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1 **Anomalous transport on regular fracture networks: Impact of conductivity**
2 **heterogeneity and mixing at fracture intersections**

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11 **Abstract**

12 We investigate transport on regular fracture networks that are characterized by heterogeneity in hydraulic
13 conductivity. We discuss the impact of conductivity heterogeneity and mixing within fracture intersec-
14 tions on particle spreading. We show the emergence of non-Fickian transport due to the interplay between
15 the network conductivity heterogeneity and the degree of mixing at nodes. Specifically, lack of mixing
16 at fracture intersections leads to subdiffusive scaling of transverse spreading but has negligible impact on
17 longitudinal spreading. An increase in network conductivity heterogeneity enhances both longitudinal and
18 transverse spreading and leads to non-Fickian transport in longitudinal direction. Based on the observed
19 Lagrangian velocity statistics, we develop an effective stochastic model that incorporates the interplay be-
20 tween Lagrangian velocity correlation and velocity distribution. The model is parameterized with a few
21 physical parameters and is able to capture the full particle transition dynamics.

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23 I. INTRODUCTION

24 Understanding transport in network systems is of critical importance in many natural and en-
25 gineered processes, including groundwater contamination and geothermal production in fractured
26 geologic media [1, 2], disease spreading through river networks [3], engineered flows and medical
27 applications in microfluidic devices [4], and urban traffic [5]. While particle spreading has tra-
28 ditionally been described using a Fickian framework, anomalous transport—characterized by the
29 nonlinear scaling with time of the mean square displacement and the non-Gaussian scaling of so-
30 lute distributions and fluxes—has been widely observed in porous and fractured media at various
31 scales from pore [6–9] to column [10–12] to field scale [13–18]. The observation of anomalous
32 transport is not limited to porous and fractured media, and has been observed in many different
33 systems from diffusion of a molecule in a single cell to animal foraging patterns [19–21]. Pre-
34 dictability of the observed anomalous transport is essential because it controls the early arrival and
35 the long residence time of particles [22–24]. This becomes especially important for environmental
36 and human health related issues, such as radionuclide transport in the subsurface [25, 26], or water
37 quality evolution in managed aquifer recharge systems [27–29].

38 Stochastic models that account for the observed non-Fickian transport behavior in porous and
39 fractured media include continuous-time random walks (CTRW) [30–34], fractional advection-
40 dispersion equations (fADE) [35, 36], multirate mass transfer (MRMT) [17, 37, 38], stochastic
41 convective stream tube (SCST) models [39], and Boltzmann equation approaches [40]. All of
42 these models have played an important role in advancing the understanding of transport through
43 porous and fractured geologic media.

44 The CTRW formalism [41, 42] offers an attractive framework to describe and model anoma-
45 lous transport through porous media and networks [30, 43, 44] because it allows incorporating
46 essential flow heterogeneity properties directly through the Lagrangian velocity distribution. The
47 CTRW approach successfully described average transport in quenched random environments from
48 purely diffusive transport [e.g., 23] to biased diffusion [e.g., 45–48]. Most studies that employ the
49 CTRW approach assume that successive particle jumps are independent of each other, therefore
50 neglecting velocity correlation between jumps [49]. Indeed, a recent study showed that CTRW
51 with independent transition times emerges as an exact macroscopic transport model when particle
52 velocities are uncorrelated [48].

53 However, recent studies based on the analysis of Lagrangian particle trajectories demonstrates

54 conclusively that particle velocities in mass-conservative flow fields exhibit correlation along their
55 trajectory [9, 40, 45, 50–53]. Mass conservation induces correlation in the Eulerian velocity field
56 because fluxes must satisfy the divergence-free constraint at each intersection. This, in turn, in-
57 duces correlation in the Lagrangian velocity along a particle trajectory. To take into account ve-
58 locity correlation, Lagrangian models based on temporal [50, 54] and spatial [9, 40, 45, 51, 52]
59 Markovian processes have recently been proposed. These models successfully capture many im-
60 portant aspects of the particle transport behavior. The importance of velocity correlation has also
61 been recently shown for a field-scale tracer transport experiment [18].

62 Following the work by Le Borgne *et al.* [45], the spatial Markov model for particle velocities at
63 Darcy-scale has been recently extended to describe multidimensional transport at both pore- and
64 network-scale [9, 51]. The model captures multidimensional features of transport via a multidimensional
65 velocity transition matrix. In these approaches, the transition matrices are constructed
66 utilizing Lagrangian velocity information obtained from direct numerical simulations. Therefore,
67 for an effective parameterization in terms of the medium geometry and the statistical characteris-
68 tics of the Eulerian velocity, a model for the velocity transition process is crucial. Furthermore, it
69 is well known that the mixing at fracture intersections and fracture conductivity distribution has
70 major impact on transport properties [55–58]. However, the impact of the interplay between the
71 network conductivity heterogeneity and the mixing dynamics at fracture intersections on anoma-
72 lous transport, and the ability of spatial Markov models to capture it, is still an open question.

73 The paper proceeds as follows. In the next section, we present the heterogeneous fracture
74 network, the flow and transport equations and details of the different mixing rules at fracture
75 intersections. In Section III, we investigate the emergence of anomalous transport by direct Monte
76 Carlo simulations of flow and particle transport. In Section IV, we analyze the statistics of the
77 Lagrangian particle velocities measured equidistantly along the particle trajectories to gain insight
78 into the effective particle dynamics and elucidate the origins of the observed anomalous behavior.
79 In Section V, we develop a spatial Markov model that is characterized by the probability density
80 function (PDF) of Lagrangian velocities and their transition PDF, which are derived from the
81 Monte Carlo simulations. The resulting correlated CTRW model is in excellent agreement with
82 Monte Carlo simulations. In Section VI, we then present a physics-based spatial Markov model for
83 the velocity transitions that is characterized by only a few parameters, which are directly related to
84 the properties of the conductivity heterogeneity and the mixing rules at fracture intersections. The
85 predictive capabilities of this model are demonstrated by comparison to the direct Monte Carlo

86 simulations. In Section VII, we summarize the main findings and conclusions.

87 II. FLOW AND TRANSPORT THROUGH REGULAR FRACTURE NETWORKS

88 We study a regular fracture network consisting of two sets of parallel, equidistant fractures
89 oriented at an angle of $\pm\alpha$ with the x -axis. The distance between the neighboring nodes is l
90 (Figure 1). Flow through the network is modeled by Darcy's law [59] for the fluid flux u_{ij} between
91 nodes i and j , $u_{ij} = -K_{ij}(\Phi_j - \Phi_i)/l$, where Φ_i and Φ_j are the hydraulic heads at nodes i and
92 j , and $K_{ij} > 0$ is the hydraulic conductivity of the link between the two nodes. Imposing mass
93 conservation at each node i , $\sum_j u_{ij} = 0$ (the summation is over nearest-neighbor nodes), leads to
94 a linear system of equations, which is solved for the hydraulic heads at the nodes. The fluid flux
95 through a link from node i to j is termed incoming for node i if $u_{ij} < 0$, and outgoing if $u_{ij} > 0$.
96 We denote by \mathbf{e}_{ij} the unit vector in the direction of the link connecting nodes i and j . A realization
97 of the random regular network is generated by assigning independent and identically distributed
98 random conductivities to each link. Therefore, the K_{ij} values in different links are uncorrelated.
99 The set of all realizations of the quenched random network generated in this way forms a statistical
100 ensemble that is stationary and ergodic. We assign a lognormal distribution of K values. We study
101 the impact of conductivity heterogeneity on transport by varying the variance of $\ln(K)$. The use of
102 this particular distribution is motivated by the fact that conductivity values in many natural media
103 can be described by a lognormal law [60].

104 We study a uniform flow setting characterized by constant mean flow in the positive x -direction,
105 by imposing no-flow conditions at the top and bottom boundaries of the network, and fixed hy-
106 draulic head at the left ($\Phi = 1$) and right ($\Phi = 0$) boundaries. Thus, the mean flow velocity is
107 given by $\bar{u} = K_g$ where $K_g = \exp(\overline{\ln K})$ is the geometric mean conductivity. The overbar in the
108 following denotes the ensemble average over all network realizations. Even though the underly-
109 ing conductivity field is uncorrelated, the mass conservation constraint together with heterogeneity
110 leads to the formation of preferential flow paths with increasing network heterogeneity. (Figure 2).

111 Once the fluxes at the links have been determined, we simulate transport of a passive tracer by
112 particle tracking. We neglect the longitudinal diffusion along links, and thus particles are advected
113 with the mean flow velocity between nodes. Here we assume that Darcy's law is valid in each
114 link, which means we assume that the links are filled with a porous medium characterized by
115 variable conductivity, but constant porosity. This is motivated also by the fact that the variability

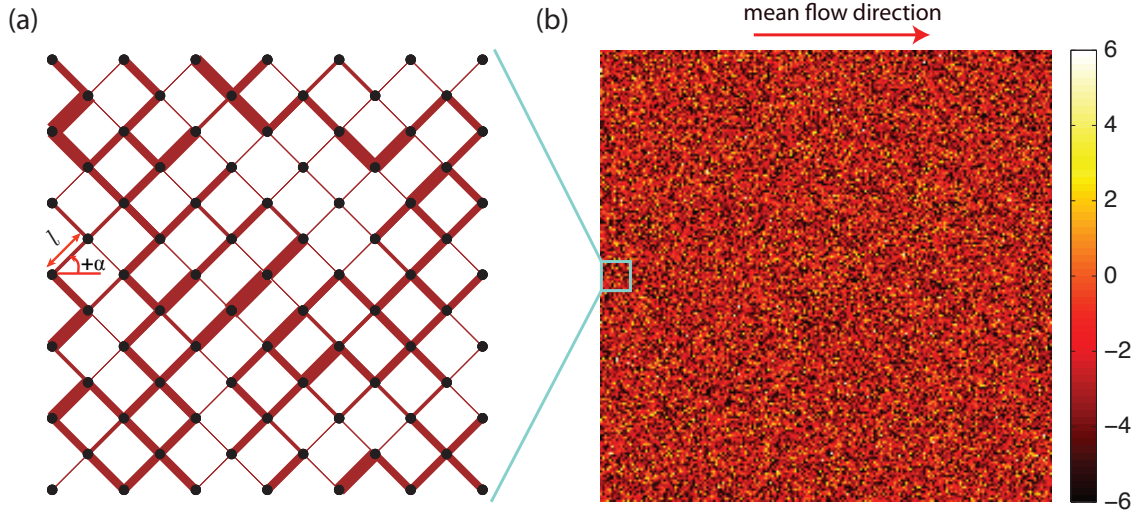


FIG. 1. (a) Schematic of the fracture network studied here, with two sets of links with orientation $\pm\alpha = \pm\pi/4$ and uniform spacing l . The conductivity values are reflected in the link thickness. We study log-normal conductivity distributions with three different conductivity variance values: $\sigma_{\ln K}^2 = 0.1, 1, 5$. (b) Map of the spatially uncorrelated conductivity field with $\sigma_{\ln K}^2 = 5$ shown in a log-scale color scheme. No flow boundary conditions on the top and bottom and constant hydraulic head on the left and right boundaries ensure a uniform mean flow.

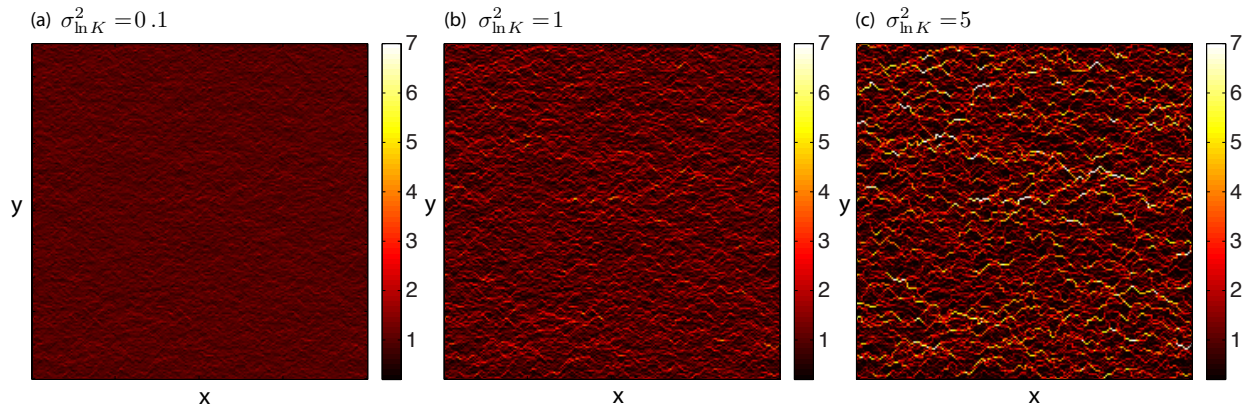


FIG. 2. (a) Normalized flow field ($|u_{ij}|/\bar{u}$) for log-normal conductivity distribution with variance 0.1. (b) Normalized flow field for log-normal conductivity distribution with variance 1. (c) Normalized flow field for log-normal conductivity distribution with variance 5. Even though the underlying conductivity field is uncorrelated, the combined effect of network heterogeneity and the mass conservation constraint at nodes leads to a correlated flow field with preferential flow paths.

116 in porosity is typically much smaller than the conductivity variability [59, 61]. When particles
 117 arrive at nodes, they follow either complete mixing or streamline routing (no mixing) rule [56–58].
 118 Complete mixing assumes that Péclet numbers at nodes are small enough that particles are well
 119 mixed within the node. Thus, the link through which the particle exits a node is chosen randomly
 120 with flux-weighted probability. Streamline routing assumes that Péclet numbers at nodes are large
 121 enough that particles essentially follow the streamlines and do not transition between streamlines.
 122 The complete mixing and streamline routing rules are two end members. In general, the local
 123 Péclet number and the intersection geometry determine the strength of mixing at nodes, which is
 124 in between these two end members.

125 In Figure 3 we illustrate the fundamental difference between the two mixing rules. When
 126 the two incoming and the two outgoing links have equal fluxes, the particles from the incoming
 127 link partition equally into the two outgoing links for the complete mixing rule. However, for the
 128 streamline routing case, all particles transit to the adjacent link. Therefore, we anticipate that the
 129 degree of mixing at the nodes will lead to a dramatically different global spreading behavior.

130 For complete mixing, the particle transfer probabilities p_{ij} from node i to node j are given by

$$p_{ij} = \frac{|u_{ij}|}{\sum_k |u_{ik}|}, \quad (1)$$

131 where the summation is over outgoing links only, and $p_{ij} = 0$ for incoming links. Particle
 132 transitions are determined only by the outgoing flux distribution. Equation (1) applies to both
 133 complete mixing and streamline routing rules for nodes with three outgoing fluxes and one in-
 134 coming. However, for nodes with two outgoing fluxes, streamline routing implies the transfer
 135 probabilities

$$p_{\text{adj}} = \begin{cases} 1, & u_{\text{adj}} > u_{\text{in}} \\ \frac{u_{\text{adj}}}{u_{\text{in}}}, & u_{\text{adj}} < u_{\text{in}} \end{cases} \quad p_{\text{opp}} = \begin{cases} 0, & u_{\text{adj}} > u_{\text{in}} \\ \frac{u_{\text{in}} - u_{\text{adj}}}{u_{\text{in}}}, & u_{\text{adj}} < u_{\text{in}}, \end{cases} \quad (2)$$

136 where p_{adj} is the probability of a particle transitioning to an adjacent link and p_{opp} is the probability
 137 of a particle transitioning to an opposite link (Figure 3).

138 The Langevin equations describing particle movements in space and time are

$$\mathbf{x}_{n+1} = \mathbf{x}_n + l \frac{\mathbf{v}(\mathbf{x}_n)}{|\mathbf{v}(\mathbf{x}_n)|}, \quad t_{n+1} = t_n + \frac{l}{|\mathbf{v}(\mathbf{x}_n)|}. \quad (3)$$

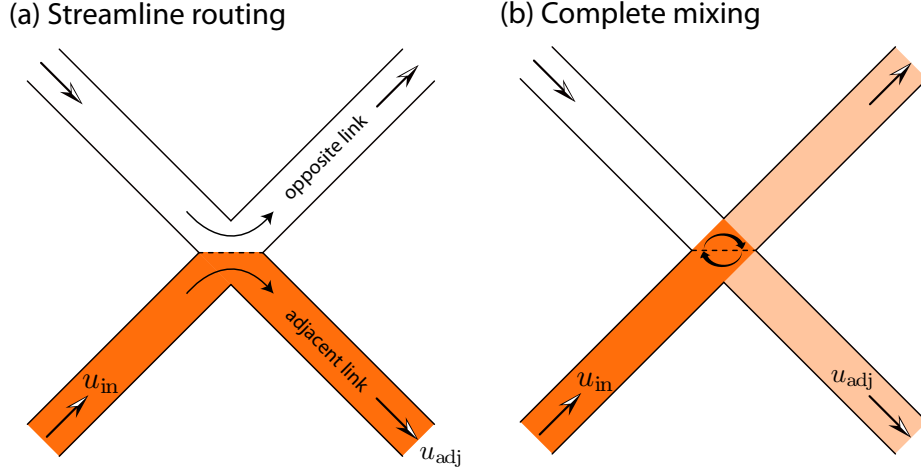


FIG. 3. Schematic for the two different mixing rules for the case in which nodes at the two incoming and the two outgoing links have equal fluxes. (a) The streamline routing rule makes all the particles transit to the adjacent link because particles cannot switch between streamlines. (b) The complete mixing rule makes half of the particles move upward and the other half downward, following flux-weighted probabilities.

139 where \mathbf{x}_n is the position of the n th node visited by the tracer particle, and t_n is the time at
 140 which the tracer particle arrives the n th node. The transition velocity is equal to $\mathbf{v}(\mathbf{x}_n) = u_{ij}\mathbf{e}_{ij}$
 141 with the transition probability p_{ij} following either Equation (1) or (2) depending on the mixing
 142 rule. The velocity vector \mathbf{v} in the following is expressed in (ν, θ) coordinates, in which $\nu =$
 143 $|v| \cos(\varphi)/|\cos(\varphi)|$ is the velocity along a link with $\varphi = \arccos(v_x/|v|)$ and $\theta = \sin(\varphi)/|\sin(\varphi)|$,
 144 so that $\mathbf{v} = [\nu \cos(\alpha), |\nu| \theta \sin(\alpha)]^T$. Superscript T denotes the transpose. Note that φ can only
 145 assume values in $\{-\alpha, \alpha, \pi - \alpha, \pi + \alpha\}$. In short, ν determines the velocity magnitude and longi-
 146 tudinal directionality and θ determines the transverse velocity directionality.

147 The system of discrete Langevin equations (3) describes coarse-grained particle transport for
 148 a single realization of the quenched random network. Particle velocities and thus transition times
 149 depend on the particle position. The particle position at time t is $\mathbf{x}(t) = \mathbf{x}_{n_t}$, where n_t denotes the
 150 number of steps needed to reach time t . The particle density in a single realization is $P(\mathbf{x}, t) =$
 151 $\langle \delta(\mathbf{x} - \mathbf{x}_{n_t}) \rangle$, where the angular brackets denote the noise average over all particles. We solve
 152 transport in a single disorder realization by particle tracking based on Equation (3) with the point-
 153 wise initial condition $\mathbf{x}_0 = \mathbf{0}$ and $t_0 = 0$. \mathbf{x}_0 is located at the center of the left boundary (marked
 154 by red star in Figure 4). As shown in Figure 4, both network heterogeneity and mixing rule at
 155 nodes have significant impact on particle spreading. An increase in network heterogeneity leads

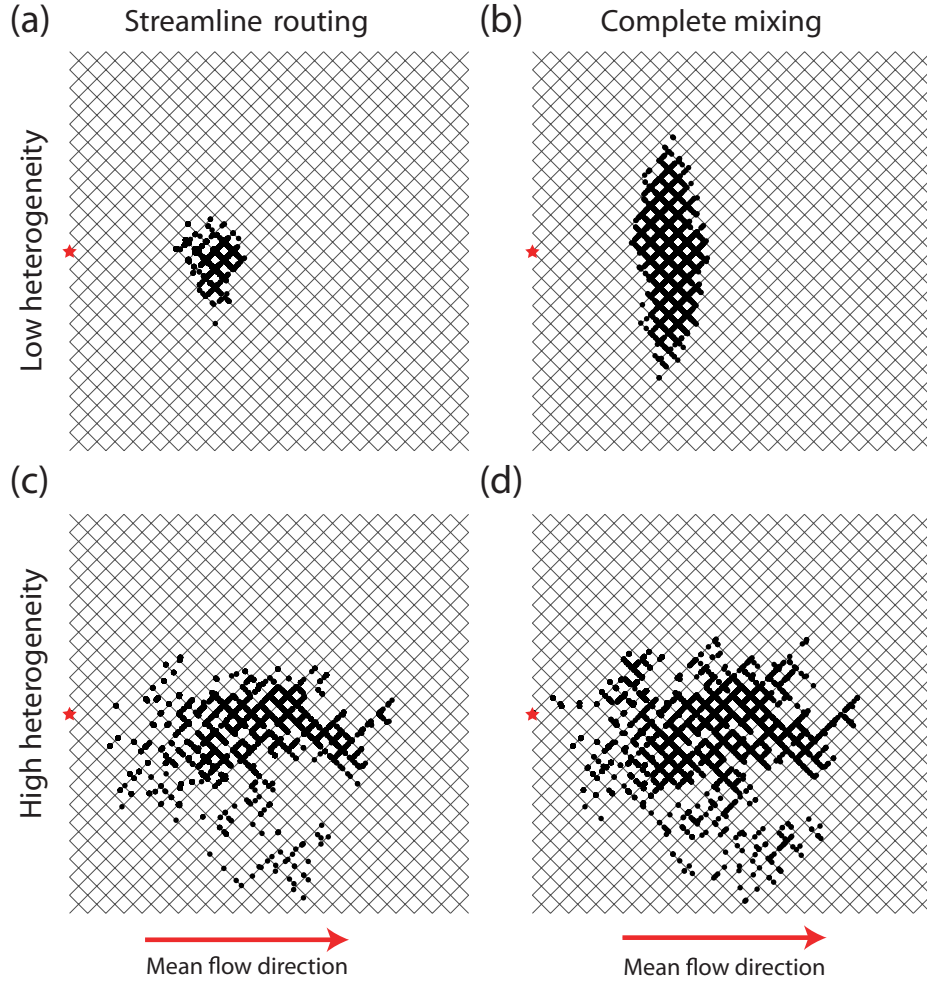


FIG. 4. Particle distribution at $t = 15\bar{t}_l$ for a given realization after the instantaneous release of particles at the origin (red star). \bar{t}_l is the mean advective time along one link. (a) Low heterogeneity ($\sigma_{\ln K}^2 = 0.1$) with streamline routing at nodes. (b) Low heterogeneity ($\sigma_{\ln K}^2 = 0.1$) with complete mixing at nodes. (c) High heterogeneity ($\sigma_{\ln K}^2 = 5$) with streamline routing at nodes. (d) High heterogeneity ($\sigma_{\ln K}^2 = 5$) with complete mixing at nodes. For low heterogeneity, complete mixing significantly enhances transverse spreading. An increase in heterogeneity significantly enhances longitudinal spreading.

156 to an increase in particle spreading in both transverse and longitudinal directions. The impact
 157 of the mixing rule has a significant impact on transverse mixing, especially for networks with
 158 low heterogeneity. Complete mixing at nodes significantly enhances transverse spreading while
 159 longitudinal spreading is much less sensitive to the mixing rule.

160 **III. AVERAGE SOLUTE SPREADING BEHAVIOR**

161 We study the average solute spreading behavior for three different conductivity variances and
 162 the two mixing rules described above. We obtain the mean particle density, $\overline{P}(\mathbf{x}, t)$, by ensemble
 163 averaging over multiple realizations,

$$\overline{P}(\mathbf{x}, t) = \overline{\langle \delta(\mathbf{x} - \mathbf{x}_{n_t}) \rangle}, \quad (4)$$

164 where the overbar denotes the ensemble average over all realizations. We run Monte Carlo
 165 particle tracking simulations for 10^2 realizations for each combination of conductivity variance
 166 and mixing rule. We consider three different $\ln(K)$ variances, $\sigma_{\ln K}^2 = 0.1, 1, 5$. The domain size
 167 is $100\sqrt{2}l \times 100\sqrt{2}l$ with 20, 201 nodes. In each realization, we release 10^4 particles at the origin
 168 (\mathbf{x}_0 , marked by a red star in Figure 4). The average particle spreading behavior is studied in terms
 169 of the mean square displacement (MSD) of average particle density, $\overline{P}(\mathbf{x}, t)$. For the longitudinal
 170 direction (x), the MSD is given by $\sigma_x^2(t) = \overline{\langle [x(t) - \langle x(t) \rangle]^2 \rangle}$ where $\langle \cdot \rangle$ denotes the average over
 171 all particles for a given realization. The same definition is applied to compute the transverse MSD,
 172 σ_y^2 .

173 In Figure 5, we show the time evolution of the longitudinal and transverse MSDs. In both
 174 directions, spreading shows a ballistic regime ($\sim t^2$) at early times, which then transitions to a
 175 different preasymptotic scaling in an intermediate regime. The transition occurs approximately at
 176 the mean advective time over one link, \overline{t}_l .

177 The Monte Carlo simulations show that, in the intermediate regime, the *longitudinal* MSD
 178 increases linearly with time for weak conductivity heterogeneity [Figure 5(a)], and faster than
 179 linearly (i.e., superdiffusively) for intermediate to strong heterogeneity [Figure 5(c)(e)]. An in-
 180 crease in $\ln(K)$ variance significantly increases the longitudinal MSD and induces a change in its
 181 temporal scaling. The Monte Carlo simulations also show that there is no noticeable difference
 182 between complete mixing and streamline routing cases on longitudinal MSD. This indicates that
 183 the network heterogeneity dictates the longitudinal spreading in regular networks.

184 The *transverse* MSD evolves linearly in time for complete mixing, and slower than linearly
 185 with time (i.e., subdiffusively) for streamline routing [Figure 5(b)(d)(f)]. In contrast with the lon-
 186 gitudinal MSD, the transverse MSD exhibits a strong dependence on the mixing rule at fracture
 187 intersections. For low heterogeneity, complete mixing induces a significantly higher transverse

188 MSD than streamline routing. This difference, however, decreases as the network heterogeneity
 189 increases. For streamline routing, the network heterogeneity is the main driver for transitions in
 190 the transverse direction, and thus we clearly observe that transverse spreading increases as hetero-
 191 geneity increases. The complete mixing rule, on the other hand, already maximizes transitions in
 192 the transverse direction so that an increase in heterogeneity has no significant impact.

193 In order to obtain complementary information on the spreading process, we also consider the
 194 first passage time distribution (FPTD) of particles at a control plane $x = \chi$, which acts like an
 195 absorbing barrier. The FPTD or, in other words, solute breakthrough curve, is obtained from the
 196 individual particle arrival times $\tau_a = \inf(t_n | |x_n - x_0| > \chi)$ as

$$f_\chi(\tau) = \overline{\langle \delta(\tau - \tau_a) \rangle}. \quad (5)$$

197 It provides an alternative measure of longitudinal spreading. Figure 6 illustrates FPTDs for
 198 different conductivity heterogeneities and mixing rules. Conductivity heterogeneity has a clear
 199 impact on the FPTD by enhancing longitudinal spreading. This is so because stronger conductivity
 200 heterogeneity leads to broader particle transition time distribution, which in turn leads to enhanced
 201 longitudinal spreading. The mixing rule, in contrast, has a negligible impact on FPTDs, and
 202 only influences transverse spreading. To understand this behavior and further quantify transverse
 203 spreading, we define the distribution of the transverse exit locations at a control plane $x = \chi$ as

$$f_\chi(\omega) = \overline{\langle \delta(\omega - y_e) \rangle}. \quad (6)$$

204 where y_e is the transverse location of a particle at the control plane at $x = \chi$. The impact of
 205 the mixing rule on transverse spreading is clearly visible in Figure 7, which compares $f_\chi(\omega)$ for
 206 different values of $\sigma_{\ln K}^2$ and different mixing rules. For small $\ln(K)$ variances, the mixing rule has
 207 a major impact on transverse spreading, which here is manifested by the width of the transverse
 208 particle distribution. The difference between the two mixing rules decreases as $\sigma_{\ln K}^2$ increases.

209 In summary, conductivity heterogeneity impacts both longitudinal and transverse spreading,
 210 whereas the mixing rule mainly impacts transverse spreading. We now analyze the Lagrangian
 211 particle statistics to understand the underlying physical mechanisms that lead to the observed
 212 anomalous particle spreading.

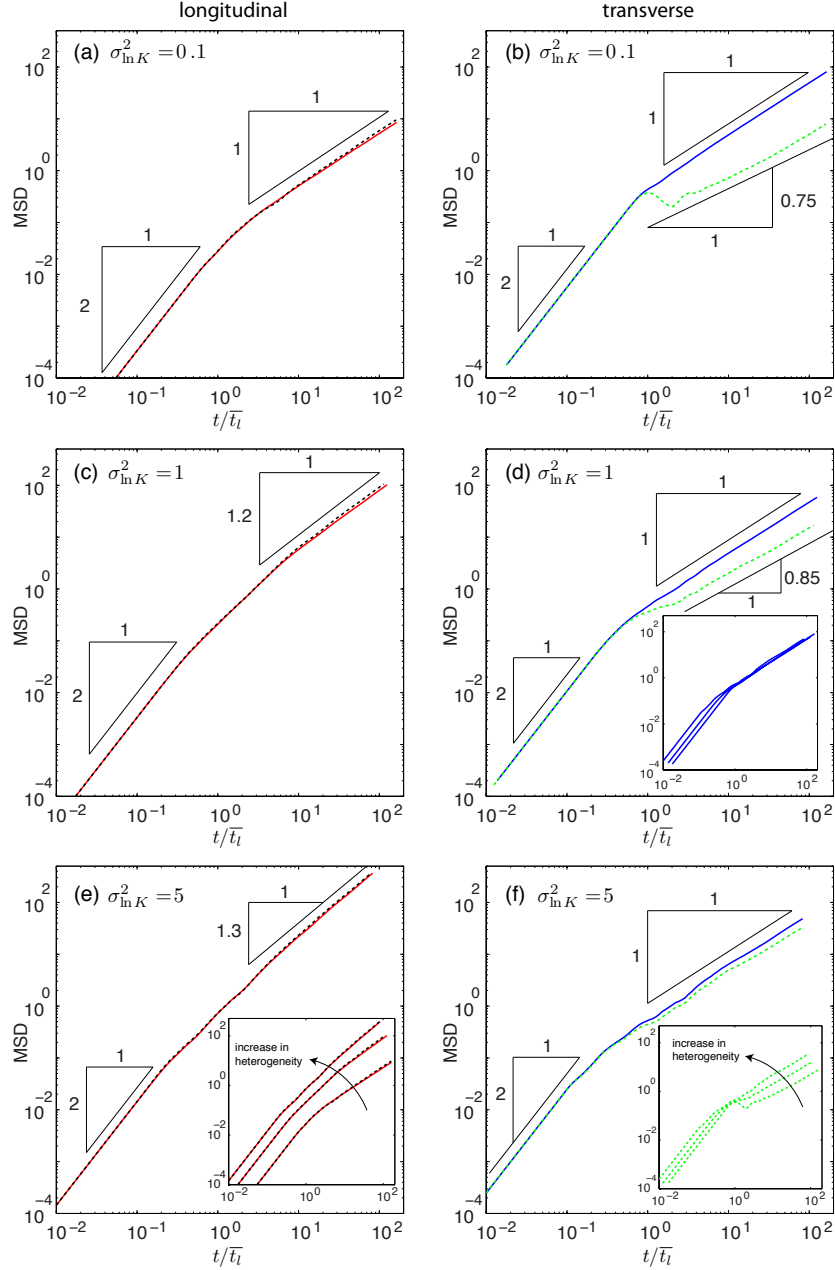


FIG. 5. Time evolution of MSDs for complete mixing (solid line) and streamline routing (dashed line). (a) Longitudinal MSD for $\sigma_{\ln K}^2 = 0.1$. (b) Transverse MSD for $\sigma_{\ln K}^2 = 0.1$. (c) Longitudinal MSD for $\sigma_{\ln K}^2 = 1$. (d) Transverse MSD for $\sigma_{\ln K}^2 = 1$. Inset: Change in the time evolution of transverse MSD for complete mixing with increasing variance. (e) Longitudinal MSD with $\sigma_{\ln K}^2 = 5$. Inset: Change in the time evolution of longitudinal MSD for complete mixing with increasing variance. (f) Transverse MSD with $\sigma_{\ln K}^2 = 5$. Inset: Change in the time evolution of transverse MSD for streamline routing with increasing conductivity variance.

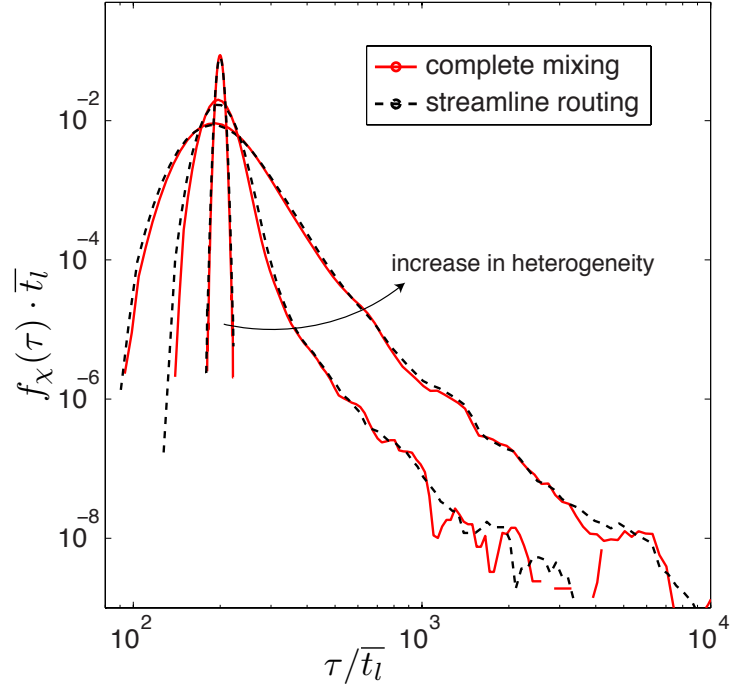


FIG. 6. First passage time distribution $f_X(\tau)$ for $\sigma_{\ln K}^2 = 0.1, 1, 5$ and different mixing rules. Conductivity heterogeneity has a major impact on particle breakthrough curves, in contrast to the mixing rules.

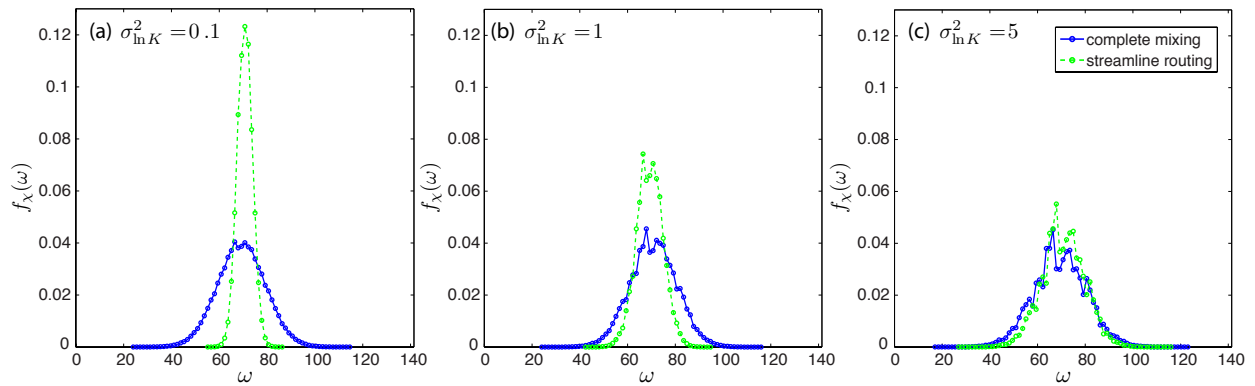


FIG. 7. Transverse breakthrough positions distribution (TBPD) at the outlet plane. Comparison between the two different mixing rules for (a) $\sigma_{\ln K}^2 = 0.1$, (b) $\sigma_{\ln K}^2 = 1$ and (c) $\sigma_{\ln K}^2 = 5$. The impact of the mixing rule on transverse spreading diminishes as the network heterogeneity increases.

213 **IV. LAGRANGIAN VELOCITY DISTRIBUTION AND VELOCITY CORRELATION STRUC-**
 214 **TURE**

215 The mechanisms leading to anomalous transport can be understood through the analysis of
 216 the statistics of Lagrangian particle velocities [9, 18, 45, 50–52]. We consider here the particle
 217 velocities at fixed positions along their trajectories. The Lagrangian velocity $\mathbf{v}_L(s_n)$ at a distance
 218 $s_n = nl$ along the particle trajectory is given by $\mathbf{v}_L(s_n) = \mathbf{v}(\mathbf{x}_n)$ with \mathbf{x}_n the particle position
 219 in the network after n steps. Its absolute value, i.e., the streamwise velocity is $v_L(s_n) = |\mathbf{v}(s_n)|$.
 220 We now analyze the Lagrangian velocity correlation structure, and the PDF of transition times
 221 between nodes along trajectories, which is given by $\tau_n = l/v_L(s_n)$.

222 This is in contrast with the classical Lagrangian viewpoint, which considers particle velocities
 223 at fixed times along trajectories, $\mathbf{u}_L(t) = \mathbf{v}(\mathbf{x}_{n_t})$ where n_t is the number of steps needed to arrive
 224 at time t through the time process in Equation (3). The distance covered along the streamline
 225 up to time t then is given by $s(t) = n_t l$, and the streamwise Lagrangian velocity is given by
 226 $u_L(t) = |\mathbf{v}(\mathbf{x}_{n_t})|$.

227 We compute the steady-state transition time and velocity distributions along streamlines, $\psi_\tau(t)$
 228 and $p_L(v)$, respectively, through sampling the transition times and velocities along all particle
 229 trajectories and among network realizations. Figure 8(a) illustrates the PDF of transition times and
 230 velocities for different $\ln(K)$ variances. As $\sigma_{\ln K}^2$ increases, the transition time and velocity PDFs
 231 become broader. The transition time follows closely a truncated power-law distribution. Both
 232 velocity and transition time distributions did not show noticeable difference between complete
 233 mixing [Figure 8(a)] and streamline routing [not shown].

234 A broad transition time distribution is known to be a source of anomalous transport behavior,
 235 and a key input parameter for the CTRW framework [31, 43]. For example, an optimal distri-
 236 bution of transition times may be inferred by interpreting first-passage time distributions [49].
 237 However, the transition time distribution alone does not have information on the spatial velocity
 238 correlation structure, which may be an important factor that controls anomalous transport behav-
 239 ior [9, 18, 45, 47, 51, 52]. To analyze the Lagrangian correlation structure, we compute the velocity
 240 autocorrelation function.

The autocorrelation function for a given lag $\Delta s = s - s'$ is defined as

$$\chi_s(s', s' + \Delta s) = \frac{\langle [v_L(s' + \Delta s) - \langle v_L(s' + \Delta s) \rangle][v_L(s') - \langle v_L(s') \rangle] \rangle}{\sigma_v(s' + \Delta s)\sigma_v(s')}, \quad (7)$$

where $\sigma_v^2(s)$ is the variance of the Lagrangian velocity at a travel distance s . It depends in general on the starting position depending on the distribution of initial particle velocities. Here, particles are injected at the origin within each realization. This implies that particles sample uniformly from the heterogeneous flow velocity. The stationary streamwise velocity distribution, in contrast, is obtained by spatial sampling along particle pathlines. As a consequence, here, the correlation function depends on the starting point s' . However, with increasing streamwise distance from the injection point, the autocorrelation becomes stationary. Thus, we define the stationary autocorrelation function $\chi_s(s - s')$ by averaging over Eq. (7) as

$$\chi_s(\Delta s) = \frac{1}{a} \int_0^a ds_1 \chi_s(s_1, s_1 + \Delta s), \quad (8)$$

241 where we use $a = 100\ell$.

242 For comparison, we also consider the correlation of Lagrangian velocities $u_L(t)$ sampled *in*
243 *time* along particle trajectories. It is defined analogously as

$$\chi_t(t - t') = \frac{1}{T} \int_0^T dt' \frac{\langle [u_L(t') - \langle u_L(t') \rangle][u_L(t' + \Delta t) - \langle u_L(t' + \Delta t) \rangle] \rangle}{\sigma_u(t' + \Delta t)\sigma_u(t')}, \quad (9)$$

244 where $\Delta t = t - t'$. Figure 8(b) illustrates the Lagrangian autocorrelation function $\chi_s(s)$ for
245 different $\ln(K)$ variances with a complete mixing rule. The correlation length scale ℓ_c is defined
246 by

$$\ell_c = \int_0^\infty ds \chi_s(s). \quad (10)$$

247 The correlation function $\chi_s(s)$ is well represented by an exponential that is characterized by ℓ_c .
248 Under the complete mixing rule, we find that ℓ_c increases as the network heterogeneity increases,
249 indicating an increase in velocity correlation ($\ell_c = 1.01, 1.34, 2.13$ for $\sigma_{\ln K}^2 = 0.1, 1, 5$, respec-
250 tively). This is mainly due to the emergence of preferential flow paths, as shown in Figure 2. The
251 inset in Figure 8(b) compares the correlation functions $\chi_s(s)$ and $\chi_t(t)$ plotted against distance
252 normalized by the link length l and time normalized by the mean advection time across a link, for
253 $\sigma_{\ln K}^2 = 5$. Velocity correlation in time is significantly stronger than velocity correlation in space,
254 and closely follows a power law with slope 0.7. The reason for this slow decay in the temporal

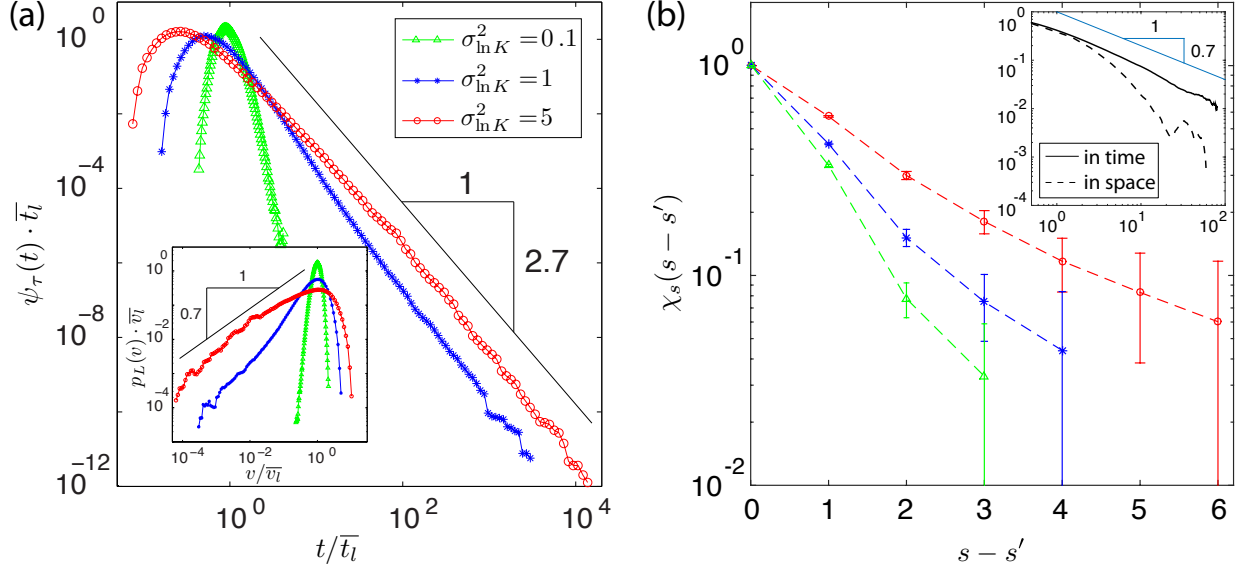


FIG. 8. (a) Lagrangian transition time distributions for $\sigma_{\ln K}^2 = 0.1, 1, 5$ and complete mixing at the nodes. Inset: Lagrangian velocity distributions for the three different values of $\sigma_{\ln K}^2$. As the network conductivity becomes more heterogeneous, both the transition time distribution and the velocity distribution become broader. (b) Velocity autocorrelation function in space. Error bars represent the coefficient of variation. An increase in network heterogeneity leads to stronger correlation. Inset: Comparison between the velocity autocorrelation in space and in time for $\sigma_{\ln K}^2 = 5$. Velocity autocorrelation in time is normalized with the mean advective time along one link, and velocity autocorrelation in space is normalized with the link length.

255 velocity correlation structure is the contribution from particles at stagnation zones (links with very
 256 small velocity values).

257 To further analyze and characterize the (spatial) Lagrangian velocity series $\{v(s_n)\}$, we com-
 258 pute the velocity transition matrix. To this end, we determine the transition probability density to
 259 encounter a velocity \mathbf{v} after $n + m$ steps given that the particle velocity was \mathbf{v}' after n steps, which
 260 in the variables (ν, θ) reads as

$$r_m(\nu, \theta | \nu', \theta') = \overline{\langle \delta[\nu - \nu(\mathbf{x}_{n+m})] \delta_{\theta, \theta(\mathbf{x}_{n+m})} \rangle}_{\nu(\mathbf{x}_n) = \nu', \theta(\mathbf{x}_n) = \theta'}. \quad (11)$$

261 To evaluate the transition probability numerically, the particle velocity distribution is dis-
 262 cretized into classes, $\nu \in \bigcup_{j=1}^N (\nu_j, \nu_{j+1}]$, with $N = 100$. We may discretize velocity equiprobably
 263 in linear or logarithmic scale. The logarithmic scale provides a better discretization for low veloc-
 264 ities, which have a decisive role for the occurrence of anomalous transport because they determine

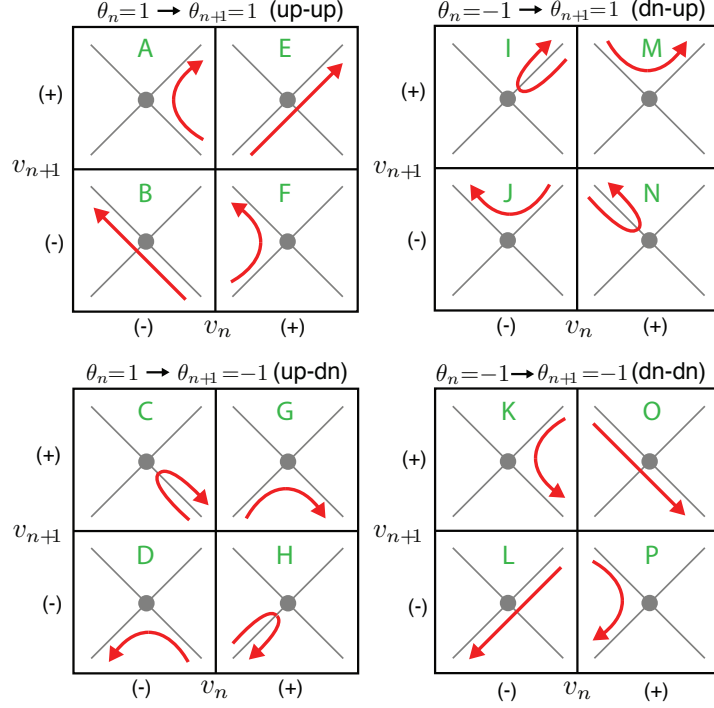


FIG. 9. Schematic of the velocity transition matrix for $d = 2$ dimensional networks. The transition matrix considers all 16 possible transitions to capture the full particle transport dynamics. The matrix has information about the one-step correlation, directionality and velocity heterogeneity.

265 the tailing behavior in FPTDs and spatial profiles. High velocities may be represented by only a
 266 few characteristic values. We define the transition probability matrix

$$T_m(i, \theta | j, \theta') = \frac{\int_{\nu_i}^{\nu_{i+1}} d\nu \int_{\nu_j}^{\nu_{j+1}} d\nu' r_m(\nu, \theta | \nu', \theta') p(\nu', \theta')}{\int_{\nu_j}^{\nu_{j+1}} d\nu' p(\nu', \theta')}, \quad (12)$$

267 where $p(\nu, \theta) = \overline{\langle \delta[\nu - \nu(\mathbf{x}_n)] \delta_{\theta, \theta(\mathbf{x}_n)} \rangle}$ is the joint single point PDF of ν and θ .

268 The transition matrices can be obtained numerically from the ensemble of particle trajectories.
 269 In $d = 2$ dimensional networks, there are sixteen possible transitions, which are described by a
 270 multi-dimensional transition matrix (Figure 9). We measure particle velocity transitions from link
 271 to link (equidistance in space) and populate the respective entries in the transition matrix. The one-
 272 step transition matrices $T_1(i, \theta | j, \theta')$ for two different heterogeneity distributions and mixing rules
 273 with equiprobable binning are shown in Figures 10 and 11. For small heterogeneity ($\sigma_{\ln K}^2 = 0.1$,
 274 Figure 10), the difference in the transition matrix for complete mixing and streamline routing at
 275 nodes is significant. This difference diminishes as heterogeneity increases ($\sigma_{\ln K}^2 = 5$, Figure 11).

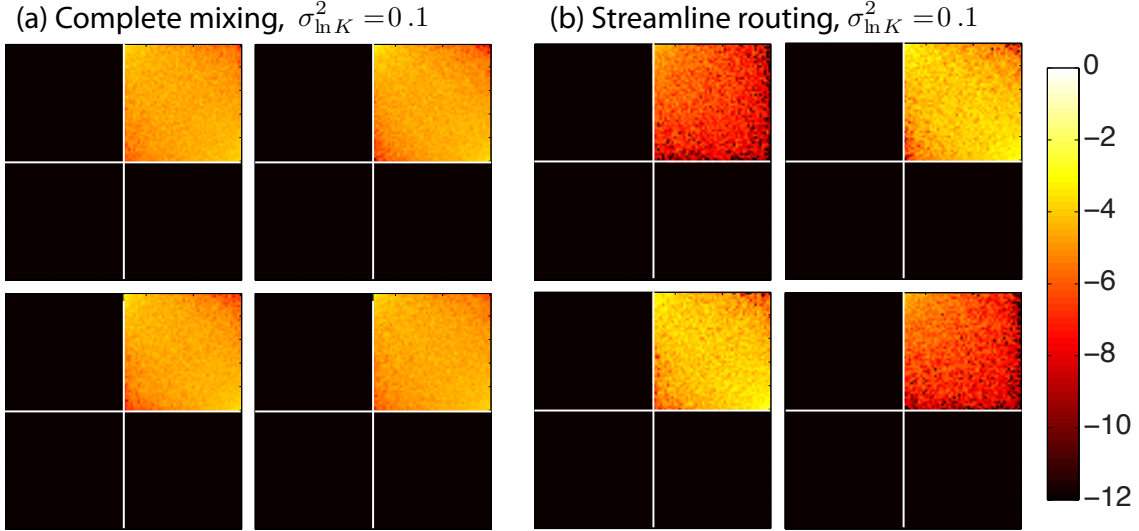


FIG. 10. (a) Velocity transition matrix with linear equiprobable binning for $\sigma_{\ln K}^2 = 0.1$ and *complete mixing* at nodes. Out of 16 transitions, only the four that have forward-forward movement in longitudinal direction (E, M, G, O) are possible. Note that the probability for each possible transition is almost identical. (b) Velocity transition matrix with linear equiprobable binning for $\sigma_{\ln K}^2 = 0.1$ with *streamline routing*. Again, only the four transitions that have forward-forward movement in longitudinal direction (E, M, G, O) are possible. Also, note that the probability for M and G transitions (0.89) is significantly higher than E and O transitions (0.11).

276 Network heterogeneity also exerts a significant impact on the particle transition matrix: as conduc-
 277 tivity distribution becomes more heterogeneous, the probability of transitions with flow reversal
 278 (negative x -direction) increases. Higher probability values along the diagonal of the transition
 279 matrix reflect the spatial velocity correlation. Similarly, the upper triangular and lower triangu-
 280 lar matrices in the transitions with backward movement (A, F, K, P) indicate that the velocity
 281 magnitude is typically smaller for backward movements than for forward movements.

282 The clear differences between transition time matrices for different mixing rules indicate the
 283 importance of taking the directionality of particle transport into account. Nonlocal theories of
 284 transport, including CTRW, are often invoked to explain the observation that the first passage
 285 time distribution (FPTD) is broad-ranged [16–18, 43]. Early arrival and slow decay of the FPTD
 286 is also observed in our model system. To develop a predictive transport model for the observed
 287 average particle density $\bar{P}(\mathbf{x}, t)$, we study average particle movements from a CTRW point of view
 288 that incorporates the velocity correlation and velocity distribution (heterogeneity). This approach

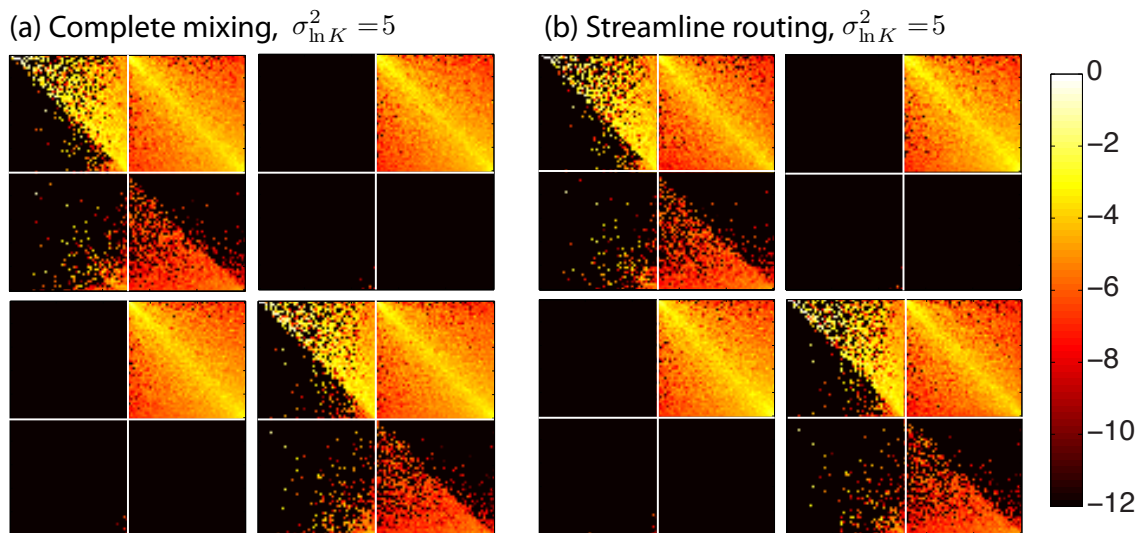


FIG. 11. (a) Velocity transition matrix with linear equiprobable binning for $\sigma_{\ln K}^2 = 5$ and *complete mixing* at the nodes. Due to strong heterogeneity, 12 different transitions, including backward movements, are possible. Also, note that up-up and down-down transitions (A, F, K, P) have triangular matrices. This indicates that velocity magnitudes mostly increase when a particle changes direction from $-x$ direction to $+x$ direction and vice versa. (b) Velocity transition matrix with linear equiprobable binning for $\sigma_{\ln K}^2 = 5$ and *streamline routing*. Since strong heterogeneity dictates particle transitions, there is no significant difference between complete mixing and streamline routing.

289 has been recently proposed for lattice fracture networks based on the finding that the series of
 290 particle velocities $\{\mathbf{v}_L(s_n)\}$ sampled spatially along a particle trajectory form in fact a Markov
 291 process [51].

292 V. SPATIAL MARKOV MODEL: A CORRELATED CONTINUOUS TIME RANDOM WALK

293 The series of Lagrangian velocities $\{\mathbf{v}_L(s_n) \equiv \mathbf{v}_n\}$ along particle trajectories can be approx-
 294 imated as a Markov process if the transition matrix satisfies the Chapman-Kolmogorov equa-
 295 tion [e.g., 62], which in matrix form reads as

$$T_n(i, \theta | j, \theta') = \sum_{i', \theta''} T_{n-m}(i, \theta | i', \theta'') T_m(i', \theta'' | j, \theta'). \quad (13)$$

296 For a Markov process, the m -step transition matrix \mathbf{T}_m is equal to the m -fold product of the

297 1-step transition matrix \mathbf{T}_1 with itself as $\mathbf{T}_m = \mathbf{T}^m$. Recent studies have shown that the spatial
 298 Markov model accurately predicts the transition probabilities, as well as the return probability for
 299 any number of steps [45, 51]. Therefore, a CTRW characterized by a Markov velocity process *in*
 300 *space* is a good approximation for describing average transport.

301 The average particle movements on the random network can be described by the following
 302 system of equations

$$\mathbf{x}_{n+1} = \mathbf{x}_n + l \frac{\mathbf{v}_n}{|\mathbf{v}_n|}, \quad t_{n+1} = t_n + \frac{l}{|\mathbf{v}_n|}. \quad (14)$$

303 The series of Lagrangian velocities $\{\mathbf{v}_n\}_{n=0}^{\infty}$ is a spatial Markov process and thus fully char-
 304 acterized by the stationary velocity density $p_s(\mathbf{v})$ and the one-step transition PDF $r_1(\mathbf{v}|\mathbf{v}') =$
 305 $\langle \delta(\mathbf{v} - \mathbf{v}_{n+1}) \rangle |_{\mathbf{v}_n=\mathbf{v}'}$. The particle density for the correlated CTRW (14) can be written as

$$P(\mathbf{x}, t) = \int d\mathbf{v} \langle \delta(\mathbf{x} - \mathbf{x}_{n_t}) \delta(\mathbf{v} - \mathbf{v}_{n_t}) \rangle, \quad (15)$$

306 in which $n_t = \max(n | t_n \leq t)$, \mathbf{x}_{n_t} is the position of the node at which the particle is at time t ,
 307 and \mathbf{v}_{n_t} is the velocity by which the particle emanates from this node. The angular brackets denote
 308 here the average over all realization of the stochastic velocity time series $\{\mathbf{v}_n\}$. Equation (15) can
 309 be recast as

$$P(\mathbf{x}, t) = \int d\mathbf{v} \int_{t-l/|\mathbf{v}|}^t dt' R(\mathbf{x}, \mathbf{v}, t'), \quad (16a)$$

in which we defined

$$R(\mathbf{x}, \mathbf{v}, t') = \sum_{n=0}^{\infty} \langle \delta(\mathbf{x} - \mathbf{x}_n) \delta(\mathbf{v} - \mathbf{v}_n) \delta(t' - t_n) \rangle. \quad (16b)$$

The latter satisfies the Kolmogorov type equation

$$R(\mathbf{x}, \mathbf{v}, t) = \delta(\mathbf{x}) p_0(\mathbf{v}) \delta(t) + \int d\mathbf{v}' r_1(\mathbf{v}|\mathbf{v}') \int d\mathbf{x}' \delta(\mathbf{x} - \mathbf{x}' - l\mathbf{v}'/|\mathbf{v}'|) R(\mathbf{x}', \mathbf{v}', t - l/|\mathbf{v}'|), \quad (16c)$$

310 where $p_0(\mathbf{v})$ denotes the distribution of initial particle velocities at step 0. For the injection con-
 311 dition applied here, the initial velocities are sampled uniformly among the network realizations.
 312 Thus, $p_0(\mathbf{v})$ is not equal to the stationary velocity PDF $p_s(\mathbf{v})$, which is obtained by sampling the

313 velocities equidistantly along a particle path, as outlined above. The correlated CTRW model (16)
 314 describes the evolution from an initial PDF $p_0(\mathbf{v})$ towards the steady state PDF through the transi-
 315 tion matrix $r_1(\mathbf{v}|\mathbf{v}')$.

316 For independent successive velocities, i.e., $r_1(\mathbf{v}|\mathbf{v}') = p(\mathbf{v})$, one recovers the CTRW model [e.g.,
 317 41]

$$P(\mathbf{x}, t) = \int_0^t dt' R(\mathbf{x}, t') \int_{t-t'}^\infty d\tau \int d\mathbf{x} \psi(\mathbf{x}, \tau), \quad (17a)$$

where $R(\mathbf{x}, t)$ satisfies

$$R(\mathbf{x}, t) = \delta(\mathbf{x})\delta(t) + \int d\mathbf{x}' \int_0^t dt' R(\mathbf{x}', t') \psi(\mathbf{x} - \mathbf{x}', t - t'), \quad (17b)$$

and the joint transition length and time density is given by

$$\psi(\mathbf{x}, t) = \int d\mathbf{v}' p(\mathbf{v}') \delta(\mathbf{x} - l\mathbf{v}'/|\mathbf{v}'|) \delta(t - l/|\mathbf{v}'|). \quad (17c)$$

318 In the following, we refer to system (16) as *correlated* CTRW because subsequent particle
 319 velocities are correlated in space, and to model (17) as *uncorrelated* CTRW because subsequent
 320 particle velocities are uncorrelated in space.

321 Based on the Markovianity assumption of particle transitions, the developed correlated CTRW
 322 model is applied to study particle transport in the random network. We compare the results ob-
 323 tained from direct Monte Carlo simulations to both the correlated and uncorrelated CTRW models.
 324 Correlated CTRW is characterized by the one-step transition matrix \mathbf{T}_1 determined from numer-
 325 ical Monte Carlo simulations [Figures 10 and 11]. Uncorrelated CTRW is characterized by the
 326 Lagrangian velocity distribution $p(\mathbf{v})$, which is obtained from Monte Carlo simulations as well.

327 The predictions of the developed correlated CTRW model show an excellent agreement with
 328 the Monte Carlo simulations for all levels of heterogeneity and mixing rules under consideration
 329 [Figures 12, 13(a)(b)(c), and 14(a)]. Note that the direct Monte Carlo simulations are performed by
 330 solving Equation (3) in 100 realizations for different mixing rules. Correlated CTRW captures the
 331 time evolution of the particle plume with remarkable accuracy, including spatial moments, first
 332 passage time distributions and distributions of transverse particle breakthrough positions. Fig-
 333 ure 12 shows the time evolution of the longitudinal and transverse MSDs. Both the scaling and the
 334 magnitude of the longitudinal spreading are captured accurately by the correlated CTRW model.
 335 The model also reproduces accurately the magnitude and evolution of the transverse MSD.

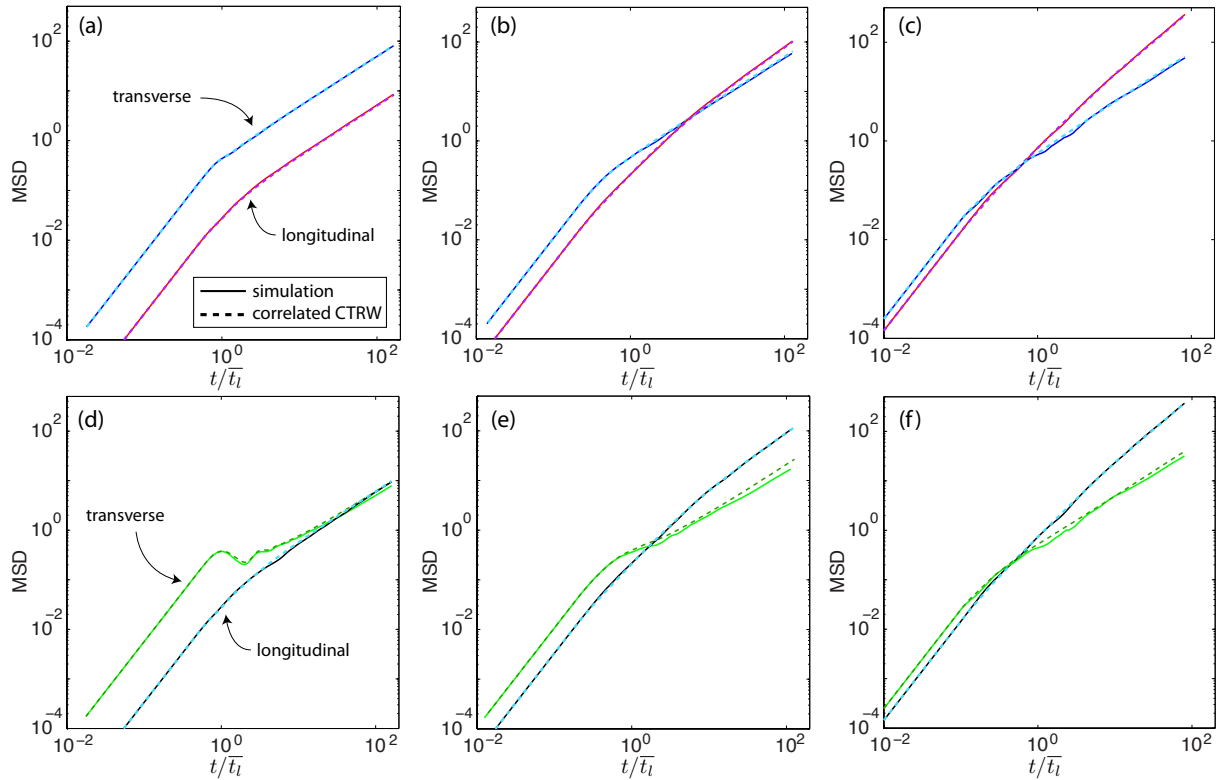


FIG. 12. Time evolution of MSDs obtained from Monte Carlo simulations (solid lines), and the model predictions from the correlated CTRW model (dashed lines). The developed correlated CTRW model is able to accurately capture the time evolution of the MSDs for all levels of heterogeneity strength and mixing rules. (a) $\sigma_{\ln K}^2 = 0.1$, (b) $\sigma_{\ln K}^2 = 1$, and, (c) $\sigma_{\ln K}^2 = 5$ with *complete mixing*, where red line is longitudinal direction and blue line is transverse direction. (d) $\sigma_{\ln K}^2 = 0.1$, (e) $\sigma_{\ln K}^2 = 1$, (f) $\sigma_{\ln K}^2 = 5$ with *streamline routing*, where black line is longitudinal and green line is transverse direction.

336 Ignoring the correlated structure of the Lagrangian velocity leads to predictions of longitudinal
 337 and transverse spreading that deviate from the direct Monte Carlo simulation [Figures 13(d)(e)(f),
 338 and 14(b)]. The uncorrelated CTRW model is not able to predict transverse spreading for the
 339 streamline routing case [Figure 13(d)(e)(f)], or the peak arrival time and spread of the FPTD
 340 [Figures 14(b)]. In contrast, these behaviors are accurately captured by the correlated model.

341 VI. PARAMETERIZATION OF THE CORRELATED CTRW MODEL

342 In the previous section, we showed that the effective particle movement can be described by
 343 a CTRW whose particle velocities, or transition times, form a spatial Markov process. The latter

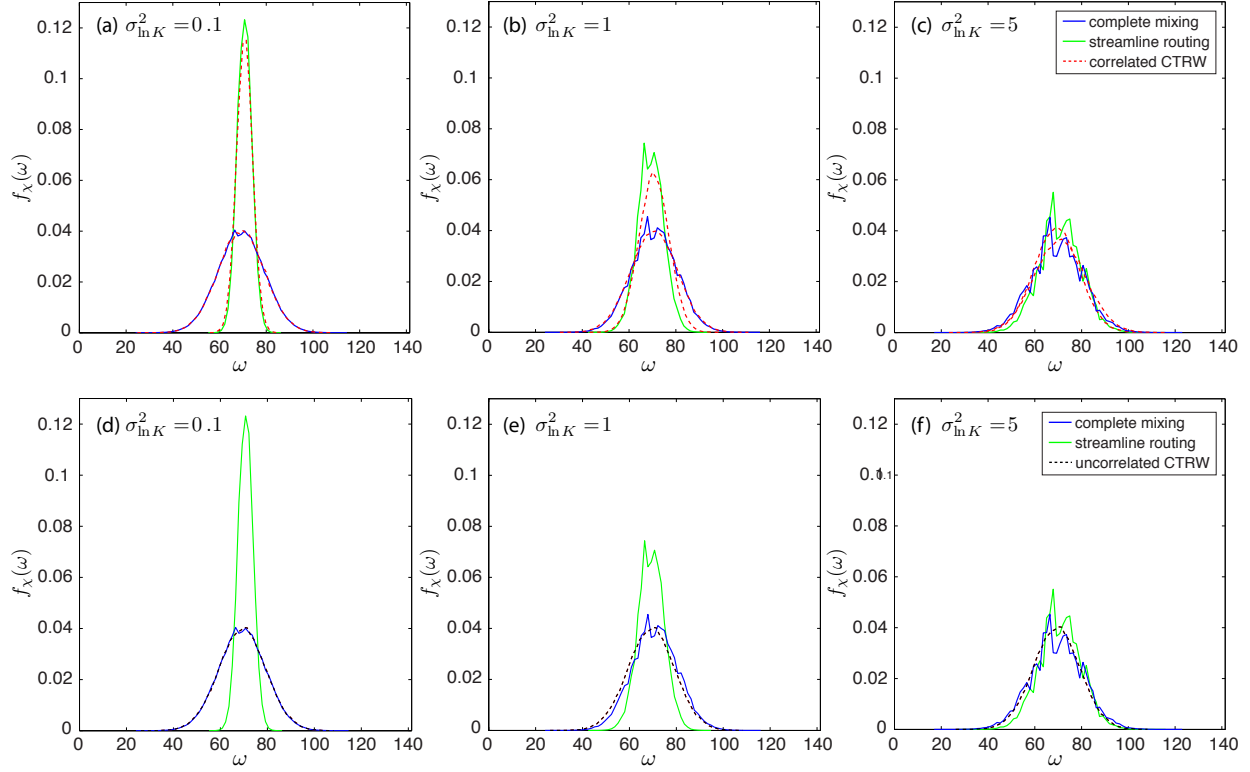


FIG. 13. Probability distributions of transverse particle breakthrough position for Monte Carlo simulations and model predictions. (a) Correlated CTRW for $\sigma_{\ln K}^2 = 0.1$. (b) Correlated CTRW for $\sigma_{\ln K}^2 = 1$. (c) Correlated CTRW for $\sigma_{\ln K}^2 = 5$. (d) Uncorrelated CTRW for $\sigma_{\ln K}^2 = 0.1$. (e) Uncorrelated CTRW for $\sigma_{\ln K}^2 = 1$. (f) Uncorrelated CTRW for $\sigma_{\ln K}^2 = 5$. Uncorrelated CTRW does not have a capability of distinguishing complete mixing and streamline routing cases.

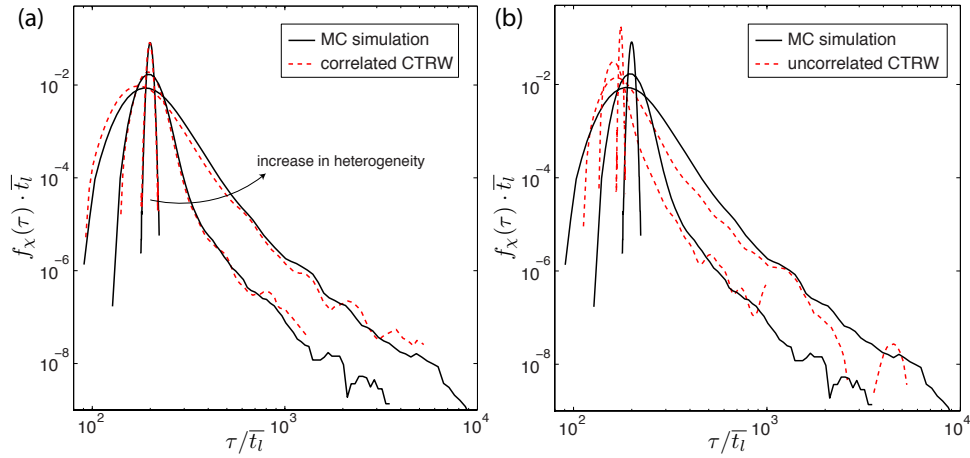


FIG. 14. Particle breakthrough curves from Monte Carlo simulations and model predictions from (a) correlated CTRW, and (b) uncorrelated CTRW.

344 has been characterized by a velocity transition PDF which has been sampled from the simulated
 345 particle velocities. While the resulting correlated CTRW describes the observed behavior well,
 346 the application of the approach to experimental data (such as tracer tests) asks for a process model
 347 that requires only a few parameters which may be estimated from the available data. Thus, here
 348 we consider an explicit Markov process model for subsequent transition times that captures the
 349 essential features of correlation with a minimal set of parameters. To accomplish this, we follow
 350 the approach of [18], who recently proposed an effective parameterization of the correlated CTRW
 351 model and applied it to the interpretation of field-scale tracer transport experiments.

352 We consider the series of streamwise particle velocities $v_n = |\mathbf{v}_L(s_n)|$ and model them as
 353 a Markov process $\{v_n\}$ through the steady state velocity PDF, $\psi_v(v)$, and the transition ma-
 354 trix \mathbf{T} [63]. First note that v is discretized into N classes, $v \in \bigcup_{i=1}^N (v_{c,i}, v_{c,i+1}]$, such that the
 355 transition probabilities between the classes are represented by the $N \times N$ transition matrix \mathbf{T} .
 356 Here, we choose equiprobable binning such that the class limits $v_{c,i}$ are given implicitly by

$$\int_{v_{c,i}}^{v_{c,i+1}} dt \psi_\tau(t) = \frac{1}{N}. \quad (18)$$

357 With this condition, \mathbf{T} is a doubly stochastic matrix, which satisfies $\sum_{i=1}^N T_{ij} = \sum_{j=1}^N T_{ij} = 1$.
 358 For a large number of transitions it converges towards uniformity

$$\lim_{n \rightarrow \infty} [\mathbf{T}^n]_{ij} = \frac{1}{N}, \quad (19)$$

359 whose eigenvalues are 1 and 0. Correlation is measured by the convergence of \mathbf{T} towards the
 360 uniform matrix. The characteristic number of steps over which the Markov chain is correlated,
 361 is determined by the decay rate of the second largest eigenvalue χ_2 of \mathbf{T} (the largest eigenvalue
 362 of a stochastic matrix is always 1). The convergence towards uniformity can be quantified by the
 363 correlation function $C(n) = \chi_2^n$, which can be written as

$$C(n) = \exp\left(-\frac{n}{n_c}\right), \quad n_c = -\frac{1}{\ln(|\chi_2|)}. \quad (20)$$

364 The transition matrix is characterized by n_c , which determines the characteristic number of
 365 steps for convergence towards uniformity. Thus, we consider here a Markov model whose transi-
 366 tion matrix is characterized by just two eigenvalues, namely 1 and χ_2 . Its transition matrix is given
 367 by

$$T_{ij} = a\delta_{ij} + (1 - a)\frac{1 - \delta_{ij}}{N - 1}. \quad (21)$$

368 It describes a Markov process that remains in the same state with probability a and changes to a
 369 different state, whose distribution is uniform, with probability $1 - a$. The diagonal value of $a \leq 1$
 370 determines the correlation strength. A value of $a = 1$ implies perfect correlation, which renders
 371 the N -dimensional unity matrix, $T_{ij} = \delta_{ij}$. For $a = 1/N$, all transitions are equally probable,
 372 and the transition matrix is equal to the uniform matrix with $T_{ij} = 1/N$. The eigenvalues of the
 373 transition matrix (21) are $\chi_1 = 1$ and

$$\chi_2 = \frac{Na - 1}{N - 1}. \quad (22)$$

374 Thus, the number n_c of correlation steps is given by

$$n_c = -\frac{1}{\ln\left(\frac{Na-1}{N-1}\right)} \stackrel{N \gg 1}{\approx} -\frac{1}{\ln(a)}. \quad (23)$$

375 It is uniquely determined by the value of a . The value of a can be estimated from the correlation
 376 function $\chi_s(s)$ of streamwise Lagrangian velocity given by Equation (8). The streamwise velocity
 377 correlation function is given in terms of the velocity time series $\{v_n\}$ as

$$\chi(s_{n+m} - s_n) = \frac{\langle v'_{n+m} v'_n \rangle}{\langle v'^2 \rangle}, \quad (24)$$

378 where we defined $v'_n = v_n - \langle v \rangle$ with $\langle v \rangle$ the mean streamwise velocity and $s_n = nl$. Using
 379 the discretization (18) of streamwise velocities into N equiprobable bins and the transition matrix
 380 \mathbf{T} , the velocity correlation can be written as

$$\chi_s(s_{n+m} - s_n) = \frac{\frac{1}{N} \sum_{i,j=1}^N v'_{c,i} [\mathbf{T}^m]_{ij} v'_{c,j}}{\frac{1}{N} \sum_{i=1}^N v'^2_{c,i}}. \quad (25)$$

381 Note that the transition matrix \mathbf{T} given by (21) is symmetric and has only the two eigenvalues,
 382 $\chi_1 = 1$ and χ_2 given by (22) with χ_2 of order $N - 1$. Thus, performing a base transformation
 383 in (25) into the eigensystem of \mathbf{T} , one sees that

$$\chi_s(s_{n+m} - s_n) = \exp\left(-\frac{|s_{n+m} - s_n|}{\ell_c}\right), \quad (26)$$

384 where the correlation length is given by $\ell_c = n_c l$. Thus, n_c is directly related to the correlation
 385 length of the streamwise Lagrangian velocity. Note that $\ell_c = 0$ for zero correlation and $\ell_c = \infty$
 386 for perfect correlation. As illustrated in Figure 8(b), $\chi_s(s)$ is well approximated by an exponential
 387 function. Thus, we obtain a from the correlation length ℓ_c as $a = \exp(-l/\ell_c)$. The transition
 388 matrix \mathbf{T} is fully parameterized in terms of the correlation length of the streamwise Lagrangian
 389 velocity.

390 To describe the observed steady state velocity distribution $\psi_v(v)$ we consider the equivalent
 391 distribution $\psi_\tau(\tau)$ of transition times $\tau = l/v$, which is illustrated in Figure 8(a). It is well
 392 described by the following truncated power-law distribution

$$\psi_\tau(t) \sim \frac{\exp(-\tau_0/t)}{(t/\tau_0)^{1+\beta}}, \quad (27)$$

393 where τ_0 determines the early time cutoff, and β the power-law slope. Note that τ_0 and β are
 394 both positive coefficients. The slope β of the power-law regime describes the heterogeneity of the
 395 velocity distribution. As β decreases, the transport becomes more anomalous because the prob-
 396 ability of experiencing large transition times increases. Therefore, smaller β can be understood
 397 to represent higher flow heterogeneity, as is well known in the CTRW modeling framework [43].
 398 Indeed, the estimated β values decrease as the conductivity distribution becomes more heteroge-
 399 neous (we obtain $\beta = 18, 2.6, 1.7$ for $\sigma_{\ln K}^2 = 0.1, 1, 5$, respectively). We estimate the parameters
 400 τ_0 and β from the measured transition time distributions [Figure 8(a)]. As pointed out recently
 401 by [64], the tail behavior of the transition time PDF as quantified by the exponent β may in prin-
 402 ciple be related to the lower end of the distribution of hydraulic conductivity.

403 The velocity PDF is obtained from the transition time PDF by $\psi_v(v) = (l/v^2)\psi_\tau(l/v)$ and
 404 quantifies together with the transition matrix \mathbf{T} the velocity heterogeneity and velocity correlation
 405 structure. To honor the network geometry and to accurately estimate transverse spreading we need
 406 one more input parameter that quantifies the velocity directionality. Since the majority of velocity
 407 transitions are forward-forward, we only consider E, M, G and O transitions. We need a additional
 408 parameter that quantifies the probability of changing (either M or G) or maintaing the direction
 409 (either E or O), and define γ as the probability of changing and $1 - \gamma$ is the probability of main-
 410 taining the direction. The measured γ values for the complete mixing rule are in all cases ~ 0.5 ,

411 independently of conductivity distributions. This is because the complete mixing rule maximizes
412 transverse excursions. However, for streamline routing γ is very sensitive to the underlying con-
413 ductivity distribution. It decreases as the conductivity distribution becomes more heterogeneous
414 (we find $\gamma = 0.89, 0.71, 0.58$ for $\sigma_{\ln K}^2 = 0.1, 1, 5$, respectively). This is because the probability of
415 transitioning to an adjacent link is higher for the streamline routing case, and as the conductivity
416 heterogeneity increases the probability of transitioning to the opposite link increases.

417 In summary, the correlated CTRW model for the random network under consideration is char-
418 acterized by four independent parameters that determine the velocity distribution and the velocity
419 correlation structure: β , which characterizes the slope of the truncated power-law distribution;
420 τ_0 , which characterizes the early time cutoff of the transition time distribution; a , which quantifies
421 the velocity correlation; and γ , which quantifies the velocity transition directionality.

422 In order to test the predictive power of the parametric correlated CTRW model, the model
423 predictions are compared to the results obtained from the direct Monte Carlo simulations. We
424 obtain an excellent agreement with the Monte Carlo results for all the conductivity distributions
425 and mixing rules that we studied (Figures 15 and 16). The model accurately captures the time
426 evolution of the particle plumes, including spatial moments, first passage time distributions and
427 the distributions of the transverse particle breakthrough positions (Figures 15 and 16).

428 The fact that the parametric correlated CTRW proves as good here as the more complex cor-
429 related CTRW model presented in the previous section is noteworthy, as the parametric model
430 involves only four parameters. In particular, the previous CTRW model quantifies explicitly the
431 transition probability of each velocity class to the others, while the parametric correlated CTRW
432 model only quantifies the probability to stay in the same velocity class and it assumes that the
433 probability to jump to any other class is independent of velocity. This assumption is likely valid
434 here since there is nearly no dependence of the velocity correlation properties on the velocity
435 (Figure 11). This assumption would break down in systems where transitions from one velocity
436 to the other is strongly dependent on velocity. For instance, in highly channelized systems, the
437 probability for particles to stay in high velocity channels may be different from their probability to
438 stay in low velocity areas (see discussion in [65]). This result represents an important step towards
439 the application of this framework to the field. As discussed in [18], the model parameters can
440 be estimated by analyzing jointly cross-borehole and push-pull tracer tests. In particular, velocity
441 correlation is key to distinguishing reversible from irreversible dispersion, which is linked to the
442 difference between spreading and mixing.

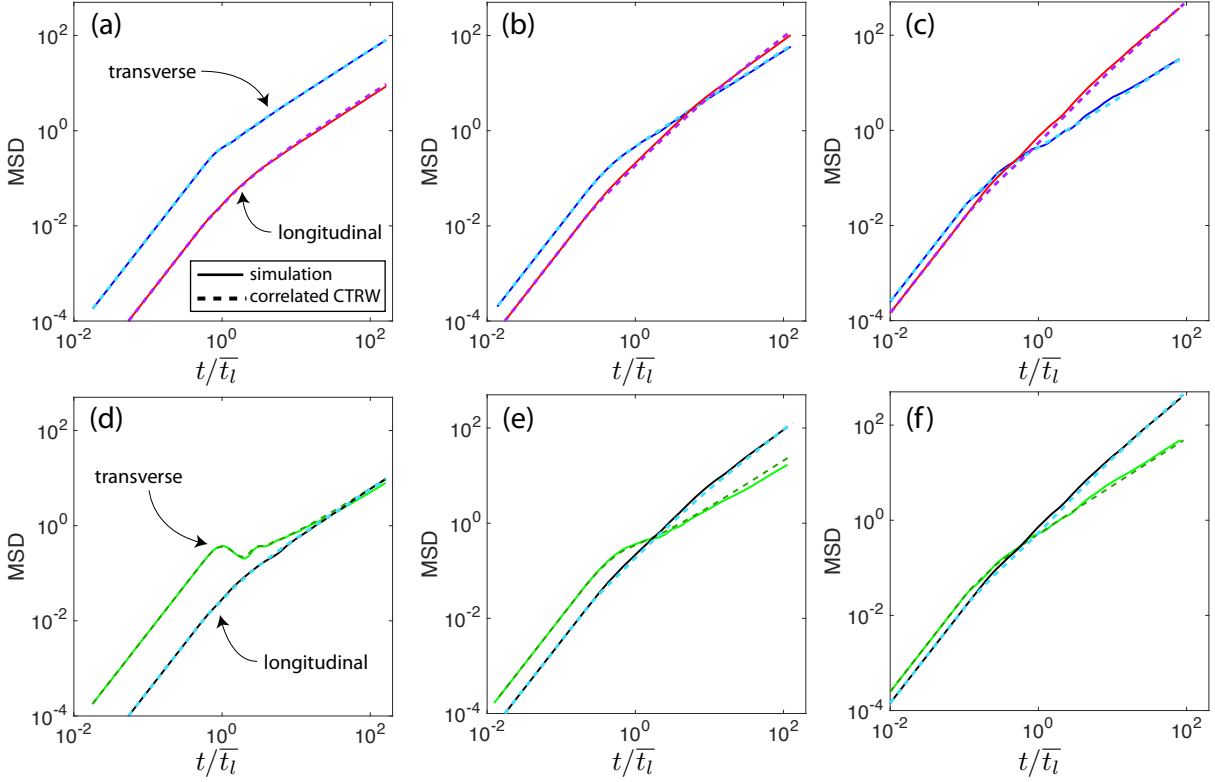


FIG. 15. Comparison between the time evolution of the MSDs obtained from the Monte Carlo simulations (solid lines) and the model predictions from the *parametric* correlated CTRW model (dashed lines). The proposed model is able to accurately capture the time evolution of the MSDs for all levels of heterogeneity and mixing rules under consideration. (a) $\sigma_{\ln K}^2 = 0.1$, (b) $\sigma_{\ln K}^2 = 1$, (c) $\sigma_{\ln K}^2 = 5$ with *complete mixing*, where red line is longitudinal direction and blue line is transverse direction. (d) $\sigma_{\ln K}^2 = 0.1$, (e) $\sigma_{\ln K}^2 = 1$, (f) $\sigma_{\ln K}^2 = 5$ with *streamline routing*, where black line is longitudinal and green line is transverse direction.

443 VII. CONCLUSIONS

444 Fracture networks characterized by conductivity heterogeneity and different mixing rules at
 445 fracture intersections lead to non-trivial transport behavior often characterized by non-Fickian
 446 dispersion properties in both longitudinal and transverse directions. The divergence-free condition
 447 arising from mass conservation leads to a correlated flow field with preferential paths, even when
 448 the underlying conductivity field is completely uncorrelated. Mixing rules at nodes are shown
 449 to have a major impact on transverse mixing. In particular, the streamline routing rule leads to
 450 subdiffusive transverse spreading behavior. While velocity distributions are mainly controlled by
 451 the underlying conductivity distributions, the velocity correlation structure is determined by the

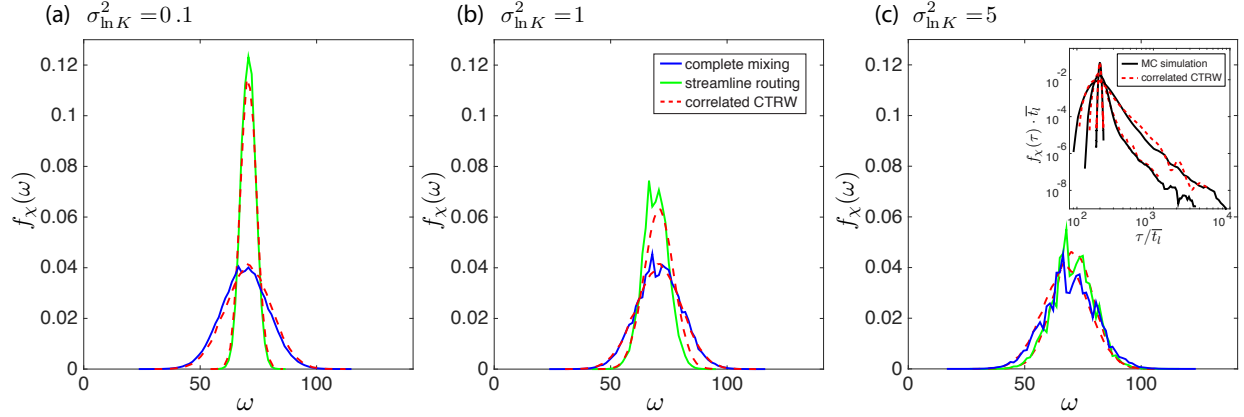


FIG. 16. Probability distributions of the transverse particle breakthrough positions obtained from Monte Carlo simulations, and predictions from the parametric correlated CTRW model. (a) $\sigma_{\ln K}^2 = 0.1$. (b) $\sigma_{\ln K}^2 = 1$. (c) $\sigma_{\ln K}^2 = 5$. Inset: Particle breakthrough curves from Monte Carlo simulations and model predictions from the parametric correlated CTRW model.

452 interplay between network heterogeneity and mixing rule at nodes.

453 Here, we propose and validate a spatial Markov model that is fully parameterized from the
 454 velocity field distribution and spatial correlation properties, and explicitly captures the multidimensional effects associated with changes in direction along the particle trajectory. In particular,
 455 we discuss the impact of spatial velocity correlations, which are typically not included in the
 456 classical CTRW framework, on the transport behavior. To make this model amenable to field applications, we develop a parametric model formulation containing a minimum set of parameters
 457 that still captures the main properties of the velocity field relevant for transport: β characterizes
 458 the slope of the truncated power-law velocity distribution, τ_0 characterizes the early time cutoff of the transition time distribution, a quantifies the velocity correlation, and γ quantifies the velocity
 459 transition directionality.
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463 The excellent agreement between the model and the numerical simulations provides a validation
 464 of this parametric correlated CTRW approach, whose parameters can be determined from field
 465 tracer tests [18] to assess the respective role of velocity distributions and velocity correlations *in situ*. It is important to note that, in its current formulation, the parametric correlated CTRW model
 466 assumes an identical correlation length over all velocity classes. This assumption allows us to
 467 quantify velocity correlation with a single parameter, but could be an oversimplified approach for
 468 certain cases. For example, correlated conductivity field with strong preferential paths may lead
 469

470 to longer velocity correlation length for high velocities compared to small velocities. This should
471 be investigated in future research and we conjecture that assigning variable correlation length as a
472 function of velocity class could be a promising approach.

473 Finally, our study shows how the interplay between fracture geometrical properties (conductiv-
474 ity distribution and network geometry) and physical transport mechanisms (the balance between
475 advection and diffusion that determines mixing at the fracture scale) controls average particle
476 transport via Lagrangian velocity statistics. We conjecture that the proposed correlated CTRW
477 model may provide an avenue to link the model parameters to geometrical and physical transport
478 mechanisms.

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