Special Section: Contaminants in the Vadose Zone

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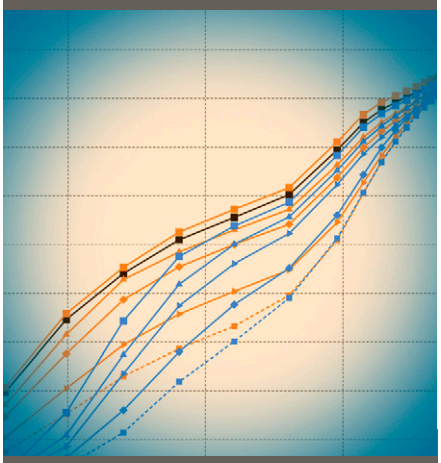

This paper describes a particle-tracking technique for dual-permeability media suitable for simulation of radionuclide transport through unsaturated, fractured rock. The model is used to conduct sensitivity analyses of radionuclide transport travel times for the Yucca Mountain unsaturated zone, from the repository horizon to the water table.

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Simulation of Radionuclide Transport through Unsaturated, Fractured Rock: Application to Yucca Mountain, Nevada

This paper describes the numerical methods developed to simulate the transport of radionuclides through the unsaturated, fractured rock below the proposed repository at the Yucca Mountain. Regardless of the disposition of the Yucca Mountain License Application, this method has general applicability for modeling contaminant transport through deep vadose zones. Using a dual-permeability solution for fluid flow as a basis, the transport model represents contaminant transport with a cell-based particle-tracking technique in which particles move from cell to cell in the three-dimensional large-scale grid of two overlapping continua. Particle movement between cells, and the residence time within a cell, is computed probabilistically based on transfer functions using numerical solutions of the transport equations for an idealized fracture-matrix system. Using this approach, the following transport phenomena are simulated: advection through fracture and matrix continua, including between these continua, dispersion, sorption of dissolved radionuclides to the matrix continuum, and within the fault zones, and molecular diffusion between the fractures and matrix. In addition to providing a realistic representation of the relevant processes, this method has the virtue of computational efficiency. After describing the numerical method, this paper presents sensitivity analyses for radionuclide transport travel time distributions from the proposed repository to the water table through the unsaturated zone for the Yucca Mountain system.

Abbreviations: ADE, advection-dispersion equation; CDF, cumulative distribution functions; CHn, Calico Hills formation; DFM, discrete fracture model; TSPA, total system performance assessment; UZ, unsaturated zone.

The focus of Yucca Mountain, Nevada by the USDOE as a potential site for the nation's first high-level radioactive waste repository is largely due to the perceived beneficial attributes related to the thick, unsaturated zone where the waste would be disposed. To investigate the suitability of the site, a large amount of site characterization and modeling activities have been performed. Accompanying these investigations was the development of numerical models of the various components of the natural system, including the vadose zone and the saturated zone. A host of models were developed investigating various aspects of the unsaturated zone (UZ) behavior, with modeling approaches selected to optimize the effectiveness of the modeling tool for specific purposes, including unsaturated flow (Pruess, 1991; Nitao, 1996; Zyvoloski et al., 1997), coupled processes (Bower and Zyvoloski, 1997; Valentine et al., 2002, Rutqvist et al., 2002; Buscheck et al., 2003), reactive transport (Viswanathan et al., 1998; Robinson et al., 2000; Xu et al., 2008), and radionuclide transport for Performance Assessment analyses (Robinson et al., 2003). For the representation of UZ radionuclide transport in Total System Performance Assessment (TSPA) models, an approach was required that enabled the complex, three-dimensional behavior of the system to be captured in a probabilistic setting in which multiple realizations of system behavior, of which UZ transport was only one part, were performed. Thus, the requirement of computational efficiency, with a competing goal of achieving a minimal degree of abstraction from more detailed models, was established as a criterion of the model.

This paper describes the modeling method for UZ transport that was implemented to achieve these objectives, and presents a sensitivity analyses for UZ radionuclide transport for the proposed Yucca Mountain repository. While the model results are specific to Yucca Mountain, the methods are developed from basic hydrologic and transport principles for vadose zones consisting of porous and fractured rocks. Therefore, regardless of the ultimate disposition of the Yucca Mountain License Application (USDOE, 2008), this modeling study is relevant

to the general problem of contaminant transport in deep vadose zones, which is important to many environmental problems, notably including several deep vadose zone sites contaminated due to the legacy of operations conducted at the USDOE facilities (e.g., Birdsell et al., 2005; Gee et al., 2007; Smith, 2004; Kwicklis et al., 2006).

Robinson et al. (2012, this issue) presented the scientific basis for a conceptual model for flow and transport through unsaturated, fractured tuff in Yucca Mountain. A fundamental feature of the system is the wide range of transport velocities depending on whether flow occurs through fractures or the surrounding rock matrix. In a complex, heterogeneous fractured rock, behavior spanning the range from fracture-dominated to matrix-dominated flow and transport may occur due to spatially varying hydrologic characteristics. Research in the field of transport in porous media has led to a variety of conceptualizations to deal with complexities brought on by flow through fractures or other fast pathways. The earliest of these was the dual porosity formulation (Barenblatt et al., 1960; Warren and Root, 1963), which treats the fractured porous media as a primary porosity but includes an adjoining rock matrix as a fluid storage medium. In this formulation, the matrix is not a continuous medium in which the full mass and energy transport equations are solved. Nevertheless, fluid, solute mass, and energy exchange between the fractures and matrix can be represented. When the principle permeability in the system is due to the fractures, this formulation is a very effective extension to traditional continuum models. Subsequent modifications to the dual porosity method have been developed to generalize the approach to include the ability to resolve the calculation within the matrix blocks through added discretization, and to improve the computational efficiency of the numerical solution (e.g., Zyvoloski, 1983; Pruess and Narasimhan, 1985; Smith and Seth, 1999; Zyvoloski et al., 2008).

A significant limitation of the dual porosity concept is that there is no provision for representing flow within the rock matrix, independent of the fracture pathways. To relax this limitation, an alternative conceptualization was advanced in which flow occurs within two overlapping continua for the fractures and matrix (Zyvoloski et al., 1992; Gerke and van Genuchten, 1993; Zimmerman et al., 1993; Tseng and Zyvoloski, 2000). These models, now referred to as dual permeability models, allow for flow with each continuum, and define interchange terms in which mass and energy transfer between the continua. In the Yucca Mountain Project modeling efforts, it was recognized that this conceptualization would lead to a practical approach for modeling flow and transport in unsaturated, fractured tuff because in such media, simultaneous flow could occur at vastly different velocities in the fractures and surrounding matrix rock. Therefore, modeling efforts increasingly employed the dual permeability approach for the UZ, culminating in the adoption of this conceptual model for most simulation models of UZ flow and transport behavior leading up to and including the analyses used in the Yucca Mountain License Application (e.g., Robinson et al., 1997; Wu et al., 1999; Bandurraga and Bodvarsson, 1999; Robinson and Bussod, 2000; Bechtel SAIC Company, 2004a, 2004b; USDOE, 2008).

This paper presents a particle tracking method developed to simulate radionuclide transport in unsaturated, fractured porous media. Herein we focus on the transport model, and presume that dual-permeability numerical flow models are available as input to the transport model. For the specific application presented, the UZ at Yucca Mountain, the origin of these flow fields are from other published and referenced studies. The transport model extends the methods initially published in Robinson et al. (2003) by developing a novel transfer function approach that accurately captures the unique aspects of unsaturated flow in dual-permeability media. The paper is organized as follows. First, we develop the particle tracking method for radionuclide transport in the UZ, starting with basic principles and assumptions, followed by a description of the overall algorithm. Next, we discuss the numerical implementation, focusing on the use of the transfer function approach to capture the processes of combined fracture and matrix flow, with advective and diffusive exchange between the media. Then, after describing some implementation details, we present model calculations illustrating the behavior of the Yucca Mountain UZ as a barrier to radionuclide migration, emphasizing the quantification of uncertainty in travel time results as a function of key transport parameters. Finally, we conclude with a summary and general discussion of the role of this model in the study of transport in deep vadose zones.

Model Development

Conventional solutions to the advection-dispersion equation (ADE), such as the continuum solvers used in most finite-element or finite-difference flow and transport codes, are versatile and allow the simultaneous solution of multiple interacting species. However, one drawback of typical continuum ADE solutions is that significant numerical dispersion may arise in the portion of a computational domain occupied by a front with steep concentration gradients (e.g., Russell, 2002). Reducing the numerical dispersion requires either increased grid resolution or higher-order approximation methods, both of which may lead to prohibitive computational costs. As a result, these models are typically very computationally expensive, often prohibitively so for applications in which large, three-dimensional models and/or extensive sensitivity analyses or multi-realization probabilistic model runs are required. In this section, we develop an alternative approach using the particle tracking method, with methods tailored to the dual permeability model formulation.

Particle Tracking Methodology

Particle-tracking transport models take a fundamentally different approach than traditional continuum solutions to the ADE. The trajectory of individual molecules or packets of fluid containing

molecules are tracked through the model domain. When the objective is to map out the fluid path lines, velocity interpolation schemes can be used to resolve the streamlines (e.g., Pollock, 1988; Lu, 1994; LaBolle et al., 1996), and a relatively small number of particles can be used. Particle tracking can also be used to simulate solute transport, in situations such as the migration of a contaminant plume or the prediction of breakthrough curves in interwell tracer experiments (e.g., Akindunni et al., 1995; Johnson et al., 1994). For these applications, a relatively large number of particles must be used to obtain accurate solutions to the transport problem. Random walk models are often combined with the streamline tracing method to implement a fully functioning method to solve the ADE (Kinzelbach, 1988; Tompson and Gelhar, 1990; Lichtner et al., 2002).

An alternate approach to the streamline tracing method is to route the particles from one computational grid cell to the next based on the portion of the fluid flux traveling from one cell to the next (e.g., Desbarats, 1990). In this method, the calculation of a spatially resolved path line at the sub-grid level is replaced with a cell-to-cell routing of the particle: for this approach, the probability of transfer to the adjacent cell is proportional to the flux to that cell, and no velocity interpolations are performed. Although in principle either a cell-based or path-line-based model could have been used in the Yucca Mountain UZ model, the computational efficiency of the cell-based method made it an attractive choice for this application. Also, with dual permeability model formulation, the overlapping grid cell of the "other" medium is simply another computational cell like any other, making the incorporation of advection between the continua straightforward. Once the particle is routed to a given cell, the residence time of a nonsorbing, nondiffusing solute at that cell equals the fluid mass residing within the cell volume divided by the total mass flux leaving the cell.

Particle-based methods are also advantageous for capturing other transport processes beyond advection, such as fracture-matrix diffusive transport and sorption. For diffusion, the simple deterministic calculation of residence time or velocity of a particle is no longer sufficient. For example, the process of matrix diffusion yields a spreading of the solute mass, which conceptually is equivalent to a distribution of travel times, rather than a single, deterministic time. Methods have been developed to simulate this behavior by determining probabilistically the transport time for each leg of the particle's path through the system. These methods, referred to alternatively as time-domain or transfer function methods, derive travel time distribution functions using idealized representations of the sub-grid system, and use these functions as probability distributions, which are sampled on the fly to compute the travel time. This approach has been used to simulate tracer transport in saturated, fractured rock using a variety of assumptions for the transfer function (Robinson, 1988; Yamashita and Kimura, 1990; Arnold et al., 2003; Painter et al., 2008; Kelkar et al., 2010; Robinson et al., 2010).

In unsaturated media, particle-based methods have also been developed, including those based on the dual permeability method (Robinson et al., 1997; Pan et al., 1999; Liu et al., 2000; Robinson and Bussod, 2000; Pan and Bodvarsson, 2002; Robinson et al., 2003). A common difficulty in the implementation of these methods is the difficulty of determining a transfer function method that is accurate over large ranges of parameter values representing all extremes of transport behavior, from fracture-dominated transport to rapid diffusion within the rock matrix. The present study develops a new approach that uses numerical simulation for an idealized fracture-matrix model geometry to extend the range over which the transfer function method can be applied in dual permeability media. Thus, the sub-grid model employed to generate the transfer functions for diffusive exchange between the continua is solved using a large number of numerical simulations spanning many orders of magnitude of the relevant parameters, and numerical interpolation is used to select the proper transfer function curve.

Other elements of the model and numerical code remain the same as in previous versions of this model (Robinson et al., 2003; Sandia National Laboratories, 2008a), and are briefly summarized herein for completeness.

Dual Permeability Particle Tracking Algorithm

This section describes the algorithm developed to represent UZ transport using the particle tracking method. In a dual-permeability system, the transfer of solute mass between fractures and matrix can occur via advection, the movement of solute via fluid flow from one medium to the other, and matrix diffusion, where molecular diffusion transports mass. For saturated flow systems, fluid flow is often dominated by fractures because of the orders-of-magnitude larger permeabilities in the fractures compared to the surrounding rock matrix. However, in vadose zones such as the UZ at Yucca Mountain, fluid can travel simultaneously in both fractures and matrix, depending on the relative hydrologic properties of each medium (e.g., Robinson et al., 2005). An additional process that allows solute to transport between the continua is molecular diffusion. The variability of transport times and accompanying spreading of solute mass through such a system is a complex function of the relative velocities in the two media, the advective flux between the media, the spacing between fracture flow paths, matrix diffusion coefficients, and sorption.

For transport in a fractured, unsaturated system at the field scale, the dual permeability flow model conceptualization consists of one matrix grid cell for each fracture cell. However, important processes associated with transport occur at scales smaller than those considered in a field-scale model, particularly in the matrix rock immediately adjacent to each flowing fracture. Therefore, incorporation of fracture-matrix interactions into the model is in essence an upscaling problem. To deal with this complexity, a suitable idealized model is required that captures important

sub-grid-scale transport processes, and allows this small-scale behavior to be readily upscaled for inclusion in the large-scale model. To accomplish the upscaling within the particle tracking transport model, we first construct an idealized transport model at the small scale that allows the transfer functions to be computed and tabulated, and then use these functions as the basis for making the probabilistic calculation of particle travel time at each computational grid cell. Because dual permeability transport behavior is vastly different depending on whether solute starts in the fracture or in the matrix, the transfer function approach must be developed to capture these effects. The new method develops transfer functions which establish the residence time distribution in a cell, and also enable the subsequent decision to be made regarding whether the particle enters the adjacent cell in the fractures or the matrix. In this implementation, the combined fracture and matrix system is treated as a unified medium in which there is a distribution of transport times, conditioned on whether the particle enters a given cell in the fractures or the matrix. The transfer functions themselves (described below and in further detail in the Supplement to this paper) are computed based on an idealized fracture-matrix transport model with parallel flow in the fractures and matrix. Two versions of this model, which we generically term the "model system" for fracture–matrix interchange, were developed to investigate the impact of different model conceptualizations on the large-scale transport results. The choice of a parallel fracture model for developing the matrix diffusion submodel was made on the grounds of practicality, in recognition of the fact that information does not exist to warrant more complex fracture geometries. This approach is in keeping with the philosophy of the aforementioned dual porosity and dual permeability models, which use relatively simple interchange conceptual models to represent processes at scales smaller than the size of a computational grid cell.

The steps of the algorithm are as follows (the algorithm starts with a known particle location, either in the fracture or matrix continuum):

Step 1. Determine probabilistically whether the particle should move to the other medium due to advective flux to that medium.

Step 2. Determine probabilistically whether the particle will leave this cell via the current medium or the other medium.

Step 3. Use the conditional transfer function to determine probabilistically the residence time of the particle.

Step 4. Determine probabilistically using the relative total flux to adjacent nodes which cell the particle moves to next (whether it starts in the fracture or matrix continuum in the next cell has been determined previously in Step 2).

This approach treats the overlapping fracture and matrix grid cells as a single porous medium through which mass travels, and apportions the particles to each continuum according to the diffusive and advective fluxes defined by the flow field and the transport parameters. The dual-permeability flow model at the mountain scale (e.g., at a scale of several hundred meters to several kilometers), presumed to be input to the transport model in this development, prescribes a net flow through the fracture continuum, a net flow through the matrix continuum, and a fracture to matrix (or matrix to fracture) fluid flux.

To implement this algorithm and allow the transfer function to be computed, Step 1 takes the fracture-matrix advective flux term and applies it immediately when the particle enters the cell. Then, after potentially shifting the particle from one medium to the other via advection (with no increase to the transport time), the subsequent transfer functions are based on parallel flow in the two continua with no advective flux between the media. This sequential approach simplifies the transfer function process by eliminating the need for an additional variable, the fracture-matrix advective flux, in the construction of the transfer function curves. Instead, a probability p_{fm} of the particle transferring to the other medium (Step 1) is assigned:

$p_{fm} = 0$ if the fracture-matrix flux term f_{fm} is *into* the medium in which the particle already resides, or

$p_{fm} = f_{fm}/f_{out}$, where f_{out} is the total flux out of the continuum in which the particle currently resides, either to adjacent cells within that continuum or to the other continuum.

Step 2 accounts for the fact that there is a finite probability that a fraction of the solute will leave the cell through the other medium due to diffusive transfer of mass. In developing the transfer functions, solute mass is introduced in the model system in either the fracture fluid or the matrix fluid. For the general case of water flowing through both fracture and matrix, mass can leave the model system via either medium. Therefore, conditional transfer functions must be generated to obtain the probabilities in Step 2. That is, for mass injected with the fracture fluid entering the discrete fracture submodel, there is a breakthrough curve for mass leaving the model via the fractures, and an accompanying breakthrough curve for mass leaving via the matrix. Similarly, there are two breakthrough curves for mass injected with the matrix fluid. The plateau value attained for a given transfer function curve represents the probability of solute mass leaving via a particular medium in Step 2. Then, the probability of a particle leaving via a given continuum equals the steady state solute mass flux (the plateau of the transfer function curve) divided by the total mass flux through the model system. This step provides a way to assign probabilities for transitioning particles between the media via the diffusion process in a system in which water flows through both continua. Note that the special case of flow only in one medium automatically results in that

particle leaving the cell in the medium that flows, even if diffusion results in a travel time delay during transport.

Step 2, which determines the medium through which the particle leaves, also dictates the selection of which conditional transfer function to apply to obtain the residence time in Step 3. As in previously developed transfer function implementations, a random number is generated between 0 and 1, and based on the value, the particle residence time is determined from the transfer function.

Finally, Step 4 routes the particle to the appropriate connecting cell in the finite volume domain, using the node-to-node routing scheme described earlier. If the particle is determined to enter an adjoining cell via the fracture continuum, then the internodal fluxes associated with the fractures are used to define the probabilities of traveling to each connected fracture cell. Similarly, for transport to an adjoining matrix cell, matrix fluxes are used.

Of course, all numerical methods have potential limitations that must be considered in any application. With respect to the routing of particles from cell to cell, there is the possibility of numerical errors when such particle transfers displace particles away from their natural streamline trajectories as may be calculated from conventional point-to-point velocity interpolation methods (e.g., Goode and Shapiro, 1991). This effectively results in an artificial lateral spreading of particles, a form of numerical dispersion, whose magnitude will depend on the grid cell orientation and its relation to the calculated streamlines. The degree of error associated with this unwanted phenomenon depends on the application. For example, for numerical studies of macrodispersion in heterogeneous aquifers, the magnitude of the error has been shown to be small (Desbarats, 1991), leading investigators to choose either the cell-to-cell routing method or a velocity interpolation scheme, based on considerations of computational efficiency or convenience (Ritzi et al., 2000).

In the results presented herein, the numerical grid consists of vertical, columnar grid cells and sloping boundaries that provide water flux pathways along the zones of lateral flow. Gravity-driven flow results in vertical, downward transport through a column of cells, which is, in some portions of the domain, interrupted by lateral diversion through a connected set of cells representing a hydrostratigraphic pathway. Because the grid-cell connections exist to capture either vertical or lateral flow, a cell-to-cell routing of particles should adequately represent the movement of solute through the model. Thus, the scheme was selected on the basis of simplicity and computational efficiency. Note, however, that if necessary to reduce numerical dispersion, the transfer function methodology employed herein could be combined with a streamline-based particle tracking algorithm.

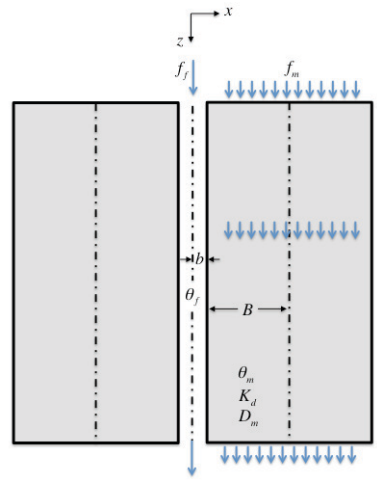

Fig. 1. Schematic diagram of the fracture-matrix interchange model system. The model domain for the fracture-matrix interchange sub-model contains half of the fracture and the half of the matrix block.

Model System for Fracture–Matrix Diffusive Interchange

The process employed in this model to obtain the transfer functions for the dual permeability transport model system consists of a series of numerical simulations on the idealized model system depicted in Fig. 1. Because each grid block in the large-scale model possesses different hydrologic and transport parameters, a procedure for deriving a non-dimensional form of the model system is required to make the method practical. In the figure and in the derivation that follows, D_m is the effective diffusion coefficient; τ_f and τ_m are the fluid transport times in the fracture and matrix, respectively; R_f and R_m are the retardation factors in the fracture and matrix, respectively; B is the half-spacing between flowing fractures; b is the fracture half-aperture; and θ_f and θ_m are the volumetric water contents of the fracture and matrix, respectively. Taking a control volume in the fracture of width b (half of the full aperture), depth d, and length Δz, the terms of the transient solute mass balance (units of each of these terms are solute mass per time) are:

Accumulation: $bd\Delta z\theta_f R_f(C - C_{prev}) / \Delta t$ [1]

where C_{prev} represents the concentration at the previous time step.

Advection: $bd\overline{V_z}(C_{z+\Delta z} - C_z)$ [2]

where $\overline{V_z}$ is the Darcy velocity in the fracture, equal to volumetric flow rate divided by the total cross-sectional area in the fracture.

Diffusion into matrix: $d\Delta z\theta_m D_m \left.\frac{\partial C_m}{\partial x}\right|_{x=b}$ [3]

These terms form the overall solute mass balance equation:

$$bd\Delta z\theta_f R_f \frac{(C-C_{prev})}{\Delta t} = -bd\bar{V}_z(C_{z+\Delta z}-C_z) + d\Delta z\theta_m D_m \left.\frac{\partial C_m}{\partial x}\right|_{x=b} \quad [4]$$

Dividing by $bd\Delta z\theta_f$, making use of the relation for the fracture interstitial pore-water velocity $V_f = \bar{V}_z / \theta_f$, and taking the limit as Δz and Δt go to 0:

$$R_f \frac{\partial C_f}{\partial t} = -V_f \frac{\partial C_f}{\partial z} + \left.\frac{\theta_m D_m}{\theta_f b}\frac{\partial C_m}{\partial x}\right|_{x=b} \quad [5]$$

Note also that the subscript "f" on the concentration is adopted to denote the fracture.

Given the assumption of one-dimensional diffusion within the matrix, perpendicular to the flow direction, the differential equation governing transport in the matrix is:

$$R_m \frac{\partial C_m}{\partial t} = D_m \frac{\partial^2 C_m}{\partial x^2} - V_m \frac{\partial C_m}{\partial z} \quad [6]$$

where V_m is the interstitial pore-water velocity in matrix. The initial and boundary conditions for the system are:

$$C_f(z,0) = 0 \quad [7]$$

$$C_m(x,z,0) = 0 \quad [8]$$

$$C_f(0,t) = C_{0,f} \quad [9]$$

$$C_m(x,0,t) = C_{0,m} \quad [10]$$

$$C_m(b,z,t) = C_f(z,t) \quad [11]$$

$$\frac{\partial C_m}{\partial x}(B,z,t) = 0 \quad [12]$$

Here the terms $C_{0,f}$ and $C_{0,m}$ for the fracture and matrix, respectively, are nonzero if mass is being introduced into that medium, and 0 if mass is being introduced in the other medium.

Nondimensionalization of these equations can be accomplished by introducing the following dimensionless variables:

$$\hat{C}_f = C_f / C_0 \quad [13]$$

$$\hat{C}_m = C_m / C_0 \quad [14]$$

$$\hat{z} = z / L \quad [15]$$

$$\hat{x} = x / B \quad [16]$$

$$\hat{t} = \frac{V_f t}{R_f L} = \frac{t}{\tau_f R_f} \quad [17]$$

where L is the length of the flow path. Equation [17] uses the definition of $\tau_f = L/V_f$; a corresponding relation is also used for the matrix fluid transport time ($\tau_m = L/V_m$). Note that because the equations are nondimensionalized with respect to transport times through the fracture and matrix, the physical dimensions of the flow path used to develop the transfer function curves, including the length, are unimportant to the final implementation in the code. Next, substituting Eq. [13] to [17] into Eq. [5] and [6]:

$$\frac{\partial \hat{C}_f}{\partial \hat{t}} = -\frac{\partial \hat{C}_f}{\partial \hat{z}} + \left.\frac{\theta_m D_m \tau_f}{\theta_f bB}\frac{\partial \hat{C}_m}{\partial \hat{x}}\right|_{\hat{x}=b/B} \quad [18]$$

$$\frac{\partial \hat{C}_m}{\partial \hat{t}} = \frac{D_m \tau_f R_f}{B^2 R_m}\frac{\partial^2 C_m}{\partial x^2} - \frac{\tau_f R_f}{\tau_m R_m}\frac{\partial \hat{C}_m}{\partial \hat{z}} \quad [19]$$

The boundary and initial conditions (Eq. [7–12]) are transformed to

$$\hat{C}_f(\hat{z},0) = 0 \quad [20]$$

$$\hat{C}_m(\hat{x},\hat{z},0) = 0 \quad [21]$$

$$\hat{C}_f(0,\hat{t}) = 1 \text{ (or 0 if injection is into the matrix)} \quad [22]$$

$$\hat{C}_m(\hat{x},0,\hat{t}) = 1 \text{ (or 0 if injection is into the fracture)} \quad [23]$$

$$\hat{C}_m(b/B,\hat{z},\hat{t}) = \hat{C}_f(\hat{z},\hat{t}) \quad [24]$$

$$\frac{\partial \hat{C}_m}{\partial \hat{x}}(1,\hat{z},\hat{t}) = 0 \quad [25]$$

Referring to Eq. [18] and [19], the system is fully characterized by three dimensionless parameters:

$$p_1 = \frac{D_m \tau_f R_f}{B^2 R_m} \quad [26]$$

$$p_2 = \frac{D_m \tau_f \theta_m}{bB\theta_f} \quad [27]$$

$$p_3 = \frac{\tau_f R_f}{\tau_m R_m} \quad [28]$$

where the retardation factor $R_k (k = f, m)$ is related to the sorption coefficient K_d using the standard form of the retardation factor:

$$R_k = 1 + \frac{\rho_b K_d}{\theta_k} \quad [29]$$

This derivation shows that a series of transfer function curves generated based on a model system with parallel flow in the fractures and matrix can capture the range of behavior of the UZ transport as long as the curves span the ranges of the parameters in the vector (p_1, p_2, p_3).

For a given parameter vector (p_1, p_2, p_3), there is a unique set of conditional transfer function curves of the form $\hat{C}$ versus $\hat{t}$, where $\hat{C}$ is the normalized breakthrough curve for the nondimensional time $\hat{t}$ given by

$$\hat{t} = \frac{t}{R_f \tau_f} \quad [30]$$

The set of conditional transfer function curves consists of a total of four normalized curves for each (p_1, p_2, p_3): mass input in fracture, output in fracture; mass input in fracture, output in matrix; mass input in matrix, output in fracture; and mass input in matrix, output in matrix. In this implementation, the method requires the pre-calculation and tabulation of the transfer functions, a method for choosing the most appropriate function [given that local values of (p_1, p_2, p_3) will never be identical to those selected to generate the transfer functions], and methods to improve accuracy and handle extreme values of parameters. For details on these important implementation details, and a discussion of the behavior of this model system, see the supplementary material associated with this paper.

One final aspect of the treatment of diffusive transport within the matrix is the level of discretization used to resolve the diffusion term in the model system. In this implementation, two options have been developed and used: the Discrete Fracture Model (DFM) and the dual-k model. The DFM uses a discretized grid, with closely spaced nodes near the fracture, and more widely spaced nodes in between fractures. This model resolves sharp concentration gradients close to the fractures, yielding an accurate solution to the idealized fracture-matrix model system throughout the entire parameter range. The dual-k method uses a single node in the matrix, giving it the virtue of consistency with the dual permeability flow model, at the expense of accuracy of the diffusion solution for relatively small amounts of diffusion. In the simulations that follow, and in the Supplement, comparisons of these two implementations of the model system are presented and described. With regard to the computations, the process for generating the breakthrough curves and transfer functions is the same for the dual-k and DFM models. Furthermore, the use of these curves in the particle tracking simulation is completely transparent, requiring only a choice of which input file of transfer functions to use. Thus, the particle tracking method facilitates the incorporation of various conceptual models for fracture-matrix interactions very easily.

Other Transport Processes and Model Features

In addition to fracture-matrix interactions via advection and diffusion, other transport processes such as dispersion, colloid-facilitated transport, and hydrologic features impacting radionuclide migration are represented in the model. Because these features have been described elsewhere (dispersion, radioactive decay and decay chains, development of numerical model for colloid-facilitated transport: Robinson et al., 2003; colloid transport conceptual model: Robinson et al. (2012), this issue; water table rise under future climate conditions, radionuclide source term input to model: Sandia National Laboratories, 2008a), the reader is referred to those publications for detailed descriptions.

Model Verification

The radionuclide transport model developed above was tested extensively against analytical and numerical solutions such as the finite fracture spacing solution of Sudicky and Frind (1982), and benchmarked against other numerical models. One example, provided in Robinson et al. (2012, this issue), shows very good agreement of the particle tracking results with a continuum solution to the ADE for the same three-dimensional geometry, boundary conditions, and source terms. Other more limited comparisons were also conducted to test the advection, diffusion, and sorption processes separately against continuum model results. In all cases, the comparisons are favorable (typically within 5% on average), indicating that the present model is suitable for use in large-scale UZ radionuclide transport simulations. Details of these comparisons are presented in Sandia National Laboratories (2008a).

Model Parameter Uncertainties for the Yucca Mountain System

The model developed in the present study is designed to enable extensive uncertainty analyses to be performed, either for the UZ transport model on its own, or as part of a probabilistic Performance Assessment (PA) analysis of the entire repository system. This requires that stochastic distributions be established for all uncertain parameters. Robinson et al. (2012, this issue) discusses some of the considerations involved in the quantification of uncertainty for the Yucca Mountain UZ, and Sandia National Laboratories (2007b) and Sandia National Laboratories (2008a) provide a complete description of the parameter distributions and their bases. In the present study, we restrict attention to a subset of those parameters, choosing particularly important uncertainties

and those that allow us to demonstrate the utility of the particle tracking model. These parameters include infiltration rate, sorption coefficient, and diffusion coefficient. See Sandia National Laboratories (2007b, 2008a) for the detailed analysis for these and other parameters used in the PA analyses presented in the Yucca Mountain License Application (USDOE, 2008).

Uncertainty in infiltration rate, which has a first-order impact on the water velocity through the UZ, was represented in the Yucca Mountain Project through the generation of multiple steady state flow fields that span the range of infiltration rates that are consistent with the available observations. A presentation of the extensive set of analyses, calibrations, and model validation activities accompanying the UZ flow model is beyond the scope of this study: see Sandia National Laboratories (2008b) for details on the development of infiltration scenarios, and Bechtel SAIC Company (2004b) for the methods used to calibrate the models. This effort led to the selection of four infiltration scenarios spanning the uncertainty range for infiltration. For each of these, model calibrations were performed. Then, by sampling from these scenarios, the uncertainty associated with infiltration was represented in the probabilistic analyses. In addition to the present-day climate and infiltration, two future climate scenarios consisting of wetter climates (the monsoon and glacial-transition climate conditions) were developed for each of the four infiltration scenarios to account for the potential for increased flow rate in the future. This approach yielded a total of 12 UZ flow models, which then became input to the UZ transport model. In the PA analysis, the probability of each of the four infiltration scenarios was established based on an analysis in which the model was compared to other data sets beyond those used in the calibration; those probabilities were the weights applied to prescribe the infiltration scenario for each realization (Sandia National Laboratories, 2007a). In the present study, we examine the influence of infiltration by presenting results for each of the UZ flow fields, based on the so-called 10th, 30th, 50th, and 90th percentile infiltration maps.

Uncertainty distributions for sorption and diffusion coefficients are assigned based on the categorization of the rocks of the UZ into distinct rock types grouped on the basis of their similarities with respect to sorption or diffusion. These categorization schemes allowed for different parameterizations to be established to account for the likely property variability due to differences in characteristics such as mineral content and pore size distributions. For sorption, these rock types were grouped based on mineralogy into vitric, devitrified, and zeolitic rocks. For diffusion, layers of similar values of the porosity and water content were grouped together to develop the parameter distributions. Other transport properties such as porosity varied layer by layer, and were assigned constant values inherited directly from the flow model.

The partition coefficient (or sorption coefficient), K_d, is defined as the ratio of the quantity of the solute adsorbed per mass of solid to the amount of the solute remaining in the solution at equilibrium. K_d values for each radionuclides were developed for each of the three rock types on the basis of geochemical conditions, rock types, and analyses of laboratory-scale sorption measurements. Sandia National Laboratories (2007b) provides the analysis for all radionuclides, and a summary is provided in Robinson et al. (2012, this issue). For the present study, the sorption coefficient distributions for the three radionuclides presented are taken from Sandia National Laboratories (2007b). For purposes of comparison, ^{99}Tc is presented as a nonsorbing radionuclide ($K_d = 0$ for all rock types); ^{237}Np is a moderately sorbing radionuclide with K_d uncertainty distributions ranging from about 0.5 to 6 mL g^{-1}; and isotopes of plutonium are considered strongly sorbing radionuclides with K_d uncertainty distributions ranging from about 10 to 200 mL g^{-1}.

The diffusion coefficient distributions were established by defining an uncertainty distribution for the tortuosity factor τ, and, after assigning a constant value of the free diffusion coefficient D^* (values for ^{99}Tc, ^{237}Np and plutonium isotopes, of 1.95×10^{-9}, 6.18×10^{-10}, and 1.30×10^{-9} m^2 s^{-1}, respectively), determining the value of D_m using the relation $D_m = \tau D^*$. Uncertainty in the base-10 log of tortuosity for each rock group was taken to follow a normal distribution, with log mean equal to the values developed from considerations of water content and rock type (log mean values in the range −1.84 to −1.15, or the geometric mean of 1.45×10^{-2} to 7.01×10^{-2}) and a standard deviation of 0.29 (see Sandia National Laboratories, 2008a for details).

Model Results and Uncertainty Analyses

In Robinson et al. (2012, this issue), some representative results were presented in the form of travel times of key radionuclides from the repository to the water table as a function of release location, and breakthrough curves at the water table. A significant finding was the large-scale spatial variability of transport behavior, as illustrated in Fig. 2. This figure plots the mean travel time without diffusion for transport to the water table as a function of release location across the footprint of the proposed repository. In this simulation, particles are released into the fracture continuum at a large number of locations across the repository footprint, and the travel time to the water table is recorded, and the results contoured to illustrate the impact of release location on travel time through the UZ. Releases from the northern section of the repository reach the water table much more rapidly than releases in the south because of the prevalence of fracture-dominated flow and transport in the north. By contrast, significant thicknesses of nonwelded Calico Hills formation (CHn) exist beneath the southern release points, resulting in matrix flow and longer travel times through that unit. For the sensitivity analyses that follow, we selected two representative release locations within the repository footprint: the Northern release location (pink dot in the figure)

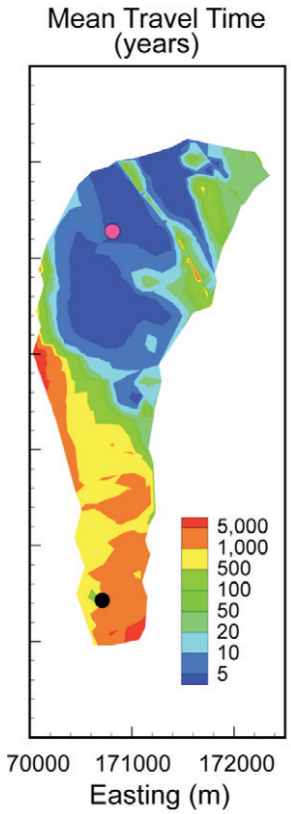

Fig. 2. Contour map the mean travel time for nonsorbing, nondiffusing particles released at all repository nodes using the glacial-transition, 10th percentile infiltration map. Two dots are taken as representative release locations within the repository footprint.

and the Southern release location (the black dot). Travel times or breakthrough curves from these two representative locations are examined to illustrate the behavior of the system. In the present study, attention is focused on travel times or radionuclides; then a companion study, issues of large scale flow pathways and lateral transport are examined more fully (Robinson et al. (2012).

In the subsections that follow, we describe the methodology used to examine the behavior of the system, then present several sensitivity results to illustrate the behavior of the model and the uncertainty of its predictions.

Uncertainty Quantification Methodology

The degree to which the unsaturated zone acts to limit the migration of radionuclides must be assessed with a full accounting of the uncertainties of key transport parameters and processes. The role of the UZ in the repository system as a whole must be assessed within the total-system model. Nevertheless, simulations of the UZ submodel on its own are also useful, particularly if an appropriate "figure of merit" is used as a proxy for unsaturated zone performance. Although more complex metrics can be used, herein we use the mean travel time for our uncertainty analysis. By comparing the mean travel time to the regulatory time period of interest, one can gauge the degree to which the unsaturated zone alone will retard a radionuclide during a period of relevance to repository analysis. In addition, it is useful to plot and compare the travel time distributions for different model parameters to gain further insight into the behavior of the UZ system.

The travel time distribution depends on a number of factors, including sorption coefficient K_d, matrix diffusion coefficient D_m, and infiltration scenario. For sorbing species in a dual permeability system with fracture-matrix diffusive interchange, combinations of K_d and D_m values must be examined to fully understand the system. For this case, we developed a method that enables two stochastic parameters to be investigated simultaneously over their entire uncertainty ranges. Parameter values were sampled from the cumulative distribution functions (CDF) for that parameter, in equal intervals of 0.1 on the CDF axis, except for the normal distribution, where the low and high ends are defined by values close to 0 and 1, namely minus and plus three standard deviations from the mean for a normal distribution (i.e., 0.00135 and 0.99865). For sorbing species examined in these calculations, a grid of 11 values of D_m and 11 values of K_d (for a total of 121 simulations) was used, and the resulting mean travel time surface was contoured to enable the parameter uncertainties to be examined. For the simpler case of a nonsorbing species, for which D_m is the only significant parameter, a line plot of mean travel time as a function of D_m, or alternatively a series of breakthrough curves, was used to examine the results.

Uncertainty Analysis Results for Nonsorbing and Sorbing Radionuclides

For these simulations, results for a representative nonsorbing (^{99}Tc), moderately sorbing (^{237}Np), and strongly sorbing (^{240}Pu) radionuclide are presented. For each species, the range of infiltration scenarios is covered by the four representative glacial-transition flow fields described earlier (called GT10, GT30, GT50, and GT90 in the figures) for the dual-k fracture-matrix interaction model.

Figure 3 plots, on a log scale, the mean travel time of ^{99}Tc as a function of diffusion coefficient D_m for particles released at northern and southern locations, for each of the four glacial-transition flow fields. As expected, the mean travel time decreases significantly with the infiltration rate for both release locations. There is roughly an order of magnitude uncertainty in travel times due to uncertainty in infiltration rate. With respect to diffusion, the mean travel time increases with D_m for the northern release node, while for the southern release node, the travel time is virtually independent of D_m. For the southern release location, longer travel times are predicted within the Calico Hills nonwelded units, which exhibit matrix-dominated flow and transport. Under these conditions, the travel times are a function of the flow velocity in the matrix, and fractures play no role in providing a short-circuit pathway. Therefore, diffusion is irrelevant under these conditions.

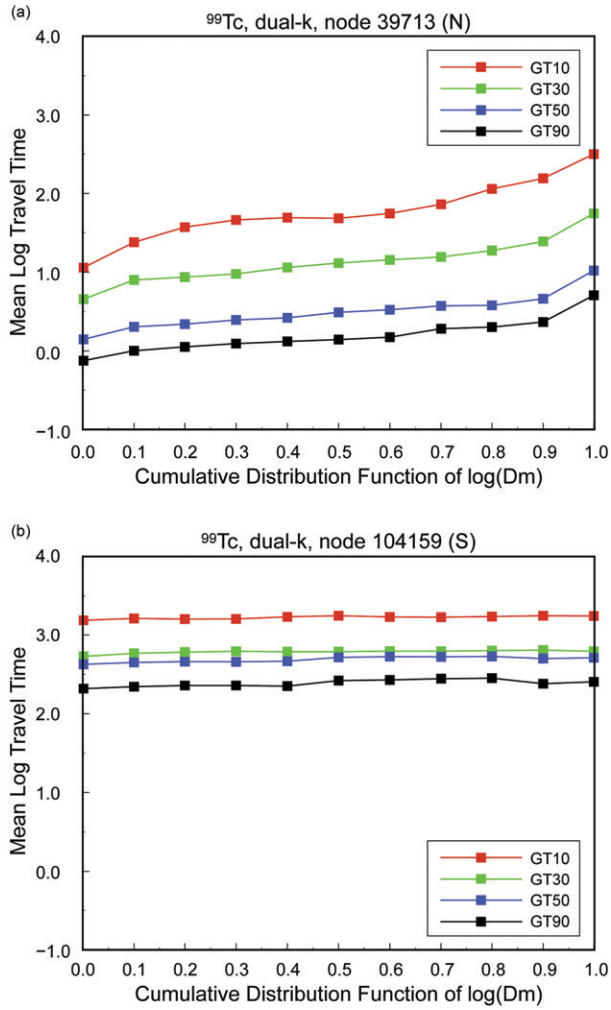

Fig. 3. Comparison of mean travel time of ^{99}Tc as a function of matrix diffusion coefficient under glacial-transition climate conditions; dual-k conceptual model for fracture-matrix interchange.

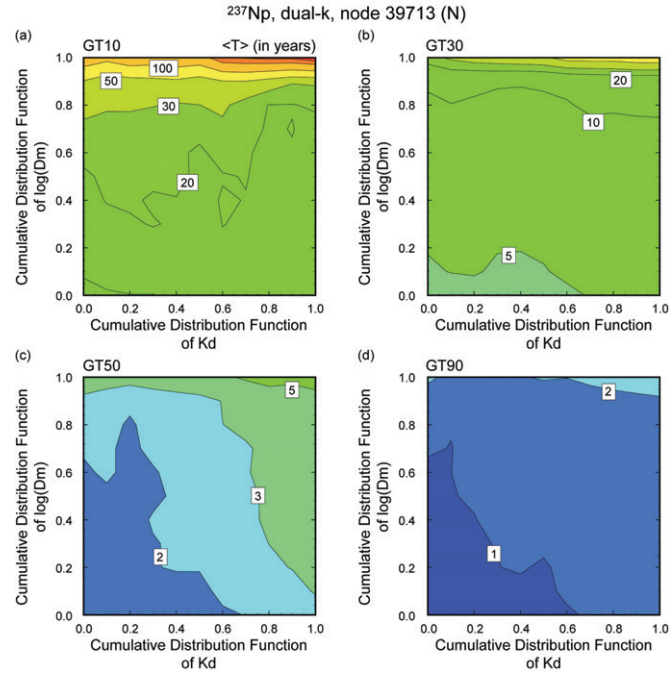

Fig. 4. Mean travel time of ^{237}Np as a function of matrix diffusion coefficient and sorption coefficient for the glacial-transition climate condition; dual-k conceptual model for fracture-matrix interchange; northern release location.

Note that matrix diffusion presumably impacts travel time in the fracture-dominated TSw even for the southern release point, but the effect is masked by the subsequent slow transport through the underlying nonwelded tuff. For this reason, the travel times are longer from the southern release location, and are virtually independent of the diffusion coefficient.

For sorbing species, both K_d and D_m have an influence on the travel time to the water table, so we use the contour plot approach described earlier to present these results. The calculated mean travel times for the weakly sorbing species ^{237}Np released at the northern (Fig. 4) and southern (Fig. 5) release points under four different flow fields (GT10, GT30, GT50, and GT90) reveal the following results:

- For the northern release location, mean travel times range from very short (approximately 1 yr) for the highest infiltration scenario to greater than 100 yr (but still short compared to the long half-life of ^{237}Np).
- The travel times from the northern release location are a function of both K_d and D_m.
- The releases from the southern location are much longer than those in the north, and the travel times depend, to first order, on the K_d but, like ^{99}Tc, are virtually insensitive to D_m.

The interplay between diffusion and sorption for transport through units with fracture-dominated flow is reflected in these results, which are in qualitative agreement with previous studies of ^{237}Np transport (e.g., Viswanathan et al., 2003). Greater diffusion leads to both diffusion and sorption-related delays in the transport time, reflected in the larger travel times for larger values of either K_d or D_m. However, for the southern release, all of these effects are masked by the significant travel time delays associated with slow matrix transport and sorption in the Calico Hills unit.

An analogous set of comparisons for the mean travel time for the strongly sorbing ^{240}Pu (reversibly sorbed species) are illustrated in Fig. 6 and 7. Similar overall trends are observed for this strongly sorbing species as were discussed for ^{237}Np, but the travel time disparity between the northern and southern release locations is more striking. A matrix-dominated transport layer in the flow path

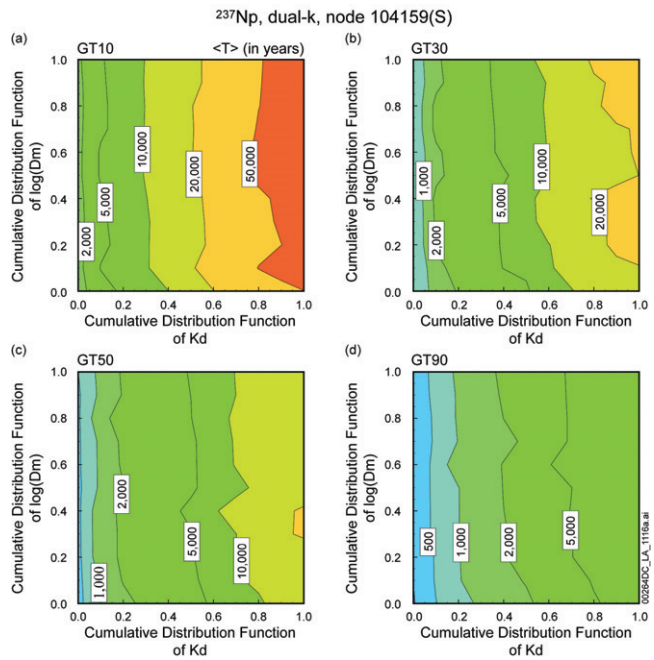

Fig. 5. Mean travel time of ^{237}Np as a function of matrix diffusion coefficient and sorption coefficient for the glacial-transition climate condition; dual-k conceptual model for fracture-matrix interchange; southern release location.

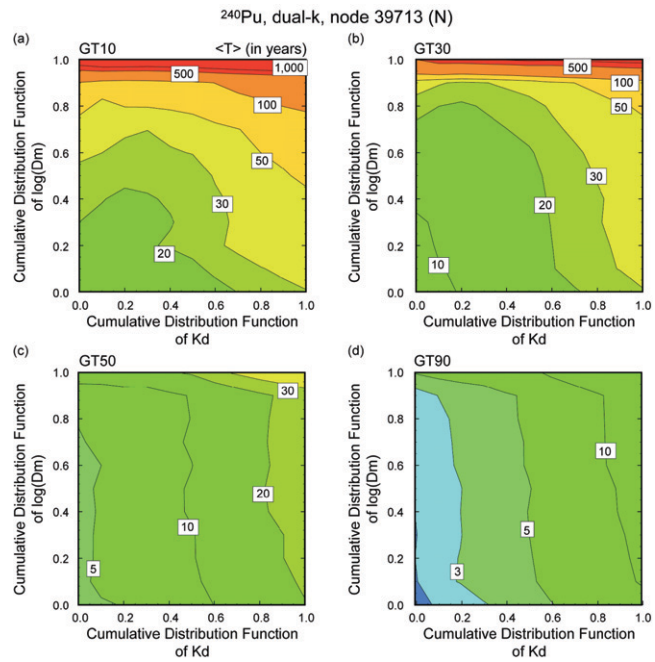

Fig. 6. Mean travel time of ^{240}Pu as a function of matrix diffusion coefficient and sorption coefficient for the glacial-transition climate condition; dual-k conceptual model for fracture-matrix interchange; northern release location.

has an extremely beneficial impact on radionuclide isolation when combined with strong sorption. Colloid transport is represented through radionuclide attachment to colloids that are either reversible or irreversible. For reversible attachment, the degree of partitioning onto colloids is a function of both the colloid concentration and the sorption coefficient for a given radioelement onto the colloid. The lower ionic strength colloid concentration distribution results in greater colloid concentrations leading to greater colloid-facilitated radionuclide transport.

Impact of Hydrologic and Physical Transport Properties

Hydrologic processes and parameters clearly exert a strong influence on the UZ radionuclide transport model results. In addition to sensitivity to the infiltration rate (reflected in the results of the previous section), the model is potentially sensitive to residual uncertainties in the calibrated hydrologic properties obtained for a given infiltration scenario. Analyses examining these model sensitivities, discussed extensively in Sandia National Laboratories (2007b, 2008a), are briefly summarized here. Of all of the hydrologic parameters in the model, uncertainty in matrix permeability appeared to have the largest influence on transport uncertainty. Increasing the matrix permeabilities, in ranges consistent with the available data and calibrated model, results in longer radionuclide transport times to the water table, due to an increased relative flow fraction in the matrix. By contrast, the van Genuchten unsaturated flow parameters and the fracture

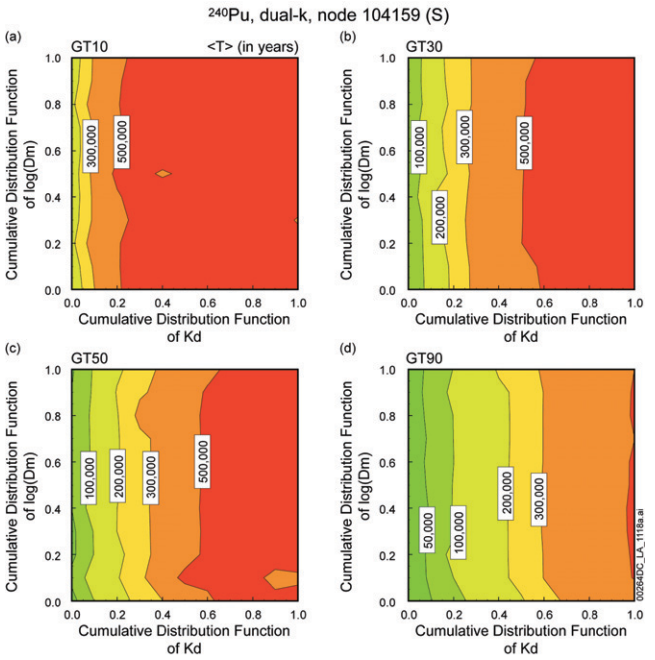

Fig. 7. Mean travel time of ^{240}Pu as a function of matrix diffusion coefficient and sorption coefficient for the glacial-transition climate condition; dual-k conceptual model for fracture-matrix interchange; southern release location.

parameters exhibited a relatively small sensitivity. This result is due to the relatively small changes to the relative fracture and matrix flow rates predicted as the hydrologic parameters are

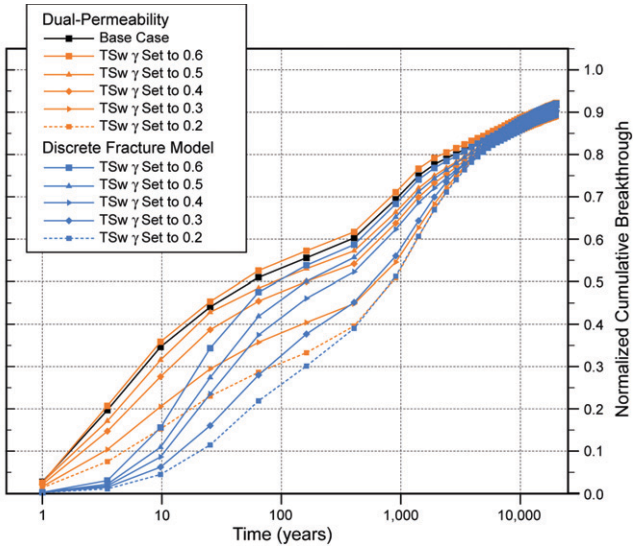

Fig. 8. Breakthrough curves of ^{99}Tc at the water table for different values of the Active Fracture Model γ parameter and different conceptual models for fracture–matrix interchange (dual-k and discrete fracture model, DFM).

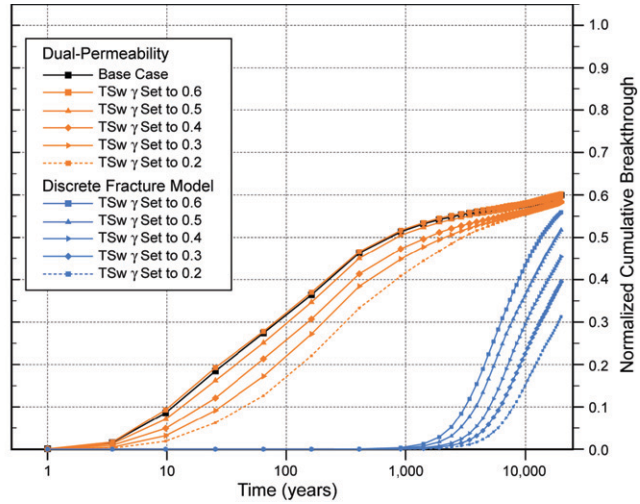

Fig. 9. Breakthrough curve of ^{242}Pu at the water table for different values of the Active Fracture Model γ parameter and different conceptual models for fracture–matrix interchange (dual-k and discrete fracture model, DFM).

changed in the model, for the ranges considered to be within the uncertainty bounds for these parameters. These results suggest that while infiltration clearly has a first order effect on transport, uncertainties in hydrologic parameters for a given infiltration scenario may not need to be propagated through a full radionuclide transport analysis.

Other physical transport properties, and the conceptual model for the fracture-matrix interchange model (dual-k versus DFM), also influence the transport results. For ^{99}Tc, Fig. 8 plots the travel times at the water table for a distributed release across the entire repository footprint, varying both the conceptual model and the model parameter (described below) that influences the wetted surface area available for diffusion. Regarding the conceptual model, the analysis yielded delayed first arrival times for the DFM compared to the dual-k model, although breakthrough curves for both conceptual models converge at later times. This figure also demonstrates that the conceptual uncertainty reflected by differences in the dual-k and DFM models is more significant than parameter uncertainty related to the γ parameter, which is varied from 0.2 to 0.6 in the sensitivity cases shown. This parameter, derived from the Active Fracture Model of Liu et al. (1998), accounts for the fact that under unsaturated flow conditions, only a fraction of the available fracture surface area is expected to be wetted; this results in channelized flow. In the model, higher values of the γ parameter result in a more channelized flow condition. Nevertheless, the impact of this uncertainty appears to be modest compared to the conceptual uncertainty in the fracture-matrix interaction model, especially for sorbing radionuclides, as illustrated in Fig. 9 for ^{242}Pu. The separation in time between breakthrough curves for the dual-k and DFM models are more dramatic for species with larger sorption coefficients. The DFM results in much longer transport times to the water table for sorbing radionuclides, due to the larger degree of contact of the radionuclide with the rock matrix (a precursor for sorption in the matrix rock) when the sharp gradient in concentration near each fracture is resolved in the fracture-matrix interchange model.

Discussion and Conclusions

This paper describes the numerical methods developed to simulate the transport of radionuclides through the unsaturated, fractured rock below the proposed repository at the Yucca Mountain. Using a dual-permeability solution for fluid flow as a basis, the transport model represents contaminant transport with a cell-based particle-tracking technique in which particles move from cell to cell in the three-dimensional large-scale grid of two overlapping continua. Particle movement from between cells, and the residence time within a cell, is computed probabilistically based on transfer functions using numerical solutions of the transport equations for an idealized fracture-matrix system. Using this approach, the following transport phenomena are simulated: advection through fracture and matrix continua, including between these continua, dispersion of both aqueous and colloidal species, sorption of dissolved radionuclides to the matrix continuum, and within the fault zones, and molecular diffusion between the fractures and matrix.

The new development in terms of computational methods is the implementation of a new transfer function method for dual permeability systems. The model derivation revealed that, for the idealized system of parallel flow in fractures and the surrounding matrix, a set of three dimensionless groups are required to fully define the parameter space. Therefore, to implement the method,

a set of normalized transfer functions are generated spanning the ranges of these three parameters, and an interpolation scheme is used to determine the appropriate transfer functions to use. These transfer functions serve as probability distributions for travel time through a given computational grid cell: when a particle is in that cell, the code samples from this distribution to obtain the travel time through the cell. Because of the vast difference in travel time depending on whether the particle resides in the fractures or the matrix, the method subdivides the problem at each cell by making the sampled travel time distribution conditional on whether the particle enters the cell via the fractures or matrix. In this way, the extremes of behavior, ranging from fracture-dominated to matrix-dominated flow and transport, can be simulated.

In addition to providing a realistic representation of the relevant processes, this method has the virtue of computational efficiency. This attribute is essential when performing many realizations in a large-scale simulation study, or in a multi-realization Monte Carlo analysis such as those used in performance or risk assessments. The application presented in this study, radionuclide transport through the UZ for the proposed Yucca Mountain repository, required a large number of individual model runs to explore the uncertainties in transport behavior, a situation that is well suited for analysis using this new method. In this study, model sensitivities were examined for uncertainties in the infiltration rate, sorption parameters, and diffusion parameters for a nonsorbing, moderately sorbing, and strongly sorbing radionuclide. Uncertainty distributions for each uncertain parameter were developed and propagated through the UZ transport model.

In a heterogeneous system like Yucca Mountain, large spatial variability in flow velocities and travel times result from significant differences in hydrologic and transport conditions within the domain of interest. In this model, fracture-dominated transport occurs in the northern repository region, whereas longer travel times are predicted in the southern region, where there is a layer of nonwelded tuff in which flow transitions to matrix flow. As expected, the travel times for both release locations are smaller for the higher infiltration rate scenarios. With respect to sorption and diffusion, the degree to which diffusion influences the model results depends on the flow and transport regime: when the transport is fracture-dominated, higher diffusion coefficients lead to greater contact of the radionuclide with the rock matrix, which results in larger travel times. However, if there is an interval in the flow path that exhibits matrix-dominated flow (as with the southern release location), travel times are controlled by the slow transport in that unit, and diffusion effects in the other units are masked by the fact that the travel times in those units are much shorter overall. Therefore, diffusion is an insensitive parameter for that situation. This result highlights the need to capture the flow regime exhibited by the system throughout the flow path. For complex vadose zones, this regime will be spatially variable, controlled by the local hydrologic conditions. Rapid transport from a release location to the water table will be the norm if all rocks in the flow path exhibit fracture-dominated flow, but significant delays may be experienced within intervening layers in which transport occurs predominantly through the rock matrix.

Another feature of the particle tracking method and its treatment of fracture-matrix diffusive interchange is the ease with which different model conceptualizations can be incorporated into the model. The code used in the present study is constructed so that different fracture-matrix interaction models can be simulated by changing only the set of transfer function curves supplied as input. In this study, a dual-k model consisting of a single node in the matrix was compared to a DFM in which the concentration gradients near the fracture were resolved with a finely discretized set of matrix grid nodes. For fracture-dominated transport, the DFM exhibited longer travel times through the system, an effect that was more prevalent for sorbing radionuclides. Clearly, the way in which small-scale processes are represented in the numerical model can have a profound impact on the large-scale predictions, a result that points to the need of gaining a fundamental understanding of these processes through experiments and theoretical considerations.

While the model results presented in this paper are specific to Yucca Mountain, this dual permeability transport modeling approach should be applicable to other vadose zone situations in similar systems. Even though unsaturated flow and transport through fractured rock is a complex problem exhibiting a wide range of behaviors, the model is capable of handling those systems efficiently and with accuracy comparable to continuum-based transport models. Therefore, regardless of the ultimate disposition of the Yucca Mountain License Application, this modeling study should be relevant to the general problem of contaminant transport in deep vadose zones, which is important to many environmental characterization and cleanup problems.

Acknowledgments

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Appendix: Nomenclature

b	fracture half-aperture
B	half-spacing between flowing fractures
C_{prev}	concentration at the previous time step
$\hat{C}$	the normalized breakthrough curve for the nondimensional time $\hat{t}$
D_{m}	effective diffusion coefficient

$d, \Delta z$	control volume depth and length, respectively
f_{fm}	fracture-matrix flux term
f_f, f_m	fluid mass fluxes for the fracture and matrix, respectively
f_{out}	total flux out of the continuum in which the particle currently resides
K_d	sorption coefficient
L	length of the flow path
m_f, m_m	fluid masses in a cell for the fracture and matrix, respectively
p_i (i: 1,2,3)	dimensionless parameters
p_{fm}	a probability of the particle transferring to the other medium
R_f, R_m	retardation factors in the fracture and matrix, respectively
$\hat{t}$	nondimentional time
V_m	interstitial pore-water velocity in matrix
$\bar{V}_z$	Darcy velocity in the fracture
ρ_b	bulk density
θ_f, θ_m	volumetric water contents of the fracture and matrix, respectively
τ_f, τ_m	fluid transport times in the fracture and matrix, respectively

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