Continuous Time Random Walks for the Evolution of Lagrangian Velocities

Marco Dentz,[1,2,*] Peter K. Kang,[3] Alessandro Comolli,[1,2,4] Tanguy Le Borgne,[5] and Daniel R. Lester[6]

[1]*Spanish National Research Council (IDAEA-CSIC), Barcelona, Spain*
[2]*Hydrogeology Group (CSIC-UPC)*
[3]*Korea Institute of Science and Technology, Seoul 136-791, Republic of Korea*
[4]*Department of Civil Engineering, Technical University of Catalonia (UPC), Barcelona, Spain*
[5]*Université de Rennes 1, CNRS, Geosciences Rennes, UMR 6118, Rennes, France*
[6]*School of Civil, Environmental and Chemical Engineering, RMIT University, 3000 Melbourne, Victoria, Australia*

We develop a continuous time random walk (CTRW) approach for the evolution of Lagrangian velocities in steady heterogeneous flows based on a stochastic relaxation process for the streamwise particle velocities. This approach describes persistence of velocities over a characteristic spatial scale, unlike classical random walk methods, which model persistence over a characteristic time scale. We first establish the relation between Eulerian and Lagrangian velocities for both equidistant and isochrone sampling along streamlines, under transient and stationary conditions. Based on this, we develop a space continuous CTRW approach for the spatial and temporal dynamics of Lagrangian velocities. While classical CTRW formulations have non-stationary Lagrangian velocity statistics, the proposed approach quantifies the evolution of the Lagrangian velocity statistics under both stationary and non-stationary conditions. We provide explicit expressions for the Lagrangian velocity statistics, and determine the behaviors of the mean particle velocity, velocity covariance and particle dispersion. We find strong Lagrangian correlation and anomalous dispersion for velocity distributions which are tailed toward low velocities as well as marked differences depending on the initial conditions. The developed CTRW approach predicts the Lagrangian particle dynamics from an arbitrary initial condition based on the Eulerian velocity distribution and a characteristic correlation scale.

I. INTRODUCTION

The dynamics of Lagrangian velocities in fluid flows are fundamental for the understanding of tracer dispersion, anomalous transport behaviors, but also pair-dispersion and intermittent particle velocity and acceleration time series, as well as fluid stretching and mixing. A classical stochastic view-point on particle velocities in heterogeneous flows is their representation in terms of Langevin models for the particle velocities [1], which accounts for temporal persistence, and the random nature of velocity through a Gaussian white noise. Such approaches assume that velocity time series form a Markov process when measured isochronically along a particle trajectory [2].

The observation of intermittency in Lagrangian velocity and acceleration time series in steady heterogeneous flow [3–5] questions the assumptions that underly the representation of Lagrangian velocity in terms of a classical random walk. Observed intermittency patterns manifest themselves in long episodes of low velocities and relatively short episodes of high velocity. This indicates an organizational principle of Lagrangian velocities that is different from the one implied in a temporal Markov processes, which assumes that velocities are persistent for a constant time interval of characteristic duration τ_c. Observed intermittency for flow through disordered media [3–5] suggests that particle velocities are persistent along a characteristic length scale ℓ_c along streamlines. Approaches that model particle velocities as Markov processes in space, assign to particle transitions a random transition time, which is given kinematically by the transition distance divided by the transition velocity. Thus, such approaches are termed continuous time random walks (CTRW) [6–9]. They are different from classical random walk approaches, which employ a constant discrete transition time.

Particle motion and particle dispersion have been shown to follow CTRW dynamics for flow through pore and Darcy-scale heterogeneous porous and fractured media [10–17], as well as turbulent flows [18, 19]. While CTRW provides an efficient framework for the quantification of anomalous dispersion and intermittency in heterogeneous flows, some key questions remain open regarding the relation of particle velocities and Eulerian flow statistics, and the stationarity of Lagrangian velocity statistics.

In classical CTRW formulations, particle velocities are non-stationary. This means, for example that the velocity mean and covariance evolve in time. This property is termed aging [20]. However, for steady divergence-free random flows, such as flow through porous media, it has been found that particle velocities may in fact be stationary [21]; specifically the Lagrangian mean velocity may be independent of time. Furthermore, it has been found for flow through random fracture networks that the Lagrangian velocity statistics depends on the initial particle distribution [22–24]. Hence, in general, Lagrangian velocities are expected to evolve from an arbitrary initial distribution toward an asymptotic stationary distribution. Quantifying this property, which is not

* E-mail: marco.dentz@csic.es

described in current CTRW frameworks, is critical for upscaling transport dynamics through disordered media, whose transport properties are sensitive to the initial velocity distribution.

In this paper, we study the evolution of Lagrangian velocities and their relation with the Eulerian velocity statistics. To this end, we discuss in the following section the concepts of Lagrangian velocities determined isochronically and equidistantly along streamlines and their relation to the Eulerian velocity. Furthermore, we recall some fundamental properties that elucidate the conditions under which they are transient or stationary. In Section III, we derive the Lagrangian velocity statistics in the classical CTRW and develops a Markov-chain CTRW approach that models the evolution of equidistant streamwise Lagrangian velocities as a stochastic relaxation process. In this framework, we derive explicit expressions for the one and two-point statistics of Lagrangian velocities, and analyze the evolution of the mean particle velocity, its covariance as well as particle dispersion in Section IV.

II. LAGRANGIAN VELOCITIES

We consider purely advective transport in a heterogeneous stationary velocity field $\mathbf{u}(\mathbf{x})$. Particle trajectories are described by the advection equation

$$\frac{d\mathbf{x}(t,\mathbf{a})}{dt} = \mathbf{v}(t,\mathbf{a}), \tag{1}$$

where $\mathbf{v}(t,\mathbf{a}) = \mathbf{u}[\mathbf{x}(t,\mathbf{a})]$ denotes the Lagrangian particle velocity. The initial particle position is given by $\mathbf{x}(t = 0,\mathbf{a}) = \mathbf{a}$. The particle motion can be described in terms of the distance $s(t,\mathbf{a})$ traveled along a trajectory, which is given by

$$\frac{ds(t,\mathbf{a})}{dt} = v_t(t,\mathbf{a}), \qquad \frac{dt(s,\mathbf{a})}{ds} = \frac{1}{v_s(s,\mathbf{a})}, \tag{2}$$

We define the t-Lagrangian particle velocity as $v_t(t,\mathbf{a}) = |\mathbf{v}(t,\mathbf{a})|$, the s-Lagrangian velocity $v_s(s,\mathbf{a}) = v_t[t(s,\mathbf{a}),\mathbf{a}]$. The initial velocities are denoted by $v_0(\mathbf{a}) \equiv v_t(t = 0,\mathbf{a}) \equiv v_s(s = 0,\mathbf{a})$.

The absolute Eulerian velocities are defined by $v_e(\mathbf{x}) = |\mathbf{u}(\mathbf{x})|$. Their probability density function (PDF) is defined through spatial sampling as

$$p_e(v) = \lim_{V\to\infty} \frac{1}{V} \int\limits_{\Omega} d\mathbf{x}\delta[v - v_e(\mathbf{x})], \tag{3}$$

where Ω is the sampling domain and V its volume. We assume here Eulerian ergodicity, this means that spatial sampling is equal to ensemble sampling such that

$$p_e(v) = \overline{\delta[v - v_e(\mathbf{x})]}, \tag{4}$$

where the overbar denotes the ensemble average. In the following, we discuss the t-Lagrangian velocities $v_t(t,\mathbf{a})$, which are sampled isochronally along particle trajectories, and the s-Lagrangian velocities $v_s(s,\mathbf{a})$, which are sampled equidistantly along particle trajectories. Here and in the following, we assume both Eulerian and Lagrangian ergodicity. As outlined below, Lagrangian ergodicity implies that the statistics of particle velocities $v_t(t,\mathbf{a})$ sampled in time along a trajectory coincide with the statistics obtained by sampling between particles.

A. Steady Lagrangian Velocity Distributions

The PDF of the t-Lagrangian velocity is defined by isochrone sampling along a particle trajectory as

$$p_t(v,\mathbf{a}) = \lim_{T\to\infty} \frac{1}{T} \int\limits_0^T dt\delta\left[v - v_t(t,\mathbf{a})\right], \tag{5}$$

Under Lagrangian ergodic conditions, it is independent of the initial particle position $\mathbf{a}$ and equal to the average over an ensemble of particles

$$p_t(v) = \lim_{V_0\to\infty} \frac{1}{V_0} \int\limits_{\Omega_0} d\mathbf{a}\delta[v - v_t(t,\mathbf{a})]. \tag{6}$$

The latter is equal to the Eulerian velocity PDF due to volume conservation,

$$p_t(v) = \lim_{V_0\to\infty} \frac{1}{V_0} \int\limits_{\Omega(t)} d\mathbf{x}\delta[v - v_e(\mathbf{x})] \equiv p_e(v), \tag{7}$$

which can be seen by performing a change of variables according to the flow map $\mathbf{a} \to \mathbf{x}(t,\mathbf{a})$ and recalling that the Jacobian is one due to the incompressibility of the flow field.

The PDF of the s-Lagrangian velocity is defined in analogy to (5) by equidistant sampling along a particle trajectory as

$$p_s(v,\mathbf{a}) = \lim_{L\to\infty} \frac{1}{L} \int\limits_0^L ds\delta[v - v_s(s,\mathbf{a})]. \tag{8}$$

Changing variables under the integral according to the kinematic relationship (2) between t and s gives immediately

$$p_s(v,\mathbf{a}) = \frac{v p_t(v,\mathbf{a})}{\langle v_t \rangle}, \tag{9}$$

this means the s-Lagrangian velocity PDF is equal to the flux weighted t-Lagrangian velocity PDF. This can also be understood intuitively by the fact that isochrone sampling as expressed through $p_t(v)$ gives a higher weight to low velocities because particles spend more time at low velocities, while equidistant sampling assigns the same weight to high and low velocities.

Under conditions of Lagrangian ergodicity, we thus have that (i) $p_s(v, \mathbf{a}) = p_s(v)$ is independent of the particle trajectory and equal to the average over an ensemble of particles and (ii) that the s-Lagrangian velocity PDF is related to the Eulerian velocity PDF through flux weighting as

$$p_s(v) = \frac{v p_e(v)}{\langle v_e \rangle}. \tag{10}$$

The latter establishes the relation between s-Lagrangian and Eulerian velocity distributions.

B. Transient Lagrangian Velocity Distributions

In the previous sections, we considered the PDFs of t- and s-Lagrangian velocities under stationary conditions. Here we focus on their transient counterparts, which are defined through a spatial average over an arbitrary normalized initial particle distribution $\rho(\mathbf{a})$.

The PDF of t-Lagrangian velocities then is defined by

$$\hat{p}_t(v, t) = \int d\mathbf{a} \rho(\mathbf{a}) \delta[v - v_t(t, \mathbf{a})]. \tag{11}$$

Its temporal average is given by

$$\lim_{T\to\infty} \frac{1}{T} \int\limits_0^T dt \hat{p}_t(v, t) = p_t(v) = p_e(v), \tag{12}$$

and thus its steady state PDF is of course given by the Eulerian velocity PDF. In analogy, we consider the PDF of s-Lagrangian velocities for an arbitrary initial PDF

$$\hat{p}_s(v, s) = \int d\mathbf{a} \rho(\mathbf{a}) \delta[v - v_s(s, \mathbf{a})]. \tag{13}$$

Its average along a streamline is given by

$$\lim_{L\to\infty} \frac{1}{L} \int\limits_0^L ds \hat{p}_t(v, s) = p_s(v) = \frac{v p_e(v)}{\langle v_e \rangle}. \tag{14}$$

The initial conditions for both the t-Lagrangian and s-Lagrangian velocity PDFs are identical,

$$\hat{p}(v, s = 0) = \hat{p}(v, t = 0) = p_0(v) \tag{15}$$

Thus, as their respective steady state PDFs are different, either one or both of them need to evolve, depending on whether the initial PDF is the flux weighted Eulerian PDF, (the steady state PDF for $\hat{p}_s(v, s)$), the Eulerian PDF (the steady state PDF for $\hat{p}_t(v, t)$), or neither of the two.

The initial velocity PDF depends on the particle injection mode. For example, a uniform in space particle injection corresponds here to an initial velocity PDF equal to the Eulerian PDF,

$$p_0(v) = \lim_{V_0\to\infty} \frac{1}{V_0} \int\limits_{\Omega_0} d\mathbf{a} \delta[v - v_0(\mathbf{a})] \equiv p_e(v) \tag{16}$$

because of Eulerian ergodicity. As this initial distribution is equal to the asymptotic steady t-Lagrangian velocity distribution, the $\hat{p}_t(v, t) = p_e(v)$ is independent of time for this initial injection condition, while the $\hat{p}_s(v)$ evolves with distance from the injection.

A flux weighted particle injection mode corresponds to an initial velocity PDF equal to the flux weighted Eulerian PDF

$$p_0(v) = \lim_{V_0\to\infty} \frac{1}{V_0} \int\limits_{\Omega_0} d\mathbf{a} \frac{v_0(\mathbf{a})}{\langle v_e \rangle} \delta[v - v_0(\mathbf{a})] \equiv \frac{v p_e(v)}{\langle v_e \rangle} \tag{17}$$

again because of Eulerian ergodicity. As this initial distribution is equal to the asymptotic steady s-Lagrangian velocity distribution, $\hat{p}_s(v, s) \equiv p_s(v)$ is independent of s for this initial injection condition, while $\hat{p}_t(v, t)$ evolves with time.

A point-like injection at the initial position $\mathbf{x}(t = 0|\mathbf{a}) = \mathbf{a}$ corresponds to the delta initial PDF

$$p_0(v) = \delta[v - v_0(\mathbf{a})]. \tag{18}$$

For this initial condition, both the t-Lagrangian and s-Lagrangian velocities are unsteady.

The evolution of Lagrangian velocities may be very slow and thus have a strong impact on the transport dynamics. This is the case in particular for heavy-tailed (towards low velocities) velocity distributions that induce long-range temporal correlations of particle velocities. In the following, we study the quantification of the evolution of the Lagrangian velocity PDFs in a Markov model in s, this means distance along streamline.

C. Lagrangian Velocity Series

We have established that the Lagrangian velocity PDFs evolve with travel time or travel distance along a streamline, unless the initial velocity distribution coincides with the respective steady state PDF. In order to quantify this evolution, we need to model the Lagrangian velocity series. As mentioned in the Introduction, a classical approach is to model the t-Lagrangian velocity as a Markov process, based on the assumption, or observation that velocities decorrelate on a characteristic time scale τ_c. Thus, the equations of motion (2) may be discretized isochronically as

$$t_{n+1} = t_n + \Delta t, \qquad s(t_{n+1}) = s(t_n) + v_t(t_n)\Delta t. \tag{19}$$

Velocity time series have been modeled by Langevin equations of the type [1]

$$\tilde{v}_t(t_{n+1}) = \tilde{v}_t(t_n) - \frac{\Delta t}{\tau_c} \tilde{v}_t(t_n) + \sqrt{\frac{2\sigma_v^2 \Delta t}{\tau_c}} \xi(t_n), \tag{20}$$

which describes an Ornstein-Uhlenbeck process for the velocity fluctuation $\tilde{v}_t(t_n) = v_t(t_n) - \langle v_t \rangle$. The noise $\xi(t_n)$

is Gaussian distributed with zero mean and unit variance. The steady state distribution $p_t(v)$ here is Gaussian with mean $\langle v_t \rangle$ variance σ_v^2. Under stationary conditions, the velocity correlation is exponential with correlation time τ_c. Evidently, this modeling framework is limited to Gaussian statistics and short range correlation in time.

Here, we consider a different modeling approach. As pointed out in the Introduction, there has been ample evidence that particle motion in the flow through random porous and fractured media may be quantified by a CTRW [9]. In fact, as a consequence of the existence of a spatial correlation length scale for, e.g., the hydraulic conductivity or pore-structure, flow velocities are expected to vary over a characteristic length scale ℓ_c. This implies for t-Lagrangian velocities that a given velocity v_t persists for a duration of ℓ_c/v_t, and specifically that small velocities are more strongly correlated in time than high velocities [25, 26]. This characteristic can explain intermittency in velocity and acceleration time series [3–5]. The existence of a characteristic length scale ℓ_c suggests discretizing the equations of motion (2) along a particle trajectory equidistantly such that

$$s_{n+1} = s_n + \Delta s, \qquad t(s_n) = t(s_n) + \frac{\Delta s}{v_s(s_n)}. \tag{21}$$

Here, the s-Lagrangian velocity series $v_s(s_n)$ is modeled as Markov process, which renders the equations of motion (21) a CTRW. In the following, we analyze the evolution of the Lagrangian velocity statistics in the setup of a classical CTRW characterized by independent s-Lagrangian velocities, and a CTRW in which the velocity series is modeled as a Markov process through a stochastic relaxation.

III. CONTINUOUS TIME RANDOM WALK

We study now the evolution of space and time Lagrangian velocities in the CTRW framework. The classical approach assigns to each particle transition a transit time τ that is sampled at each step from its PDF $\psi(t)$. The transition times are related to the characteristic transition length ℓ_c and s-Lagrangian velocities v_s as $\tau = \ell_c/v_s$. Thus, independence of subsequent transit times implies indepence of subsequent s-Lagrangian velocities. In the following, we first consider the evolution of t-Lagrangian velocities in this classical CTRW formulation. The velocity statistics turn out to be non-stationary at finite times. We then study a CTRW formulation that is based on a Markov process for the s-Lagrangian velocities that allows for an evolution of both the s- and t-Lagrangian velocities.

A. Independent s-Lagrangian Velocities

Particle motion along a particle trajectory is quantified in the framework of a classical CTRW by the recursion relations

$$s_{n+1} = s_n + \ell_c, \qquad t_{n+1} = t_n + \tau_n, \tag{22}$$

where the transition length ℓ_c denotes a characteristic length scale on which streamwise velocities $v_n \equiv v_s(s_n)$ decorrelate. In this framework, the particle velocity is constant between turning points. Thus, the transition times $\tau_n = \ell_c/v_n$ are independent identically distributed random variables. Their PDF is given by $\psi(\tau)$. It is related to the distributions of s-Lagrangian and Eulerian velocities by

$$\psi(\tau) = \frac{\ell_c}{\tau^2} p_s(\ell_c/\tau) = \frac{\ell_c \tau_v}{\tau^3} p_e(\ell_c/\tau), \tag{23}$$

where we defined the advection time scale $\tau_v = \ell_c/\langle v_e \rangle$. Note that the mean transit time $\langle \tau \rangle = \tau_v$ is equal to the characteristic advection time.

In this framework, the t-Lagragian velocity is given by

$$v_t(t) = v_{n_t}, \tag{24}$$

where the renewal process $n_t = \sup(n|t_n \leq t)$ denotes the number of steps needed to arrive at time t. The PDF of the t-Lagrangian velocity is given by

$$\hat{p}_t(v,t) = \langle \delta[v - v_{n_t}] \rangle. \tag{25}$$

This expression can be expanded as

$$\hat{p}_t(v,t) = p_s(v) \int\limits_0^{\ell_c/v} dz R(t-z), \tag{26}$$

for $t > \ell_c/v$ and $\hat{p}_t(v,t) = p_s(v)$ for $0 < t \leq \ell_c/v$; $R(t)$ is the probability per time that a particle arrives at a turning point at time t, see Appendix A. Thus, the t-Lagrangian velocity PDF is determined by the sampling of the steady s-Lagrangian PDF $p_s(v)$ between turning points. The right side of (26) expresses the probability $p_s(v)$ of encountering velocity v at a turning point times the probability that the particle has arrived within an interval of length ℓ_c/v before the observation time. The arrival time frequency $R(t)$ at a turning point satisfies the Kolmogorov-type equation

$$R(t) = \delta(t) + \int\limits_0^t dt' R(t') \psi(t - t'). \tag{27}$$

The probability per time to just arrive at a turning point is equal to the probability to be at a turning point at any time t' times the probability to make a transition of duration $t - t'$ to arrive at the next turning point. The t-Lagrangian velocity PDF (26) is non-stationary.

From (27), the Laplace space solution for $R^*(\lambda)$ is

$$R^*(\lambda) = \frac{1}{1 - \psi^*(\lambda)}. \tag{28}$$

In the limit $\lambda\tau_v \ll 1$, it can be approximated by $R^*(\lambda) = (\lambda\tau_v)^{-1} + \dots$ and therefore for $t \gg \tau_v$, we approximate $R(t) = \tau_v^{-1} + \dots$. Thus, in the limit in the limit of $t \gg \tau_v$, we obtain from (26)

$$\hat{p}_t(v, t) = p_e(v) + \dots. \tag{29}$$

Thus asymptotically, $\hat{p}_t(v, t)$ converges toward the Eulerian velocity PDF $p_e(v)$.

Similarly, we obtain for the two-point PDF of the t-Lagrangian velocity the equation

$$\hat{p}_t(v, t; v', t') = p_s(v') \times \int\limits_0^{\ell_c/v'} dz' \hat{p}(v, t - t' + z') R(t' - z'), \tag{30}$$

where $t > t'$, see Appendix A. It is non-stationary as indicated by its explicit dependence on t'. Again, in the limit $t, t' \gg \tau_v$, we approximate

$$\hat{p}_t(v, t; v', t') = p_e(v') \hat{p}(v, t - t'). \tag{31}$$

It is therefore asymptotically stationary.

In summary, the classical CTRW describes the evolution of the t-Lagrangian velocity PDF from the flux weighted Eulerian to the Eulerian velocity PDF. The t-Lagrangian velocities are non-stationary [27]. This property is also called aging in the literature [20]. In the following, we analyze a CTRW formulation that allows for stationary t-Lagrangian statistics and accounts for the evolutions of the t- and s-Lagrangian velocity PDFs from any initial distribution.

B. Markov Process of s-Lagrangian Velocities

In order to introduce correlations between subsequent particle velocities, and thus quantify the evolution of Lagrangian velocity statistics, we describe the velocity series $v_s(s)$ measured equidistantly along a streamline as a Markov process [2, 13, 15, 28]. The evolution of the s-Lagrangian velocity PDF is now given by the Chapman-Kolmogorov equation

$$\hat{p}_s(v, s + \Delta s) = \int\limits_0^\infty dv' r(v, \Delta s | v') \hat{p}_s(v', s), \tag{32}$$

where we assume that the transition PDF $r(v, s | v, s') \equiv r(v, s - s' | v')$ is stationary in s. The evolution of particle time in this CTRW is given by

$$t(s + \Delta s) = t(s) + \frac{\Delta s}{v_s(s)}. \tag{33a}$$

The joint Markov process $[v_s(s), t(s)]$ of streamwise velocity and time is characterized by the joint transition density

$$\psi(v, t - t', \Delta s | v') = r(v, s | v') \delta(t - t' - \Delta s / v'). \tag{33b}$$

Note that a Markov-chain may be characterized by the convergence rate of the transition PDF $r(v, n\Delta s | v')$ toward its steady state, which here is given by

$$\lim_{n \to \infty} r(v, n\Delta s | v') = p_s(v). \tag{33c}$$

The (spatial) convergence rate is given by the inverse of the correlation distance ℓ_c along the streamline. We consider now a process that is uniquely characterized by the steady state PDF $p_s(v)$ and the streamwise correlation distance ℓ_c, and model the s-Lagrangian velocity series by the stochastic relaxation process

$$v_s(s + \Delta s) = [1 - \xi(s)] v(s) + \xi(s) \nu(s). \tag{33d}$$

The random velocities $\nu(s)$ are identical independently distributed according to the steady s-Lagrangian velocity PDF $p_s(\nu)$. The $\xi(s)$ are identical independently distributed Bernoulli variables that take the value 1 with probability $1 - \exp(-\Delta s / \ell_c)$ and 0 with probability $\exp(-\Delta s / \ell_c)$. Thus, its PDF is

$$p_\xi(\xi) = \exp(-\Delta s / \ell_c) \delta(\xi) + [1 - \exp(-\Delta s / \ell_c)] \delta(\xi - 1). \tag{33e}$$

The initial velocity distribution is given by $p_0(v)$. The transition probability $r(v, s | v')$ for the process (33d) is given by

$$r(v, s | v') = \exp(-s / \ell_c) \delta(v - v') + [1 - \exp(-s / \ell_c)] p_s(v). \tag{33f}$$

The velocity process is fully defined by the transition PDF (33f) and the PDF $p_0(v)$ of initial velocities.

1. Space-Lagrangian Velocity Statistics

Using the explicit expression (33f) in (32) and performing the continuum limit $\Delta s \to 0$, we obtain the following Master equation for the streamwise evolution of $\hat{p}_s(v, s)$,

$$\frac{\partial \hat{p}_s(v, s)}{\partial s} = \ell_c^{-1} [p_s(v) - \hat{p}_s(v, s)] \tag{34}$$

subject to the initial condition $\hat{p}_s(v, s = 0) = p_0(v)$. Its solution

$$\hat{p}_s(v, s) = p_s(v) + \exp(-s / \ell_c) [p_0(v) - p_s(v)] \tag{35}$$

converges exponentially from $p_0(v)$ toward the steady state distribution $p_s(v)$, and for $p_0(v) = p_s(v)$ it is stationary. The mean s-Lagrangian velocity is defined by

$$\langle v_s(s) \rangle = \int\limits_0^\infty dv v \hat{p}_s(v, s), \tag{36}$$

and from (35) we obtain the explicit expression

$$\langle v_s(s)\rangle = \langle v_s\rangle + \exp(-s/\ell_c)\left[\langle v_0\rangle - \langle v_s\rangle\right], \quad (37)$$

Under stationary conditions, this means for $v_0 = v_s$, it is constant equal to $\langle v_s\rangle$.

The velocity covariance is then defined by

$$C_s(s,s') = \langle v_s(s)v_s(s')\rangle - \langle v_s(s)\rangle\langle v_s(s')\rangle, \quad (38)$$

where the velocity cross-moment is

$$\langle v_s(s)v_s(s')\rangle = \int_0^\infty dv \int_0^\infty dv' v v' r(v, s-s'|v')p_s(v',s'), \quad (39)$$

for $s > s'$. Using (34) and (33f), we obtain for $s > s'$ the explicit expression

$$C_s(s,s') = \left(\langle v_0\rangle - \langle v_s\rangle\right)^2 \exp(-s/\ell_c)\left[1-\exp(-s'/\ell_c)\right] + \sigma_{v_s}^2 \exp[-(s-s')/\ell_c] + \left(\sigma_{v_0}^2 - \sigma_{v_s}^2\right)\exp(-s/\ell_c). \quad (40)$$

For stationary initial velocities $v_0 = v_s$, it reduces to $C_s(s,s') \equiv C_s(s-s') = \sigma_{v_s}^2 \exp[-(s-s')/\ell_c]$.

2. *Time-Lagrangian Velocity Statistics*

Here we quantify the temporal evolution of the Lagrangian velocity distribution. The existence of a spatial correlation length entails short range correlation in space and long range correlation in time for the Lagrangian velocities, which we quantify in the following.

In the continuum limit of $\Delta s \to 0$, the time process (33a) becomes

$$\frac{dt(s)}{ds} = \frac{1}{v_s(s)}. \quad (41)$$

The conjugate process $s(t)$, which is the distance traveled along the streamline until time t is defined by $s(t) = \sup\{s|t(s) \le t\}$. The t-Lagragian velocities $v_t(t)$ are now given in terms of $v_s(s)$ as

$$v_t(t) = v_s[s(t)], \quad (42)$$

a. One-Point Statistics Thus, the t-Lagrangian velocity PDF reads now as

$$\hat{p}_t(v,t) = \langle\delta\left(v - v_s[s(t)]\right)\rangle. \quad (43)$$

Using the properties of the Dirac-delta, we can expand this equation into

$$\hat{p}(v,t) = \int_0^\infty ds v^{-1} R(v,t,s), \quad (44)$$

where we defined the probability density $R(v,t,s)$ that a particle has the velocity v and the time t at a distance s along the trajectory as

$$R(v,t,s) = \langle\delta[v - v(s)]\delta[t - t(s)]\rangle. \quad (45)$$

Note that $R(v,t,s)$ is the density of the joint Markov process (33) for $[v_s(s), t(s)]$. Thus, it satisfies the Chapman-Kolmogorov equation

$$R(v,t,s+\Delta s) = \int_0^\infty dv' \int_0^t dz \psi(v, t-z, \Delta s|v')R(v',z,s). \quad (46)$$

Inserting (33b) and (33f) into the right side of (46) and taking the limit $\Delta s \to 0$ gives the Master equation (see Appendix B)

$$\frac{\partial R(v,t,s)}{\partial s} = -\ell_c^{-1}R(v,t,s) - v^{-1}\frac{\partial R(v,t,s)}{\partial t} + \ell_c^{-1}p_s(v)\int_0^\infty dv' R(v',t,s), \quad (47)$$

with the initial condition $R(v,t,s=0) = p_0(v)\delta(t)$. Integrating this equation over s according to (44) gives for the t-Lagrangian velocity PDF the integro-differential equation

$$\frac{\partial \hat{p}_t(v,t)}{\partial t} = -\frac{v}{\ell_c}\hat{p}_t(v,t) + p_s(v)\int_0^\infty dv' \frac{v'}{\ell_c}\hat{p}_t(v',t) \quad (48)$$

with the initial condition $\hat{p}(v,t=0) = p_0(v)$. Its solution in Laplace space is given by (see Appendix B)

$$\hat{p}_t^*(v,\lambda) = p_0(v)g_0^*(v,\lambda) + \frac{v}{\langle v_e\rangle}\frac{p_e(v)g_0^*(v,\lambda)\psi_0^*(\lambda)}{1-\psi_s^*(\lambda)}, \quad (49)$$

where we defined the propagator

$$g_0(v,t) = \exp(-tv/\ell_c), \quad (50)$$

whose Laplace transform is given by $g_0^*(v,\lambda) = (\lambda + v/\ell_c)^{-1}$. We define the transit time distributions $\psi_0(t)$, $\psi_s(t)$, and $\psi_e(t)$ through

$$\psi_i(t) = \tau_v^{-1}\int_0^\infty dv g_0(v,t)\frac{vp_i(v)}{\langle v_e\rangle} \quad (51)$$

with $i = 0, s, e$. Note that its initial value is $\psi_i(t=0) = \langle v_i\rangle/\ell_s$. Its Laplace transform is given by

$$\psi_i^*(\lambda) = \tau_v^{-1}\int_0^\infty dv \frac{vp_i(v)}{(\lambda + v/\ell_c)\langle v_e\rangle}. \quad (52)$$

It can be seen from (49) that $\hat{p}(v,t)$ is steady for the initial condition $p_0(v) = p_e(v)$ and is unsteady for any other initial condition by noting that $1 - \psi_s^*(\lambda) = \lambda\tau_v\psi_e^*(\lambda)$.

Expression (49) quantifies the evolution of the t-Lagrangian velocity distribution through potentially long-range temporal correlations reflected by the transit time distributions (51). Note that the transition time PDFs (51) are different from definition (23) for the classical s–discrete CTRW framework discussed in Section III A.

b. Two-Point Statistics The two-point velocity density is defined here by

$$\hat{p}(v,t;v',t') = \langle \delta(v - v[s(t)])\delta(v' - v[s(t')])\rangle. \tag{53}$$

Along the same lines as above, we derive by using the properties of the Dirac-delta

$$\hat{p}(v,t;v',t') = \int_0^\infty ds \int_0^\infty ds' v^{-1} R(v, t-t', s-s'|v') \times v'^{-1} R(v',t',s'). \tag{54}$$

The conditional PDF $R(v,t-t',s-s'|v')$ describes the joint distribution of $[v_s(s), t(s)]$ conditional to $v_s(s') = v'$ and $t(s') = t'$. It satisfies the Master equation (47) with the initial condition $R(v,t,s=0|v') = \delta(v-v')\delta(t)$. Note that $R(v,t-t',s-s'|v')$ is stationary in t and s due to the stationarity of the velocity and time processes as expressed by the transition PDF (33b). Using definition (44), we can now write (54) as

$$\hat{p}(v,t;v',t') = \hat{p}(v,t-t'|v')\hat{p}_t(v',t'). \tag{55}$$

where we defined

$$\hat{p}(v,t|v') = v^{-1}\int_0^\infty ds R(v,t,s|v'). \tag{56}$$

It satisfies the integro-differential equation (48) for the initial condition $\hat{p}(v,t=0|v') = \delta(v-v')$. Its Laplace space solution is obtained from (49) by setting $p_0(v) = \delta(v-v')$ as

$$\hat{p}_t^*(v,\lambda|v') = g_0^*(v,\lambda)\delta(v-v') + \frac{vv'}{\langle v_e\rangle^2\tau_c}\frac{p_e(v)g_0^*(v,\lambda)g_0^*(v',\lambda)}{1-\psi_s^*(\lambda)}, \tag{57}$$

where we note that here $\psi_0^*(\lambda) = g_0^*(v',\lambda)v'/\ell_c$. Recall that the one-point PDF $\hat{p}(v,t)$ is stationary and equal to $p_e(v)$ for the initial condition $p_0(v) = p_e(v)$. Under these conditions, the two-point density (55) is then

$$\hat{p}(v,t;v',t') \equiv \hat{p}(v,t-t',v') = \hat{p}(v,t-t'|v')p_e(v'), \tag{58}$$

and so is stationary. In the following, we determine the mean and covariance of the t-Lagrangian velocities as well as the corresponding particle dispersion.

IV. VELOCITY MEAN, COVARIANCE AND DISPERSION

We study here the t-Lagrangian mean velocity, its covariance and the particle dispersion for the CTRW model presented in Section III B. We investigate these quantities for the following Γ–distribution of Eulerian velocities

$$p_e(v) = \frac{(v/v_0)^{\alpha-1}\exp(-v/v_0)}{v_0\Gamma(\alpha)} \tag{59}$$

for $\alpha > 0$, which provides a parametric model for the low end of Eulerian velocity distributions in porous media both on the pore and on the Darcy scale [5, 9]. As initial conditions we consider either the Eulerian (59) or steady s-Lagrangian velocity PDF (10), which is obtained from the Eulerian velocity PDF through flux weighting

$$p_s(v) = \frac{(v/v_0)^{\alpha}\exp(-v/v_0)}{v_0\Gamma(\alpha+1)}. \tag{60}$$

Note that the Eulerian and flux-weighted mean and mean square velocities are

$$\langle v_e\rangle = \alpha v_0, \qquad \langle v_e^2\rangle = \alpha(\alpha+1)v_0^2 \tag{61}$$

$$\langle v_s\rangle = v_0(\alpha+1), \qquad \langle v_s^2\rangle = v_0^2(\alpha+1)(\alpha+2). \tag{62}$$

Inserting (59) into (51), we obtain for the transit time distribution $\psi_e(t)$

$$\psi_e(t) = \frac{\alpha}{\tau_0(1+t/\tau_0)^{1+\alpha}}. \tag{63}$$

where $\tau_0 = \ell_c/v_0$. For the transit time distribution $\psi_s(t)$, we obtain analogously

$$\psi_s(t) = \frac{\alpha+1}{\tau_0(1+t/\tau_0)^{2+\alpha}}. \tag{64}$$

The Laplace transforms of $\psi_e(t)$ and $\psi_s(t)$ can be expanded by using Tauberian theorems. For $0 < \alpha < 1$, $\psi_e^*(\lambda)$ is

$$\psi_e^*(\lambda) = 1 - a_\alpha(\lambda\tau_0)^\alpha, \tag{65}$$

where $a_\alpha = \Gamma(1-\alpha)$. For $\alpha = 1$, we have

$$\psi_e^*(\lambda) = 1 + \lambda\tau_0\ln(\lambda\tau_0). \tag{66}$$

In the range $0 < \alpha < 1$, we obtain for $\psi_s^*(\lambda)$ the expansion

$$\psi_s^*(\lambda) = 1 - \lambda\tau_v + b_\alpha(\lambda\tau_0)^{1+\alpha}, \tag{67}$$

where $\tau_v = \ell_c/(\alpha v_0)$ and $b_\alpha = \Gamma(2-\alpha)$. For $\alpha = 1$, one obtains

$$\psi_s^*(\lambda) = 1 - \lambda\tau_0 - (\lambda\tau_0)^2\ln(\lambda\tau_0). \tag{68}$$

Note that the case $\alpha = 1$ corresponds to an exponential distribution of Eulerian velocities.

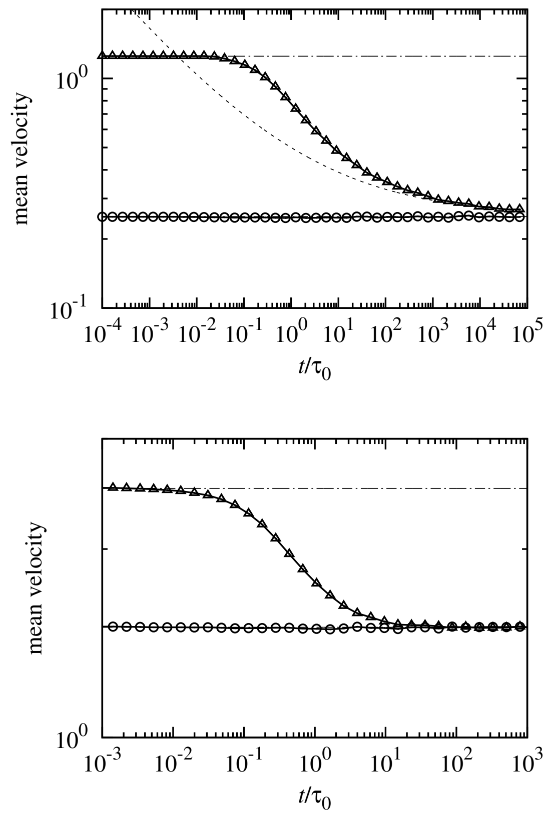

Figure 1. Evolution of the mean velocity under stationary and non-stationary conditions for (circles) $p_0(v) = p_e(v)$ and (triangles) $p_0(v) = p_s(v)$ for (top panel) $\alpha = 1/4$ and (bottom panel) $\alpha = 3/2$. The dashed line in the top panel indicates the asymptotic behavior (75). The dash-dotted lines indicate the average stationary s-Lagrangian and Eulerian velocities. The numerical random walk simulation to produce these data are based on (33) for $\Delta s = 10^{-2}\ell_c$ for 10^5 particles.

For $\alpha > 1$, both the first and second moments of $\psi_s(t)$ exist, such that $\psi_s^*(\lambda)$ can be expanded as

$$\psi_s^*(\lambda) = 1 - \lambda\tau_v + \frac{\langle\tau_s^2\rangle}{2}\lambda^2. \tag{69}$$

In the following, we will discuss the mean t-Lagrangian velocity, the velocity covariance and particle dispersion. We present general Laplace space expressions based on the explicit expressions for the one- and two point velocity PDFs derived in Section III B 2, and study their temporal behavior for the Eulerian velocity PDF given by the Γ–distribution (59). To this end, we perform random walk particle tracking simulations based on (33) and derive explicit expressions for the early and late time behaviors using the expansions (67)–(69) of the Laplace transform of the streamwise transition time PDF $\psi_s(t)$.

A. Mean Velocity

The mean particle velocity is equal to the one-point t-Lagrangian velocity moment

$$m_1(t) = \int_0^\infty dv v\hat{p}_t(v,t). \tag{70}$$

Using (49), we obtain for the Laplace transform of $m_1(t)$

$$m_1^*(\lambda) = \ell_c\psi_0^*(\lambda) + \int_0^\infty dv \frac{v^2}{\langle v_e\rangle}\frac{p_e(v)g_0^*(v,\lambda)\psi_0^*(\lambda)}{1-\psi_s^*(\lambda)}. \tag{71}$$

For the stationary initial conditions, $p_0(v) = p_e(v)$, the particle velocity is constant, $m_1(t) = \langle v_e\rangle$ and equal to the mean Eulerian velocity.

For the non-stationary initial conditions $p_0(v) = p_s(v)$ we obtain at short times $t \ll \tau_v$

$$m_1(t) = \ell_c\psi_s(t). \tag{72}$$

This means it decreases from its initial value $\langle v_s\rangle$ as $\psi_s(t)$. For times $t \gg \tau_v$ and $0 < \alpha < 1$, we use the expansion (67) in (71), which gives in leading order

$$m_1^*(\lambda) = \frac{\langle v_e\rangle}{\lambda} + \frac{\langle v_e\rangle\tau_0 b_\alpha}{b_1}(\lambda\tau_0)^{\alpha-1}. \tag{73}$$

For $\alpha = 1$ we obtain

$$m_1^*(\lambda) = \frac{\langle v_e\rangle}{\lambda} - \ell_c\ln(\lambda\tau_0). \tag{74}$$

Thus, the long-time behavior of $m_1(t)$ for $0 < \alpha \leq 1$ is

$$m_1(t) = \langle v_e\rangle + c\langle v_e\rangle(t/\tau_0)^{-\alpha}, \tag{75}$$

where we defined $c = b_\alpha/[\Gamma(1-\alpha)b_1]$ for $0 < \alpha < 1$ and $c = 1$ for $\alpha = 1$. This means, the mean velocity converges as a power-law toward its asymptotic value, which is given by the Eulerian mean velocity.

For $\alpha > 1$, we use (69) in order to obtain in leading order for $\lambda \ll \tau_0$

$$m_1^*(\lambda) = \frac{\langle v_e\rangle}{\lambda} + \ell_c + \langle v_e\rangle\frac{\langle\tau_s^2\rangle}{2\tau_v}. \tag{76}$$

This means, for $t \gg \tau_v$, $m_1(t)$ can be written as

$$m_1(t) = \langle v_e\rangle + \left(\ell_c + \langle v_e\rangle\frac{\langle\tau_s^2\rangle}{2\tau_v}\right)\delta(t). \tag{77}$$

Note that the Dirac-delta indicates that the convergence toward its asymptotic value is faster than $1/t$. These behaviors are illustrated in Figure 1, which shows the evolution of the t-Lagrangian mean velocity with time under Eulerian and flux-weighted Eulerian initial conditions for $\alpha = 1/4$ and $\alpha = 3/2$.

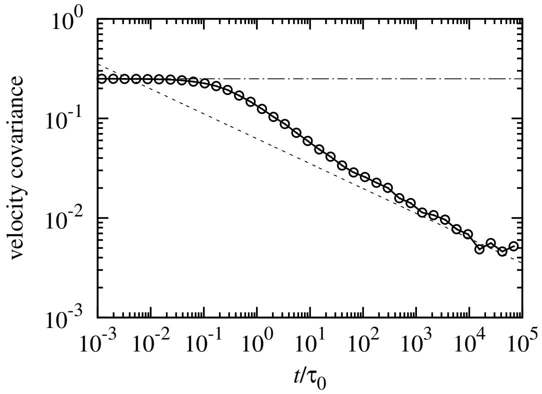

Figure 2. Covariance of the t-Lagrangian velocity under the stationary condition $p_0(v) = p_e(v)$ for $\alpha = 1/4$. The dashed line indicates the asymptotic behavior (85). The dash-dotted lines indicates the velocity variance. The numerical random walk simulation to produce these data are based on (33) for $\Delta s = 10^{-2}\ell_c$ for 10^5 particles.

B. Velocity Covariance

The t-Lagrangian velocity covariance is given by

$$C_t(t,t') = m_2(t,t') - m_1(t)m_1(t'), \tag{78}$$

where we defined the two-point velocity moment by

$$m_2(t,t') = \int\limits_0^\infty dv \int\limits_0^\infty dv' vv' \hat{p}_t(v,t;v',t'), \tag{79}$$

which can be written in terms of (55) for the two-point velocity PDF as

$$m_2(t,t') = \int\limits_0^\infty dv' m_1(t-t'|v')v'\hat{p}_t(v',t'), \tag{80}$$

where we defined the conditional velocity moment as

$$m_1(t|v') = \int\limits_0^\infty dv v \hat{p}_t(v,t|v'). \tag{81}$$

The Laplace transform of (81) is then obtained from (57) as

$$m_1^*(\lambda|v') = v' g_0^*(v',\lambda) + \int\limits_0^\infty dv \frac{v^2 v'}{\langle v_e\rangle^2 \tau_v} \frac{p_e(v) g_0^*(v,\lambda) g_0^*(v',\lambda)}{1-\psi_s^*(\lambda)}. \tag{82}$$

We first consider the case $0 < \alpha \leq 1$. For times $t \gg \tau_v$, this means for $\lambda\tau_v \ll 1$, we find by using (67) and (68) in (82) that the leading order of $m_1^*(\lambda|v')$ is given by (73) for $0 < \alpha < 1$ and (74) for $\alpha = 1$. Specifically, this implies that $m_1(t|v')$ is independent of v'. Using (75) in (80), we obtain

$$m_2(t,t') = \left[\langle v_e\rangle + \frac{c\langle v_e\rangle\tau_0^\alpha}{(t-t')^\alpha}\right] m_1(t'). \tag{83}$$

Under stationary conditions, $p_0(v) = p_e(v)$, $m_1(t) = \langle v_e\rangle$ and $m_2(t,t') \equiv m_2(t-t')$, hence

$$m_2(t-t') = \langle v_e\rangle^2 + \frac{c\langle v_e\rangle^2\tau_0^\alpha}{(t-t')^\alpha}. \tag{84}$$

Thus, the velocity covariance is stationary and behaves for $(t-t') \gg \tau_v$ and $0 < \alpha \leq 1$ as

$$C_t(t-t') = \frac{c\langle v_e\rangle^2\tau_0^\alpha}{(t-t')^\alpha}. \tag{85}$$

This behavior is illustrated in Figure 2.

Under the non-stationary condition with $p_0(v) = p_s(v)$, we use the fact that $m_1(t|v') = m_1(t)$ in the limit $t \gg \tau_v$ in order to write

$$m_2(t,t') = m_1(t-t')m_1(t'). \tag{86}$$

Accordingly, we obtain for the covariance in the limit $(t-t') \gg \tau_v$

$$C_t(t,t') = m_1(t')\left[m_1(t-t') - m_1(t)\right]. \tag{87}$$

We now consider the case $\alpha > 1$. For $\lambda\tau_v \ll 1$, we expand (82) by using (69) to leading order, which gives

$$m_1(\lambda|v') = \frac{\langle v_e\rangle}{\lambda} + \frac{\langle v_e\rangle\langle\tau_s^2\rangle}{2\tau_v} - \frac{\langle v_2\rangle\ell_c}{v'}. \tag{88}$$

Thus, we obtain for $m_2(t,t')$

$$m_2(t,t') = \left[\langle v_e\rangle + \frac{\langle v_e\rangle\langle\tau_s^2\rangle}{2\tau_v}\delta(t-t')\right] m_1(t') - \ell_c\langle v_e\rangle\delta(t-t'). \tag{89}$$

For $t - t' \gg \tau_v$, we obtain for the covariance under both stationary and non-stationary conditions the expression

$$C_t(t-t') = \ell_c\langle v_e\rangle\left(\frac{\langle\tau_s^2\rangle}{2\tau_v^2} - 1\right)\delta(t-t'). \tag{90}$$

Again note that the Dirac-delta indicates here that the covariance decays faster than $1/t$. These expression allow studying the dynamics of dispersion as a function of the Eulerian velocity distribution and the initial injection, as discussed in the following.

C. Dispersion

The time-dependent dispersion coefficient $\mathcal{D}(t)$ is obtained from the Green-Kubo relation [29] as the integral of the t-Lagragian velocity correlation as

$$\mathcal{D}(t) = \int\limits_0^t dt' C_t(t,t'). \tag{91}$$

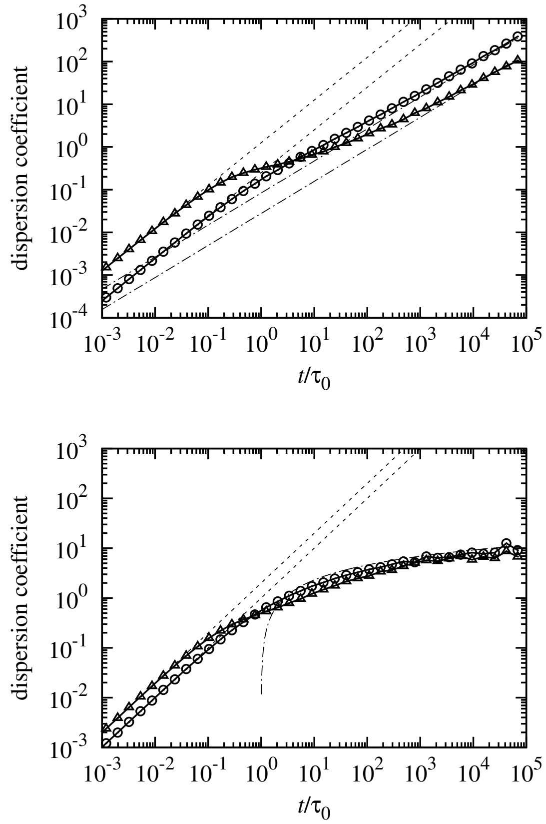

Figure 3. Evolution of the dispersion coefficient under stationary and non-stationary conditions for (circles) $p_0(v) = p_e(v)$ and (triangles) $p_0(v) = p_s(v)$ for (top panel) $\alpha = 1/4$, (bottom panel) $\alpha = 1$. The dashed lines indicate the ballistic behaviors (92) at short times, dash-dotted lines the asymptotic power-law behaviors (93) and (94) for $\alpha = 1/4$, and the logarithmic behavior (95) for $\alpha = 1$. The numerical random walk simulation to produce these data are based on (33) for $\Delta s = 10^{-2}\ell_c$ for 10^5 particles.

At time $t \ll \tau_v$, particle velocities are strongly correlated. As a consequence, the dispersion coefficient grows ballistically as

$$\mathcal{D}(t) = \langle (v_0 - \langle v_0 \rangle)^2 \rangle t. \tag{92}$$

Thus, for the initial condition $p_0(v) = p_s(v)$ the ballistic initial growth is faster than for the stationary condition $p_0(v) = p_e(v)$, because the variance of the flux weighted $p_s(v)$ is larger than the variance of the Eulerian $p_e(v)$. For times $t > \tau_v$, particle velocities decorrelate from their initial values. High velocities decorrelate faster than low velocities because the characteristic time at which a particle of velocity v makes a velocity transition is given by ℓ_c/v. Thus, at time τ_v most of the particles with $v > \langle v_e \rangle$ have experienced a velocity transition, which particles with $v < \langle v_e \rangle$ persist in their initial velocities. The dispersion coefficient $\mathcal{D}(t)$ then crosses over to its asymptotic long time behavior, which we study in the following.

We first consider the case $0 < \alpha \leq 1$. Under stationary conditions, this means for $p_0(v) = p_e(v)$, we obtain from (85) for $t \gg \tau_v$ and $0 < \alpha < 1$

$$\mathcal{D}(t) = \langle v_e \rangle \ell_c \frac{c\alpha}{1-\alpha} (t/\tau_0)^{1-\alpha}. \tag{93}$$

Thus, the dispersion behavior is superdiffusive. In the non-stationary case, for $p_0(v) = p_s(v)$, we obtain from (87) and (75) at $t \gg \tau_v$

$$\mathcal{D}(t) = \langle v_e \rangle \ell_c \frac{c\alpha^2}{(1-\alpha)^2} (t/\tau_0)^{1-\alpha}. \tag{94}$$

It grows asymptotically with the same power-law, but slower then in the stationary case. Thus, while the growth rate of particle dispersion is initially larger for the non-stationary initial condition, asymptotically its growth is slower than for the stationary initial velocity PDF. For $\alpha = 1$, we obtain for both stationary and non-stationary intial conditions the behavior

$$\mathcal{D}(t) = \langle v_e \rangle \ell_c \ln(t/\tau_0). \tag{95}$$

Figure 3 illustrates the evolution of $\mathcal{D}(t)$ for $\alpha = 1/4$ and $\alpha = 1$ under stationary and non-stationary initial conditions. For times $t \ll \tau_v$, we observe the ballistic behavior (92), which persists until particle velocities start decorrelating from their initial velocity. Then an intermediate time regime develops which marks the cross-over to the super-diffusive long-time behavior. In this regime, the $\mathcal{D}(t)$ for the non-stationary initial velocity distribution grows slower than for stationary. The dispersion behavior here is due to the fluctuations of fast velocities, which have already decorrelated, and low velocity particles that persist in the ballistic mode. The stationary, Eulerian initial distribution $p_e(v)$ has a stronger weight on low velocities than the flux-weighted $p_s(v)$. Thus, dispersion for the former is higher in the intermediate time regime than for the latter. The end of the intermediate regime is characterized by the decorrelation of most particles from their initial velocities. In the long time regime, we observe for $0 < \alpha < 1$ the power-law behaviors (93) and (94), for stationary and non-stationary initial conditions. The difference persists and the dispersion coefficient for stationary initial conditions is larger than for non-stationary. The power-law scalings (93) and (94) are consistent with the ones observed in the CTRW for uncorrelated particle velocities [30, 31]. For $\alpha = 1$, we observe the logarithmic behavior (95) for both stationary and non-stationary initial conditions.

For $\alpha > 1$, the dispersion coefficient converges for $t \gg \tau_v$ both for stationary and non-stationary initial conditions towards the constant asymptotic long-time value

$$\mathcal{D}^e = \langle v_e \rangle \ell_c \left(\frac{\langle \tau_s^2 \rangle}{2\tau_v^2} - 1 \right). \tag{96}$$

Figure 4 illustrates the evolution of the dispersion coefficient toward the asymptotic value for $\alpha = 3/2$. At

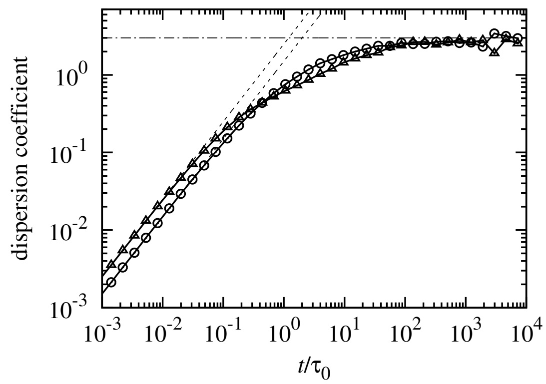

Figure 4. Evolution of the dispersion coefficient for (circles) steady and (triangles) unsteady initial velocity PDFs for $\alpha = 3/2$. The dashed lines indicate the ballistic behaviors (92) at short times, dash-dotted lines the asymptotic long time value (96). The numerical random walk simulation to produce these data are based on (33) for $\Delta s = 10^{-2}\ell_c$ with 10^5 particles.

short times $t \ll \tau_v$, both dispersion coefficients evolve ballistically, again, the one for the non-stationary initial condition evolves faster. Then for $t > \tau_v$, the dispersion coefficient for stationary initial conditions grows faster than for non-stationary. As pointed out above, the dispersion behavior is due to the fluctuations of decorrelated fast velocities, and persistent low velocity. As the stationary, Eulerian initial velocity distribution gives a higher probability to low velocities than the flux-weighted, the contrast between particle positions increases faster. The asymptotic regime is reached as the particle velocities fully decorrelate from their initial values. For times $t \gg \tau_v$ the dispersion coefficients for both stationary and non-stationary initial conditions converge to the same asymptotic long-time value (96).

V. SUMMARY AND CONCLUSIONS

We develop a CTRW approach for the evolution of Lagrangian velocities based on a Markov model for the streamwise equidistant Lagrangian velocities in the form of a stochastic relaxation process. The CTRW framework provides a natural formalism to quantify the impact of the persistence of particle velocities in space on the Lagrangian velocity statistics in time. It has been used to quantify intermittent particle velocities and accelerations for flow through pore- and Darcy-scale porous media, in which flow velocities vary on a characteristic length scale. The velocity statistics in CTRW formulations based on independent successive particle velocities are in general non-stationary. This however, is not necessarily the case for particle motion through heterogeneous flow fields. Specifically, under Eulerian and Lagrangian ergodicity, the stationarity of the Lagrangian velocity series depends on the initial velocity distribution.

In order to shed light on these dynamics, we first discuss the relation between the Eulerian flow properties and the t-Lagrangian and s-Lagrangian velocities. The t-Lagrangian velocities are defined as the particle velocities sampled isochrone along a streamline, the s-Lagrangian velocities accordingly through equidistant sampling. We find that the PDFs of s- and t-Lagrangian velocities are related through flux weighting. This can be understood by the fact that isochrone sampling gives a higher weight to low velocities because particles spend more time at low velocities, while equidistant sampling assigns the same weight to high and low velocities. Under Eulerian and Lagrangian ergodicity and for volume conserving flows, the Eulerian velocity PDF is equal to the t-Lagrangian PDF. This gives a direct relation between the s-Lagrangian velocity PDF, a transport property, to the Eulerian PDF, a flow property, via flux weighting. We then show that t-Lagrangian velocities are stationary if their initial distribution is equal to the Eulerian, while s-Lagrangian velocities are stationary if their initial distribution is given by the flux-weighted Eulerian distribution.

Based on these considerations, we first analyze the t-Lagrangian velocity statistics in the s-discrete CTRW characterized by independent velocities with a unique velocity distribution. In classical CTRW approaches, the velocity statistics are in general non-stationary. Thus, we introduce a CTRW that is defined through a Markovian velocity process, for which we use a stochastic relaxation relation that is characterized by the steady state s-Lagrangian velocity PDF and the correlation length along the streamlines. Based on this we define a CTRW approach that models the evolution of Lagrangian velocities from arbitrary initial conditions and yields stationary and non-stationary s- and t-Lagrangian velocity series. Specifically, this CTRW is s-continuous, this means the streamwise s-Lagrangian velocity is defined at any point along the streamline and its distribution evolves continuously in s. We determine the evolution equations and solutions for the Lagrangian one-and two-point statistics and discuss the evolution of the mean particle velocity, covariance and dispersion under stationary and non-stationary initial conditions. We apply these results to a Γ-distribution of Eulerian velocities, which serves as a model for heavy-tailed flow-statistics through porous media. The low-end of the velocity spectrum here scales as $p_e(v) \propto v^{\alpha-1}$. For $0 < \alpha \leq 1$ we find strong velocity correlations and anomalous dispersion, this means here a power-law or logarithmic evolution of the dispersion coefficient with time, while for $\alpha > 1$ it evolves toward a constant. These behaviors are fully determined by the Eulerian velocity PDF and the streamwise correlation length. The asymptotic scalings for dispersion are similar as the ones obtained in a corresponding discrete CTRW, as they are attained when particle velocities decorrelate. Their evolution, however, depends on the initial velocity distributions and can be quite different under stationary and non-stationary conditions.

The developed approach sheds light on the modeling and understanding of Lagrangian velocity series in heterogeneous flows, and their evolution under stationary and non-stationary conditions. It provides a bridge between CTRW based modeling approaches of particle transport, and stochastic transport approaches that start from the representation of the Eulerian velocity field, or the medium structure as spatial random fields. The developed CTRW is fully characterized in terms of the Eulerian velocity PDF and the streamwise correlation length, which allows to predict Lagrangian particle dynamics based on the flow or medium properties.

Acknowledgments

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Appendix A: Velocity Statistics for Uncorrelated s-Lagrangian Velocities

The one-point t-Lagrangian velocity PDF (25) can be expanded as

$$\hat{p}(v,t) = \int_0^t dt' \sum_{n=0}^{\infty} \langle \delta(v - v_n)\delta(t' - t_n)\delta_{n,n_t}\rangle, \qquad \text{(A1)}$$

where δ_{ij} denotes the Kronecker-delta. Note that $\delta_{n,n_t} \equiv \mathbb{I}(t_n \leq t < t_{n+1})$. Thus, we can write (A1) as

$$\hat{p}(v,t) = \int_0^t dt' \sum_{n=0}^{\infty} \langle \delta(v - v_n)\delta(t' - t_n)\rangle \times \mathbb{I}(0 \leq t - t' < \ell_c/v), \qquad \text{(A2)}$$

where we used that t_n is independent of v_n, and that per the Dirac-delta, the v_n in the indicator function is set equal to v. We further obtain

$$\hat{p}(v,t) = \int\limits_{t-\ell_c/v}^{t} dt' \sum_{n=0}^{\infty} \langle \delta(v - v_n)\rangle\langle \delta(t' - t_n)\rangle \equiv p_s(v) \int\limits_{t-\ell_c/v}^{t} dt' \sum_{n=0}^{\infty} R_n(t'). \qquad \text{(A3)}$$

for $t > \ell_c/v$; $R_n(t)$ is the PDF of t_n. As t_n is a Markov process in step number, we have the Chapman-Kolmogorov equation for the conditional PDF $R_{n,n'}(t|t')$

$$R_{n+1,n'}(t|t') = \int\limits_{t'}^{t} dz \psi(t - z) R_{n,n'}(z|t'). \qquad \text{(A4)}$$

As the process is homogeneous in n and in t, we have that $R_{n'+m,n'}(t|t') \equiv R_m(t - t')$. The sum over $R_n(t)$,

$$R(t) = \sum_{n=0}^{\infty} R_n(t') \qquad \text{(A5)}$$

satisfies the integral equation (27).

For the two-point PDF, we obtain in analogy to (A3)

$$\hat{p}(v,t;v',t') = p_s(v)p_s(v') \times \int\limits_{t-\ell_c/v}^{t} dz \int\limits_{t'-\ell_c/v'}^{t'} dz' \sum_{n=0}^{\infty}\sum_{n'=0}^{\infty} R_{n,n'}(z,z'), \qquad \text{(A6)}$$

where $R_{n,n'}(z,z')$ is the joint density of t_n and $t_{n'}$, which can be written as

$$R_{n'+m,n'}(z,z') = R_m(z - z')R_{n'}(z'). \qquad \text{(A7)}$$

We used the stationarity of the conditional PDF discussed above. Thus, we obtain now

$$\hat{p}(v,t;v',t') = p_s(v)p_s(v') \times \int\limits_{t-\ell_c/v}^{t} dz \int\limits_{t'-\ell_c/v'}^{t'} dz' \sum_{m=0}^{\infty}\sum_{n'=0}^{\infty} R_m(z - z')R_{n'}(z'), \equiv p_s(v)p_s(v') \int\limits_{t-\ell_c/v}^{t} dz \int\limits_{t'-\ell_c/v'}^{t'} dz' R(z - z')R(z'). \qquad \text{(A8)}$$

Shifting $z \to t - z$ and $z' \to t' - z'$ gives

$$\hat{p}(v,t;v',t') = p_s(v)p_s(v') \times \int\limits_{0}^{\ell_c/v} dz \int\limits_{0}^{\ell_c/v'} dz' R(t - t' + z' - z)R(t' - z'). \qquad \text{(A9)}$$

Using now expression (26) gives (30).

Appendix B: Velocity Statistics for Markov Process of s-Lagrangian Velocities

The Master equation (47) for $R(v,t,s)$ follows from the Chapman-Kolmogorov equation (46) in the limit $\Delta s \to 0$. In fact, inserting (33b) and (33f) gives

$$R(v,t,s+\Delta s) = \exp(-\Delta s/\ell_c)R(v,t - \Delta s/v, s) + [1 - \exp(-\Delta s/\ell_c)]p_s(v) \int\limits_{0}^{\infty} dv' R(v', t - \Delta s/v', s). \qquad \text{(B1)}$$

Expanding the left hand right side for small Δs gives

$$
\begin{aligned}
&R(v,t,s) + \Delta s \frac{\partial R(v,t,s)}{\partial s} + \cdots = R(v,t,s) \\
&- \frac{\Delta s}{v}\frac{\partial R(v,t,s)}{\partial t} - \frac{\Delta s}{\ell_c} R(v,t,s) + \\
&\frac{\Delta s}{\ell_c} p_s(v) \int_0^\infty dv' R(v',t,s) + \ldots,
\end{aligned} \tag{B2}
$$

where the dots denote higher order contributions in Δs. Dividing by Δs and taking the limit $\Delta s \to 0$ gives (47).

We now derive the solution of Equation (48). To this end, we perform the Laplace transform, which gives

$$
\lambda \hat{p}_t^*(v,\lambda) = -\frac{v}{\ell_c}\hat{p}_t^*(v,\lambda) + p_s(v)\int_0^\infty dv' \frac{v'}{\ell_c}\hat{p}_t^*(v',\lambda). \tag{B3}
$$

This is a Fredholm equation of the second kind with degenerate kernel [32]. It can be written as

$$
\begin{aligned}
\hat{p}_t^*(v,\lambda) &= g_0^*(v,\lambda) p_0(v) \\
&+ g_0^*(v,\lambda) p_s(v) \int_0^\infty dv' \frac{v'}{\ell_c} \hat{p}^*(v',\lambda).
\end{aligned} \tag{B4}
$$

where we defined

$$
g_0^*(v,\lambda) = \frac{1}{\lambda + v/\ell_c}. \tag{B5}
$$

The solution of (B4) has the form

$$
\hat{p}_t^*(v,\lambda) = g_0^*(v,\lambda)\left[p_0(v) + p_s(v) A(v,\lambda)\right]. \tag{B6}
$$

Inserting the latter into (B4) gives for $A(v,\lambda)$

$$
A(v,\lambda) = \frac{\psi_0^*(\lambda)}{1 - \psi_s^*(\lambda)} \tag{B7}
$$

where we defined

$$
\psi_i^*(\lambda) = \int_0^\infty dv' g_0^*(v',\lambda)\frac{v'}{\ell_c} p_i(v) \tag{B8}
$$

with $i = 0, s$. Inserting (B7) into (B6) and setting $p_s(v) = v p_e(v)/\langle v_e \rangle$ gives (49).

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