Transition from Non-Fickian to Fickian longitudinal transport through 3-D rough fractures: Scale-(in)sensitivity and roughness dependence

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Abstract

Understanding transport in rough fractures from non-Fickian to Fickian regimes and the prediction of non-Fickian transport is critical for the development of new transport theories and many practical applications. Through computational experiments that fall within the macrodispersion regime, we first simulated and analyzed solute transport through synthetic rough fractures with stationary geometrical properties (i.e., fracture roughness $\sigma_b/\langle b \rangle$ and correlation length λ , where b refers to aperture with its standard deviation σ_b and arithmetic mean $\langle b \rangle$) across increasing fracture longitudinal transport domain length L , with L/λ ranging from 2.5 to 50. The results were used to determine how solute transport behavior evolves with increasing scale in the longitudinal direction. Moreover, a set of correlated fractures with aperture fields following normal and log-normal distributions was created to further identify and quantify the dependence of non-Fickian transport on roughness. We found that although persistent intermittent velocity structures were present, the breakthrough curves (BTCs) and residence time distributions showed diminishing early arrival and tailing, features of non-Fickian transport, with increasing longitudinal L/λ , ultimately converging to a Fickian transport regime given $\sigma_b/\langle b \rangle$ remained constant. Inverse analysis of the experimental BTCs with the

advection-dispersion equation (ADE) model showed that the dispersion coefficient (D) was non-trivially scale-dependent. Simulation results for rough fractures with varying $\sigma_b/\langle b \rangle$ and L/λ indicated that the ratio of fluid velocity to transport velocity fitted to the ADE model depends on $\sigma_b/\langle b \rangle$ and L/λ . The continuous time random walk (CTRW) performed much better across all transport scales, and resulted in scale-insensitive fitted parameters, i.e., β in the memory function. The fitted β is proportional to $\sigma_b/\langle b \rangle$ but is insensitive to L/λ . Therefore, bulk longitudinal solute transport across the pre-asymptotic and asymptotic regimes can be estimated based on the CTRW model parameterized by measurable fracture physical properties.

1. Introduction

Solute transport through fractured media is subject to complex physical and chemical processes, characterized by preferential flow in fractures and slow/stagnant flow in the porous matrix or eddies, flow and solute exchange between the fracture and matrix (Roubinet et al., 2012), and fluid-rock interactions (Mukhopadhyay et al., 2013). The broad variations of velocity and chemical reactions consequently lead to non-Fickian transport across all scales of fractured media (Berkowitz, 2002). Unfortunately, accurate prediction of complex non-Fickian transport processes remains a challenge due to limitations in detailed description of single fractures, let alone discrete fracture networks and dual-porosity media (Neuman, 2005). As a result, numerous studies have focused on non-Fickian transport through single fractures (Bauget and Fourar, 2008; James et al., 2005; James and Chrysikopoulos, 2000; Nowamooz et al., 2013; Wang and Cardenas, 2014).

Non-Fickian transport is due to two underlying mechanisms, both due to local

heterogeneity in the flow field. First, the transport observation occurs at spatial and temporal scales that are too short or small, and therefore the solute fails to experience or sample the complete heterogeneity of the medium after where or when transport reaches the Fickian (Taylor) regime (e.g., Bolster et al., 2014; Bouquain et al., 2012; Koch and Brady, 1987; Majdalani et al., 2015). Second, incomplete local mixing resulting from a complex flow field takes place (Koch and Brady, 1987), including for example a growing eddy (Cardenas et al., 2007; Cardenas et al., 2009; Sund et al., 2015). Even though the effect of heterogeneity (e.g., variance and correlation length) on the flow field has been documented extensively for decades, recent studies continue to quantify the impact of micro-scale heterogeneity on non-Fickian transport (Aubeneau et al., 2014; Heidari and Li, 2014; Kang et al., 2014).

Numerous studies have shown that fracture roughness (another indicator of heterogeneity for fractures) has a remarkable impact on the non-Fickian transport through single fractures (e.g., Bauget and Fourar, 2008; Cardenas et al., 2007; Keller et al., 1999). Fracture roughness typically refers to the roughness of fracture surfaces (Brown, 1987; Candela et al., 2012), but in this study, we define roughness ($\sigma_b/\langle b \rangle$) as the variation of aperture field, where σ_b and $\langle b \rangle$ are standard deviation and arithmetic mean of aperture b , respectively. The non-Fickian transport behavior can be approximated based on physical properties ($\sigma_b/\langle b \rangle$) of two-dimensional (2-D) rough fractures following our previous study (Wang and Cardenas, 2014). However, transport through 2-D fractures does not allow for channelization which could occur in 3-D rough fractures, thus adding a mechanism for non-Fickian behavior. Therefore, the information needed for predicting non-Fickian transport through three-dimensional (3-D) rough fractures remains mostly unknown, particularly with increasing fracture scales.

Although simulations of flow and transport through simple pores at increasing pore scales have shown that the transport transitions from non-Fickian to Fickian regimes (Cardenas, 2009; Zhang and Lv, 2007), the effect of scale on non-Fickian transport through fractures needs to be further studied in terms of identifying transition time and length scales, such that transport transition can be properly predicted (Bolster et al., 2014). Moreover, the determination of transition scales can help in the design of observation networks for experimental sampling or even field monitoring (Majdalani et al., 2015).

To capture transport behavior, the classical advection-dispersion equation (ADE) with constant velocity and dispersion coefficient D is widely used, even though its shortcomings for capturing non-Fickian behavior are widely recognized. Therefore a suite of nonlocal representations of advective and dispersive solute transport has emerged that more accurately describes non-Fickian transport (Bauget and Fourar, 2008; Berkowitz et al., 2006; Neuman and Tartakovsky, 2009). The continuous time random walk (CTRW) framework is one such approach for modeling non-Fickian transport which has been applied extensively to fractures and fracture networks (Berkowitz et al., 2006; Cortis et al., 2004; Dentz et al., 2004; Le Borgne et al., 2008). Moreover, the inclusion of the truncated power law (TPL) in time within the CTRW framework led to improved agreement between theoretical breakthrough curves and observations for systems with a finite scale (Cortis and Berkowitz, 2004). However, although the theoretical prediction of transport across non-Fickian to Fickian transition is possible by using the CTRW-TPL, there are few physical and even computational experiments that quantify such a transitional transport phenomenon across increasing scales. Only a few studies directly assess and directly connect the parameters of the CTRW-TPL to measurable physical properties

(McInnis et al., 2014; Wang and Cardenas, 2014).

The CTRW-TPL, whose ability to represent macroscopic non-Fickian transport has been verified several times (Berkowitz, 2002; Berkowitz et al., 2006; Dentz et al., 2004; Kang et al., 2011), assumes that the temporal and spatial transitions are decoupled. This decoupled partial differential equation form of CTRW performs well regardless of whether the velocity field is correlated or not, as long as the spatial transitions are compact (Berkowitz and Scher, 2010). Recent studies integrated information about velocity correlation via a spatial Markov method into the classic CTRW; this framework is sometimes called correlated CTRW (Bolster et al., 2014; Kang et al., 2014; Kang et al., 2011; Le Borgne et al., 2008). The correlated CTRW, for some cases with appropriate discretization of transition path, can accurately capture the early arrival and long tailing features (de Anna et al., 2013; Kang et al., 2011; Le Borgne et al., 2008). However, the correlated CTRW requires prior knowledge of particle distribution, which is normally unknown a priori, thus limiting its practical application. Therefore, instead of using the correlated CTRW, we investigated whether the classic CTRW-TPL framework is able to predict solute transport transitioning from non-Fickian to Fickian when velocity field is correlated; this was achieved by identifying an appropriate truncated time and other relevant parameters, and then relating the model parameters to the physical properties of 3-D rough fractures. We also compared the analysis with the CTRW-TPL framework to more conventional inverse modeling with the ADE.

In this study, we first investigated solute transport through single rough-walled fractures with increasing scales (fracture length) in order to quantify the effect of scale in potentially transforming transport from non-Fickian to Fickian regimes, and to further assess

the usefulness and appropriateness of the CTRW-TPL framework for prediction across transitional transport phenomenon but confined within the macrodispersion regime (Detwiler et al., 2000). Then, we attempted to establish a relationship between the parameter β in the CTRW-TPL model and physical properties of 3-D rough fractures by changing fracture roughness and correlation length; β represents the nature of the transport, i.e., the degree of deviation from Fickian transport, and encapsulates the degree of heterogeneity. We addressed the following questions: How do the ADE and CTRW parameters change with fracture scale across, presumably, the pre-asymptotic to asymptotic regimes in rough fractures? What is the asymptotic longitudinal scale for transport through rough fractures, if any? How robust is CTRW-TPL for modeling the transport transition across scales? Is the relationship between β in the CTRW-TPL model and fracture roughness as established for the 2-D cases applicable for the 3-D cases? To this end, we first created synthetic fractures with aperture fields following typical distributions as observed in natural fractures. Then we simulated solute transport through the fractures using a computationally efficient quasi-3-D particle tracking random walk (PTRW) algorithm which uses a 2-D velocity field computed through a modified Local Cubic Law. The resultant 'experimental' breakthrough curves were then analyzed with the classical one-dimensional (1-D) ADE and the CTRW-TPL. The statistical details of the heterogeneous flow fields were examined in order to interpret the transport observations.

2. Theoretical Background

2.1. Advection dispersion equation

The solute transport in shear flow through fractures can be upscaled to a 1-D advective-dispersive process by using a bulk velocity and an effective dispersion coefficient, which reads

132 as:

$$\frac{\partial C}{\partial t} = -U_{ADE} \nabla C + D_{ADE} \nabla^2 C \quad (1)$$

133 where C is upscaled solute concentration, t is time, U_{ADE} is constant mean velocity, and D_{ADE} is
134 constant hydrodynamic dispersion coefficient. The inherent assumption behind the ADE is the
135 applicability of the Taylor (Fickian) dispersion theory (Taylor, 1953). That is, the travel time of
136 the solute is long enough so that the transverse gradients in solute concentration become
137 negligible; this threshold time is often called the “asymptotic time” (Koch and Brady, 1987). For
138 an idealized fracture with perfectly smooth surfaces (i.e., parallel plates), the asymptotic time
139 can be calculated from the aperture (Wang et al., 2012). However, fracture roughness or
140 heterogeneity may significantly lengthen the asymptotic time because it takes longer for the
141 transverse mixing to smooth out the longitudinal concentration gradients (Wang and Cardenas,
142 2014).

143 **2.2. Continuous time random walk theory**

144 The CTRW is fundamentally derived from the Master equation of statistical mechanics
145 with appropriate memory functions to represent non-Fickian behavior (Berkowitz et al., 2006;
146 Cortis and Berkowitz, 2004). In 1-D Laplace space, the CTRW model reads as:

$$p\tilde{C}(x, p) - C_0(x) = -\tilde{M}(p) \left[U_{CTRW} \frac{\partial}{\partial x} \tilde{C}(x, p) - D_{CTRW} \frac{\partial^2}{\partial x^2} \tilde{C}(x, p) \right] \quad (2)$$

147 where the tilde represents a variable or function in the Laplace space, p is the Laplace variable,
148 U_{CTRW} and D_{CTRW} are transport velocity and dispersion coefficient in the context of the CTRW,
149 respectively, which may vary temporally according to the variations of local velocity fields
150 caused by local heterogeneity (Berkowitz et al., 2006). Note that the U_{CTRW} and D_{CTRW} are not

equivalent to U_{ADE} and D_{ADE} except under special cases. $\tilde{M}(p) = \bar{t}p \frac{\tilde{\psi}(p)}{1-\tilde{\psi}(p)}$ represents the memory function where $\tilde{\psi}(p)$ is the transition rate probability. The memory function connects the micro-scale physics, e.g., the variation of the velocity field, to macro-scale non-Fickian transport phenomena. This is because $\tilde{\psi}(p)$ represents micro-scale particle movements dependent on the probability density function of the velocity field; it can also be viewed as the origin for the non-Fickian transport.

The CTRW framework has been extensively applied to various environments including, for example, to characterize non-Fickian transport through porous media (Cortis and Berkowitz, 2004; Dentz et al., 2004), single fractures (Bauget and Fourar, 2008; Wang and Cardenas, 2014), and fracture networks (Berkowitz and Scher, 1998). The cornerstone of CTRW is the determination of $\tilde{\psi}(p)$. Cortis and Berkowitz (2004) considered three models for $\tilde{\psi}(p)$: the asymptotic model, the truncated power law model (TPL), and the modified exponential model. Since the TPL reduces to the asymptotic model when the cut-off time is small, and since the modified exponential model is not suitable for this study because it primarily describes a random molecular diffusion system, the TPL was exclusively used here to model non-Fickian solute transport.

The TPL model is described by:

$$\tilde{\psi}(p) = (1 + \tau_2 p t_1)^\beta \exp(t_1 p) \frac{\Gamma(-\beta, \tau_2^{-1} + t_1 p)}{\Gamma(-\beta, \tau_2^{-1})} \quad (3)$$

where t_1 is the time for the onset of the power-law, t_2 is cut-off or truncation time corresponding to when large-scale Fickian behavior begins or dominates, $\tau_2 = t_2/t_1$, $\Gamma()$ is the

incomplete Gamma function, and β represents the degree of deviation from Fickian behavior (Berkowitz et al., 2006; Cortis and Berkowitz, 2005; Dentz et al., 2004). The transport regimes can be divided into (Berkowitz et al., 2006): (1) strongly non-Fickian transport where $0 < \beta < 1$, (2) moderate non-Fickian transport where $1 < \beta < 2$, and (3) Fickian transport where $\beta \geq 2$ and the CTRW-TPL converges to the ADE.

The CTRW-TPL potentially allows for capturing the transport process from non-Fickian (pre-asymptotic) to Fickian (asymptotic) regimes while using consistent or scale-invariant parameters, including the truncation time. However, recent studies highlight the non-uniqueness of $\tilde{\psi}(p)$ during inverse modeling based on breakthrough curves (Hansen and Berkowitz, 2014), thus requiring a careful manipulation of initial guesses of parameters and assessment of the rationality of results calculated from the CTRW-TPL (Aubeneau et al., 2014; Bauget and Fourar, 2008; Wang and Cardenas, 2014).

3. Methods

In this section, we first discuss the generation of the fractures and the calculation of flow and transport fields within these fractures. This is followed by the methods we used for analyzing the resulting fields.

3.1 Numerical simulations

3.1.1 Two-dimensional flow modeling through synthetic fractures

Synthetic self-affine fractures with increasing lengths were generated through the program SynFrac (Figure 1), which has been shown to reproduce the aperture distribution of real fractures (Ogilvie et al., 2006). The synthetic fractures were prescribed with the same arithmetic mean $\langle b \rangle$, standard deviation σ_b ($\sim 0.49 \langle b \rangle$), and correlation length λ ($= 20 \langle b \rangle$),

where b is aperture field. The smallest fracture size we considered was 50×50 with b defined over a regular grid of 100×100 points over this fracture area. We increased the total fracture length L in increments of 100 until the maximum of 1000, i.e., the largest fracture size is 1000×50 with 2000×100 points (Figure 1).

Note that with increasing fracture scales, the transverse mixing and spreading maybe limited by the fracture lateral boundaries. The idealized case would be to maintain the domain aspect ratio by increasing fracture width proportionally with increasing fracture length, but this drastically increases the computational burden and requires resources that are beyond our current computational capability. Moreover, simply increasing fracture length instead of width is consistent with the CTRW theory and its previous classic validation studies using column experiments (Cortis and Berkowitz, 2004). That is, the focus in this study agrees with the central idea of CTRW of representing transitional transport behavior in the longitudinal direction.

A predictive model for non-Fickian transport requires relating β in the framework of CTRW to fracture roughness; consequently, we generated synthetic fractures with aperture field following both normal (Figure 2) and log-normal (Figure 3) distributions following random fracture generation method (Brush and Thomson, 2003), because these types of distributions are typically observed in natural fractures (Berkowitz, 2002). The correlation length λ increases from $10 \sim 100$ with fixed L (200), and $\sigma_b/\langle b \rangle$ varies with a broad range $0 \sim 2$ by changing the magnitude of σ_b or $\sigma_{\log(b)}$ while keeping $\langle b \rangle$ or $\langle \log(b) \rangle$ constant. The small fraction of contact areas ($b = 0$) when $\sigma_b/\langle b \rangle = 2$ was artificially increased to be a constant ($b = \langle b \rangle / 100$). The variability of $\sigma_b/\langle b \rangle$ corresponds to that used in previous studies (Brush and Thomson, 2003; Mourzenko et al., 1995). We then calculated the flow field through solving the

modified Local Cubic Law and solute transport by implementing a particle tracking algorithm.

For computational ease and efficiency, the Local Cubic Law (LCL), which is a simplified version of the Navier-Stokes equation, has been extensively applied to simulate vertically- or aperture-integrated flow fields (Detwiler et al., 2002; Mourzenko et al., 1995). Though the LCL offers computational advantages, it always overestimates volumetric flow rates (Brush and Thomson, 2003; Wang et al., 2015; Zimmerman et al., 2004). To circumvent the shortcomings of the LCL, here we used a recently developed modified LCL (MLCL) to simulate the 2-D flow fields (Wang et al., 2015), where flow was driven by a pressure gradient in the longitudinal direction, and no-flux was applied for the other two transverse boundaries. The MLCL accounts for fracture roughness, tortuosity, and weak inertial force and is described as:

$$\nabla \cdot \left[\frac{T_x}{M} \frac{\partial p}{\partial x} \cos(\phi_x) \vec{i} + \frac{T_y}{M} \frac{\partial p}{\partial y} \cos(\phi_y) \vec{j} \right] = 0 \quad (4)$$

where T is transmissivity, p is pressure, ϕ is the flow orientation angle estimated from the definition of local fracture tortuosity, and M is a correction coefficient that accounts for local fracture roughness and weak inertial force which have been previously calculated and tabulated from results of equivalent-flux models of flow through wedges. Details of the MLCL development and formulation are available in Wang et al. (2015) where it has also been shown that solutions based on the MLCL are hydraulically equivalent to that from directly solving the Navier-Stokes equations for 3-D fractures. The resultant 2-D MLCL-based velocity field served as input for particle tracking simulations for solute transport.

3.1.2. Quasi-3-D solute transport modeling through particle tracking

A quasi-3-D particle tracking random walk (PTRW) algorithm was used here to model transport through the rough fractures, where the advection and diffusion of particles occur in

the horizontal direction, while only diffusion occurs in the vertical (i.e., z) direction (James et al., 2005; Zheng et al., 2009). However, the exclusion of advection in the z direction can be complemented by including up/down movements according to the mean flow direction. Moreover, the number of particles released at the inlet boundary is proportional to the local flow rate, i.e., an idealized parabolic velocity profile. In the particle tracking method, the no-flux condition is equivalent to an elastic reflection when a particle rebounds when it encounters the lower, upper, and two lateral walls. In order to balance solution accuracy with a small time step and computational efficiency with a large time step, we applied an adaptive time stepping approach such that these two competing objectives can be achieved simultaneously as explained in our previous study (Wang and Cardenas, 2015).

The details of our PTRW approach and its accuracy compared to direct numerical simulations were described in Wang and Cardenas (2015); our previous study showed that the PTRW using a vertically-integrated 2-D flow field solved through the MLCL accurately simulated solute transport through 3-D rough and tortuous fractures (Wang and Cardenas, 2015), while the PTRW with the conventional LCL may miss the overall transport behavior. Therefore, we take advantage of our accurate and efficient approach and implemented the PTRW with MLCL by instantaneously releasing about $N = 1 \times 10^4$ particles at the inlet of increasingly larger fractures. Simulation results were insensitive to the N as we further increased N (results were not shown here). The modeling was implemented through MATLAB using parallel computing. The forward transport modeling took 1 week $\sim$ 3 months to accomplish, where simulation time depended on fracture scale, on a workstation with 16 processor cores and 80 Gb shared memory. The PTRW made computation of BTCs possible even for the largest fracture; this

would have been prohibitive with direct numerical simulation, i.e., solving the 3-D Navier-Stokes equations and the advection-diffusion-equation. In our simulations, the Peclet number ($P_e = \langle u \rangle \langle b \rangle / D_m$) ranges from 5.7 to 10.6 based on Table 1, where $\langle u \rangle$ is mean flow velocity, $\langle b \rangle$ is arithmetic mean aperture, and D_m is the diffusion coefficient. The transport corresponds to the macrodispersion regime as defined by Detwiler et al. (2000). This study was designed to eliminate strong Peclet number-dependence and thus exclusively identified the roughness' effect on transport process across different length scales.

3.2. Flow and solute transport analysis

3.2.1. Statistical analysis of the flow field

Once the velocity field was computed through the MLCL, we calculated its λ by fitting an exponential variogram model with nugget effect to the experimental variogram. The typical exponential variogram $\gamma(h)$ model follows (Li et al., 2010):

$$\gamma(h) = A \left[1 - \exp\left(-\frac{h}{\lambda}\right) \right] + \gamma_0 \quad (5)$$

where γ_0 is the nugget, A is the sill, h is the lag distance.

The spatial pattern of acceleration (a_t) over time was estimated based on the ratio of velocity difference to the travel time of individual particles:

$$a_t = \frac{u_{n+1} - u_n}{t_{n+1} - t_n} \quad (6)$$

where n represents the current time step, and $n+1$ is future time step. The acceleration was further used to analyze the probability density of its normalized standard deviation.

3.2.2. Calculation of breakthrough curves and residence time distributions

To determine breakthrough curves, we continuously recorded the cumulative number of particles ($N_a(t)$) and arrival time at the outlet boundary. Dividing $N_a(t)$ by N yielded the cumulative fraction of particles (i.e., C) over time:

$$C(t) = \frac{N_a(t)}{N} \quad (7)$$

where $C(t)$ represents the ‘real’ or ‘experimental’ breakthrough curves (BTCs).

Residence time distributions (RTDs) provide an additional metric for examining non-Fickian transport features: early arrival and heavy tailing. The RTD is calculated by taking the time derivative of the BTC.

3.2.3. Inverse modeling of breakthrough curves

Here, we employed the 1-D ADE and CTRW-TPL models to analyze the transport process transitioning from non-Fickian to Fickian regimes, and to establish a predictive model based on the physical properties of fractures. Two parameters (U_{ADE} and D_{ADE}) in the ADE, and five parameters (U_{TPL} , D_{TPL} , t_1 , t_2 , and β) in the CTRW-TPL were estimated through the 1-D inverse modeling module in the CTRW toolbox (Cortis and Berkowitz, 2005). Each BTC was analyzed independently. That is, no two or more BTCs were fit simultaneously.

The initial guess for unknowns in the ADE model were assumed to be constant, and the inversion was insensitive to the initial input. However, the initial guesses have a large impact in the inversion of the CTRW-TPL because the CTRW toolbox can easily arrive at a local error minimum rather than the global minimum. This potentially leads to parameter non-uniqueness and identifiability issues with the CTRW-TPL. We therefore not only iteratively altered the initial guesses based on the parameters from the ADE model to find the global minimum error, but also maintained the same initial guesses across increasing scales to minimize this non-

uniqueness issue.

The root mean square error (E) is used as a metric to determine the goodness-of-fit of the curve fitting to the BTCs. The E is:

$$E_i = \sqrt{\sum_{n=1}^N (C_{1D}^i - C_n)^2} \quad (8)$$

where N is the number of particles in the BTCs, C_{1D}^i represents the 1D BTCs fitted through the ADE and TPL models, where i signifies either the ADE or the TPL, and C is from the particle tracking. Errors from the ADE and TPL models are denoted as E_{ADE} and E_{TPL} .

4. Results and Discussions

4.1. Flow field spatial and statistical structure for fractures with increasing scales

Non-Fickian transport fundamentally results from the spatial structure of the velocity field (Heidari and Li, 2014; Kang et al., 2014). And for fractures these are determined by the fracture roughness ($\sigma_b/\langle b \rangle$) and the aperture correlation length λ . For fractures with increasing scales, we eliminated the effects of these factors by using a fixed $\sigma_b/\langle b \rangle$ and aperture λ . Thus, the velocity field structure should also remain similar. As expected, the velocity variograms showed a well-defined sill particularly for fractures with $L/\lambda > 5$. The λ of the velocity field consistently agreed with that prescribed for the aperture field ($\sim 20 \langle b \rangle$) (Figure 4a), indicating that the 2-D velocity fields were determined by the correlated aperture fields, a result one expects.

The statistical distributions (PDFs) of the particle velocities were also consistent for all fractures (Figure 4b). This is because $\langle b \rangle$ and σ_b , or roughness, was kept constant. The velocity PDFs exhibited a sharp and narrow peak about zero acceleration and two broad power-law tails;

these features comprise a type of transition (intermittent) velocity structure (de Anna et al., 2013; Kang et al., 2014). The sharp peak about zero acceleration suggests that the particles would most likely encounter stagnant or low velocity zones, and long power-law tails indicate the likelihood of encountering velocity jumps from low to high values, or vice versa. Taken together, these illustrate the degree and nature of the flow and transport heterogeneity. Note that some transition velocity structures inherently control the temporal transition structure and thus generate non-Fickian transport behaviors (de Anna et al., 2013; Kang et al., 2014).

4.2. Transport progression from Non-Fickian to Fickian regimes with increasing scales

Early arrival and long tailing are two typical features of non-Fickian transport; these are clearly shown for fractures with $L/\lambda < 30$ (Figures 5a-5d). Perfect Fickian transport corresponds to an “S”-shaped curve centered about 1 pore volume (PV), where/when the normalized solute concentration $C' = 0.5$ is expected to be observed. This was not the case for most of the studied fractures. The experimental BTCs showed the progression of non-Fickian to Fickian transport as fracture scale longitudinally, i.e., the early arrivals and long tailing gradually disappeared at a log-log scale (Figures 5b and 5d). This is also demonstrated by the observation at a linear scale, since the expectation that $C' = 0.5$ at dimensionless time = 1 PV was progressively satisfied and the ADE became more valid for fitting BTCs as fracture scale increased (Figures 5a-5b).

The RTDs further confirmed the transport progression from non-Fickian to Fickian with increasing L/λ (Figure 5e). The mode for the RTD was not only < 1 PV for the relatively short fractures, but also increased with increasing L/λ , until this converged to 1 PV when the fracture was long enough which satisfied the Fickian transport condition. Moreover, bimodal RTDs were observed for the relatively short fractures, and these gradually evolved into inverted ‘U’-shaped

curves. That is, the RTDs became more symmetric and tended to follow a Gaussian distribution with increasing fracture scales. Rigorously fitting certain portion of RTDs would yield different power law exponents (Figure 5e). The above-mentioned phenomena provided clear evidence that the early arrival and long tail disappear as fracture length increases.

The computational experiments clearly captured the transition from non-Fickian (pre-asymptotic) to Fickian (asymptotic) regimes in the presence of persistent intermittent velocity structure. These observations are in contrast to the expectation of persistent non-Fickian transport behavior with infinite Peclet number that needs longer time or length for transitioning into Fickian transport. For example, non-Fickian transport was persistently observed even after many hundreds of pore volumes for a porous medium in Kang et al. (2014). The different scaling effect on the earlier transport transition occurrence in this study is due to the inclusion of molecular diffusion in our PTRW algorithm (Berkowitz et al., 2006), which was ignored by Kang et al. (2014). We thus showed that the stationary intermittent velocity field within fractures can lead to Fickian transport once a threshold length scale is surpassed.

The length scale for Fickian transport has been observed numerically through cubic-packed pores (Cardenas, 2009; Zhang and Lv, 2007), and is expected to be dependent on the magnitude of Pe and correlation length based on studies of porous media transport (Salles et al., 1993), but this has been rarely documented in rough fractures. Although the asymptotic time and length scales through fractures for the transport transition can be theoretically predicted through an idealized fracture (i.e., for the parallel plates model (Wang et al., 2012)), this prediction is complicated by fracture roughness which increases this length scale (Wang and Cardenas, 2014). Here, we determined based on visual inspection of the fits and the reduction

of fitting errors (discussed further below) that rough-walled fractures should be at least of $\sim 35\lambda$ long in order to transition to Fickian transport. This factor is greater than a previous experimentally determined value of 20λ by Detwiler et al. (2002); the difference may be largely due to the different Pe , which is fundamentally determined by the roughness of the fractures, considered in previous studies compared to ours. Moreover, this factor is smaller than the theoretical value ($10Pe\lambda$) estimated for porous media (Salles et al., 1993). Future research efforts should systematically explore the combined roles of Pe and aperture λ on controlling the length scale for transport regime transition through fractures.

4.3. Theoretical prediction and assessment of transport transition

The ADE model as expected failed to capture typical non-Fickian transport features (early arrival and long tails) for the relatively short fractures which led to large fitting errors. But the ADE model was gradually able to match the experimental BTCs as L/λ increased (Figures 5a-5b) with concurrent reductions in error (Table 1). The fitted velocity (U_{ADE}) agreed well with mean flow velocity (U), which is calculated as the ratio of L to the PV (Table 1). The fitted D_{ADE} increased initially with increasing L/λ but then decreased with further increases in L/λ (Figure 6 and Table 1). Thus, the inverse modeling analysis with the ADE led to somewhat erratic behavior in D_{ADE} and did not show a clear progression towards the asymptotic regime where D_{ADE} should be stationary.

Compared to the ADE, the CTRW-TPL framework was able to fit the experimental BTCs across scales and transport regimes from non-Fickian to Fickian (Figures 5c-5d, Table 1). The errors remained small except for the shortest fracture with $L/\lambda=2.5$. For all other fracture length cases, the CTRW-TPL showed similar if not smaller errors than the smallest error

achieved with the ADE even in the asymptotic regime. At each scale E_{TPL} is always less than E_{ADE} . However, for the shortest fracture, errors were large for both cases. This is because the ADE and the CTRW-TPL framework completely miss and in fact cannot handle the bimodal RTDs (Figure 5e). Unlike the ADE which would require adaption to include an integro-differential term to model the progression of non-Fickian to Fickian transport, the CTRW-TPL reproduced the bulk transport behavior with a more or less consistent set of parameters that was insensitive to the scales (Table 1), because the exponential function dominates over the power law at large scales as shown in equation (3). That is, the power law plays a nominal role even though it may be still applicable for certain portion of RTDs with varying power law exponents (Figure 5e).

Non-uniqueness during the inversion process can be a problem with CTRW (Hansen and Berkowitz, 2014), especially when transport converges to the Fickian regime. However, here we observed the stabilization of β and t_2 for fractures with increasing L/λ (Figure 6) across Fickian and non-Fickian transport regimes given that fracture roughness remains constant. In our cases, the CTRW-TPL framework was sensitive to these two parameters and less so to t_1 , since the latter changed a little bit with increasing scales but remained small compared to t_2 . β fluctuated slightly about ~ 1.13 . Moreover, t_2 converged and stabilized quickly to around $10^{3.4}$ s (disregarding the fracture $L/\lambda=2.5$). Thus the β and t_2 that resulted from the inverse modeling are truly reflective of the local micro-scale processes and is insensitive to the domain size. This implies that incorporation of transport process at one scale may improve the prediction of β at other scales if fracture roughness is stationary. Moreover it showed that β may be insensitive to the fracture correlation length as we previously showed for 2-D fractures (Wang and Cardenas,

2014). Based on the magnitude of β , a moderate non-Fickian transport was observed; this indicated that the transport velocity is constant and that the dispersion coefficient scales as a power law over time. Therefore, U_{TPL} remains fairly constant but D_{TPL} varies with increasing L/λ for our numerical results (Table 1).

4.4. Impact of fracture roughness and aperture correlation length on effective solute transport velocity

Fluid velocity is known to be different from transport velocity in fractures (Silliman, 1989; Tsang, 1992; Zheng et al., 2008). Depending on the anisotropy of the aperture field, transport velocity could be either greater or smaller than fluid velocity if flow direction is parallel to or perpendicular to the aperture variations, respectively (Silliman, 1989). For an isotropic case, theoretical analysis indicates that transport velocity is retarded by fracture roughness ($\sigma_b/\langle b \rangle$) compared to fluid velocity, and the retardation coefficient could be solely predicted based on $\sigma_b/\langle b \rangle$ (Silliman, 1989). Our results were consistent with this theory that transport velocity is always less than fluid velocity as our synthetic fractures were isotropic (Figure 7). Unlike previous studies, we found that the deviations between fluid and transport velocities are dependent on both $\sigma_b/\langle b \rangle$ and λ . For short-correlation fractures, the ratio of fluid velocity to transport velocity increases exponentially with $\sigma_b/\langle b \rangle$. However, this incremental rate of velocity ratio with respect to $\sigma_b/\langle b \rangle$ decreases as λ/L increases, and eventually become insensitive to $\sigma_b/\langle b \rangle$ especially when $\lambda/L \geq 0.8$ (Figure 7). This is most likely due to the fact that the aperture field with a large λ/L tends to create preferential paths, and these fast flow paths are spatially invariant across a broad range of $\sigma_b/\langle b \rangle$. Future research could explore a quantitative relationship between velocity ratio and $\sigma_b/\langle b \rangle$ and λ/L , such that the effective

transport velocity can be empirically predicted based on fluid velocity.

4.5. Dependence of non-Fickian magnitude on 3-D fracture heterogeneity

Non-Fickian transport can be attributed to incomplete local mixing or to mixing failing to reach time and length scales that satisfy Fickian dispersion theory (Dentz et al., 2011; Le Borgne et al., 2011). Rough fracture walls (i.e., $\sigma_b/\langle b \rangle$) create immobile zones which induce incomplete local mixing and lengthen the residence time of solutes. Moreover, λ affects the length scales that satisfy Fickian dispersion as discussed in section 4.3. In any case, non-Fickian transport through 2-D fractures can be accurately captured by the CTRW-TPL model (Wang and Cardenas, 2014), where the degree of deviation of non-Fickian from Fickian transport can be represented by β .

Our previous study on non-Fickian transport through 2-D fractures showed that β is proportional to $\sigma_b/\langle b \rangle$, but it is insensitive to λ/L (Wang and Cardenas, 2014). Yet, the prediction of non-Fickian transport through 3-D fractures remains elusive particularly with respect to $\sigma_b/\langle b \rangle$ and λ . To this end, we inversely estimated β based on the “experimental” breakthrough curves through 3-D rough and tortuous fractures, where aperture fields follow either normal (Figure 2) or log-normal distributions (Figure 3). Similar to transport behavior through 2-D fractures, for 3-D cases, β is found to be linearly proportional to $\sigma_b/\langle b \rangle$ despite of variability in λ/L (Figure 8). Note that the slope and intercept of the regressed linear equation are different from that for 2-D cases. This might be due to additional transverse dispersion occurring in 3-D rough fractures. Specifically, β in a 3-D case was observed to be smaller than that in a 2-D case for given $\sigma_b/\langle b \rangle$, and resulted in non-Fickian transport with relatively heavier tailing. Moreover, β decreased rapidly when $\sigma_b/\langle b \rangle < 0.1$ ($\beta = 2$ for parallel plates since Fickian

transport occurs within a relatively short length) which was not observed in a 2-D case (Figure 8). This shows that a small change in $\sigma_b/\langle b \rangle$ may significantly change the transverse dispersion process which in turn contributes to longer solute residence time through 3-D fractures.

As expected, β is more sensitive to $\sigma_b/\langle b \rangle$ rather than λ for both 2-D (Wang and Cardenas, 2014) and 3-D (Figure 8) fractures. This is because β is naturally a scale-insensitive parameter, as discussed in the section 4.3. As long as L/λ is greater than 2.5, which is when β becomes stable (Figure 6), the variation of λ will not significantly affect β (Figure 8). Our study further showed the robustness and applicability of CTRW-TPL model for predicting non-Fickian transport by directly linking it to measurable physical properties of fractures across different scales.

4.6. Limitations of current study

Although the ability of the CTRW for predicting the transport transition from non-Fickian to Fickian with increasing scales has been validated here with measurable properties of single fractures, the scaling effects on transport behavior in a fractured system could be further complicated by fracture density and orientations; these present an additional challenge in characterizing non-Fickian transport at increasing scales (Berkowitz, 2002; Brown, 1987; Neuman, 2005). Moreover, our analysis takes into account only the effect of the longitudinal scale of transport. Ideally, the fractures can be made wider in the transverse direction as the longitudinal L is increased. Thus, our analysis assumes that the transverse length is sufficiently large such that it does not add additional pre-asymptotic mixing. Given that β apparently stabilizes at $L/\lambda > 2.5$, this assumption is reasonable since asymptotic mixing in the transverse direction for a solute released uniformly is typically achieved even sooner.

The effect of inertial force (i.e., advective flux) on transport behavior was not investigated here. Increasing inertial force not only leads to the failure of Darcy' law in describing fluid flow (Zhou et al., 2016; Zimmerman et al., 2004), but also results in the failure of the ADE in describing transport, which can be attributed to the growth of the immobile zones as shown by 2-D studies (Bolster et al., 2014; Cardenas et al., 2009). Unlike 2-D cases, recent experimental results further showed a shortened tailing with increasing Reynolds number (Re) within laterally extensive 3-D rough fractures (Lee et al., 2015). However, transport across increasing Re in natural 3-D rough fractures is still poorly understood. Therefore, appropriate prediction of non-Fickian behavior across a broad range of Re remains an open challenge. The aspects mentioned above are beyond the scope of the current study.

5. Summary and Conclusions

Flow and transport through fractures is complex with seemingly ubiquitous non-Fickian transport. In this study, we first illustrated the progression of transport from pre-asymptotic (non-Fickian) to asymptotic (Fickian) transport through 3-D rough-walled synthetic fractures by modeling experiments conducted at incrementally increasing scales but where fracture roughness and aperture correlation length remained constant. The solute transport was simulated through a quasi-3-D particle tracking random walk algorithm with a 2-D flow field calculated through a modified local cubic law. This resulted in a series of experimental breakthrough curves corresponding to each fracture scale. The solute breakthrough curves and residence time distributions clearly showed the transition from non-Fickian to Fickian transport in the presence of an intermittent velocity structure; the transport transition is expected to occur later when molecular diffusion is ignored. The Fickian advection-dispersion equation

expectedly failed to capture transport for the relatively short fractures and resulted in erratic estimates for the dispersion coefficient. Analysis of the breakthrough curves with the continuous time random walk (CTRW) with truncated power law resulted in a set of transport parameters that were insensitive to fracture scale, even for the shorter fractures and through the transport regime transition. Moreover, we implemented additional particle tracking experiments through fractures with varying roughness and correlation length, where aperture fields follow either normal or log-normal distributions. We found that the parameter β in the CTRW model is linearly dependent on fracture roughness but is insensitive to aperture correlation length. Therefore, the CTRW model parameters determined from measurable physical properties of fractures at various scales can be used for transport prediction at other scales.

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Tables

Table 1. Curve fitting parameters with corresponding fitting errors (E) for fractures with increasing scales (as shown in Figure 1) through two inverse models: the advection-dispersion equation (ADE), and continuous time random walk with truncated power law (TPL). U is velocity, D is dispersion coefficient, t_1 is onset of power law, and t_2 is the truncation time after when Fickian transport dominates. Moreover, mean flow velocity (U) was computed following $U=L/PV$, where L is total length of fractures as shown in Figure 1, PV denotes pore volume.

Relative length L/λ	U_{ADE} $\times 10^{-5}$ [m/s]	D_{ADE} $\times 10^{-7}$ [m ² /s]	E_{ADE} [-]	$U=L/PV$ $\times 10^{-5}$ [m/s]	U_{TPL} $\times 10^{-4}$ [m/s]	D_{TPL} $\times 10^{-7}$ [m ² /s]	β [-]	t_1 [s]	t_2 [s]	E_{TPL} [-]
2.5	2.28	1.10	3.37	3.19	0.38	1.80	1.30	$10^{-1.63}$	$10^{-0.11}$	3.37
5.0	4.23	1.65	1.56	4.41	2.91	0.22	1.00	$10^{-0.20}$	$10^{3.33}$	0.54
10	4.25	1.44	2.05	4.23	1.87	0.29	1.15	$10^{-0.21}$	$10^{3.61}$	0.25
15	3.39	1.98	0.75	3.37	1.49	3.31	1.14	$10^{-0.06}$	$10^{3.49}$	0.47
20	3.93	2.58	0.71	3.97	1.74	6.39	1.17	$10^{-0.73}$	$10^{3.53}$	0.73
25	3.99	2.57	2.48	3.76	1.99	0.71	1.14	$10^{-0.36}$	$10^{3.89}$	0.36
30	3.79	1.55	1.51	3.65	1.62	0.70	1.17	$10^{-0.44}$	$10^{3.68}$	0.46
35	3.95	1.30	0.91	4.09	1.76	0.89	1.14	$10^{-0.46}$	$10^{3.25}$	0.22
50	3.03	1.04	0.86	3.08	1.52	1.09	1.12	$10^{-0.69}$	$10^{3.36}$	0.19

Figure captions

Figure 1. Normally distributed synthetic aperture fields used in this study. The fractures have the same arithmetic mean $\langle b \rangle$, standard deviation of aperture field b , and correlation length λ across varying fracture total length L . The fractures were generated through the SynFrac program (Ogilvie et al., 2006).

Figure 2. Examples of fractures with aperture field following normal distribution with different roughness ($\sigma_b/\langle b \rangle$) and correlation length (λ), where b is apparent aperture with standard deviation σ_b of aperture and arithmetic mean aperture $\langle b \rangle$, L is the total length of fractures.

Figure 3. Examples of fractures with aperture field following log-normal distribution with different roughness ($\sigma_b/\langle b \rangle$) and correlation length (λ), where b is apparent aperture with standard deviation σ_b of aperture and arithmetic mean aperture $\langle b \rangle$, L is the total length of fractures.

Figure 4. Statistical properties of the flow field for fractures with different length. (a) Experimental variogram $\gamma(h)$ for the longitudinal velocity field for the studied fractures; h is lag distance and $\langle b \rangle$ is the arithmetic mean of aperture field b . (b) Probability distribution function (PDF) of normalized Lagrangian acceleration of particles in the x direction ($a_t(x)$), where a_t is acceleration with its standard deviation (σ_{at}), L is total length, and λ is the correlation length of aperture field.

Figure 5. Experimental breakthrough curves (BTCs) generated from particle tracking (hollow circles), and inverse or fitted BTCs from solving the advection-dispersion equation (a-b, solid lines) and continuous random walk (c-d, solid lines) for fractures with increasing scales (total length L /correlation length λ). Each BTC group was shifted by 1 pore volume τ with each incremental increase in scale for visual purposes in a) and c). The corresponding residence time distributions (RTDs) are shown in (e) with the fitted power law shown as black lines. C' represents normalized concentration.

Figure 6. The scale-dependence of transport parameters: 1-D ADE dispersion coefficient (D_{ADE}), CTRW-TPL β , and CTRW-TPL truncation time t_2 .

Figure 7. The ratio of fluid velocity (V_{Fluid}) to effective transport velocity ($V_{Transport}$) is a function of fracture roughness ($\sigma_b/\langle b \rangle$) and aperture correlation length (λ), where $V_{Transport}$ is obtained by fitting breakthrough curves to the advection-dispersion equation, b is apparent aperture with standard deviation σ_b of aperture and arithmetic mean aperture $\langle b \rangle$, L is the total length of fracture.

Figure 8. Dependence of parameter β in the CTRW-TPL model on fracture roughness ($\sigma_b/\langle b \rangle$), where b is apparent aperture with standard deviation σ_b of aperture and arithmetic mean aperture $\langle b \rangle$, L is the total length of studied fracture. Color represents different aperture correlation length (λ) following normal and log-normal distributions as shown by hollow circle

701 and asterisk, respectively, filled blue circle refers to the ideal parallel plates model. The black
702 line is the linear fit for β with $\sigma_b/\langle b \rangle$.

Figure 1. Normally distributed synthetic aperture fields used in this study. The fractures have the same arithmetic mean $\langle b \rangle$, standard deviation of aperture b , and correlation length λ across varying fracture total length L . The fractures were generated through the SynFrac program [Ogilvie *et al.*, 2006].

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Figure 5. Breakthrough curves (BTCs) generated from particle tracking random walk (hollow circles), and inverse BTCs from solving the advection-dispersion equation (5a-5b, solid curves) and continuous random walk (5c-5d, solid curves) for fractures with increasing scales (total length L / correlation length λ). Each BTC group was shifted by 1 pore volume τ with each incremental increase in scale in 5a and 5c. The corresponding residence time distributions (RTD) are shown in 5e with fitted power law shown as black lines. C' represents normalized concentration.

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Figure 8. Dependence of parameter β in the TPL model on fracture roughness ($\sigma_b / \langle b \rangle$), where b is apparent aperture with standard deviation σ_b of aperture and arithmetic mean aperture $\langle b \rangle$, L is the total length of studied fracture. Color represents different aperture correlation length (λ) following normal and log-normal distributions as shown by hollow circle and asterisk, respectively, filled blue circle refers to the ideal parallel plates model. The black line is the linear fit for β with $\sigma_b / \langle b \rangle$.