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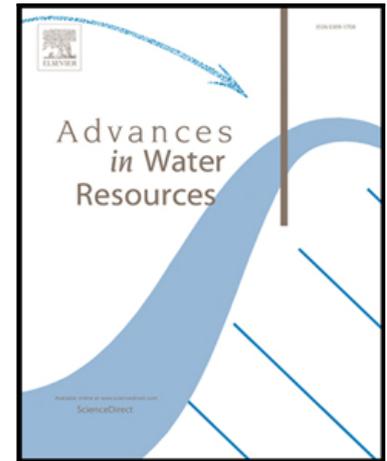
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Highlights

- Injection modes have major impact on anomalous transport in DFNs
- Evolution of the Lagrangian velocity distribution is governed by injection modes
- Spatial velocity Markov model for variable injection modes
- Equivalence between spatial Markov model and Boltzmann equation

Anomalous transport in disordered fracture networks:
spatial Markov model for dispersion with variable
injection modes

Peter K. Kang

Korea Institute of Science and Technology, Seoul 02792, Republic of Korea

*Massachusetts Institute of Technology, 77 Massachusetts Ave, Building 1, Cambridge,
Massachusetts 02139, USA*

Marco Dentz

*Institute of Environmental Assessment and Water Research (IDEA), Spanish National
Research Council (CSIC), 08034 Barcelona, Spain*

Tanguy Le Borgne

Université de Rennes 1, CNRS, Geosciences Rennes, UMR 6118, Rennes, France

Seunghak Lee

Korea Institute of Science and Technology, Seoul 02792, Republic of Korea

Ruben Juanes

*Massachusetts Institute of Technology, 77 Massachusetts Ave, Building 1, Cambridge,
Massachusetts 02139, USA*

Abstract

We investigate tracer transport on random discrete fracture networks that are characterized by the statistics of the fracture geometry and hydraulic conductivity. While it is well known that tracer transport through fractured media can be anomalous and particle injection modes can have major impact on dispersion, the incorporation of injection modes into effective transport modelling has remained an open issue. The fundamental reason behind

this challenge is that—even if the Eulerian fluid velocity is steady—the Lagrangian velocity distribution experienced by tracer particles evolves with time from its initial distribution, which is dictated by the injection mode, to a stationary velocity distribution. We quantify this evolution by a Markov model for particle velocities that are equidistantly sampled along trajectories. This stochastic approach allows for the systematic incorporation of the initial velocity distribution and quantifies the interplay between velocity distribution and spatial and temporal correlation. The proposed spatial Markov model is characterized by the initial velocity distribution, which is determined by the particle injection mode, the stationary Lagrangian velocity distribution, which is derived from the Eulerian velocity distribution, and the spatial velocity correlation length, which is related to the characteristic fracture length. This effective model leads to a time-domain random walk for the evolution of particle positions and velocities, whose joint distribution follows a Boltzmann equation. Finally, we demonstrate that the proposed model can successfully predict anomalous transport through discrete fracture networks with different levels of heterogeneity and arbitrary tracer injection modes.

Keywords: Discrete Fracture Networks, Injection Modes, Anomalous Transport, Stochastic Modelling, Lagrangian Velocity, Time Domain Random Walks, Continuous Time Random Walks, Spatial Markov Model

1. Introduction

Flow and transport in fractured geologic media control many important natural and engineered processes, including nuclear waste disposal, ge-

4 ologic carbon sequestration, groundwater contamination, managed aquifer
5 recharge, and geothermal production in fractured geologic media [e.g., 1,
6 2, 3, 4, 5]. Two dominant approaches exist for simulating flow and trans-
7 port through fractured media: the equivalent porous medium approach [6, 7]
8 and the discrete fracture network approach (DFN) [8, 9, 10, 11, 12, 13, 14,
9 15, 16, 17, 18, 19, 20, 21]. The DFN approach explicitly resolves individ-
10 ual fractures whereas the equivalent porous medium approach represents
11 the fractured medium as a single continuum by deriving effective parame-
12 ters to include the effect of the fractures on the flow and transport. The
13 latter, however, is hampered by the fact that a representative elementary
14 volume may not exist for fractured media [22, 23]. Dual-porosity mod-
15 els are in between these two approaches, and conceptualize the fractured-
16 porous medium as two overlapping continua, which interact via an exchange
17 term [24, 25, 26, 27, 28, 29, 30, 31, 32].

18 DFN modelling has advanced significantly in recent years with the in-
19 crease in computational power. Current DFN simulators can take into ac-
20 count multiple physical mechanisms occurring in complex 3D fracture sys-
21 tems. Recent studies also have developed methods to explicitly model ad-
22 vection and diffusion through both the discrete fractures and the permeable
23 rock matrix [33, 34, 35, 36]. In practice, however, their application must
24 account for the uncertainty in the subsurface characterization of fractured
25 media, which is still an considerable challenge [37, 38, 39]. Thus, there is a
26 continued interest in the development of upscaled transport models that can
27 be parameterized with a small number of model parameters. Ideally, these
28 model parameters should have a clear physical interpretation and should be

29 determined by means of field experiments, with the expectation that the
30 model can then be used for predictive purposes [40, 41].

31 Developing an upscaled model for transport in fractured media is espe-
32 cially challenging due to the emergence of anomalous (non-Fickian) trans-
33 port. While particle spreading is often described using a Fickian framework,
34 anomalous transport—characterized by scale-dependent spreading, early ar-
35 rivals, long tails, and nonlinear scaling with time of the centered mean
36 square displacement—has been widely observed in porous and fractured
37 media across multiple scales, from pore [42, 43, 44, 45, 46] to single frac-
38 ture [47, 48, 49, 50] to column [51, 52] to field scale [53, 54, 55, 56, 57, 41, 58].
39 The ability to predict anomalous transport is essential because it leads to fun-
40 damentally different behavior compared with Fickian transport [59, 60, 61].

41 The continuous time random walk (CTRW) formalism [62, 63] is a frame-
42 work to describe anomalous transport through which models particle motion
43 through a random walk in space and time characterized by random space
44 and time increments, which accounts for variable mass transfer rates due
45 to spatial heterogeneity. It has been used to model transport in heteroge-
46 neous porous and fractured media [64, 65, 66, 67, 68, 49, 69] and allows
47 incorporating information on flow heterogeneity and medium geometry for
48 large scale transport modelling. Similarly, the time-domain random walk
49 (TDRW) approach [70, 71, 72] models particle motion due to distributed
50 space and time increments, which are derived from particle velocities and
51 their correlations. The analysis of particle motion in heterogeneous flow
52 fields demonstrate that Lagrangian particle velocities exhibit sustained cor-
53 relation along their trajectory [73, 70, 74, 75, 76, 77, 78, 79, 45]. Volume

54 conservation induces correlation in the Eulerian velocity field because fluxes
55 must satisfy the divergence-free constraint. This, in turn, induces correlation
56 in the Lagrangian velocity along a particle trajectory. To take into account
57 velocity correlation, Lagrangian models based on Markovian processes have
58 been proposed [70, 71, 74, 76, 77, 78, 45, 41, 80]. Spatial Markov models
59 are based on the observation that successive velocity transitions measured
60 equidistantly along the mean flow direction exhibit Markovianity: a parti-
61 cle's velocity at the next step is fully determined by its current velocity. The
62 spatial Markov model, which accounts for velocity correlation by incorporat-
63 ing this one-step velocity correlation information, has not yet been extended
64 to disordered (unstructured) DFNs.

65 The mode of particle injection can have a major impact on transport
66 through porous and fractured media [81, 82, 83, 84, 85, 86, 58, 87]. Two
67 generic injection modes are uniform (resident) injection and flux-weighted
68 injection with distinctive physical meanings as discussed in Frampton and
69 Cvetkovic [84]. The work by Sposito and Dagan [82] is one of the earliest
70 studies of the impact of different particle injection modes on the time evolu-
71 tion of a solute plume spatial moments. The significance of injection modes
72 on particle transport through discrete fracture networks has been studied for
73 fractured media [84, 58]. Dagan [87] recently clarified the theoretical relation
74 between injection modes and plume mean velocity. Despite recent advances
75 regarding the significance of particle injection modes, the incorporation of
76 injection methods into effective transport modelling is still an open issue
77 [84]. The fundamental challenge is that the Lagrangian velocity distribution
78 experienced by tracer particles evolves with time from its initial distribution

79 which is dictated by the injection mode to a stationary velocity distribution
80 [73, 84, 75, 88]. In this paper, we address these fundamental questions, in
81 the context of anomalous transport through disordered DFNs.

82 The paper proceeds as follows. In the next section, we present the studied
83 random discrete fracture networks, the flow and transport equations and
84 details of the different particle injection rules. In Section 3, we investigate the
85 emergence of anomalous transport by direct Monte Carlo simulations of flow
86 and particle transport. In Section 4, we analyze Eulerian and Lagrangian
87 velocity statistics to gain insight into the effective particle dynamics and
88 elucidate the key mechanisms that lead to the observed anomalous behavior.
89 In Section 5, we develop a spatial Markov model that is characterized by the
90 initial velocity distribution, probability density function (PDF) of Lagrangian
91 velocities and their transition PDF, which are derived from the Monte Carlo
92 simulations. The proposed model is in excellent agreement with direct Monte
93 Carlo simulations. We then present a parsimonious spatial Markov model
94 that quantifies velocity correlation with a single parameter. The predictive
95 capabilities of this simplified model are demonstrated by comparison to the
96 direct Monte Carlo simulations with arbitrary injection modes. In Section 6,
97 we summarize the main findings and conclusions.

98 **2. Flow and Transport in Discrete Random Fracture Networks**

99 *2.1. Random Fracture Networks*

100 We numerically generate random DFNs in two-dimensional rectangular
101 regions, and solve for flow and tracer transport within these networks. The
102 fracture networks are composed of linear fractures embedded in an imper-

103 meable rock matrix. The idealized 2D DFN realizations are generated by
 104 superimposing two different sets of fractures, which leads to realistic discrete
 105 fracture networks [89, 67]. Fracture locations, orientations, lengths and hy-
 106 draulic conductivities are generated from predefined distributions, which are
 107 assumed to be statistically independent: (1) Fracture midpoints are selected
 108 randomly over the domain size of $L_x \times L_y$ where $L_x = 2$ and $L_y = 1$; (2) Frac-
 109 ture orientations for two fracture sets are selected randomly from Gaussian
 110 distributions, with means and standard deviation of $0^\circ \pm 5^\circ$ for the first set,
 111 and $90^\circ \pm 5^\circ$ for the second set; (3) Fracture lengths are chosen randomly
 112 from exponential distributions with mean $L_x/10$ for the horizontal fracture
 113 set and mean $L_y/10$ for the vertical fracture set; (4) Fracture conductivities
 114 are assigned randomly from a predefined log-normal distribution. An exam-
 115 ple of a random discrete fracture network with 2000 fractures is shown in
 116 Figure 1.

The position vector of node i in the fracture network is denoted by \mathbf{x}_i .
 The link length between nodes i and j is denoted by l_{ij} . The network is char-
 acterized by the distribution of link lengths $p_l(l)$ and hydraulic conductivity
 K . The PDF of link lengths here is exponential

$$p_l(l) = \frac{\exp(-l/\bar{l})}{\bar{l}}. \quad (1)$$

117 Note that the link length and orientation are independent. The character-
 118 istic fracture link length is obtained by taking the average of a link length
 119 over all the realizations, which gives $\bar{l} \approx L_x/200$. A realization of the ran-
 120 dom discrete fracture network is generated by assigning independent and
 121 identically distributed random hydraulic conductivities $K_{ij} > 0$ to each link
 122 between nodes i and j . Therefore, the K_{ij} values in different links are un-

123 correlated. The set of all realizations of the spatially random network gener-
 124 ated in this way forms a statistical ensemble that is stationary and ergodic.
 125 We assign a lognormal distribution of K values, and study the impact of
 126 conductivity heterogeneity on transport by varying the variance of $\ln(K)$.
 127 We study log-normal conductivity distributions with four different variances:
 128 $\sigma_{\ln K} = 1, 2, 3, 5$. The use of this particular distribution is motivated by the
 129 fact that conductivity values in many natural media can be described by a
 130 lognormal law [90, 91].

131 2.2. Flow Field

132 Steady state flow through the network is modeled by Darcy's law [22] for
 133 the fluid flux u_{ij} between nodes i and j , $u_{ij} = -K_{ij}(\Phi_j - \Phi_i)/l_{ij}$, where Φ_i
 134 and Φ_j are the hydraulic heads at nodes i and j . Imposing flux conservation
 135 at each node i , $\sum_j u_{ij} = 0$ (the summation is over nearest-neighbor nodes),
 136 leads to a linear system of equations, which is solved for the hydraulic heads
 137 at the nodes. The fluid flux through a link from node i to j is termed
 138 incoming for node i if $u_{ij} < 0$, and outgoing if $u_{ij} > 0$. We denote by \mathbf{e}_{ij} the
 139 unit vector in the direction of the link connecting nodes i and j .

We study a uniform flow setting characterized by constant mean flow
 in the positive x -direction parallel to the principal set of fractures. No-flow
 conditions are imposed at the top and bottom boundaries of the domain,
 and fixed hydraulic head at the left ($\Phi = 1$) and right ($\Phi = 0$) boundaries.
 The overbar in the following denotes the ensemble average over all network
 realizations. The one-point statistics of the flow field are characterized by the
 Eulerian velocity PDF, which is obtained by spatial and ensemble sampling

of the velocity magnitudes in the network

$$p_e(u) = \frac{\sum_{i>j} \overline{l_{ij} \delta(u - u_{ij})}}{N_\ell \bar{l}}. \quad (2)$$

where N_ℓ is the number of links in the network. Link length and flow velocities here are independent. Thus, the Eulerian velocity PDF is given by

$$p_e(u) = \frac{1}{N_\ell} \sum_{i>j} \overline{\delta(u - u_{ij})}. \quad (3)$$

140 Even though the underlying conductivity field is uncorrelated, the mass con-
 141 servation constraint together with heterogeneity leads to the formation of
 142 preferential flow paths with increasing network heterogeneity [92, 80]. This
 143 is illustrated in Figures 2a and b, which show maps of the relative velocity
 144 magnitude for high velocities in networks with $\log-K$ variances of 1 and 5. As
 145 shown in Figures 2c and d, for low heterogeneity most small flux values occur
 146 along links perpendicular to the mean flow direction, whereas low flux values
 147 do not show directionality for the high heterogeneity case. This indicates
 148 that fracture geometry dominates small flux values for low heterogeneity and
 149 fracture conductivity dominates small flux values for high heterogeneity. An
 150 increase in conductivity heterogeneity leads to a broader Eulerian velocity
 151 PDF, with significantly larger probability of having small flux values as illus-
 152 trated in Figure 3, which shows $p_e(u)$ for networks of different heterogeneity
 153 strength.

154 2.3. Transport

Once the fluxes at the links have been determined, we simulate transport of a passive tracer by particle tracking. Particles are injected along a line

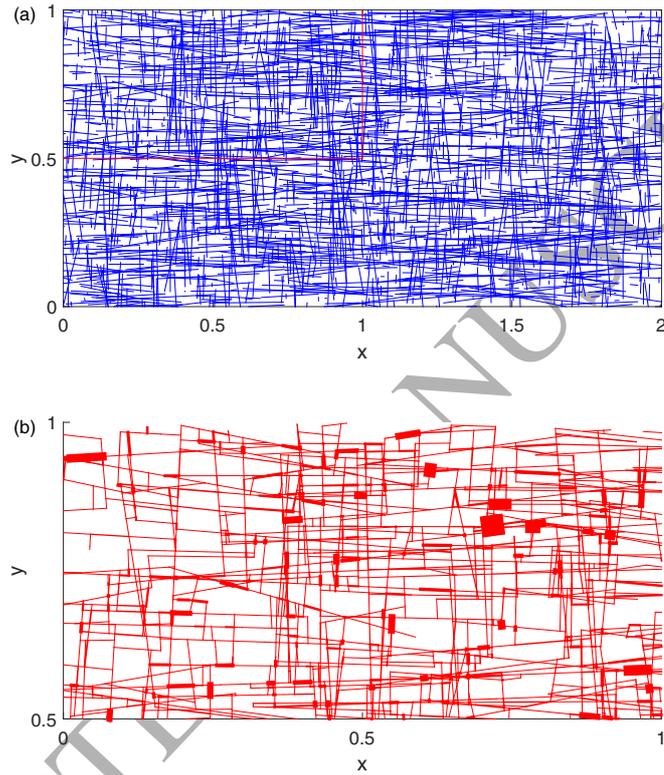


Figure 1: (a) Example of a two-dimensional DFN studied here, with 2000 fractures (1000 fractures for each fracture set). (b) Subsection of a spatially uncorrelated conductivity field between $0 \leq x \leq 1$ and $0.5 \leq y \leq 1$. Conductivity values are assigned from a lognormal distribution with $\sigma_{\ln K} = 1$. Link width is proportional to the conductivity value; only connected links are shown.

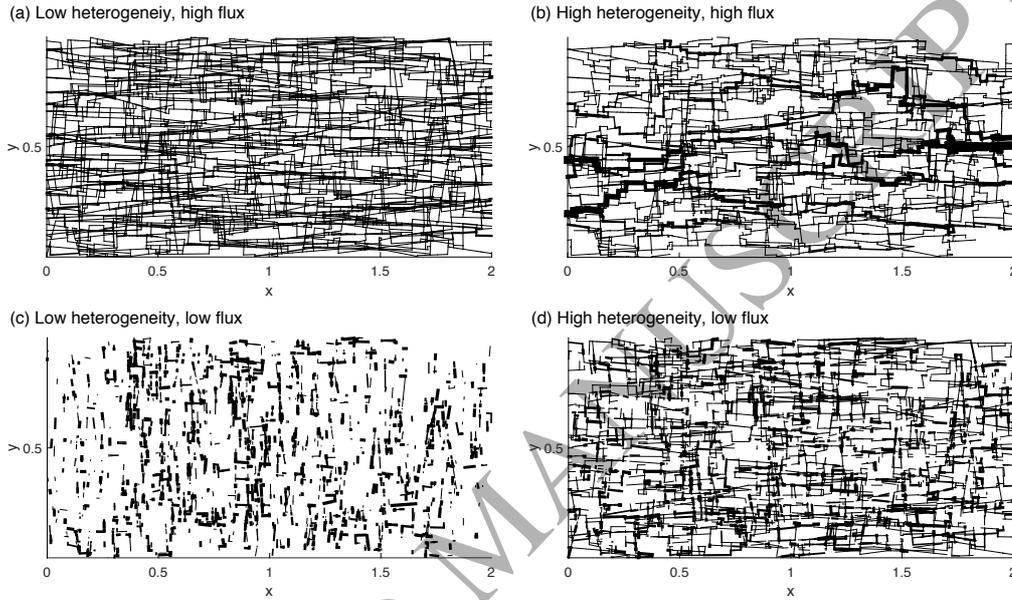


Figure 2: Normalized flow field ($|u_{ij}|/\bar{u}$) showing high and low flux zones for a log-normal conductivity distribution with two different heterogeneities. Link width is proportional to the magnitude of the normalized flow. (a) $\sigma_{\ln K} = 1$. Links with the flux value *smaller* than $\bar{u}/5$ are removed. (b) $\sigma_{\ln K} = 5$. Links with the flux value *smaller* than $\bar{u}/5$ is removed. Preferential flow paths emerge as conductivity heterogeneity increases. (c) $\sigma_{\ln K} = 1$. Links with the flux value *larger* than $\bar{u}/5$ are removed. Most of low flux values occur at the links perpendicular to the mean flow direction. (d) $\sigma_{\ln K} = 5$. Links with the flux value *larger* than $\bar{u}/5$ are removed. Low flux values show less spatial correlation than high flux values.

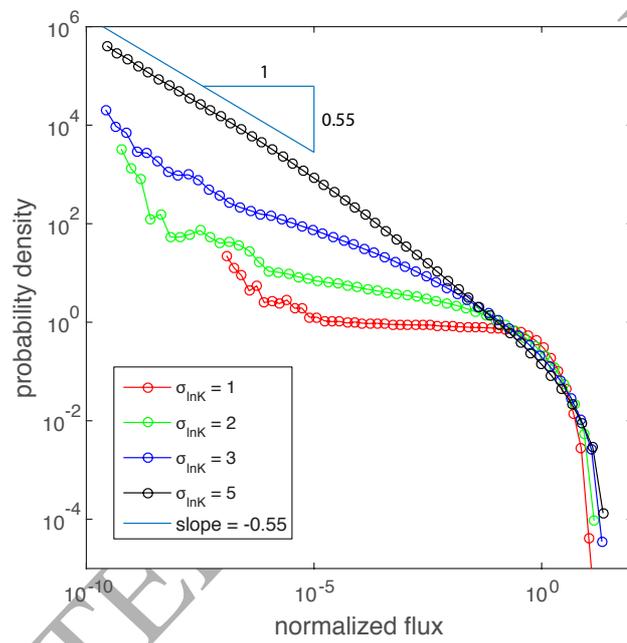


Figure 3: Eulerian flux probability density functions for four different levels of conductivity heterogeneity. Increase in conductivity heterogeneity significantly increases the probability of small flux values.

at the inlet, $x = 0$, with two different injection methods: (1) uniform injection, and (2) flux-weighted injection. *Uniform (resident) injection* introduces particles uniformly throughout the left boundary; this means that an equal number of particles is injected into each inlet node i_0 ,

$$N_{i_0} = \frac{N_p}{\sum_{i_0}}, \quad (4)$$

where N_{i_0} is the number of particles injected at node i_0 , N_p is the total number of injected particles. *Flux-weighted injection* introduces particles proportional to the total incoming flux Q_{i_0} at the injection location i_0

$$N_{i_0} = N_p \frac{Q_{i_0}}{\sum_{i_0} Q_{i_0}}. \quad (5)$$

155 Uniform injection simulates an initial distribution of tracer particles extended
 156 uniformly over a region much larger than the characteristic heterogeneity
 157 scale, and flux-weighted injection simulates a constant concentration pulse
 158 where the injected mass is proportional to the local injection flux at an inlet
 159 boundary that is much larger than the heterogeneity scale. For the uniform
 160 injection, the initial velocity distribution is then equal to the distribution of
 161 the Eulerian velocities. For the flux-weighted injection, the initial velocity
 162 distribution is equal to the flux-weighted Eulerian distribution. In general the
 163 initial velocity distribution may be arbitrary and depends on the conditions
 164 at the injection location. More detailed discussions can be found in section 4
 165 and section 5.

166 Injected particles are advected with the flow velocity u_{ij} between nodes.
 167 To focus on the impact of conductivity variability on particle transport, we
 168 assume porosity to be constant. This is a reasonable assumption because

169 the variability in porosity is significantly smaller than the the variability in
 170 conductivity [22, 93].

171 At the nodes, we apply a complete mixing rule [94, 95, 96]. Complete mix-
 172 ing assumes that Péclet numbers at the nodes are small enough that particles
 173 are well mixed within the node. Thus, the link through which the particle
 174 exits a node is chosen randomly with flux-weighted probability. A different
 175 node-mixing rule, streamline routing, assumes that Péclet numbers at nodes
 176 are large enough that particles essentially follow the streamlines and do not
 177 transition between streamlines. The complete mixing and streamline routing
 178 rules are two end members. The local Péclet number and the intersection
 179 geometry determine the strength of mixing at nodes, which is in general be-
 180 tween these two end-members. The impact of the mixing rule on transverse
 181 spreading can be significant for regular DFNs with low heterogeneity [97, 80].
 182 However, its impact is much more limited for random DFNs [96]. Since our
 183 interest in this study is the longitudinal spreading in random DFNs, we focus
 184 on the case of complete mixing. Thus, the particle transition probabilities
 185 p_{ij} from node i to node j are given by

$$p_{ij} = \frac{|u_{ij}|}{\sum_k |u_{ik}|}, \quad (6)$$

186 where the summation is over outgoing links only, and $p_{ij} = 0$ for incoming
 187 links. Particle transitions are determined only by the outgoing flux distribu-
 188 tion.

The particle pathways and times are obtained by the following recursion

relations

$$\mathbf{x}_{n+1} = \mathbf{x}_n + \ell_n \mathbf{e}_n, \quad (7a)$$

$$t_{n+1} = t_n + \frac{\ell_n}{u_n}, \quad (7b)$$

where $\mathbf{x}_n \equiv \mathbf{x}_{i_n}$ is the particle position after n random walk steps, $\ell_n \equiv \ell_{i_n i_{n+1}}$ the particle displacement and $\mathbf{e}_n \equiv \mathbf{e}_{i_n i_{n+1}}$ its orientation; the particle velocity at the n th step is denoted by $u_n \equiv |u_{i_n i_{n+1}}|$. The particle displacement, orientation and velocity determined by the transition probability $p_{i_n j}$ from node i_n to the neighboring nodes j given by Eq. (6). Equations (7) describe coarse-grained particle transport for a single realization of the spatial random network. Particle velocities and thus transition times depend on the particle position. The particle position at time t is $\mathbf{x}(t) = \mathbf{x}_{i_{n_t}}$, where

$$n_t = \sup(n | t_n \leq t) \quad (8)$$

denotes the number of steps needed to reach time t . We solve transport in a single disorder realization by particle tracking based on Eq. (7) with the two different injection rules (4) and (5) at the inlet at $x = 0$. The particle density in a single realization is

$$p(\mathbf{x}, t) = \langle \delta(\mathbf{x} - \mathbf{x}_{n_t}) \rangle, \quad (9)$$

189 where the angular brackets denote the average over all injected particles. As
 190 shown in Figure 4, both network heterogeneity and injection rule have signifi-
 191 cant impact on particle spreading. An increase in network heterogeneity leads
 192 to an increase in longitudinal particle spreading, and the uniform injection
 193 rule significantly enhances longitudinal spreading compared to flux-weighted

194 injection. The impact of network heterogeneity and injection method can be
 195 clearly seen from projected concentration profiles, $f_\tau(\omega)$. Arbitrary injection
 196 modes are also studied and discussed in section 5.2.

197 3. Average Solute Spreading Behavior

We first study the average solute spreading behavior for the four different levels of conductivity heterogeneity and the two different injection methods described above. We first illustrate the persistent effect of the particle injection method on particle transport, with the two different injection modes. To investigate the average spreading behavior, we average over all particles and network realizations. The average particle density is given by

$$\overline{P}(\mathbf{x}, t) = \overline{\langle \delta(\mathbf{x} - \mathbf{x}_{n_i}) \rangle}, \quad (10)$$

198 where the overbar denotes the ensemble average over all realizations. We
 199 run Monte Carlo particle tracking simulations for 100 realizations for each
 200 combination of conductivity heterogeneity and particle injection rule. In each
 201 realization, we release 10^4 particles at the inlet ($x = 0$) with the two different
 202 injection methods.

203 3.1. Breakthrough Curves

204 The average particle spreading behavior is first studied with the first
 205 passage time distribution (FPTD) or breakthrough curve (BTC) of particles
 206 at a control plane located at $x = x_c$. The FPTD is obtained by averaging
 207 over the individual particle arrival times $\tau_a(x_c) = \inf(t_n | |x_n - x_0| > x_c)$ as

$$f(\tau, x_c) = \overline{\langle \delta[\tau - \tau_a(x_c)] \rangle}. \quad (11)$$

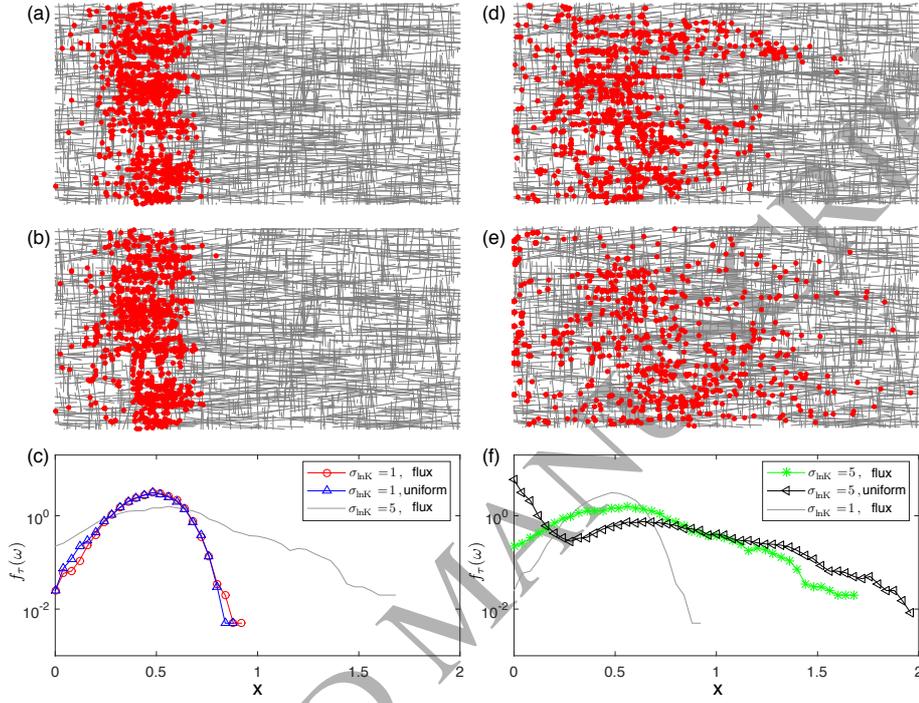


Figure 4: Particle distribution at $t = 20\bar{t}_l$ for a given realization after the instantaneous release of particles at the inlet, $x = 0$. \bar{t}_l is the median transition time to travel $L_x/100$. (a) The low heterogeneity case ($\sigma_{\ln K} = 1$) with flux-weighted injection. (b) The low heterogeneity case ($\sigma_{\ln K} = 1$) with uniform injection. (c) The projected particle distribution in the longitudinal direction for the low heterogeneity case ($\sigma_{\ln K} = 1$). (d) The high heterogeneity case ($\sigma_{\ln K} = 5$) with the flux-weighted injection. (e) The high heterogeneity ($\sigma_{\ln K} = 5$) with the uniform injection. (f) The projected particle distribution in the longitudinal direction for the high heterogeneity case ($\sigma_{\ln K} = 5$). For the high heterogeneity case, the injection method has significant impact on particle spreading. The uniform injection method leads to more anomalous spreading.

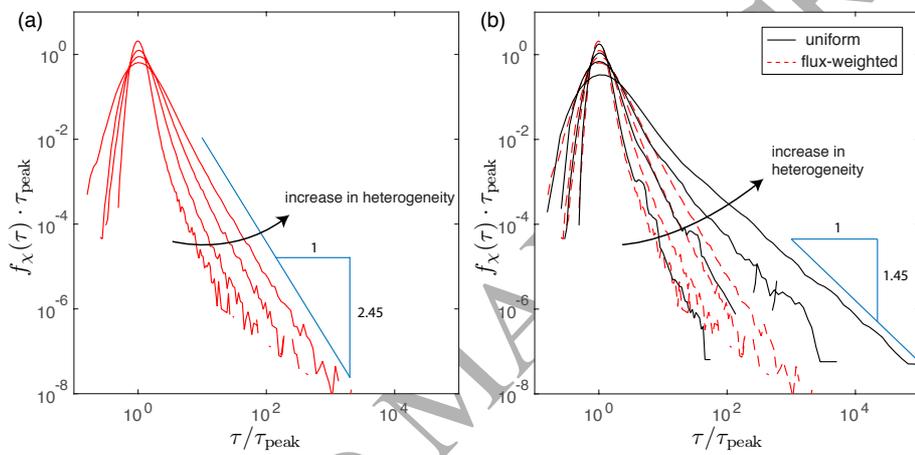


Figure 5: (a) FPTDs for $\sigma_{\ln K} = 1, 2, 3, 5$ with flux-weighted injection at $x_c = 200\bar{l}$. Increase in conductivity heterogeneity leads to larger dispersion and stronger late-time tailing. (b) FPTDs for $\sigma_{\ln K} = 1, 2, 3, 5$ with uniform injection (black solid lines). Uniform injection leads to significantly larger dispersion and late-time tailing compared to the flux-weighted injection (red dashed lines). FPTDs are normalized with the peak arrival time.

208 Figure 5 shows FPTDs at the outlet, $f(\tau, x_c = 200\bar{l})$, for different con-
 209 ductivity heterogeneities and injection rules. Conductivity heterogeneity has
 210 a clear impact on the FPTD by enhancing longitudinal spreading. This
 211 is so because stronger conductivity heterogeneity leads to broader particle
 212 transition time distribution, which in turn leads to enhanced longitudinal
 213 spreading. The injection rule also has a significant impact on FPTDs espe-
 214 cially for high conductivity heterogeneity. FPTDs between the two different
 215 injection rules are similar for $\sigma_{\ln K} = 1$, but uniform injection shows sig-
 216 nificantly stronger tailing for $\sigma_{\ln K} = 2, 3, 5$ [Figure 5(b)]. As conductivity
 217 heterogeneity increases, the flux values at the inlet also becomes broader.
 218 For flux-weighted injection, most of particles are injected at the nodes with
 219 high flux values. However, for uniform injection, particles are uniformly in-
 220 jected across the injection nodes and relatively large number of particles are
 221 released at the nodes with low flux values. This leads to notable difference
 222 between the two injection rules and the difference grows as the conductivity
 223 heterogeneity increases.

224 3.2. Centered Mean Square Displacement

225 We also study longitudinal spreading in terms of the centered mean square
 226 displacement (cMSD) of average particle density, $\bar{P}(\mathbf{x}, t)$. For the longitudi-
 227 nal direction (x), the cMSD is given by $\sigma_x^2(t) = \overline{[x(t) - \langle x(t) \rangle]^2}$ where $\langle \cdot \rangle$
 228 denotes the average over all particles for a given realization. In Figure 6,
 229 we show the time evolution of the longitudinal cMSDs. The time axis is
 230 normalized with the mean travel time along the characteristic fracture link
 231 length, \bar{l} . For both injection methods, spreading shows a ballistic regime
 232 ($\sim t^2$) at early times, which then transitions to a preasymptotic scaling in

233 an intermediate regime and finally to a final asymptotic regime. The time
 234 evolutions of cMSDs for the two injection cases are notably different as con-
 235 ductivity heterogeneity increases, while the asymptotic late-time scalings are
 236 very similar.

The asymptotic power-law scaling can be understood in the framework of a continuous time random walk (CTRW) description of dispersion. At large times the Lagrangian velocity distributions are in their steady states and subsequent particle velocities are independent. Thus, at large times horizontal particle dispersion can be described by the CTRW

$$x_{n+1} = x_n + \ell_0, \quad t_{n+1} = t_n + \tau_n, \quad (12)$$

with the transition time $\tau_n = \ell_0/v_n$. The velocities v_n are distributed according to $p_s(v)$ which is space Lagrangian velocity PDF, and ℓ_0 is a distance along the streamline that is sufficiently large so that subsequent particle velocities may be considered independent. Thus, the distribution of transit times τ_n is given in terms of the space Lagrangian and Eulerian velocity PDFs as [88]

$$\psi(\tau) = \frac{\ell_0}{\tau^2} p_s(\ell_0/\tau) = \frac{\ell_0}{\tau^3 \bar{v}} p_e(\ell_0/\tau), \quad (13)$$

where \bar{v} is the average Eulerian velocity, see also Section 4. Specifically, for the scaling $p_e(v) \propto v^\alpha$ at small velocities, the transit time PDF scales as

$$\psi(\tau) \propto \tau^{-1-\beta}, \quad \beta = 2 + \alpha. \quad (14)$$

237 From Figure 3, we estimate for $\sigma_{\ln K} = 5$ that $\alpha \approx -0.55$, which corresponds
 238 to $\beta = 1.45$. CTRW theory [66, 65] predicts that the cMSD scales as $t^{3-\beta}$,
 239 which here implies $t^{1.55}$. This is consistent with the late-time scaling of the
 240 cMSD shown in 6 for $\sigma_{\ln K} = 5$.

241 The Monte Carlo simulations show that, in the intermediate regime (t/\bar{t}_i
 242 approximately between 1 and 100), the *longitudinal* cMSD increases linearly
 243 with time for flux-weighted injection [Figure 6(a)]. For uniform injection,
 244 cMSD increases faster than linearly (i.e., superdiffusively) for intermediate to
 245 strong heterogeneity in the intermediate regime [Figure 6(b)]. The stronger
 246 heterogeneity led to the increase in the late-time temporal scaling for both
 247 flux-weighted and uniform injection cases. The Monte Carlo simulations also
 248 show that there is no noticeable difference between the uniform injection
 249 and the flux-weighted injection for the low heterogeneity case whereas the
 250 difference increases as heterogeneity increases [Figure 6(b), inset].

251 In summary, both the increase in conductivity heterogeneity and the uni-
 252 form injection method enhance longitudinal spreading. For low heterogeneity,
 253 the two different injection rules do not affect particle spreading significantly.
 254 The difference, however, becomes significant as the conductivity heterogene-
 255 ity increases. Both the magnitude of the cMSD and the super-diffusive scaling
 256 behavior are notably different for the two different injection rules at high het-
 257 erogeneity. We now analyze the Lagrangian particle statistics to understand
 258 the underlying physical mechanisms that lead to the observed anomalous
 259 particle spreading.

260 4. Lagrangian Velocity Statistics

261 The classical CTRW approach [64, 65]—see Eq. (12)—relies on the in-
 262 dependence of particle velocities at subsequent steps and thus spatial po-
 263 sitions. Recent studies, however, have shown that the underlying mecha-
 264 nisms of anomalous transport can be quantified through an analysis of the

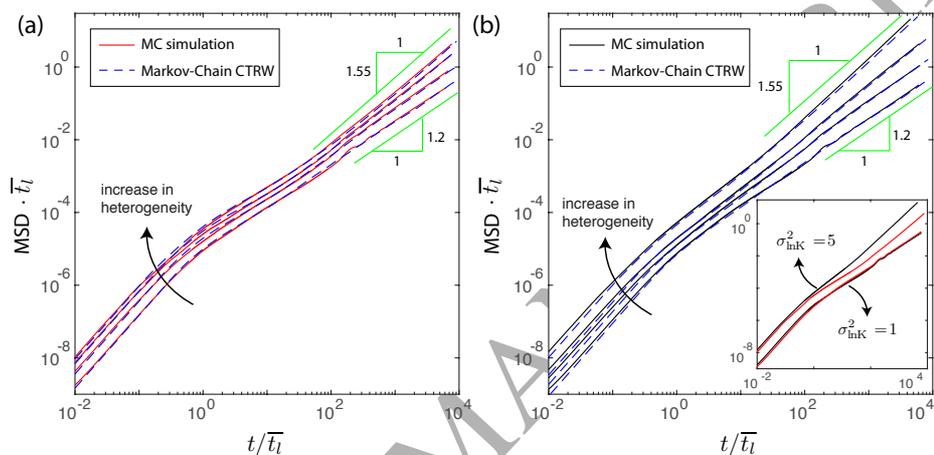


Figure 6: Time evolution of longitudinal MSDs for $\sigma_{\ln K} = 1, 2, 3, 5$ obtained from Monte Carlo simulations (solid lines), and the model predictions from the Markov-Chain CTRW (32) and (35) with the full transition PDF (dashed lines). Increase in conductivity heterogeneity leads to higher dispersion, and the Markov-chain CTRW model is able to accurately capture the time evolution of the MSDs for all levels of heterogeneity and injection rules. (a) Flux-weighted injection. (b) Uniform injection. Inset: Comparison between flux-weighted and uniform injection for $\sigma_{\ln K} = 1, 5$. Impact of injection rule is significant for high conductivity heterogeneity.

265 statistics of Lagrangian particle velocities such as velocity distribution and
 266 correlation [74, 76, 98, 77, 78, 45, 80]. In the following, we briefly intro-
 267 duce two viewpoints for analyzing Lagrangian velocities—equidistantly and
 268 isochronally along streamlines—and the relation between them [88]. We then
 269 proceed to a detailed analysis of the Lagrangian velocity statistics measured
 270 equidistantly along streamlines.

271 4.1. Lagrangian Velocities

Particle motion is described here by the recursion relations (7). In this framework, we consider two types of Lagrangian velocities. The t(ime)-Lagrangian velocities are measured at a given time t ,

$$v_t(t) = u_{n_t}, \quad (15)$$

where n_t is defined by (8). The s(pace)-Lagrangian velocities are measured at a given distance s along the trajectory. The distance s_n traveled by a particle along a trajectory after n steps is given by

$$s_{n+1} = s_n + \ell_n. \quad (16)$$

The number of steps needed to cover the distance s is described by $n_s = \sup(n | s_n \leq s)$. Thus, the particle velocity at a distance s along a trajectory is given by

$$v_s(s) = u_{n_s}. \quad (17)$$

The PDF of t-Lagrangian velocities sampled along a particle path is given by

$$p_t(v) = \lim_{n \rightarrow \infty} \frac{\sum_{i=1}^n \tau_i \delta(v - u_i)}{\sum_{i=1}^n \tau_i}, \quad (18)$$

where we defined the transit time,

$$\tau_i = \frac{\ell_i}{u_i} \quad (19)$$

The PDF of s-Lagrangian velocities sampled along a particle path are defined analogously as

$$p_s(v) = \lim_{n \rightarrow \infty} \frac{\sum_{i=1}^n \ell_i \delta(v - u_i)}{\sum_{i=1}^n \ell_i}. \quad (20)$$

Note the difference with respect to Eq. (2), which samples velocities in the network uniformly, while in Eq. 20 velocities are sampled along trajectories. Using the definition of the transit time τ_i in (19), the PDFs of the s- and t-Lagrangian velocities are related through flux weighting as [88]

$$p_s(v) = \frac{vp_t(v)}{\int dv vp_t(v)}. \quad (21)$$

Furthermore, for flux-preserving flows and under ergodic conditions, the Eulerian and t-Lagrangian velocity PDFs are equal,

$$p_e(v) = p_t(v). \quad (22)$$

Thus, under these conditions, the s-Lagrangian and Eulerian velocity PDFs are related as [88]

$$p_s(v) = \frac{vp_e(v)}{\int dv vp_e(v)}. \quad (23)$$

272 This means that the *stationary* s-Lagrangian velocity PDF can be determined
 273 from the Eulerian velocity PDF. Figure 7 illustrates this relation by compar-
 274 ing the s-Lagrangian velocity PDFs measured from the numerical simulation
 275 to the flux-weighted Eulerian velocity PDFs shown in Figure 3.

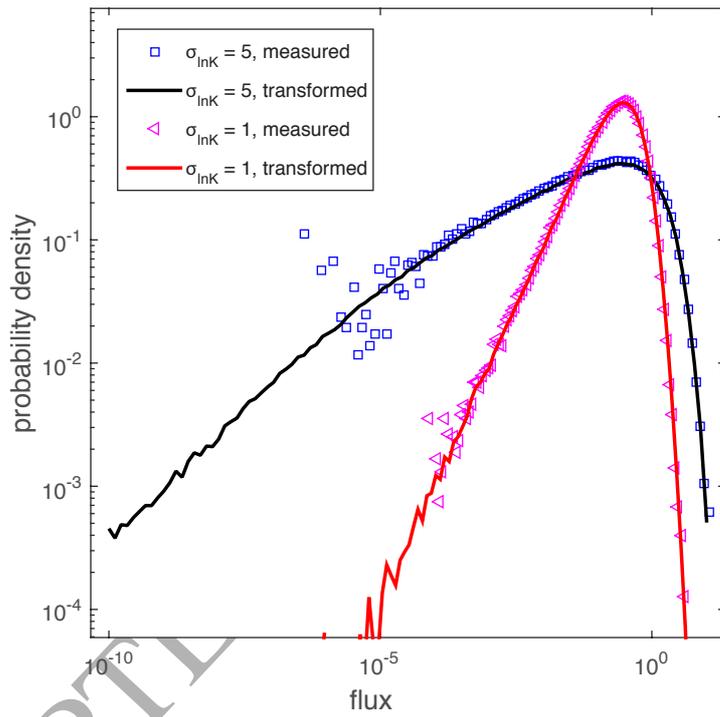


Figure 7: s-Lagrangian velocity PDFs for $\sigma_{\ln K} = 1$ and $\sigma_{\ln K} = 5$. The measured s-Lagrangian velocity PDF agrees very well with the PDF obtained by transforming the Eulerian velocity PDF using Eq. (23).

276 4.2. Evolution of Lagrangian Velocity Distributions

It is important to emphasize that the above definitions of the Lagrangian velocity PDFs refer to stationary conditions. We now define the PDFs of t- and s-Lagrangian velocities through sampling between particles and network realizations at a given time (t-Lagrangian) or space (s-Lagrangian) velocities

$$\hat{p}_t(v, t) = \overline{\langle \delta[v - v(t)] \rangle}, \quad \hat{p}_s(v, s) = \overline{\langle \delta[v - v(s)] \rangle}. \quad (24)$$

In general, these quantities evolve in time and with distance along the streamline and are sensitive to the injection conditions because evidently for $t = 0$ and $s = 0$ both are equal to the PDF of initial particle velocities $\hat{p}_t(v, t = 0) = \hat{p}_s(v, s = 0) = p_0(v)$, but their respective stationary PDFs are different, namely

$$p_t(v) = \lim_{t \rightarrow \infty} \hat{p}_t(v, t), \quad p_s(v) = \lim_{s \rightarrow \infty} \hat{p}_s(v, s). \quad (25)$$

Let us consider some further consequences of these properties. First, we notice that under (Eulerian) ergodicity the *uniform injection* condition (4) corresponds to an initial velocity PDF of

$$p_0(v) = p_e(v) = p_t(v), \quad (26)$$

277 that is, the initial velocity PDF is equal to the Eulerian and thus t-Lagrangian
 278 velocity PDFs. This means that for the uniform injection method, the t-
 279 Lagrangian velocity PDF is steady, $\hat{p}_t(v, t) = p_t(v)$, while the s-Lagrangian
 280 velocity PDF is not. It evolves from its initial distribution $p_s(v, s = 0) =$
 281 $p_e(v)$ to the steady state distribution (23).

The *flux-weighted injection* condition, on the other hand, corresponds to the the initial velocity PDF

$$p_0(v) = p_s(v), \quad (27)$$

282 due to relation (21). The initial velocity PDF is equal to the s-Lagrangian
 283 velocity PDF. This means that under flux-weighting, the s-velocity PDF is
 284 steady, $\hat{p}_s(v, s) = p_s(v)$. Under these conditions, the t-Lagrangian velocity
 285 PDF $\hat{p}_t(v, t)$ evolves from the initial distribution $\hat{p}_t(v, t = 0) = p_s(v)$ towards
 286 the asymptotic $p_t(v) = p_e(v)$, which is equal to the Eulerian velocity PDF.
 287 These are key insights for the qualitative and quantitative understanding of
 288 the average transport behavior.

289 4.3. Space-Lagrangian Velocity Statistics

290 We analyze particle velocities along their projected trajectories in the
 291 longitudinal direction. Spatial particle transitions may be characterized by
 292 the characteristic fracture link length, \bar{l} . The Lagrangian velocity $v_s(s_n)$ at
 293 a distance $x_n = n\bar{l}$ along the projected trajectory is approximated by the
 294 average velocity $v_n \equiv \bar{l}/\tau_n$ where τ_n is the transition time for the distance
 295 \bar{l} at step n . In the following, we investigate the statistical characteristics of
 296 the s-Lagrangian velocity series $\{v_n\}$. For the uniform flow conditions under
 297 consideration here, the projected distance x_n is a measure for the streamwise
 298 distance s_n , and v_n for the s-Lagrangian velocity $v_s(s_n)$. Spatial Lagrangian
 299 velocities have been studied by Cvetkovic et al. [73] and Gotovac et al. [75]
 300 for highly heterogeneous porous media and by Frampton and Cvetkovic [16]
 301 for 3D DFNs in view of quantifying particle travel time statistics and thus
 302 modelling effective particle motion.

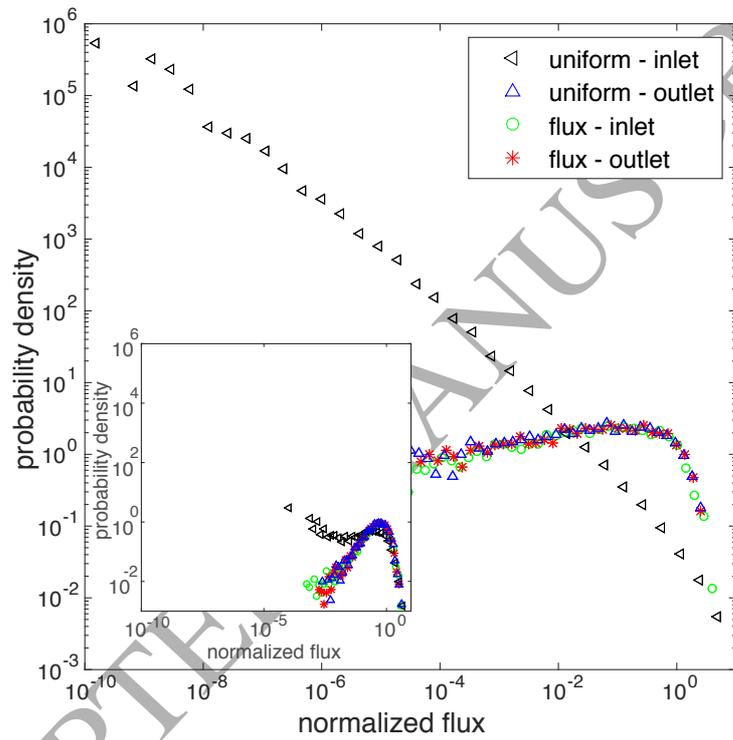


Figure 8: Lagrangian flux distributions at the inlet and outlet for uniform and flux-weighted injection rules, for $\sigma_{\ln K} = 5$. Note that Lagrangian flux distributions at the outlet are identical regardless of the injection method. Inset: the same plot for $\sigma_{\ln K} = 1$.

303 We first study the convergence of the s-Lagrangian velocity PDFs towards
 304 a stationary distribution and the invariance of $\hat{p}_s(v, s)$ for a stationary (flux-
 305 weighted) initial velocity PDF. We consider the two injection conditions (4)
 306 and (5) and record the distribution of particle velocities at a line located at
 307 the control point x_c . Under ergodic conditions, we expect $\hat{p}_s(v, s)$ to converge
 308 towards its steady state distribution (23) for uniform injection and to remain
 309 invariant for the flux-weighted injection. Figure 8 shows $\hat{p}_s(v, s = 0)$ and
 310 $\hat{p}_s(v, s = x_c)$ for uniform and flux-weighted injection conditions and two
 311 different heterogeneity strengths. We clearly observe that $\hat{p}_s(v, x_c) = p_s(v)$
 312 is invariant for flux-weighted injection. For uniform injection, $\hat{p}_s(v, x_c)$ has
 313 already evolved towards its steady limit after $x_c = 200\bar{l}$. This is an indication
 314 that the flow and transport system is in fact ergodic. Note that in terms
 315 of computational efficiency, this observation gives a statistically consistent
 316 way of continuing particle trajectories through reinjection at the inlet. If
 317 the outlet is located at a position x_c large enough so that $\hat{p}_s(v, s = x_c) =$
 318 $p_s(v)$, particles are reinjected at the inlet with flux-weighted probability, this
 319 means that the velocity statistics are preserved. Furthermore, this method
 320 ensures that the domain is large enough to provide ergodic conditions. In
 321 the following, we analyze the statistical properties of streamwise velocity
 322 transitions with the aim of casting these dynamics in the frame of a Markov
 323 model for subsequent particle velocities.

324 We first consider the distribution $\psi_\tau(t)$ of transition times along particle
 325 trajectories through sampling the transition times along all particle trajecto-
 326 ries and among network realizations. To this end, we consider a flux-weighted
 327 injection because it guarantees that the s-Lagrangian velocities are station-

328 ary. Figure 9 illustrates the PDF of transition times for different variances
 329 of $\ln(K)$. As $\sigma_{\ln K}$ increases, the transition time PDFs become broader. The
 330 transition time closely follows a truncated power-law distribution.

Next we consider two-point velocity statistics to gain insight into the velocity correlations along a streamline. To this end, we consider the velocity auto covariance for a given lag $\Delta s = s - s'$. As pointed out above, for flux-weighted injection, the streamwise velocities here are stationary and therefore

$$C_s(s - s') = \overline{\langle [v_s(s) - \langle v_s(s) \rangle][v_s(s') - \langle v_s(s') \rangle] \rangle}. \quad (28)$$

In order to increase the statistics, we furthermore sample along streamlines over a distance of $10^2 \bar{l}$. The velocity variance is $\sigma_v^2 = C_s(0)$. The velocity autocorrelation function $\chi_s(s) = C_s(s)/\sigma_v^2$. The correlation length scale ℓ_c is defined by

$$\ell_c = \int_0^{\infty} ds \chi_s(s). \quad (29)$$

331 The inset in Figure 9 shows the increase in the velocity correlation length
 332 scale with increasing $\ln(K)$ variances for a flux-weighted injection case. This
 333 can be attributed to the emergence of preferential flow paths, as shown in
 334 Figure 2. Painter and Cvetkovic [71] and Frampton and Cvetkovic [16] also
 335 reported the existence of clear velocity correlation between successive jumps
 336 in DFNs and showed that this correlation structure should be captured for
 337 effective transport modelling.

The existence of a finite correlation length along the particle trajectories indicates that subsequent velocities, when sampled at a distance much larger than the correlation length ℓ_c , may be considered independent. In order to

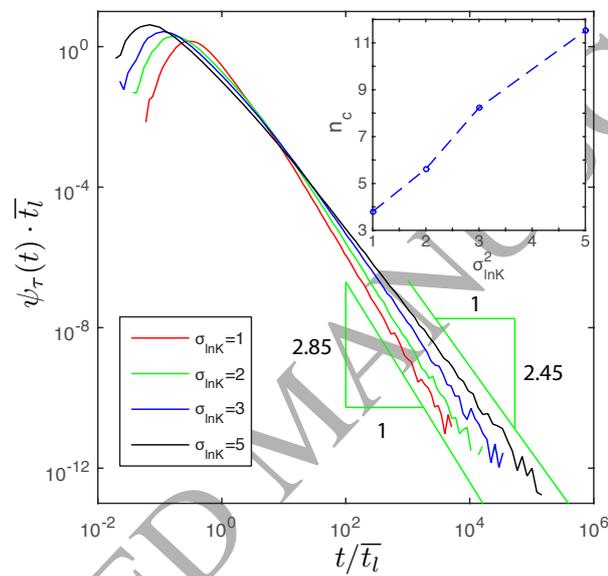


Figure 9: (a) Lagrangian transition time distributions for $\sigma_{\ln K} = 1, 2, 3, 5$ with flux-weighted injection. As the network conductivity becomes more heterogeneous, the transition time distribution becomes broader. Inset: the effective correlation length increases with increasing network heterogeneity: $3.8\bar{l}, 5.6\bar{l}, 8.2\bar{l}, 11.5\bar{l}$. The correlation step (n_c) is computed by integrating velocity autocorrelation function in space.

study this feature, we characterize the series of s-Lagrangian velocities $\{v_n\}$ in terms of the transition probabilities to go from velocity v_m to velocity v_{m+n} . We determine the transition probabilities under flux-weighted particle injection because, as detailed above, under these conditions, the s-Lagrangian velocity is stationary. Thus, the transition probability is only a function of the number n of steps,

$$r_n(v|v') = \langle \delta(v - v_{m+n}) \rangle |_{v_m=v'} \quad (30)$$

Numerically, the transition probability is determined by discretizing the s-Lagrangian velocity PDF into N velocity classes $\mathcal{C}_i = (v_{s,i}, v_{s,i} + \Delta v_{s,i})$ and recording the probability for each class given the previous velocity class. This procedure gives the transition matrix $T_n(i|j)$ from class j to i after n steps such that $r_n(v|v')$ is approximated numerically as

$$r_n(v|v') = \sum_{i,j=1}^N \frac{\mathbb{I}_{\mathcal{C}_i}(v) T_n(i|j) \mathbb{I}_{\mathcal{C}_j}(v')}{\Delta v_i}, \quad (31)$$

338 where the indicator function $\mathbb{I}_{\mathcal{C}_i}(v)$ is 1 if $v \in \mathcal{C}_i$ and 0 otherwise.

339 Figure 10 shows the one-step transition matrix $T_1(i|j)$ for equidistant and
 340 logarithmically equidistant velocity classes for different network heterogene-
 341 ity. Higher probabilities along the diagonal than in the off-diagonal positions
 342 indicate correlation between subsequent steps, which, however, decreases as
 343 the number of steps along the particle trajectory increases, as indicated by
 344 the existence of a finite correlation scale ℓ_c .

345 5. Stochastic Particle Motion and Effective Transport Model

346 In the following, we describe the evolution of the s-Lagrangian velocities
 347 by a Markov-chain, which is motivated by the existence of a finite spatial

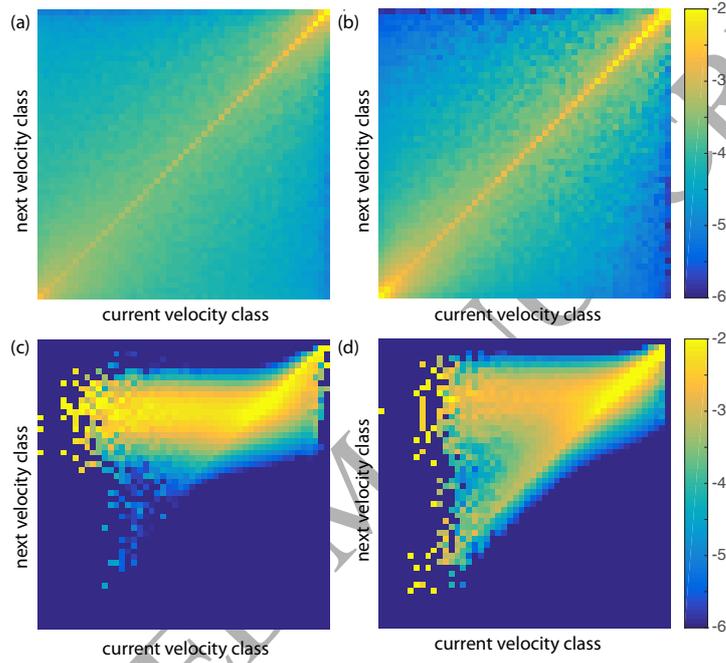


Figure 10: (a) One-step velocity transition matrix $T_1(i|j)$ with linear equiprobable binning for $N = 50$ velocity classes for $\sigma_{\ln K} = 1$. (b) Velocity transition matrix with linear equiprobable binning for $\sigma_{\ln K} = 5$. The color-bar shows the logarithmic scale. (c) Velocity transition matrix with logarithmic binning for $\sigma_{\ln K} = 1$. (d) Velocity transition matrix with logarithmic binning for $\sigma_{\ln K} = 5$. Increase in conductivity heterogeneity leads to higher probability close to diagonal entries.

348 correlation scale (see inset of figure 9). This leads to a spatial Markov-chain
 349 random walk (which we also termed spatial Markov Model) formulation of
 350 particle dispersion that is valid for any initial velocity distribution, and thus
 351 for any injection protocol. This modelling approach is in line with the time-
 352 domain random walk (TDRW) and continuous time random walk (CTRW)
 353 approaches discussed in the Introduction and below.

354 5.1. Markovian Velocity Process

Along the lines of Le Borgne et al. [74] and Kang et al. [77], we model the velocity series $\{v_n\}$ as a Markov-chain, which is a suitable model to statistically quantify the evolution of the s-Lagrangian velocities based on the existence of a finite correlation length. In this framework, the n -step transition probability $r_n(v|v')$ satisfies the Chapman–Kolmogorov equation [99]

$$r_n(v_n|v_0) = \int dv_k r_{n-k}(v_n|v_k) r_k(v_k|v_0). \quad (32a)$$

The velocity process is fully characterized in terms of the one-step transition PDF $r_1(v|v')$ and the steady state PDF $p_s(v)$ of the s-Lagrangian velocity. Consequently, the evolution of the s-Lagrangian velocity PDF $\hat{p}_s(v, s_n)$ is given by

$$p_s(v, s_n) = \int dv' r_1(v|v') p_s(v', s_n), \quad (32b)$$

355 with the arbitrary initial PDF $p_s(v, s_0 = 0) = p_0(v)$. The number of steps
 356 to decorrelate this Markov-chain is given by $n_c = \ell_c/\bar{l}$. Figure 11 shows the
 357 evolution of the PDF of s-Lagrangian velocities for the uniform injection (4).
 358 Recall that the uniform injection mode corresponds to the initial velocity

359 PDF $p_e(v)$. Thus, the numerical Monte Carlo simulations are compared to
 360 the predictions of (32b) for the initial condition $\hat{p}_s(v, s_0 = 0) = p_e(v)$. The
 361 transition PDF $r_1(v|v')$ is given by (31) with the velocity transition matrix
 362 shown in Figure 10. As shown in Figure 11, the prediction of the Markovian
 363 velocity model and the Monte Carlo simulation are in excellent agreement,
 364 which confirms the validity of the Markov model (32) for the evolution of
 365 s-Lagrangian velocities. Velocity transition dynamics are independent of the
 366 particular initial conditions and thus allow predicting the evolution of the
 367 Lagrangian velocity statistics for any initial velocity PDF and thus for any
 368 injection protocol.

As mentioned above, the Markov-chain $\{v_n\}$ is fully characterized by
 the stationary PDF of the s-Lagrangian velocities and the transition PDF
 $r_1(v|v')$. The behavior of the latter may be characterized by the number of
 steps n_c needed to decorrelate, i.e., the number of steps n_c such that $r_n(v|v')$
 for $n > n_c$ converges to the stationary PDF $r_n(v|v') \rightarrow p_s(s)$. The number
 of steps for velocities to decorrelate can be quantified by

$$n_c = \frac{\ell_c}{\bar{l}}, \quad (33)$$

The simplest transition PDF that shares these characteristics is [41, 80, 50,
 88]

$$r_1(v|v') = a\delta(v - v') + (1 - a)p_s(v), \quad (34)$$

369 with $a = \exp(-\bar{l}/\ell_c)$. This transition PDF is thus fully determined by one
 370 single parameter n_c . Note that the latter increases with the level of hetero-
 371 geneity, as illustrated in the inset of figure 9. This parameter is estimated
 372 here from the simulated Lagrangian velocities. It may also be measured in

373 the field from multiscale tracer tests [41]. In the following, we study particle
 374 dispersion in the Markovian velocity model for the full transition PDF shown
 375 in Figure 10 and the reduced-order Markov model (34).

376 5.2. Particle Dispersion and Model Predictions

We consider particle motion along the mean pressure gradient in x -
 direction, which is described by the stochastic regression

$$x_{n+1} = x_n + \bar{l}, \quad t_{n+1} = t_n + \frac{\bar{l}}{v_n}. \quad (35)$$

The velocity transitions are determined from the Markovian velocity pro-
 cess (32). Note that the $\{v_n\}$ process describes equidistant velocity tran-
 sitions along particle trajectories, while (35) describes particle motion pro-
 jected on the x -axis. In this sense, (35) approximates the longitudinal travel
 distance x_n with the distance s_n along the streamline, which is valid if the
 tortuosity of the particle trajectories is low. As indicated in Section 3.2, for
 travel distances ℓ_0 larger than ℓ_c , or equivalently, step numbers $n \gg n_c \equiv$
 ℓ_c/\bar{l} , subsequent velocities may be considered independent and particle dis-
 persion is fully characterized by the recursion relation (12) and the transition
 time PDF (13). Thus, as shown in Section 3.2, the CTRW of Eq. (12) cor-
 rectly predicts the asymptotic scaling behavior of the centered mean square
 displacement. This is not necessarily so for the particle breakthrough and the
 preasymptotic behavior of the cMSD. As seen in Section 3.1, the late time
 tailing of the BTC depends on the injection mode and thus on the initial
 velocity PDF. In fact, the slope observed in Figure 5 for uniform particle in-
 jection can be understood through the persistence of the initial velocity PDF.
 The first random walk steps until decorrelation at $n = n_c$ are characterized

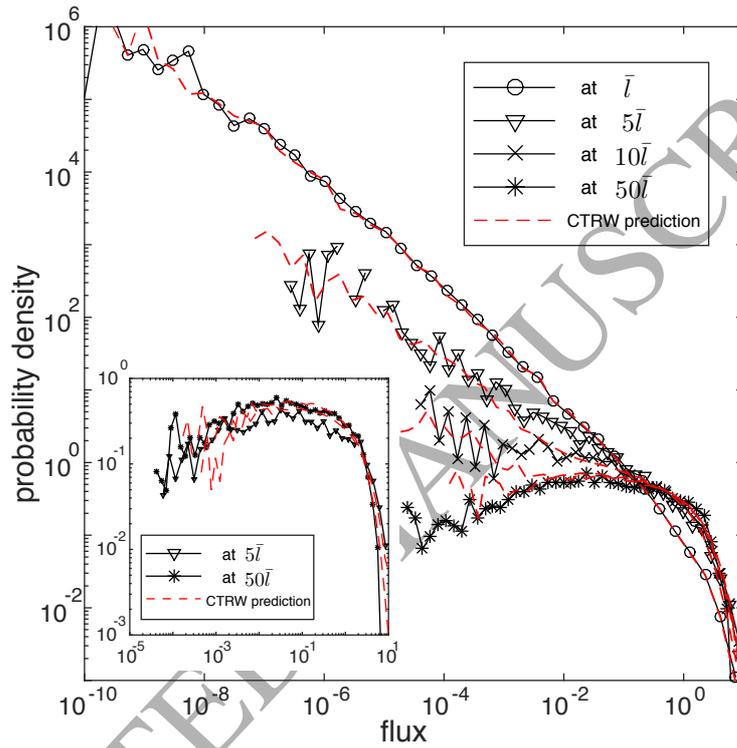


Figure 11: Evolution of the PDF of s-Lagrangian velocities for *uniform injection*, i.e., for an initial velocity PDF $\hat{p}_s(v, s_0 = 0) = p_e(v)$. The symbols denote the data obtained from the direct numerical simulation, the dashed lines show the predictions of (32b) with the transition matrix shown in Figure 10. Inset: Evolution of the PDF of s-Lagrangian velocities for a *flux-weighted injection*. In this case, the initial velocity PDF is identical to the stationary s-Lagrangian velocity PDF.

by the transit time PDF

$$\psi_0(t) = \frac{\bar{l}}{t^2} p_0(\bar{l}/t). \quad (36)$$

Thus, for an initial velocity PDF $p_0(v) = p_e(v)$, the initial transit time PDF is given in terms of the Eulerian velocity PDF, which is characterized by a stronger probability weight towards low velocities than the PDF of the s-Lagrangian velocities, which is given by (23). The space-time random walk (35) together with the Markov model (32b) is very similar to the TDRW approach [70, 71] and can also be seen as a multi-state, or correlated CTRW approach because subsequent particle velocities and thus transition times are represented by a Markov process [100, 101, 74, 102]. The joint distribution $p(x, v, t)$ of particle position and velocity at a given time t is given by [74]

$$p(x, v, t) = \int_0^t dt' H(\bar{l}/v - t') R(x - vt', v, t - t'), \quad (37)$$

where $H(t)$ is the Heaviside step function; $R(x, v, t)$ is the frequency by which a particle arrives at the phase space position (x, v, t) . It satisfies

$$R(x, v, t) = R_0(x, v, t) + \int dv' r_1(v|v') R(x - \bar{l}, v', t - \bar{l}/v'), \quad (38)$$

where $R_0(x, v, t) = p_0(x, v)\delta(t)$ with $p_0(x, v) = p(x, v, t = 0)$. Thus, the right side of (37) denotes the probability that a particle arrives at a position $x - vt'$ where it assumes the velocity v by which it advances toward the sampling position x . Equations (37) and (38) can be combined into the Boltzmann equation

$$\frac{\partial p(x, v, t)}{\partial t} = -v \frac{\partial p(x, v, t)}{\partial x} - \frac{v}{\bar{l}} p(x, v, t) + \int dv' \frac{v'}{\bar{l}} r_1(v|v') p(x, v', t), \quad (39)$$

377 see Appendix A. This result provides a bridge between the TDRW ap-
 378 proach [70, 71] and the correlated CTRW approach.

379 As illustrated in Figure 3, the Eulerian velocity PDF can be characterized
 380 by the power-law $p_e(v) \propto v^\alpha$. Thus, the first CTRW steps until the decor-
 381 relation at $n = n_c$ are characterized by the transit time PDF $\psi_0(t) \propto t^{-2-\alpha}$.
 382 The corresponding tail of the BTC is $f(t, x_c) \propto t^{-2-\alpha}$. The observed value
 383 of $\alpha = -0.55$ explains the tailing of the BTC in Figure 5 as $t^{-1.45}$, which
 384 shows the importance of the initial velocity distribution. We also observe
 385 decrease in BTC tailing (larger absolute slope) with travel distance as initial
 386 velocity distribution converges to stationary Lagrangian velocity distribution
 387 and as tracers sample more velocity values. This implies that one needs to be
 388 careful when inferring a β from single BTC measurement because the slope
 389 can evolve depending on the injection method, velocity PDF and velocity
 390 correlation.

391 First, we compare the results obtained from Monte Carlo simulation in
 392 the random DFN to the predictions of the Markov-chain CTRW (32) and
 393 (35) with the full transition PDF of Figure 10. Figure 6 shows the evolu-
 394 tion of the cMSD for different levels of heterogeneity and different injection
 395 modes. As expected from the ability of the Markov model to reproduce
 396 the evolution of the s-Lagrangian velocity PDF for both uniform and flux-
 397 weighted injection conditions, the predictions of particle spreading are in
 398 excellent agreement with the direct numerical simulations. In Figure 12 we
 399 compare breakthrough curves obtained from numerical simulations with the
 400 predictions by the Markov model for the uniform and flux-weighted injection
 401 modes. Again, the impact of the injection mode and thus initial velocity

402 PDF is fully quantified by the Markov model.

403 We now apply the Markov model (32)–(35), i.e., employing a parsimo-
404 nious parameterization of the velocity transition PDF, with a single param-
405 eter n_c (equation (33)), which is estimated here from velocity correlations
406 along streamlines (see inset of figure 9). We first compare the reduced-order
407 Markov model to the cases of uniform and flux-weighted injection, and con-
408 clude that the proposed parsimonious stochastic model provides an excellent
409 agreement with the direct numerical simulations (Figure 13). This implies
410 that the simple correlation model (34) can successfully approximate the ve-
411 locity correlation structure. Hence it appears that high order correlation
412 properties, quantified from the full transition probabilities (figure 10), are
413 not needed for accurate transport predictions in the present case. This sug-
414 gests promising perspective for deriving approximate analytical solutions for
415 this Markov-chain CTRW model [88]. Furthermore, as discussed in [41], the
416 velocity correlation parameter n_c can be estimated in the field by combining
417 cross-borehole and push-pull tracer experiments.

418 Finally, we consider the evolution of the particle BTC and the cMSD for
419 *arbitrary injection modes*. For real systems both flux-weighted and uniform
420 injections are idealizations. A flux-weighted condition simulates a constant
421 concentration pulse where the injected mass is proportional to the local in-
422 jection flux at an inlet boundary that is extended over a distance much larger
423 than the correlation scale during a given period of time. A uniform injection
424 represents an initial concentration distribution that is uniformly extended
425 over a region far larger than the correlation length. In general, the initial
426 concentration distribution may not be uniform, and the injection boundary

427 may not be sufficiently large, which leads to an arbitrary initial velocity dis-
428 tribution, biased maybe to low or high flux zones, as for example in the
429 MADE experiments, where the solute injection occurred into a low perme-
430 ability zone [103]. For demonstration, we study two scenarios representing
431 injection into low and high flux zones: uniform injections into regions of the
432 20-percentile highest, and 20-percentile lowest velocities. The initial velocity
433 PDF for the low velocity mode shows the power-law behavior which is the
434 characteristic for the Eulerian PDF, and the initial velocity PDF for the high
435 velocity mode shows narrow initial velocity distribution (Figure 15). Even-
436 tually, the s-Lagrangian PDFs evolve towards the stationary flux-weighted
437 Eulerian PDF as discussed in the previous section.

438 Figure 15 shows the predictive ability of the effective stochastic model
439 for these different injection conditions. The reduced-order Markov velocity
440 model compares well with the direct Monte Carlo simulation in the random
441 networks. As expected, the BTCs for injection into low velocity regions
442 have a much stronger tailing than for injection into high velocity regions.
443 In fact, as the initial velocity shows the same behavior at low velocities as
444 the Eulerian velocity PDF, the breakthrough tailing is the same as observed
445 in Figure (12). We also observed that the reduced-order Markov velocity
446 model can capture important features of the time evolution of cMSDs. This
447 demonstrates that the proposed model can incorporate arbitrary injection
448 modes into the effective modelling framework.

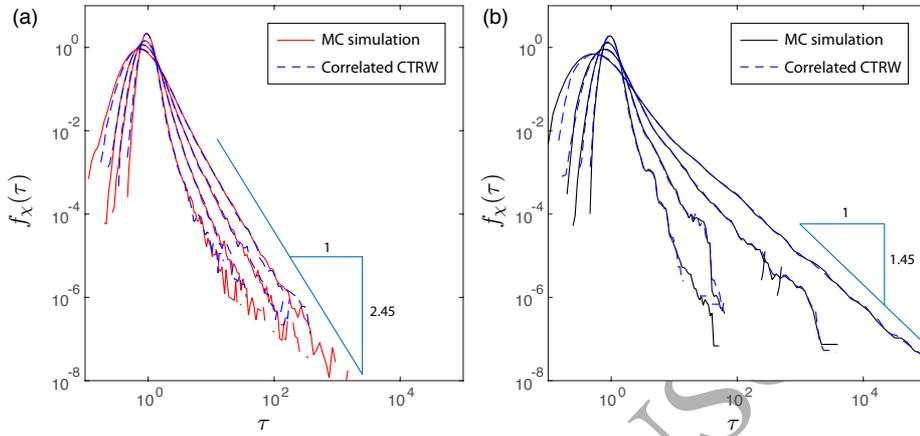


Figure 12: Particle BTCs from Monte Carlo simulations and the predictions from the Markov-chain CTRW model with the *full* velocity transition matrix for (a) flux-weighted injection, and (b) uniform injection at $x_c = 200\bar{l}$.

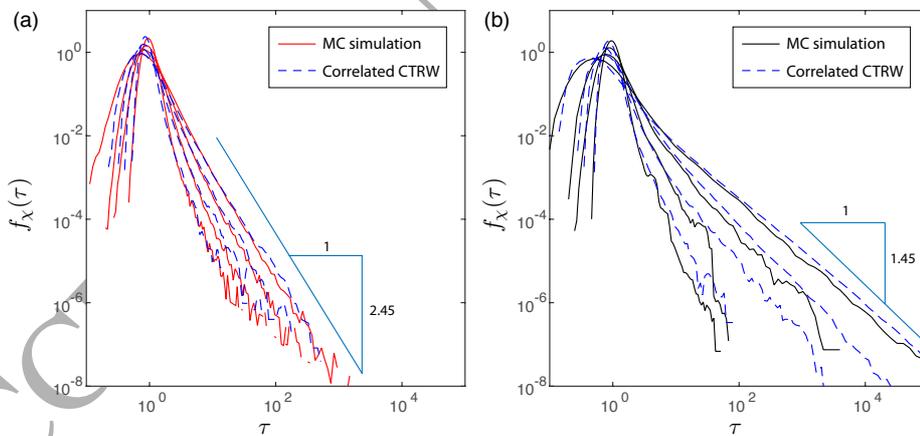


Figure 13: Particle BTCs from Monte Carlo simulations and predictions from the Markov-chain CTRW model with the *reduced-order* velocity transition matrix for (a) flux-weighted injection, and (b) uniform injection at $x_c = 200\bar{l}$.

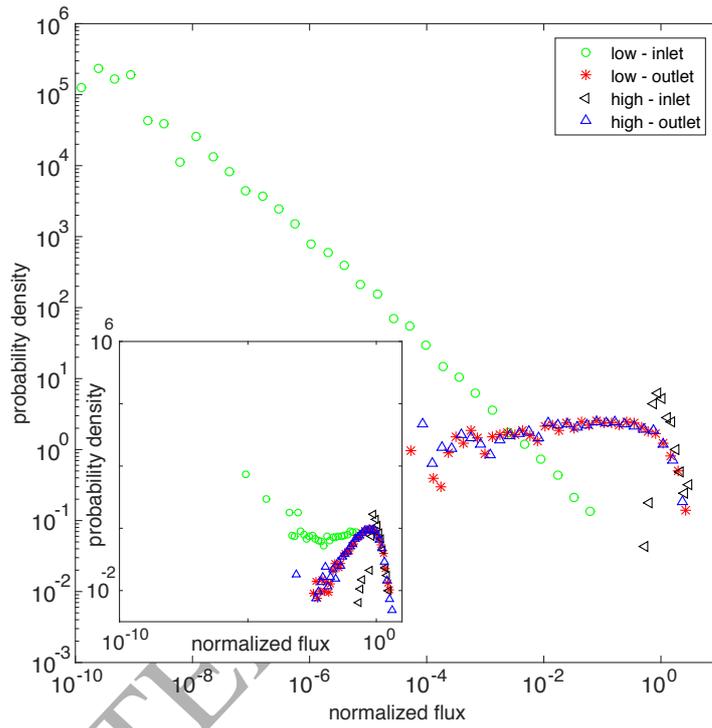


Figure 14: Lagrangian flux distributions at the inlet and outlet with two arbitrary initial velocity distributions for $\sigma_{\ln K} = 5$. The two initial velocity distributions come from uniform injections into regions of the 20-percentile highest, and 20-percentile lowest velocities. Flux values are normalized with the mean flux value. Note that flux distributions at outlet are identical regardless of the initial velocity distribution. Inset: same plot for $\sigma_{\ln K} = 1$.

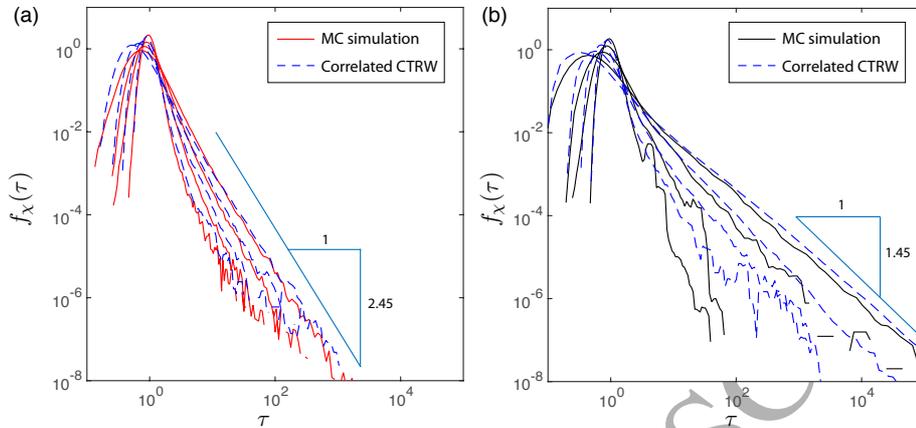


Figure 15: (a) Particle BTCs from Monte Carlo simulations for injection into *high-velocity* regions (solid line) and predictions from Markov-chain CTRW model with the reduced-order velocity transition matrix (dashed line). (b) Corresponding results for injection into *low-velocity* regions.

449 6. Conclusions

450 This study shows how the interplay between fracture geometrical prop-
 451 erties (conductivity distribution and network geometry) and tracer injection
 452 modes controls average particle transport via Lagrangian velocity statistics.
 453 The interplay between fracture heterogeneity and tracer injection methods
 454 can lead to distinctive anomalous transport behavior. Furthermore, the in-
 455 jection conditions, for example, uniform or flux-weighted, imply different
 456 initial velocity distributions, which can have a persistent impact on particle
 457 spreading through DFNs. For uniform injection, the s-Lagrangian velocity
 458 distribution evolves from an Eulerian velocity distribution initially to a sta-
 459 tionary s-Lagrangian distribution. In contrast, for flux-weighted injection,
 460 the s-Lagrangian velocity distribution remains stationary.

461 We have presented a spatial Markov model to quantify anomalous trans-
462 port through DFNs under arbitrary injection conditions. We derive an ana-
463 lytical relation between the stationary Lagrangian and the Eulerian velocity
464 distribution, and formally incorporate the initial velocity distribution into the
465 spatial Markov model. The proposed model accurately reproduces the evo-
466 lution of the Lagrangian velocity distribution for arbitrary injection modes.
467 This is accomplished with a reduced-order stochastic relaxation model that
468 captures the velocity transition with a single parameter: the effective ve-
469 locity correlation ℓ_c . The agreement between model predictions and direct
470 numerical simulations indicates that the simple velocity correlation model
471 can capture the dominant velocity correlation structure in DFNs.

472 In this study, we investigated the particle transport and the impact of
473 the injection condition for idealized 2D DFN using a Markov velocity model.
474 These findings can be extended to 3D DFNs, for which similar behaviors
475 regarding the injection mode have been found [58]. Also, Frampton and
476 Cvetkovic [16] reported similar velocity correlation structures for 3D DFNs
477 as in 2D, which suggests that a velocity Markov model such as the one
478 presented in this work can be used for the modelling of particle motion in
479 3D DFNs.

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491 Appendix A. Boltzmann Equation

The time derivative of (37) gives

$$\frac{\partial p(x, v, t)}{\partial t} = -v \frac{\partial p(x, v, t)}{\partial x} + R(x, v, t) - R(x - \bar{l}, v, t - \bar{l}/v). \quad (\text{A.1})$$

Note that $R(x, v, t)$ denotes the probability per time that a particle has the velocity v at the position x . It varies on a time scale of \bar{v} . Thus we can approximate (37) as

$$p(x, v, t) \approx \frac{\bar{l}}{v} R(x - \bar{l}, v, t - \bar{l}/v). \quad (\text{A.2})$$

Using this approximation and combining (A.1) with (38) gives for $t > 0$

$$\frac{\partial p(x, v, t)}{\partial t} = -v \frac{\partial p(x, v, t)}{\partial x} - \frac{v}{\bar{l}} p(x, v, t) + \int dv' r_1(v|v') \frac{v'}{\bar{l}} p(x, v', t). \quad (\text{A.3})$$

492

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