Influence of Small-Scale Heterogeneities on Contaminant Transport in Fractured Crystalline Rock
by Ralph Mettier1,2, Georg Kosakowski3, and Olaf Kolditz2

Abstract
We present a sequence of purely advective transport models that demonstrate the influence of small-scale geometric inhomogeneities on contaminant transport in fractured crystalline rock. Special weight is placed on the role of statistically generated variable fracture apertures. The fracture network geometry and the aperture distribution are based on information from an in situ radionuclide retardation experiment performed at Grimsel test site (Swiss Alps). The obtained breakthrough curves are fitted with the advection dispersion equation and continuous-time random walks (CTRW). CTRW is found to provide superior fits to the late-arrival tailing and is also found to show a good correlation with the velocity distributions obtained from the hydraulic models. The impact of small-scale heterogeneities, both in fracture geometry and aperture, on transport is shown to be considerable.

Introduction
Flow and transport in crystalline rock mainly takes place within single or multiple fractures, or even complex networks of fractures. Due to the complexity and spatial inhomogeneity, flow and transport in fractured crystalline rock are much less well understood and modeled than in porous media. A good understanding of these processes is, however, key to a solid performance evaluation with regard to underground facilities, such as nuclear waste repositories (Heer and Smith 1998; Jakob et al. 2003). Several nations are planning on creating nuclear waste repositories in crystalline rock formations (e.g., Sweden [Backblom 1998], Finland [Enescu et al. 2004]). This provides a strong need for furthering the understanding of flow and transport processes in such media.

Several approaches to modeling flow and transport through single fractures (Moreno et al. 1988; Maloszewski and Zuber 1993; Oron and Berkowitz 1998; Lunati et al. 2003) and through more complex systems (Long et al. 1982; Hsieh et al. 1985; Geiger et al. 2004) of several, often interconnected, fractures have been used. Among these approaches are methods such as the use of equivalent porous media models (Endo et al. 1984), with single and dual/multiple porosities, representation of the fracture networks through networks of tubes of varying diameter (Dershowitz and Fidelibus 1999), reduction of the network to a single fracture or to several parallel fractures (Hadermann and Heer 1996), and also the use of statistically generated discrete fracture network models (Cacas et al. 1990; Berkowitz and Scher 1997; de Dreuzy et al. 2001a; Park et al. 2003). Common to all the mentioned approaches is that they are frequently used as a rather general representation of reality and are only rarely conditioned to represent a specific case, such as a fracture network. This is a consequence of the common absence of detailed data sets portraying the internal geometric structure of the studied rock volume. In most cases, “hard” data is limited to the breakthrough curves, with additional “soft” data coming from the borehole walls, surrounding geology, water chemistry, etc.

The excavation project (EP) provides us with a rare data set containing information not only of overall flow and transport characteristics but also on small- and medium-scale geometry and several geological properties of the studied rock volume (Alexander et al. 2003a). The data set was obtained by performing a number of transport tracer experiments through a section of a shear zone in the Grimsel test site in the Swiss Alps, followed by
impregnation of the fracture network with fluorescent resin. Subsequent overcoring with two slightly overlapping 30-cm-diameter cores, parallel to the main flow direction, along the shear zone, and extraction of the resulting cores enabled a large percentage of the rock volume involved in the transport tests to be recovered intact and cut into slices of ~3-cm thickness. The surfaces of these slices were then photographed in visible and ultraviolet (UV) light. The resulting set of 304 digital images presents a unique view of the interior structure of such a fracture network.

Related to EP, further transport experiments with various tracers have been performed in other parts of the same shear zone within the framework of the Radionuclide Retention Programme (RRP) project (Hadermann and Heer 1996; Smith et al. 2001; Alexander et al. 2003; Biggin et al. 2003) and Colloid Retention and Retardation (CRR) project (Möri 2003; Geckeis et al. 2004). The breakthrough curves obtained from CRR have recently been studied and fitted using the advection dispersion equation (ADE) and continuous-time random walks (CTRW) (Kosakowski 2004). It has therein been shown that neither uranine nor colloid tracers produce a breakthrough curve that can be adequately fitted using plain ADE. Both types of tracers show extended tailing, indicating some type of non-Fickian transport. Kosakowski (2004) showed that the introduction of matrix diffusion was sufficient to fit the breakthrough curves of uranine, but not those of the colloids, and suggested that colloids may not be party to matrix diffusion and that the prolonged tailing in the breakthrough curves stems from the influence of geometric small-scale heterogeneities. Similar observations concerning tailing effects in fractured media, which are not explainable by matrix diffusion, have been reported by Sidle et al. (1998), Kosakowski et al. (2000), and Becker and Shapiro (2000). Various related observations in other geological media are listed in Cortis et al. (2004). Based on this interpretation, one is led to suppose that factors such as matrix diffusion and geometric heterogeneities may be interchangeable to a degree in their role as tailing-producing retardation influences.

With this work, we attempt to study the impact of the small-scale geometric heterogeneity, including varying fracture apertures, upon the macroscopic advective transport. We base this on the geometric information from the EP project and do this with the following steps.

1. Create a simplified fracture network model, based on the geometric data obtained from EP
2. Quantify the spatial variability of fracture apertures and add this information to the fracture network model
3. Calculate purely advective transport in different conceptual models (different network geometries, with and without variable apertures) to see the influence of these fractures on the breakthrough curves
4. Compare the capability of fitting analytically obtained solutions to the modeled breakthrough curves by ADE and CTRW
5. Test the hypothesis that geometric heterogeneities can produce late-arrival tailing in purely advective transport, which may be mistaken for matrix diffusion or similar processes.

Figure 1. UV photography of one of the ~30-cm-diameter core slices obtained from the EP experiment. Resin-filled fractures show up well in the UV lighting. F1 is a main fracture, consisting of a large number of individual small fractures strongly interconnected and interspersed with mylonitic substance. F2 is a connecting fracture. F3 marks the beginning of a second main fracture.

Fracture Planes

The sequence of images provided by the EP data set allows us to identify three main fractures with a number of connections visible between them, in a model volume that encompasses two overlapping 30-cm-diameter cores in a rectangular prism box of 2.6-m length and 0.6-m width/depth. Of the interconnecting fractures, 18 were deemed significant for the flow and transport in the model. It must be noted that these main fractures are not always simple, continuous open fractures as normally imagined. In some areas, a single large aperture fracture dominates the structure; in other areas, the structures consist of many individual fractures of various sizes forming a continuous network of roughly planar shape. This type of fracture is typical of shear zone geometries. As Figure 1 illustrates, connecting fractures are much more like individual fractures in the classical geological sense, only rarely branching out into small network-like areas.

The possibility of further interconnections between these main fractures on a scale too small to be identified in the images cannot be excluded. The resolution of the images limits the size of recognizable fractures to one pixel, or ~0.6 mm. Based on visual inspection of a few select samples, and also from a qualitative, geological point of view, there is little doubt that smaller fractures of sizes possibly orders of magnitude smaller exist and function as connections between the main fractures. However, due to their small apertures, their influence on the overall fluid flow is minor. Further, the possibility of additional flow field–relevant fractures and intersections...
outside the extracted rock volume can also not be excluded. Consideration of the full complexity of the fracture network is beyond our numerical capabilities, as well as beyond the level of detail that can be extracted from the data set. Therefore, our models shall be limited to the main fractures of the fracture network. As mentioned previously, we consider 18 connections between the three main fractures to be significant. These coincide with structures that are clearly identifiable in the images.

The main fractures are represented as single planes, extended to intersect the model borders on all sides. The connections between them are also portrayed as planar single fractures; however, their planes terminate where they intersect a main fracture or where they are no longer visible in the data.

As for degrees of complexity, we choose three variations. In case A, a single fracture with varying apertures represents the modeled volume, whereas the connections between the main fractures are ignored. In case B, the model consists of three parallel fractures and the connections between them. All fractures have a constant aperture of \(1.3 \times 10^{-4}\) m. The variation with the highest degree of complexity, and also the strongest resemblance to the actual system, is case C, a model consisting of three parallel main fractures, with connections and varying apertures on all fractures. Further possible geometric refinements of the model with regard to realism would consist of adding additional classes of fractures, smaller than the connecting fractures.

**Apertures**

All the planar representations of fractures, including main fractures and connections, are modeled as two-dimensional (2D) objects in three-dimensional space. As a first approximation, a unit aperture can be assigned. From RRP, an average transport aperture on the order of \(10^{-4}\) m for each of the three parallel fractures was calculated. This is roughly equal to the constant aperture of \(1.3 \times 10^{-4}\) m in case B. As the average aperture is much smaller than the other dimensions of the fractures (0.3 to 0.6 m), the simplification as a 2D object appears justified, not only with regard to the massive reduction in the number of necessary mesh elements and thus computational power but also as such a model was suggested by Bossart and Mazurek (1991) in their original study of the geological structure of this shear zone.

Image resolution does not allow for a conclusive study of fracture apertures. Nonetheless, a study of some large fractures can yield some information toward correlation length and reasonable aperture distribution. The apertures of a section with fairly constant large apertures (>5 mm) over a vertical distance of 20 images were measured by a simple line counting method: horizontal rows of pixels across the open fracture are considered, and the pixels that are clearly part of the fracture are counted. Each image has a size of \(~500 \times 500\) pixels, and the studied section shows a length between 0.5 and 1 core diameter (30 cm) in the images. This gives us a statistical data set of 8315 samples, large enough for a geostatistical analysis. The measured apertures were observed to have a distribution strongly reminiscent of a lognormal distribution; the histogram is shown in Figure 2. This appears credible as lognormally distributed apertures are known to commonly exist in fractured rock systems (see, e.g., Bianchi and Snow 1969; Keller 1998; de Dreuzy et al. 2001b; Vandersteen et al. 2003). Our working hypothesis is therefore that this observed lognormal distribution shall also be valid for smaller fractures.

As part of a geostatistical analysis, a variogram for the measured large fracture apertures was calculated. As shown in Figure 2, a theoretical variogram model is fitted to the data, allowing us to generate any number of aperture distributions, not only with the same statistical distribution but also with the same average rate of change in aperture over a given distance.

A satisfactory fit was found to be a combination of two spherical models (nugget: 0.005; sill 1: 0.014, range 1: 0.05 m; sill 2: 0.015, range 2: 0.25 m). The two spherical models incorporated into our model variogram cannot without a doubt be connected to specific directions or fractures. The first model, with a range of 5 cm may reflect an artifact, caused by the uncertainty of the image positions along the core axis. In spite of the uncertainties, this distribution, along with the given mean of \(1.3 \times 10^{-4}\) m, sufficiently fixes the parameters needed to generate suitable apertures. Using sequential Gaussian simulation, we generated 50 realizations of aperture distributions for model cases A and C, one of which is illustrated in Figure 3. Apertures are binned into 100 logarithmically spaced bins, varying from a low-end cutoff of \(10^{-5}\) m to an upper-end cutoff of \(2 \times 10^{-2}\) m. The upper cutoff was determined by the largest fracture aperture to be identifiable in the images, while the lower cutoff was chosen rather arbitrarily to reduce the number of sparsely filled material bins. The resulting distributions show a standard deviation of 0.80 ± 0.02 for the decadic logarithms of the

![Figure 2](image_url)
apertures. It should be noted that, unlike a natural fracture, there are no elements with zero aperture. However, due to the use of a particle tracking method, a flow volume below a certain amount results in zero particles, which is equivalent to a closure point.

All geostatistical calculations were performed using R, particularly the gstat library for R (Pebesma 2004, http://www.gstat.org).

Calculations

Flow Calculations

We used Rockflow (Kolditz et al. 1999), a standard Finite Element Galerkin Solver, to solve the hydraulic equation for flow $Q_f$ in a fracture:

$$Q_f = -T_f \nabla \bar{\phi}$$  \hspace{1cm} (1)

where $\bar{\phi}$ is the average piezometric head across a fracture’s width and $T_f$ is the transmissivity of the fracture, determined via the cubic law (Equation 2), and thereby calculate the static flow field before turning to transport calculations.

$$T_f = \frac{\rho g b^3}{\mu 12}$$ \hspace{1cm} (2)

where $\rho$ is fluid density, $g$ is gravitational acceleration, $b$ is half the fracture aperture, and $\mu$ is the fluids coefficient of dynamic viscosity.

At the upper end of the models ($z = 2.8$ m), a Dirichlet boundary with constant zero hydraulic head was set as the outflow boundary. Intersections of all fractures with outside boundaries except the top and bottom were set as no-flow boundaries. The bottom boundary ($z = 0.2$ m) was set as an inflow boundary, by using a Neumann constant flow boundary (note: due to our choice of axis orientation, flow is from bottom to top). To determine a suitable flow rate at the inflow boundary, the models were first run with a Dirichlet inflow boundary of 1, representing a head difference of 1 m between inflow and outflow boundary. The total resulting influx volume was then distributed evenly among the number of vertices on the inflow boundary for the Neumann condition. This provides us with a constant and equal flux volume at each inflow node, without taking into account the variability of the apertures along the inflow boundary.
Transport Calculations

We employed a flow line following particle tracking algorithm, running on top of the hydraulic solutions, to model purely advective transport. Diffusion processes are not considered. This may lessen the degree of realism in the model as it is not equivalent to slow solute transport. It more closely resembles the fast transport of large particles with a relatively low diffusion constant. This facilitates the investigation into the mechanism of advective transport. Due to the use of a Neumann boundary with equal inflow at each node at the inflow edge, regardless of aperture, we can set the number of particles released at each inflow element edge proportional to the volume flux at that edge to achieve an equal release rate across the inflow boundary. As we also record particle arrivals at any point along the outflow edge, our transport calculations can be considered as pseudo–one dimensional (1D) even though the model geometries are not. Breakthrough curves for cases A and C were obtained by concatenating the arrival time lists of particles at the outflow boundary from the 50 realizations, and then binning them into 30 logarithmically spaced time windows. As case B has no varying aperture, a single realization was used.

Boundary Conditions

Our choice of the inflow boundary conditions may lessen the degree of realism in the model, but it also avoids some computational problems that can arise if the boundary is set using constant hydraulic heads. Foremost among these problems is the possibility of backflow due to the contrast in aperture between elements, which runs over several orders of magnitude.

In the case of elements with small apertures, this approach will cause a larger number of particles to be forced into the elements than is to be expected with a Dirichlet boundary setting. However, the velocity in these elements will also be higher than normal, causing these particles to quickly move into adjacent elements with larger apertures.

After short distances (and times), ~5% to 10% of the overall model length, the flow field has recovered from the disturbance of the flux boundary and a flow regime equivalent to one caused by a Dirichlet boundary condition has been reached. After these distances, particles are mainly contained in a number of channels. As can be seen in Figure 3, the heterogeneous velocity field has indeed normalized, and channels of higher velocities are visible, after ~15 cm.

Continuous-Time Random Walks

CTRW is an approach to modeling transport processes by representing them with a series of particle movements on a regular grid. Mathematically, CTRW performs this by numerically solving the 1D Fokker-Plank Equation with Memory for the Laplace transform of the probability distribution function of the waiting time and step length. The memory function, which is the fundamental solution of the CTRW equation, is the Laplace transform of the probability density function of the waiting time.

The CTRW algorithm, running on top of the hydraulic solutions, to model purely advective transport. Diffusion processes are not considered. This may lessen the degree of realism in the model as it is not equivalent to slow solute transport. It more closely resembles the fast transport of large particles with a relatively low diffusion constant. This facilitates the investigation into the mechanism of advective transport. Due to the use of a Neumann boundary with equal inflow at each node at the inflow edge, regardless of aperture, we can set the number of particles released at each inflow element edge proportional to the volume flux at that edge to achieve an equal release rate across the inflow boundary. As we also record particle arrivals at any point along the outflow edge, our transport calculations can be considered as pseudo–one dimensional (1D) even though the model geometries are not. Breakthrough curves for cases A and C were obtained by concatenating the arrival time lists of particles at the outflow boundary from the 50 realizations, and then binning them into 30 logarithmically spaced time windows. As case B has no varying aperture, a single realization was used.

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$$u \tilde{c}(x, u) - c_0 = \tilde{M}(u) \left[ \frac{\partial}{\partial x} \tilde{c}(x, u) - D_b \frac{\partial^2}{\partial x^2} \tilde{c}(x, u) \right]$$

where

$$\tilde{M}(u) = \tilde{r} u - \frac{\tilde{\psi}(u)}{1 - \tilde{\psi}(u)}$$

is a memory function that accounts for the contained unknown heterogeneities. $\tilde{r}$ is some characteristic time. $v_\phi$ and $D_\phi$ are the center of mass velocity and generalized dispersion coefficient, respectively, the details of which are discussed extensively in Dentz et al. (2004) and Cortis et al. (2004). The variable $u$ is the Laplace variable.

The function $\tilde{M}(u)$ can take on several expressions depending on the form for $\tilde{\psi}(u)$. For the case of

$$\tilde{\psi}(u) = \frac{1}{1 + u}$$

Equation 3 reduces to the classical ADE, whereas for

$$\tilde{\psi}(u) = (1 + t_2/u_1)^{t^2} \exp(t_1 u) \Gamma(-\beta, \tau_2^{-1} + t_1 u) / \Gamma(-\beta, \tau_2^{-1})$$

we get a truncated power law (TPL) model, which requires the three input parameters $\beta$, $t_1$, and $t_2$ where $\tau_2 = t_1 u_2$.

The parameters $t_1$ and $t_2$ determine the upper and lower limits of the linear (on a log-log scale) or power law section of $\tilde{\psi}(u)$, whereas $\beta$ determines the slope of the linear section. In addition, $\beta$ determines if the flow type is Fickian ($\beta > 2$), non-Fickian (1 < $\beta < 2$), or strongly non-Fickian ($\beta < 1$).

All CTRW calculations for this project were performed using the CTRW Matlab toolbox (Cortis and Berkowitz 2005).

Advection Dispersion Equation

The ADE fits were calculated using the Matlab curve fitting tool, with the Ogata-Banks solution of the transport equation (Bear 1972)

$$C(x,t) = \frac{\text{d}M}{\text{d}x^2} \exp \left\{ - \frac{[x - g(t-t^2)]/n^2}{4D(t-t^2)} \right\}$$

where $g$ is Darcy velocity and $n$ is porosity, which is 1 in our case of unobstructed fractures. The total injected tracer mass $\text{d}M$ is set as 1 to reflect our normalized breakthrough curves. This leaves us with a simplified version of

$$C(x,t) = \frac{1}{4\pi D} \exp \left\{ - \frac{[x - Vt]^2}{4Dn} \right\}$$

where velocity $V$ is $q/n$ and $t = t'$. $V$ and dispersion coefficient $D$ are then obtained by fitting this equation to our breakthrough curves.

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Results and Discussion

An important observation to be made in the results of the numerical models is the predominance of particle transport through a complex network of well-defined channels. This is in agreement with the findings of Moreno and Neretnieks (1985), Tsang and Tsang (1987), and Johns and Roberts (1991), who postulated that fluid flow should be unevenly distributed in a fracture plane with rough surfaces.

The breakthrough curves obtained via particle tracking can be observed in Figure 4, together with the fits from ADE and CTRW. All three curves show late-time tailing, which is usually considered to be typical of non-Fickian transport, and cannot be characterized by plain ADE. In order to directly compare the breakthrough curves, they must be normalized for a common Darcy flux of $1 \times 10^{-3} \text{m/s}$ (fluid volume per area, where area is the fracture aperture multiplied by the width of the fracture). This is easily possible as it is evident from Equations 1 and 2 that flux, velocity, and head gradient are linearly related. To facilitate fitting by ADE and CTRW, the breakthrough curves were normalized to ensure that the peak of the case A breakthrough curve was located at $t = 2.6$ s, which gives us an average peak velocity of 1 m/s over the 2.6-m length of the models. It is evident that this normalization gives unrealistic high velocities, but due to the linear relationship in Equation 1, it is easily possible to backtransform all parameters to realistic values. In addition, the particle concentrations were normalized with the injected amount of particles to have an integral area of 1 for each curve.

For all three cases, ADE is unable to provide a satisfying fit over the whole lengths of the curves. The ADE fits to the peak regions of the curves, presented in Table 1, are good, but they cannot reproduce the late-arrival tailing. CTRW can produce a fit that takes both peak and tailing into account. Besides the difference in tailing, the CTRW fits also represent the general shape of the curves much better, especially for cases A and C. Case B proves harder to fit than cases A and C as the breakthrough curve of case B is markedly different from that of the other two. It consists of a narrow peak with broad tailing.

Table 1

<table>
<thead>
<tr>
<th>Case</th>
<th>$V$ (m/s)</th>
<th>$D$ (m$^2$/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.93</td>
<td>$1.1 \times 10^{-1}$</td>
</tr>
<tr>
<td>(single fracture, variable aperture)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>1.77</td>
<td>$3.1 \times 10^{-3}$</td>
</tr>
<tr>
<td>(network, constant aperture)—peak</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>1.20</td>
<td>$1.4 \times 10^{-1}$</td>
</tr>
<tr>
<td>(network, variable aperture)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
only a small amount of tailing visible. We attempted to fit the peak region of case B with CTRW, and also with ADE. It is important to note that for the cases where transport is Fickian, CTRW and ADE are equivalent (Dentz et al. 2004). Further, the late-arrival flank, where non-Fickian transport is dominant, was also fitted by CTRW. However, all three fits to case B are fair at best.

In order to compare the breakthrough curves and their fits, it is necessary to consider the reasons and implications of the differences. As can be seen in Figure 3, the fracture network geometry consists mainly of three large parallel fractures, with two regions where the interconnection is substantially denser. These two “connected” regions mainly influence two of the major fractures, while flow and transport in the third major fracture is undisturbed after the initial intersection. A simple case of three parallel fractures without any interconnections would obviously result in a pulse arrival with only very little numerical diffusion. Fracture geometries consisting of multiple interconnected fractures are known to exhibit a retardation effect that also causes late-arrival tailing (Berkowitz and Scher 1998; Becker and Shapiro 2000). It stands to reason that, if isolated, the small networked region would show an answer function to a pulse tracer injection that also shows substantial tailing. The result in case B is a superposition of a typical pulse breakthrough and a breakthrough with some tailing from the geometrically complex region. As the majority of tracer particles is transported directly through the major fractures, and only a small portion enters the complex network region, the influence of the tailing is smaller than the influence of the direct pulse-like breakthrough. This larger-scale heterogeneity with regard to geometric complexity makes it impossible to find a “representative elementary volume” in this model, with all the known consequences (Berkowitz and Scher 1995); thus, a single curve cannot produce a suitable fit. Fitting two CTRW curves, as shown in Figure 4, can be interpreted as fitting either the influence of the strongly networked region (flank) or that of the regions without any strong influences from the interconnected main fractures (peak).

The introduction of variable fracture apertures has a strong impact, as can be seen when comparing case B to case C and also when considering that case A would show a simple pulse breakthrough without the varying apertures. Case C (Figure 4) shares the underlying geometry with case B, but the effect of this large-scale heterogeneity is essentially invisible due to the much stronger influence of the variable apertures. This shows that the retardation effects stemming from lengthened flow paths and/or local decreases of flow velocity, while basically comparable in function, are not present in a sufficient magnitude to present a visible impact in these models.

The fitting parameters obtained from ADE are shown in Table 1, and those from CTRW in Table 2. When comparing the values for V and D for the two approaches, it is apparent that CTRW delivers consistently higher values for V and lower values for D. This is consistent with similar cases where both types of fit have been compared (Jiménez-Hornero et al. 2005) and therefore does not seem to be an artifact of these particular two cases. The lower D returned by CTRW seems expectable, considering that in ADE, dispersion is the only component contributing to widening the breakthrough curve, whereas in CTRW, the effects of small-scale heterogeneity are also considered and incorporated in the memory term. The V parameters from CTRW and from ADE are directly comparable. Both values for V describe the average velocity, in the sense of the velocity of the center of mass of the increasingly spread out tracer cloud respectively the first spatial moment of the tracer distribution (Cortis et al. 2004).

The β parameters obtained from our fits to cases A and C (Table 2) are below 2, thus indicating non-Fickian transport. In case B, CTRW can fit either the peak, using a high value for β, thus representing Fickian transport as the ADE fit does, or the late-arrival flank, using a β below 2, representing non-Fickian transport.

When we consider our models to be “black boxes” between the two planes of inflow and outflow boundaries, the breakthrough curves are transit time distributions on a global or macroscopic scale: the distance between the boundaries. The applied finite-element method provides each model element with a fixed velocity vector. These vectors represent a velocity distribution on the scale of an individual element. By weighting the inverse of this velocity distribution with the corresponding element sizes, and with the number of particles that traversed each element, we obtained a local or microscopic transit time distribution. This distribution describe the probability of a given particle to transverse a standard element in a given time and is thus equivalent to the ψ(t) function in CTRW. The local transit time distributions of all three models, shown in Figure 5, are fairly similar. On a

Table 2
The Values for Velocity V, Diffusion Coefficient D, and Fitting Parameters from the CTRW-TPL Fits to the Breakthrough Curves Seen in Figure 4. In the Case of the Peak CTRW Fit to Case B, a Non-TPL Was Used, Resulting in No Applicable Values for t1 and t2

<table>
<thead>
<tr>
<th>Case</th>
<th>V (m/s)</th>
<th>D (m²/s)</th>
<th>β</th>
<th>t1 (s)</th>
<th>t2 (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case A</td>
<td>1.34</td>
<td>1.19 x 10⁻²</td>
<td>1.54</td>
<td>3.16 x 10⁻²</td>
<td>1.01 x 10⁶</td>
</tr>
<tr>
<td>Case Bpeak</td>
<td>1.85</td>
<td>1.51 x 10⁻³</td>
<td>2.0</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Case Btail</td>
<td>1.72</td>
<td>8.30 x 10⁻³</td>
<td>1.8</td>
<td>9.54 x 10⁻⁵</td>
<td>3.05 x 10¹</td>
</tr>
<tr>
<td>Case C</td>
<td>1.24</td>
<td>2.45 x 10⁻²</td>
<td>1.71</td>
<td>8.81 x 10⁻²</td>
<td>9.45 x 10⁴</td>
</tr>
</tbody>
</table>

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log-log scale they all have a strongly asymmetric shape: for small times they show approximately lognormal behavior, whereas for larger times, they show pronounced power law behavior. The power law slopes of the local transit time distributions as shown on a log-log scale in Figure 5 are presented in Table 3. The comparison between the slopes of the local transit distributions and the \( \beta \) parameters obtained from the CTRW fits provide additional evidence that an approximation of the observed transport behavior by CTRW with a TPL-determined \( \psi(t) \) function is suitable in these cases. As for the TPL fits, the lower limiting time \( t_1 \) is small and the upper limiting time \( t_2 \) is larger by many orders of magnitude, and the fits essentially approximate the transit time distribution with a non-TPL. This is equivalent to the method using a non-TPL based \( \psi(t) \) in Berkowitz et al. (2001). A \( \psi(t) \) that also fits the lognormal section of the transit time distributions would be ideal; however, this approximation is considered sufficient for our purposes and is clearly superior to ADE, which is limited to exponential distributions of local transit times.

![Figure 5. Microscopic or local transit time distributions of the three models. Straight lines fitted to the transit time distributions indicate the log-log slopes, with parameters presented in Table 3. Case B represents one realization, and cases A and C are stacks of 50 realizations each of which explains the smoother appearance of cases A and C. Cases B and C have been shifted along the y-axis by three and six decades, respectively, to allow for better viewing. Case B shows two linear segments, with differing slopes.](image)

### Table 3: CTRW Fitting Parameter \( \beta \) Compared to the Slopes of the 1/\( \psi \) Distributions Shown in Figure 5

<table>
<thead>
<tr>
<th>Case</th>
<th>( \beta )</th>
<th>( -1 - \beta )</th>
<th>Slope of 1/( \psi )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case A</td>
<td>1.54</td>
<td>-2.54</td>
<td>-2.53</td>
</tr>
<tr>
<td>Case Bpeak</td>
<td>2.0</td>
<td>-3.0</td>
<td>-6.03</td>
</tr>
<tr>
<td>Case Bfull</td>
<td>1.8</td>
<td>-2.8</td>
<td>-3.08</td>
</tr>
<tr>
<td>Case C</td>
<td>1.71</td>
<td>-2.71</td>
<td>-2.86</td>
</tr>
</tbody>
</table>

Besides the fitting parameters for \( V \) from CTRW and ADE, we can also calculate a flow velocity from the hydraulic boundary conditions, by simply dividing normalized flux \( Q \) by the cross-sectional area of the inflow edges. For case B, with a constant aperture of \( 1.3 \times 10^{-4} \) m, this gives a precise calculation of the flow velocity at the inflow edge. For cases A and C, ergodicity dictates that we could also use an aperture of \( 1.3 \times 10^{-4} \) m as this is the mean forced on the generated aperture distribution. In fact, however, the actual mean apertures over all 50 realizations of cases A and C were used \( (1.32 \times 10^{-4} \) and \( 1.29 \times 10^{-4} \) m, respectively). As the flux was normalized to be equal over equal inflow edge lengths, all models have a near-identical theoretical hydraulic velocity, which is compared to the velocities obtained from CTRW and ADE in Table 4.

### Conclusions and Summary

We present a group of flow and transport models of varying geometric complexity, all based on the data extracted from EP.

These models all produce breakthrough curves with steep early-arrival flanks; wide, rounded peaks; and strong late-arrival tailing. Overall, these are comparable to curves obtained by transport experiments performed in situ on the rock volume described by EP.

These breakthrough curves cannot be fitted satisfactorily by the classical transport equation; they can, however, be fitted very well by CTRW. The fact that the breakthrough curve from case B cannot be fitted even with CTRW stems from the larger-scale geometric heterogeneity. A larger model, taking into account the statistical relevance of such “networked” areas, or a similar-sized model with additional smaller-scale geometric heterogeneities may provide a better approach for this case.

CTRW can be linked with the numeric transport models via the relationship between local transit time distributions, which directly portray the heterogeneity in fracture aperture and geometry, and \( \psi(t) \), which describes the duration of the CTRW “hops.” This indicates that it is conceivable to make limited predictions about transport behavior for a model case where nothing more than the local velocity distribution is known.

Comparison of our three models shows that the impact of varying fracture apertures on transport behavior is larger than that of the network geometry. However, a more
complex fracture network geometry would also have a larger impact on the velocity distribution, also leading to a widening of the breakthrough curves. Additional models, incorporating another “layer” of geometric complexity, on a smaller scale remain as a future task that promises better quantification of the role of network geometry.

Compared to the actual network structure studied in EP, our results are based on oversimplified fracture network geometries. Especially, the low complexity of the macroscopic heterogeneity presented by the network structure of the model does not allow us to fully quantify the possible influence of this characteristic on the breakthrough curves. They can offer no quantitative information about the roles of fracture aperture and network complexity in the EP transport experiments. It does, however, seem clear that both factors have the potential to enhance tailing in breakthrough curves. In the light of these results, the hypothesis presented by Kosakowski (2004) that certain tracers may show long late-arrival tailing caused by geometric complexity and small-scale heterogeneities, and not by matrix diffusion, seems very credible.

The main drawback of our scenarios is the lack of representation of diffusion processes. In a follow-up project, it is intended to include and investigate the influence of both chemical diffusion (from one flowpath to another) and diffusion into an adjacent porous rock (matrix diffusion).

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