a zeolitic layer shown in Figure 12-1a, whereas, the lens case is used to illustrate the case of a discontinuous zeolite distribution as shown in Figure 12-1b. In these block models, care must be taken to choose boundary conditions which will simulate a single element of the site-scale model. In the site-scale model, the water table serves as the bottom boundary condition and is thus simply set to a saturation of one. At steady state, the capillary pressure gradient and gravitational forces equilibrate to establish a saturation profile throughout the model domain. Due to the large size of the site-scale model, the capillary pressure gradient across a typical element is small. The balance between capillary pressure, which acts to retain water in the pores, and gravitational forces leads to a net downward percolation.

Figure 12-2. Two possible zeolitic geometries at the subgrid scale (10 m by 10 m).

In the block models, the bottom boundary cannot be set to a saturation of one as in site-scale models, because the block represents an element from the Calico Hills unit which is hundreds of meters above the water table. Therefore, we set the top and bottom boundaries of the block model to a constant capillary pressure, which results in a steady infiltration rate through the block. Lateral flow across an element can be important in the site-scale model but is not considered in this

example. The left and right sides are no-flow boundaries. We assume that these blocks are in a region of the site-scale model where downward percolation dominates.

Table 12-2 shows the properties of the vitric and zeolitic tuffs, and Figure 12-3 shows the relative permeability characteristic curves obtained from the van Genuchten model (van Genuchten, 1980). Model parameters (Flint, 1996) are given in Table 12-2.

Table 12-2. : Hydrologic and chemical properties of vitric and zeolitic tuff

Parameter	Vitric Tuff	Zeolitic Tuff
saturated hydraulic conductivity @25°C, (m/s)	4.3×10^{-9}	7.03×10^{-11}
saturated permeability, k_{sat} (m^2)	4.4×10^{-16}	7.17×10^{-18}
α of van Genuchten model (1/m)	0.0267	0.0035
n of van Genuchten model	1.386	1.495
porosity, ϕ	0.27	0.29

The quantity $k_{sat} * R_1$ is the effective permeability of the unsaturated medium. The characteristic curves of $k_{sat} * R_1$ versus capillary pressure show that, as expected, zeolitic tuff is less permeable than vitric tuff for capillary pressures less than 100 m. However, for capillary pressures greater than 100 m, the zeolitic tuff is actually more permeable than the vitric tuff. The permeability trends can be explained by the saturation-capillary pressure plot (Figure 12-3). At low capillary pressures, the zeolitic and vitric tuffs are highly saturated and the permeabilities of the media approach the saturated permeability values. However, at a high capillary pressure, both media are drier, allowing the zeolitic tuff to actually conduct as much flow (or a little more) as the vitric tuff.

At steady state, the capillary pressure and gravitational forces equilibrate, leading to a net downward infiltration through the media. The capillary pressure at the model boundaries can therefore be chosen to obtain a given infiltration rate throughout the media. For example, setting the capillary pressure to 24 m results in an infiltration rate of about 4 mm/yr, whereas, a capillary pressure of 155 m results in an infiltration rate of about 0.1 mm/yr. Note that the infiltration rates

Figure 12-3. Comparison of the characteristic curves for zeolitic and vitric tuff.

are slightly different for the lens and layer cases for a given capillary pressure but are close enough for this illustrative example.

Three simulations were conducted at infiltration rates of 4 mm/yr, 1 mm/yr and 0.1 mm/yr (Figure 12-4). For the 0.1 mm/yr ($P_{cap} = 155$ m) simulation, the velocities are actually higher through the zeolites than through the vitric tuff, resulting in preferential flow through the zeolites. At 1 mm/yr ($P_{cap} = 88$ m), a slight bypass occurs around the zeolites with preferential flow through the vitric tuff. Again, the velocities through both media are similar. However, at 4 mm/yr ($P_{cap} = 24$ m), velocities are much higher through the vitric tuff than the zeolitic tuff. These trends are a direct result of the $k_{sat} \cdot R_l$ vs. capillary pressure curve (Figure 12-3). Interestingly, the unsaturated permeabilities of the media change drastically over the range of infiltration rates that are expected at Yucca Mountain.

After a steady state flow field is obtained, ^{237}Np is injected uniformly along the top boundary to simulate the downward migration of ^{237}Np from the repository. For simplicity, ^{237}Np is assumed to linearly sorb to the zeolitic tuff with an equivalent K_d of $2 \text{ cm}^3/\text{g}$, and to not sorb to

Figure 12-4. Velocity profile for the zeolitic lens case at three different infiltration rates.

the vitric tuff. In both blocks zeolites comprise 25% of the domain with the difference being the geometry of the zeolite (Figure 12-2). Table 12-3 shows the parameters used to conduct the simulations. The dispersivity and diffusion parameters are hypothetical values but are appropriate for ^{237}Np in Calico Hills tuffs.

Table 12-3. : Parameters used in the simulations

Molecular Diffusion, D^*	$1 \times 10^{-10} \text{ m}^2/\text{s}$
Longitudinal Dispersivity, a_l	0.01 m
Transverse Dispersivity, a_t	0.001 m
Grid spacing, Δx	0.3125 m
Time step, Δt	5,000 days

Figure 12-5 compares the cumulative mass of ^{237}Np arriving at the bottom boundary for the layered and the lens cases. At each infiltration rate, a pulse of ^{237}Np was injected into the system at the beginning of each simulation. Specifically, 166 moles of ^{237}Np at 4 mm/yr, 60 moles at 1 mm/yr and 11 moles at 0.1 mm/yr were injected over a 10 year period. For cases of lower infiltration rates, the layered case and lens case behave similarly since there are no major preferential pathways. However, for the 4 mm/yr case, the ^{237}Np is retarded to a greater extent for the layered case than for the lens case. This set of simple simulations shows that at relatively high infiltration rates, more extensive bypassing will occur if there are breaks in the zeolitic layers. Specifically, flow will preferentially avoid the zeolitic areas at relatively high infiltration rates. Section 12.7 will show that zeolitically altered portions of tuff are actually correlated over length scales much larger than 10 m. Therefore, the size of the idealized flow block used in later flow and transport simulations will need to be modified. In this example, rock was assigned either zeolitic or vitric properties. This assumption is analogous to using the 10% zeolitic cutoff value. One of the questions we try to address in the rest of this chapter is whether using a zeolitic cutoff value is reasonable assumption.

12.6 Relationships Between Physical and Chemical Heterogeneity at Yucca Mountain

Figure 12-1 shows how a 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

Figure 12-5. Comparison of ^{237}Np breakthrough curves for the layer and lens cases. 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%. In this section, we examine 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.