Spatial behavior of anomalous transport

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We present a general derivation of one-dimensional spatial concentration distributions for anomalous transport regimes. Such transport can be captured in the framework of a continuous time random walk with a broad transition time distribution. This general theory includes a Fokker-Planck equation as a particular limiting case. All of the concentration profiles, as well as the associated temporal first passage time distributions, can be written in terms of a single special function (that belongs to the class of Fox functions). In addition, we consider the first two moments of the spatial concentration distributions, and determine not only their scaling behavior with time but also the coefficients and correction terms.

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I. INTRODUCTION

The well-known continuous time random walk (CTRW) framework, initially proposed by Montroll and Weiss [1], Scher and Lax [2], Montroll and Scher [3], and Scher and Montroll [4], is inherently suited to characterizing and quantifying anomalous (non-Fickian) transport. Such transport cannot be described by classical Gaussian models. In a CTRW framework one assumes that a moving particle (tracer) undergoes random transitions in space according to, in general, a coupled space-time probability density function (PDF). Here, as elsewhere [4,5], we concentrate on a wide transition time PDF $\psi(t)$, with an asymptotic algebraic tail for long times: $\psi(t)\sim t^{-1-\beta}$ with constant $\beta>0$. The case $\beta>2$ leads to Gaussian transport while the intervals $0<\beta<1$ and $1<\beta<2$ lead to different anomalous transport regimes [5,6]. In the discussion below, we shall refer to this temporal formulation of the governing PDF as ''standard'' CTRW, in contrast to Lévy flight formulations (which arise for wide transition length distributions; it is argued in [7] that Lévy flight formulations are inadequate in some physical applications, e.g., groundwater hydrology), or a mixed case of wide distributions in both space and time. In analogy to a commonly used derivation of a Fokker-Planck equation from a master equation, Berkowitz *et al.* [7] consider a general CTRW and obtain a Fokker-Planck equation as a particular case.

A physical picture of particle transport, emphasizing temporal aspects at small length scales, has been applied successfully to laboratory and field experimental data, as well as to numerical simulations of particle transport in geological media [8–12]. To date, known results of CTRW provide temporal distributions [i.e., so-called first passage time distributions (FPTDs)] for all possible values of β [4,5] and spatial concentration distributions for some specific waiting time PDFs and values of $\beta=\frac{1}{2}$ and $\frac{3}{2}$ [3]. McLean and Ausman [13] developed simple empirical formulas for spatial concentrations, for $0<\beta<1$. These authors used approximations (so-called no backflow, see Sec. II) similar to what we present here, but neglected the steepest descent asymptotic formula derived in [4]. This formula enables one to approximate an exact solution for large values of the argument (see also Appendix A). Thus the accuracy of the McLean and Ausman [13] empirical formulas could not be checked rigorously and was assumed to be reasonable. Moreover, with the development of fast computers there is today no reason not to use the exact solutions developed here.

Spatial profiles have also been obtained for $0<\beta<1$ using a fractional derivatives approach (e.g., [14]). The fractional derivatives formalism is mathematically equivalent to generalized random walks, if one focuses on algebraic long-tailed transition time and distance distributions (e.g., [7]). We observe that an unfortunate, ''propagating error'' appears in many papers dealing with CTRW and fractional derivatives: it is often suggested that the case $1<\beta<2$ is already in the domain of attraction of the central limit theorem (see [14–17] and references therein) and thus need not be considered separately. As we show in Sec. II, this is only true in the case of no spatial bias, i.e., for symmetric random walks. In general, the infinite second temporal moment of $\psi(t)$ when $1<\beta<2$ leads to a transport behavior that is different from the Gaussian solution distributions and their temporal evolution.

Multidimensional aspects of CTRW are analyzed in [16,18]. As we show in Sec. III B, the conclusion of [16] regarding the peak position at the origin is inexact.

In this paper we develop one-dimensional (1D) spatial concentration distributions (SCDs) for all possible values of β, which follow from ''standard'' CTRW. We show their interconnection and present all the distributions, including FPTDs, in terms of one special function. We note here that the SCD is a residence-averaged concentration while the FPTD is a flux-averaged concentration (in the case of no backflow). Spatial profile concentrations assuming no backflow are developed in Sec. III, while SCDs with backflow are discussed in Sec. IV. One of the major characteristics of the tracer transport is the apparent (or effective) dispersivity and its behavior in the course of time. We present and develop the ''standard'' CTRW-based predictions of this quantity in Sec. V.

II. NON-GAUSSIAN BEHAVIOR FOR $1<\beta<2$

In this section we show that non-Gaussian propagators arise in cases where $1<\beta<2$ if there is a bias ($\bar{l}\neq 0$)

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present, i.e., we show that the first moment $\overline{t}$ of $\psi(t)$ being finite is not sufficient to have Gaussian behavior. The correct (up to the leading term) behavior of the standard deviation of the propagator $\sigma_l(t)$ was first calculated by Shlesinger [6], showing that, for $1>\beta>2$, $\sigma_l(t)\sim\overline{l}\,t^{(3-\beta)/2}$ grows faster than $t^{1/2}$. However, this result remained unnoticed later; here we present an argument given in one such paper [15] and show its error. First, we note that it is reasonable to assume that if two similar values of β are used to describe the same transport process, with one value slightly smaller than 1 and the other value slightly larger than 1, the resulting behavior, including the scaling of the spatial moments with time, should be similar. Since for $0<\beta<1$, the scaling is $\sigma_l(t)\sim t^{\beta}$, the exponent of t should be close to 1 for both $\beta\nearrow 1$ and $\beta\searrow 1$; in contrast, for a Gaussian we have $t^{1/2}$.

We reproduce two paragraphs from Shlesinger *et al.* [15]; p. 500, replacing some original notation by ours:

"An important random variable representing the random walk is the sum S_N, where

$$S_N = X_1 + \cdots + X_N, \tag{1}$$

and the X_i are identically distributed random variables each with mean $\overline{l}$ and variance σ_l^2. If the variance is finite then the central limit theorem can be invoked to obtain the Gaussian probability density, say in one dimension,

$$f(x) = \lim_{N\to\infty} \mathrm{Prob}[x<S_N/\sqrt{N}<x+dx] = (2\pi\sigma_l^2)^{-1/2}\exp(-x^2/2\sigma_l^2). \tag{2}$$

We have set $\overline{l}=0$ and will do so for the rest of the analysis because one can define a new variable $Y=X-\overline{l}$ that has a zero mean. Also we will only discuss one-dimensional cases here.

One may further introduce a probability density $\psi(t)$ governing the time between the events X_i, and study the sum $S_{N(t)}$, where

$$S_{N(t)} = X_1 + \cdots + X_{N(t)}. \tag{3}$$

The random variable $N(t)$ represents the number of events that have occurred in the time interval [0, t]. If $\psi(t)$ has a finite first moment $\overline{t}$ and $\sigma_l^2<\infty$, then again the central limit theorem can be used to show that [6]

$$f(x,t) = \lim_{t\to\infty} \mathrm{Prob}[x<S_{N(t)}/\sqrt{t}<x+dx] = (4\pi Dt)^{-1/2}\exp(-x^2/4Dt) \tag{4}$$

where $D=\sigma_l^2/2\overline{t}$."

The problem with this argument is that if one considers Eq. (3) with $\overline{l}\neq 0$, one recognizes that there are two different mechanisms of dispersion: one is due to the positive $\sigma_{\overline{l}}$ and the other is due to the uncertainty in determining the number of transitions N for a given time t, because $N(t)$ is not a deterministic (single-valued) function. Even if $\sigma_{\overline{l}}=0$ [leading to $D=0$ and a δ function in Eq. (4)], then $S_{N(t)}=\overline{l}N(t)$ is not deterministic and will display some spreading depending on $\psi(t)$. Of course, for $\overline{l}=0$ no such problem arises and one indeed recovers Eq. (4) but for $\overline{l}\neq 0$ the "correction" to the Gaussian is the leading term. The calculations for the spatial moments in the case $1<\beta<2$ for the decoupled space-time single transition function are given in Appendix B. It can be seen there that even for the case of a finite second temporal moment of $\psi(t)$, when indeed the Gaussian is approached asymptotically, there are two distinct contributions to the variance $\sigma_l^2(t)$ of the propagator—one comes from the distribution of single transition lengths and the other from the distribution of single transition times. We thus conclude that the case $1<\beta<2$ with $\overline{l}\neq 0$ does not lead to a Gaussian distribution and constitutes another anomalous regime lying between the self-similar case of $0<\beta<1$ and the Gaussian $(\beta>2)$. In other words, in the presence of a bias, in the case $\beta<2$ the contribution to the dispersion coming from $\sigma_{\overline{l}}>0$ is of secondary importance for long times. Furthermore, the formula $S_{N(t)}=\overline{l}N(t)$ is correct for any $\psi(t)$, including those with $0<\beta<1$, as long as $\sigma_{\overline{l}}=0$; when we determine the SCDs for a no-backflow approximation (see below) we actually calculate the distribution of $\overline{l}N(t)$, i.e., F_{SCD} $(L\equiv S_{N(t)};t)$.

III. SPATIAL CONCENTRATION DISTRIBUTIONS WITHOUT BACKFLOW

A. Derivation of SCD

When tracer enters a medium, and is subject to a 1D flow field (i.e., transport is under the influence of a potential gradient), it spreads initially in all directions, but then eventually migrates along the preferred flow direction (decreasing potential). After some time, most of the tracer will be displaced forward of the starting point, i.e., at this time, there is effectively no backflow. The same is obviously true for any position in the medium: most of the particles found at some position will advance after some time in the forward direction.

When neglecting backflow, it is in fact quite straightforward to obtain a SCD of a tracer at a given time. For the case $\beta<1$, neglecting backflow is a good approximation for sufficiently long times: the first and centered second spatial moments both scale with time as t^{β} (e.g., [6]), which means that there is, on average, no back movement because the forward advection compensates for the backward spread. The extent of the backflow is defined by paths having velocities opposite to the mean flow direction. It is natural to expect these paths to have limited length (otherwise, there must be a "macroscopic" gradient opposite to the mean flow direction), which defines a backflow distance. We observe also that molecular diffusion is of limited extent in the presence of the gradient. This condition is easily checked in an experiment: if the backflow distance from the input point is small compared to the forward tracer extent, then the no-backflow approximation is appropriate.

For the case $1<\beta<2$, the no-backflow assumption is even simpler to justify. Here, the mean displacement scales as t while the standard deviation scales as $t^{(3-\beta)/2}$. With increasing time, the advection forward will overcome the backward spread, as in the Gaussian case.

We emphasize that the derivations and notation in this section are similar to those in [5]; the reader is referred there for more detailed explanations. We define $\langle l\rangle$ as a (single) transition distance. Then in the interpretation of $\psi(t)$ given in [5], and in the no-backflow approximation, we have that the mean single transition distance $\overline{l}=\langle l\rangle$ since no transitions back are allowed.

In the no-backflow approximation, there are two ways to derive the SCD of the propagator. First, in a direct approach, we note that the spatial profile is a residence-weighted mass distribution. It is well known that the waiting time before the next transition takes place is given (in Laplace space) as $(1-\psi^*(u))/u$ where $\psi(t)$ is a single transition time distribution and the asterisk denotes Laplace transform with variable u. Thus the spatial concentration distribution as a function of distance from the origin (number of transitions l) is given by

$$\mathcal{L}F_{\rm SCD}(l;t)=[\psi^*(u)]^l\frac{1-\psi^*(u)}{u},\tag{5}$$

where $\mathcal{L}F_{\rm SCD}$ denotes the Laplace transform of $F_{\rm SCD}$. Using the usual long-time approximation of $\psi(t)$ in the two different cases $0<\beta<1$ and $1<\beta<2$ leads to the desired solutions, given below.

A second approach, which yields (obviously) the same result, is to use conservation of mass. For any $l\geqslant 0$ and $\tau\geqslant 0$

$$\sum_{j=0}^{l}F_{\rm SCD}(j;\tau)+\int_0^{\tau}F_{\rm FPTD}(\tau';l)d\tau'\equiv 1.$$

Defining the cumulative FPTD (CFPTD) as $F_{\rm CFPTD}(\tau;l)\equiv\int_0^{\tau}F_{\rm FPTD}(\tau';l)d\tau'$ it follows that $F_{\rm SCD}(l;\tau)=-(\partial/\partial l)F_{\rm CFPTD}(\tau;l)$.

In the case $0<\beta<1$,

$$\psi^*(u)\approx 1-c_\beta u^\beta\approx e^{-c_\beta u^\beta},\tag{6}$$

for small u (see [5]; c_β is a constant), and the result of using either of the two approaches is

$$lF_{\rm SCD}(l)=-\frac{1}{\pi}\sum_{n=1}^{\infty}\left(-\frac{g}{\tau^\beta}\right)^n\frac{\Gamma(\beta n)}{\Gamma(n)}\sin\pi\beta n\tag{7}$$

and, asymptotically,

$$lF_{\rm SCD}(l)\simeq\frac{\left(\frac{\beta g}{\tau^\beta}\right)^{1/[2(1-\beta)]}\exp\left\{-\frac{1-\beta}{\beta}\left(\frac{\beta g}{\tau^\beta}\right)^{1/(1-\beta)}\right\}}{\beta\sqrt{2\pi(1-\beta)}},$$

$$\frac{g}{\tau^\beta}\gg 1,\tag{8}$$

where $\overline{t}\equiv\overline{l}/\overline{v}$, $\tau\equiv t/\overline{t}$, $g\equiv lb_{\beta,\overline{l}}$, $b_{\beta,\overline{l}}\equiv c_\beta/\overline{t}^\beta$, and $l\equiv L/\overline{l}$, where $\overline{v}$ is some fixed characteristic velocity (for example, the carrier fluid velocity), L is the distance from the input position, and t is time elapsed (see [5]). Note that the first temporal moment of $\psi(t)$ is infinite and $\overline{t}$ is defined through the (arbitrary) choice of $\overline{v}$. One can use these connections to the dimensional variables measured in or fitted to experiments, for data analysis. Further, it can be seen that

$$x\equiv\frac{g}{\tau^\beta}=\frac{B}{\overline{v}^\beta}\frac{L}{t^\beta}=\left(\frac{L}{tv_L}\right)^\beta,$$

where v_L is the effective tracer velocity at the distance L, and we used the relation $v_L=\overline{v}b_{\beta,L}^{-1/\beta}$. Also $B\equiv b_{\beta,L}L^{\beta-1}$ is a constant of motion independent of L (for a fixed $\overline{v}$); one can see that $C\equiv B/\overline{v}^\beta$ is independent of the choice of $\overline{v}$ (changing $\overline{v}$, while keeping v_L constant, will change $b_{\beta,L}$ and thus B, exactly to make C invariant) (see [5,11]). To relate C to the mean travel distance $\langle l(t)\rangle$ we note that Eq. (B3) yields, using the above definitions, $\langle l(t)\rangle=t^\beta/[C\Gamma(\beta+1)]$.

Similar results are obtained for $1<\beta<2$. In this case [5],

$$\psi^*(u)\approx 1-\overline{t}u+c_\beta u^\beta\approx e^{-\overline{t}u+c_\beta u^\beta}\tag{9}$$

and the result is

$$F_{\rm SCD}=\frac{1}{\pi\beta g^{1/\beta}}\left[\frac{\tau-l}{\beta l}+1\right]\sum_{n=1}^{\infty}(-h)^{n-1}\frac{\Gamma(n/\beta)}{\Gamma(n)}\sin\frac{\pi n}{\beta},\tag{10}$$

where $h\equiv(l-\tau)/g^{1/\beta}$. There are two approximations: as $h\nearrow\infty$,

$$F_{\rm SCD}\approx\left[\frac{1}{l-\tau}-\frac{1}{\beta l}\right]\frac{\exp\left\{-(\beta-1)\left(\frac{h}{\beta}\right)^{\beta/(\beta-1)}\right\}}{\sqrt{\frac{2\pi(\beta-1)}{\beta}\left(\frac{\beta}{h}\right)^{\beta/(\beta-1)}}},\tag{11}$$

while as $h\searrow-\infty$

$$F_{\rm SCD}\approx\frac{1}{\pi}\left[\frac{1}{l-\tau}-\frac{1}{\beta l}\right]\sum_{n=1}^{\infty}(-h)^{-n\beta}\frac{\Gamma(\beta n+1)}{\Gamma(n+1)}\sin\pi\beta n.\tag{12}$$

We use the definitions $\tau\equiv t/\overline{t}$, $l\equiv L/\overline{l}$, $w\equiv\overline{l}/\overline{t}$, and $g\equiv lb_{\beta,\overline{l}}$ to transform to dimensional variables. Here, however, $\overline{t}$ is the (finite) first moment of $\psi(t)$.

For $\beta>2$, we find that

$$F_{\rm SCD}|_{\beta>2}=\frac{e^{-h^2/4}}{4\sqrt{\pi g}}\left(1+\frac{\tau}{l}\right)=\overline{l}\frac{e^{-h^2/4}}{4\sqrt{\pi BL}}\left(1+\frac{wt}{L}\right)$$

since here $g=BL/\overline{l}^2$. Note that $\overline{l}$ should be dropped from the last expression to obtain the PDF form of the SCD.

We observe that for $1<\beta<2$ there is also a possibility to obtain a different, converging asymptotic expression when $h\searrow-\infty$, instead of the diverging asymptotic series presented above. Then the FPTD approximation for this case can be obtained also from considering mass conservation. From Eq. (5)

$$F_{\mathrm{SCD}} \approx \frac{1}{2\pi i}\int_{C_s} ds e^{s\tau} e^{-ls+gs^{\beta}} e^{-b_{\beta}s^{\beta-1}}, \qquad (13)$$

and for large $(\tau-l)\nearrow\infty$ (i.e., $h\searrow-\infty$),

$$F_{\mathrm{SCD}} \approx \frac{1}{2\pi i}\int_{C_s} ds \exp[(\tau-l)s-b_{\beta}s^{\beta-1}]$$
$$=\frac{1}{2\pi i(\tau-l)}\int_{C_z} dz\, e^{z-\eta z^{\beta-1}}$$
$$=\frac{1-\beta}{\pi(\tau-l)}\sum_{n=1}^{\infty}\eta^{n}\frac{\Gamma(n\beta-n)}{\Gamma(n)}\sin\pi\beta n, \qquad (14)$$

where $z\equiv s(\tau-l)$, $\eta\equiv b_{\beta}/(\tau-l)^{\beta-1}$ and C_s and C_z are corresponding complex-plane contours (see, e.g., [5]). This series is convergent and for large enough time η becomes small. The leading term $(n=1)$ is $[-1/(\tau-l)^{\beta}][b_{\beta}\Gamma(\beta)/\pi]\sin\pi\beta$ and is identical to the leading term of Eq. (12).

B. Unified expressions for SCD and FPTD

The solutions derived above are similar to the FPTD solutions (see, e.g., [5]). Below we present the SCD and FPTD solutions in the form of probability density functions. We stress that corresponding SCD and FPTD solutions are expressed in terms of the same function (but with different arguments). The no-backflow approximation for the SCD (which is a residence average in contrast to the flux averaged FPTD) in the case of $0<\beta<1$, denoted F_{SCD01}, can be written as

$$F_{\mathrm{SCD01}}(L)=\frac{1}{R}f\left(\frac{L}{R};\beta\right), \qquad (15)$$

where $R\equiv t^{\beta}/C\simeq\langle l(t)\rangle\Gamma(\beta+1)$, $\langle l(t)\rangle$ is the mean displacement and C is the constant of motion defined preceding Eq. (10). The corresponding cumulative SCD (CSCD) for a δ pulse of tracer is

$$F_{\mathrm{CSCD01}}(L)=f_c\left(\frac{L}{R};\beta\right). \qquad (16)$$

The functions f and f_c are defined in Appendix A. Only non-negative x are possible and thus Eq. (A4) is relevant. The FPTDs are also expressed in Appendix A through this function $f(x;\nu)$, with $x=(t_{\mathrm{mean,eff}}/t)^{\beta}$, where $t_{\mathrm{mean,eff}}$ is the effective mean time, or a shift factor along the temporal axis to be defined when transforming the solution to dimensional time. Thus, for any fixed length L and time t, we have that $x\equiv(t_{\mathrm{mean,eff}}/t)^{\beta}\equiv CL/t^{\beta}$ so that $t_{\mathrm{mean,eff}}=(CL)^{1/\beta}$.

One can see that there is an essentially exponential (actually, a stretched exponential) decrease in the concentration distribution for long distances. This is the result of the assumed normal spatial behavior of single transitions, i.e., the narrow spatial distribution of a single transition distance, when all spatial moments are finite, as in a Gaussian. We chose above a δ-function distribution, allowing single transitions only in one direction and of equal length. It is also easily found using Eqs. (A1) and/or (A2) that the peak of the distribution is at the origin for $\beta<\frac{1}{2}$ and starts moving for $\beta>\frac{1}{2}$. Finally, we remark looking at Eqs. (15) and (A1), that the amount of tracer at the input point decreases with time as $t^{-\beta}$.

The no-backflow approximation for the SCD in the case of $1<\beta<2$ yields

$$F_{\mathrm{SCD12}}(L)=\frac{E}{\beta A}f\left(h;\frac{1}{\beta}\right), \qquad (17)$$

where $h\equiv(L-R)/A$, $R\equiv wt$, $A\equiv(LB)^{1/\beta}$, $B\equiv L^{\beta-1}b_{\beta,L}$ is a constant of motion independent of L, and $E\equiv(tw-L)/(\beta L)+1\equiv1-hA/(\beta L)$. The corresponding CSCD is

$$F_{\mathrm{CSCD12}}(L)=\frac{1}{\beta}f_c\left(h;\frac{1}{\beta}\right). \qquad (18)$$

Note that because $(E/A)dL=dh$, from Eq. (A4) we have that

$$\int_{wt}^{\infty}F_{\mathrm{SCD12}}(L)dL=\frac{1}{\beta}.$$

Since $\int_0^{\infty}F_{\mathrm{SCD12}}(L)dL=1$, the relation (A5) appears. In this case, for any positive time, h is defined from minus to plus infinity, for $L\geqslant0$. Given β, B, and R, the spatial profile can be plotted as a function of L. It should be noted that in order for the plume distribution at time zero to resemble a δ pulse, we require that $L_0\ll L$ for all lengths L of interest, where $L_0\equiv B^{1/(\beta-1)}$. This can be seen by looking at h as $t=0$. Finally, at the injection point,

$$F_{\mathrm{SCD12}}(L=0)=\frac{\beta-1}{\Gamma(2-\beta)}\left(\frac{L_0}{R}\right)^{\beta}\frac{1}{L_0}$$

decreases with time as $t^{-\beta}$, as in the case $0<\beta<1$.

Figure 1 presents the temporal evolution of SCDs for several values of β. It can be seen that for smaller β more particles stay close to the origin, while as β increases a backward (''heavy'') tail appears. This tail becomes less distinct as β approaches the value of 2. Such profiles of the SCDs have been observed in studies of chemical transport in geological formations [19].

We note here parenthetically that graphs of the spatial profile (15) are similar to the Poisson-like distribution

$$P(L)=\frac{\mu^{L/R_1}e^{-\mu}}{NR_1\Gamma(1+L/R_1)}, \qquad (19)$$

in the case $\beta\leqslant\frac{1}{2}$. By equating the two expressions at $L=0$, we can choose $R_1=R/N$ and $\mu=\ln\Gamma(1-\beta)$, where N is a normalization factor depending on μ and growing to 1 as μ increases. At $\beta=\frac{1}{2}$ when the slope of Eq. (15) at the origin is zero, $\mu\approx0.572\,365$; this value is very close to the value

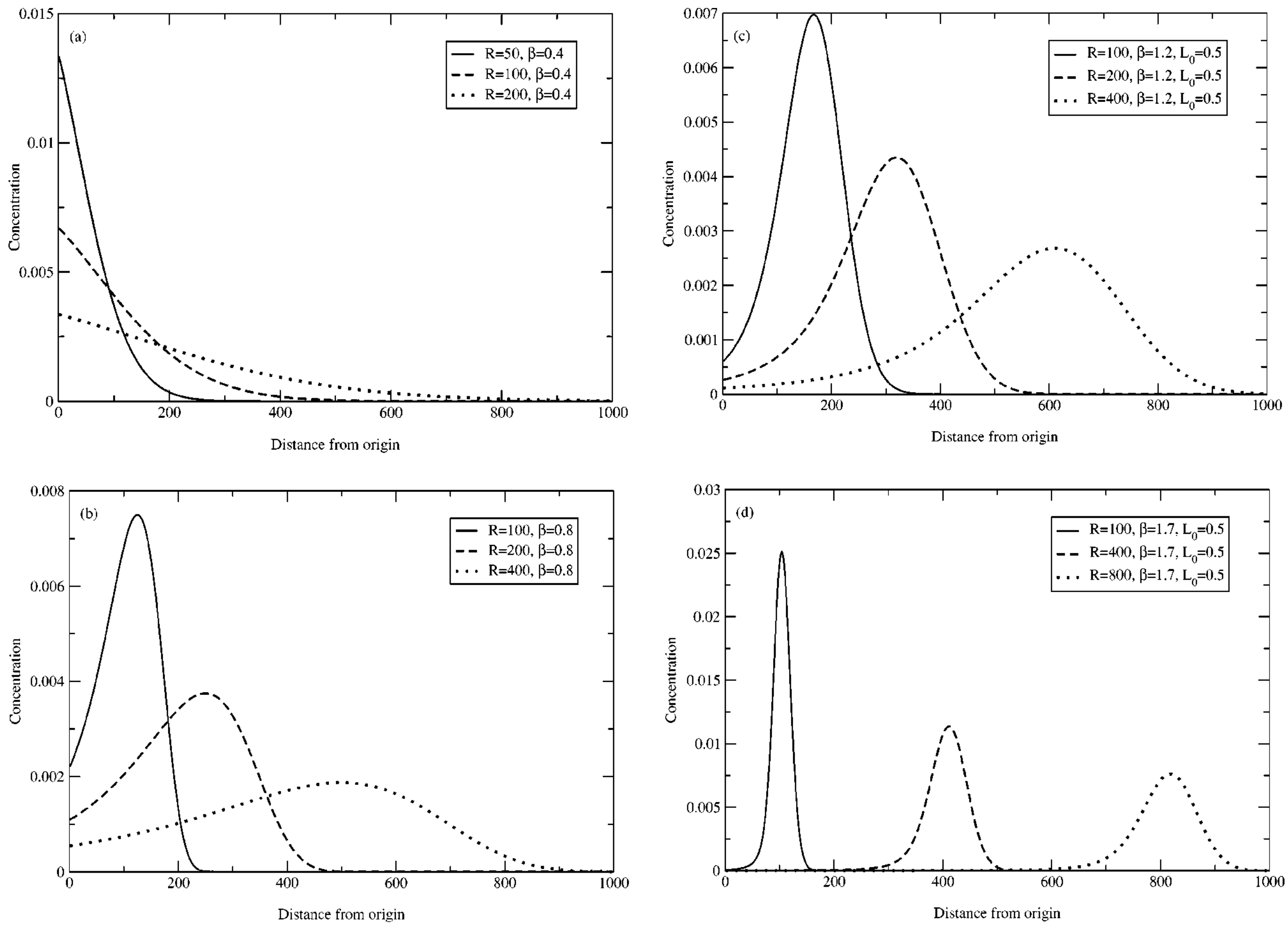

FIG. 1. Temporal evolution of spatial concentration distributions for different values of β. Here, $R \equiv t^{\beta}/C \cong \langle l(t) \rangle \, \Gamma(\beta+1)$ for $0<\beta<1$ and $R \equiv wt$ for $\beta>1$, where t is time, C and L_0 are constants of motion, $\langle l(t) \rangle$ is the mean displacement, Γ is the gamma function and w is the (asymptotic) velocity of the tracer (in the case $\beta>1$). (a) $\beta=0.4$, (b) $\beta=0.8$, (c) $\beta=1.2$, (d) $\beta=1.7$.

$\mu = e^{-\gamma_E} \approx 0.561\,459$ (γ_E being Euler gamma), for a zero slope of Eq. (19). This similarity breaks down for $\beta > \frac{1}{2}$. Note, however, that both the Poisson distribution and the SCD derived here tend to the Gaussian distribution as μ and β grow. The reason why Eq. (19) is similar to Eq. (15) for $\beta \leqslant \frac{1}{2}$ might be that the transition probabilities are very small, while for $\beta > \frac{1}{2}$ they increase. It can be shown, using a lognormal transition time PDF [12], that the value of $\beta = \frac{1}{2}$ is identified with reaching a time equal to the mean single transition time; this fact also explains why the peak of the distribution advances for $\beta > \frac{1}{2}$, while for $\beta < \frac{1}{2}$ it remains at the origin.

IV. DERIVATION OF SCD WITH BACKFLOW

We now consider the SCD solutions when backflow is not neglected. Such solutions are of particular value for early (relatively) times and/or SCD's near the particle inlet.

If there is a probability p to the forward transition and q for the backward transition (obviously, $p \geqslant q$) then the nondimensional displacement $l \equiv L/\langle l \rangle$ of the particle from the origin after n transitions is given by the binomial distribution

$$P_l(l;n) = \begin{pmatrix} n \\ \dfrac{n-l}{2} \end{pmatrix} p^{(n+l)/2} q^{(n-l)/2}.$$

Also, if we want to consider backflow, the Laplace transform of the probability $\mathcal{P}$ of making n transitions in time t is given by Eq. (5)

$$\mathcal{L}\mathcal{P}(n;\tau) = [\psi^*(u)]^n \frac{1-\psi^*(u)}{u}, \tag{20}$$

where $\tau \equiv t/\overline{t}$ is a nondimensional time (as in Sec. III) and by $\psi(t)$ we mean a single transition PDF over a distance $\langle l \rangle$. Now $\overline{l} = (p-q)\langle l \rangle$. Thus

$$F_{\mathrm{SCD}}(l;\tau) = \sum_{n=|l|}^{\infty} P_l(l;n)\mathcal{P}(n;\tau). \tag{21}$$

The Laplace transform of this expression is

$$\mathcal{L}F_{\rm SCD}(l;\tau)=\frac{1-\psi^*}{s}\left(\frac{p}{q}\right)^{l/2}\sum_{n=|l|}^{\infty}\left(\begin{array}{c}n\\ \frac{n-l}{2}\end{array}\right)[\psi^*\sqrt{pq}]^n$$

$$=\frac{1-\psi^*}{s}\left(\frac{p}{q}\right)^{l/2}x^{|l|}\,{}_2F_1\left(\frac{1+|l|}{2},\frac{2+|l|}{2};1+|l|;4x^2\right), \tag{22}$$

where $\psi^*\equiv\psi^*(u)$, $s\equiv\overline{t}u$ is the nondimensional Laplace variable, $x\equiv\psi^*\sqrt{pq}$ and ${}_2F_1$ is a hypergeometric function. Formula (15.1.14) of [20] gives ${}_2F_1(a,\frac{1}{2}+a;2a;z)=2^{2a-1}(1-z)^{-1/2}[1+\sqrt{1-z}]^{1-2a}$, so that in our case the spatial profile for the positive forward displacement l can be obtained by calculating

$$F_{\rm SCD}(l>0;\tau)=\mathcal{L}^{-1}\frac{1-\psi^*}{sy}\left(\frac{2p\psi^*}{1+y}\right)^l, \tag{23}$$

where $y=\sqrt{1-4pq(\psi^*)^2}$.

The same formula can be obtained using [3]. Formula (37) of [3] reads

$$\mathcal{L}F_{\rm SCD}(l;t)=\frac{1-\psi^*(u)}{u}G[l,\psi^*(u)], \tag{24}$$

while formula (12) of [3] is

$$G(l,z)=\frac{1}{N}\sum_{s=1}^{N}\frac{e^{2\pi isl/N}}{1-\lambda(k)z},$$

where $k\equiv2\pi s/N$, $\lambda(k)=\Sigma_l p(l)e^{-ikl}$. In our case, $p(-1)=q$, $p(1)=p$, and all other probabilities are zero. Using relations in Appendix C of [3] to calculate G, one obtains

$$G(l,z)\to\frac{\left[\frac{1-\sqrt{1-4pqz^2}}{2zq}\right]^l}{\sqrt{1-4pqz^2}}$$

in the limit $N\to\infty$, so that

$$G[l,\psi^*(u)]\to\frac{1}{y}\left\{\frac{1-y}{2q\psi^*}\right\}^l=\frac{1}{y}\left\{\frac{1-y^2}{2q\psi^*(1+y)}\right\}^l=\frac{1}{y}\left\{\frac{2p\psi^*}{1+y}\right\}^l$$

and from Eq. (24) we again recover Eq. (23).

It can be seen from Eq. (22) also that for any $l>0$ it follows that

$$F_{\rm SCD}(l;\tau)/F_{\rm SCD}(-l,\tau)=(p/q)^l,$$

where $p\equiv p_{\langle l\rangle}$ and $q\equiv q_{\langle l\rangle}$ are the probabilities for transitions of length $\langle l\rangle$. Thus, we can write

$$\left(\frac{p_{\langle l\rangle}}{q_{\langle l\rangle}}\right)^l=\left(\frac{p_{n\langle l\rangle}}{q_{n\langle l\rangle}}\right)^{l/n},$$

which can be brought into the form

$$p_{n\langle l\rangle}=\frac{1}{1+\left(\frac{1}{p_{\langle l\rangle}}-1\right)^n}.$$

For example, if we start at $p_{\langle l\rangle}=0.6$ then $p_{8\langle l\rangle}\approx0.962$ and $p_{16\langle l\rangle}\approx0.998$.

Consider the case $0<\beta<1$. Using a first-order small s expansion in powers of s^β, for $bs^\beta\ll1$ it is well known that $\psi^*\simeq1-bs^\beta$ (cf. Sec. III). Thus $(\psi^*)^2\simeq1-2bs^\beta$ and

$$y\simeq\mu\left[1+\frac{8pqbs^\beta}{\mu^2}\right]^{1/2},$$

where $b\equiv b_{\beta,\langle l\rangle}$ and $\mu\equiv\sqrt{1-4pq}\equiv2p-1$ for $p\geqslant1/2$. Here two cases arise:

(a) $8pqbs^\beta/\mu^2\gg1$ so that $y\simeq\sqrt{8pqbs}^{\beta/2}\simeq\sqrt{2bs}^{\beta/2}$ (since in this case $p\approx q$). Since $bs^\beta\ll1$ we ignore it compared to $\sqrt{2bs}^{\beta/2}\gg bs^\beta$, and so obtain $1/(1+y)\approx\exp[-\sqrt{2bs}^{\beta/2}]$. Then Eq. (23) becomes

$$F_{\rm SCD}(l>0;\tau)\simeq\tfrac{1}{2}\mathcal{L}^{-1}\{\sqrt{2bs}^{\beta/2-1}\exp[-l\sqrt{2bs}^{\beta/2}]\}. \tag{25}$$

This is exactly the form of the no-backflow case, with $\beta/2<\frac{1}{2}$ standing for β in the expression that can be obtained trivially by substituting Eq. (6) into Eq. (5). From there it is known that the peak of the distribution will in this case stay at the origin. The factor of $\frac{1}{2}$ in front of the formula appears because here the forward part is identical to the backward one and is one-half of the total. Note that setting $\beta=1$ in Eq. (25) leads to a regular diffusionlike transport in this approximation (with zero slope at the origin). Thus, at relatively short times, transport will be diffusionlike dominated and anomalous, i.e., the diffusion is a particular case of the CTRW approach used. As time advances the transition to the next case will occur and $8pqbs^\beta/\mu^2\approx1$ can be used to determine this transition time.

(b) $8pqbs^\beta/\mu^2\ll1$ so that $y\simeq\mu(1+4pqbs^\beta/\mu^2)$. This is correct for any $p>q$ as $t\to\infty$ and will be considered further below. We obtain $1/y\simeq\exp(-\lambda s^\beta)/\mu$, where $\lambda\equiv4pqb/\mu^2$ and $1/(1+y)\simeq\exp(-\nu s^\beta)/(1+\mu)$, where $\nu\equiv4pqb/[\mu(1+\mu)]$. Upon substitution into Eq. (23)

$$F_{\rm SCD}(l>0;\tau)\approx\frac{b}{(p-q)\omega}\mathcal{L}^{-1}\{\omega s^{\beta-1}e^{-l\omega s^\beta}\},$$

where $\omega\equiv b+\nu+\lambda/l\equiv b(1+2q/(p-q)+4pq/[(p-q)^2l])$ is a function of l. The last inverse Laplace transform is already known from the no-backflow approximation [see Sec. II, Eqs. (5) and (6)].

For $1<\beta<2$ a similar analysis yields $\psi^*\approx1-s+bs^\beta$ for $s\ll1$ and $bs^\beta<s$, $(\psi^*)^2\approx1-2s+2bs^\beta$ and

$$y\approx\mu\left(1+\frac{8pqs(1-bs^{\beta-1})}{\mu^2}\right)^{1/2}.$$

Again two cases arise:

(a) $8pqs(1-bs^{\beta-1})/\mu^2\gg1$ so that

$$y \approx \sqrt{2} s^{1/2}\left(1-\frac{bs^{\beta-1}}{2}\right) \approx \sqrt{2} s^{1/2}$$

(here we also assumed $bs^{\beta-1} \ll 1$, this is always true when the small-s expansion $\psi^* \approx 1-s+bs^{\beta}$ is valid). This case is identical to the case (a) for $0<\beta<1$, if one substitutes there $\beta=b=1$ [see Eq. (25)].

(b) $[8pqs(1-bs^{\beta-1}]/\mu^2 \ll 1$ so that

$$y \approx \mu\left(1+\frac{4pqs(1-bs^{\beta-1})}{\mu^2}\right).$$

We obtain

$$\frac{1}{y} \simeq \frac{\exp[-\lambda s(1-s^{\beta-1})]}{\mu},$$

where $\lambda \equiv 4pq/\mu^2$ and

$$\frac{1}{1+y} \simeq \frac{\exp[-\nu s(1-bs^{\beta-1})]}{1+\mu},$$

where $\nu \equiv 4pq/[\mu(1+\mu)]$. Substituting these expressions into Eq. (23) gives

$$F_{\mathrm{SCD}}(l>0;\tau) \approx \frac{1}{p-q}\mathcal{L}^{-1}\{(1-bs^{\beta-1}) \times \exp[-l\omega s[1-bs^{\beta-1}]]\} \approx \frac{1}{p-q}\mathcal{L}^{-1}\left\{\frac{1-\psi^*}{s}(\psi^*)^{l\omega}\right\}$$

where

$$l\omega \equiv l+\nu l+\lambda \equiv l+\frac{2ql}{p-q}+\frac{4pq}{(p-q)^2}.$$

The last transform is already known from the no-backflow approximation [see Sec. III, Eqs. (5) and (9)].

We now return to Eq. (21) and look at it from a different point of view. First note that in the case $l \gg 1$, which is of interest here, the binomial distribution converges to a Gaussian and since

$$\binom{n}{k} p^k q^{n-k} \to \frac{1}{\sqrt{2\pi npq}}\exp\left(-\frac{(k-np)^2}{2npq}\right)$$

as $n\to\infty$, with $k \equiv (n+l)/2$, it follows that

$$P_l(l;n) \approx \frac{1}{\sqrt{8\pi npq}}\exp\left(-\frac{[l-n(p-q)]^2}{8npq}\right).$$

Transforming to dimensional variables and using $\overline{l}=\langle l\rangle(p-q)$, $\overline{l^2}=\langle l\rangle^2$, $\sigma_l^2 \equiv \overline{l^2}-\overline{l}^2=4pq\langle l\rangle^2$, $l=L/\langle l\rangle$, and $P_l dl = P_L dL$ yields

$$P_L(L;n)=\frac{1}{\sqrt{2\pi n\sigma_l^2}}\exp\left(\frac{(L-n\overline{l})^2}{2n\sigma_l^2}\right). \tag{26}$$

The expression (21) can be rewritten in the form

$$F_{\mathrm{SCD}}(L;\tau) \approx \int_0^\infty dn\, P_L(L;n)\mathcal{P}(n;\tau), \tag{27}$$

which together with Eqs. (20) and (26), and for $0<\beta<1$ is identical to formulas (37)–(39) in [14]; these latter formulas follow from one of the Galilei variant transport models discussed therein. We note that if n in Eq. (27) is considered to be some parameter, not necessarily the number of transitions, then Eq. (27) may represent a general expression for the spatial profile concentration, with a coupled space-time transition function. Some particular cases include using Eq. (26) or a more general expression for a Levy flight, or δ functions of different arguments in place of $\mathcal{P}(n,\tau)$ and/or $P_L(L;n)$. As many of the fractional derivative equations considered in the literature admit solution by the separation of variables method (see, e.g., [14,17]) then Eq. (27) will be a general solution of such equations with n being related to eigenvalues of the problem.

V. SPATIAL MOMENTS

We consider now the first and second spatial moments of an evolving particle plume, as a functions of traveling time (cf. [6]; also our Appendix B). Using the definitions of velocity $v(t) \equiv d\langle l(t)\rangle/dt$ and dispersion $D(t) \equiv \frac{1}{2}\{d\sigma^2(t)/dt\}$, where $\langle l(t)\rangle$ denotes the mean travel distance and $\sigma^2(t)$ is the variance of the particle plume, the quantity D/v can be calculated. In the biased Brownian motion picture this quantity D/v will be approximately equal (neglecting molecular diffusion) to the dispersivity constant and is thus called apparent (or effective) dispersivity. For $0<\beta<1$ the result for the leading term is (see Appendix B)

$$\frac{D}{v} \simeq \langle l(t)\rangle\left[\frac{2\Gamma^2(\beta+1)}{\Gamma(2\beta+1)}-1\right], \tag{28}$$

i.e., the apparent dispersivity grows linearly with the mean particle displacement. The proportionality coefficient (in the square brackets) decreases from 1 for $\beta=0$ to 0 for $\beta=1$. In some cases, the full expression (containing an additional term) might be needed, as discussed in Appendix B.

For $1<\beta<2$ the leading term is (see Appendix B)

$$\frac{D}{v} \simeq \frac{B(\beta-1)}{\Gamma(3-\beta)}\langle l(t)\rangle^{2-\beta}, \tag{29}$$

i.e., in this case the dispersivity also grows with the mean displacement, but sublinearly. As usual, for $\beta>2$ we substitute $\beta=2$ [5] and find that the dispersivity $D/v \simeq B = b_{\beta>2,L}L=$const. We stress here that the correction terms, given in Appendix B, may be critically important particularly as $\beta \searrow 1$.

VI. CONCLUDING REMARKS

A physically based approach for describing transport phenomena in heterogeneous media exists, which generalizes a random walk formalism. This CTRW approach includes the Fokker-Planck equation as a specific case of a more general picture. The CTRW is relatively simple to treat, yet it provides qualitative and quantitative explanation and prediction of experiments because the initial model concept captures the main features of ''anomalous'' transport.

In this paper we presented a straightforward and easy derivation of spatial concentration distributions of a tracer as it travels through highly heterogeneous media. The temporal and spatial profiles are all written in terms of the same special function. The first two spatial moments of the concentration distribution were also calculated. While here we discussed 1D solutions, multidimensional generalizations are conceptually similar.

We have focused on wide *temporal* distributions (the ''standard'' CTRW, which also might be called ''Lévy flights in time''), but note that the mathematics remain similar if we assume wide *spatial* and normal (narrow) temporal distributions (i.e., the usual Lévy flights). In this case (in the presence of spatial bias) the spatial and temporal concentration formulas presented in this paper must be interchanged.

The interested reader is invited to download the computer codes performing spatial and temporal distribution calculations developed and described here from the web: http://www.weizmann.ac.il/ESER/People/Brian/CTRW. This website also includes other solutions. We note also that generalized FPTDs and SCDs have been developed for the case $0.5<\beta<1$ [12].

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APPENDIX A: FUNCTIONS $f(x;\nu)$ AND $f_c(x;\nu)$

It is convenient to represent the spatial profile distributions and also the FPTD distributions derived and presented in [5,11] for all values of $0<\beta<2$, $\beta\neq 1$ in terms of the following functions. By definition,

$$f(x;\nu)\equiv\frac{1}{\pi}\sum_{n=1}^{\infty}(-x)^{n-1}\frac{\Gamma(\nu n)}{\Gamma(n)}\sin\pi\nu n \tag{A1}$$

and it has the two approximations. As $x\nearrow+\infty$,

$$f(x;\nu)\simeq\frac{(\nu x)^{(2\nu-1)/[2(1-\nu)]}\exp\left\{-\left(\frac{1}{\nu}-1\right)(\nu x)^{1/(1-\nu)}\right\}}{\sqrt{2\pi(1-\nu)}}, \tag{A2}$$

and as $x\searrow-\infty$,

$$f(x;\nu)\simeq\frac{1}{\pi\nu^2 x}\sum_{n=1}^{\infty}(-x)^{-n/\nu}\frac{\Gamma(n/\nu)}{\Gamma(n)}\sin\frac{\pi n}{\nu}. \tag{A3}$$

At $\nu=\frac{1}{2}$ Eq. (A2) becomes exact, while Eq. (A3) will be zero. This last series (A3) is, in general, diverging in the interval $0<\nu<1$ and defines an asymptotic series. The function f is considered here only in this interval and is normalized:

$$\int_0^{\infty}f(x;\nu)dx=1 \tag{A4}$$

and

$$\int_{-\infty}^{\infty}\nu f(x;\nu)dx=1. \tag{A5}$$

Similarly,

$$f_c(x;\nu)\equiv\int_x^{\infty}f(x';\nu)dx'$$
$$\equiv 1+\frac{1}{\pi}\sum_{n=1}^{\infty}(-x)^n\frac{\Gamma(\nu n)}{\Gamma(n+1)}\sin\pi\nu n. \tag{A6}$$

As $x\nearrow+\infty$,

$$f_c(x;\nu)\simeq\frac{\exp\left\{-\left(\frac{1}{\nu}-1\right)(\nu x)^{1/(1-\nu)}\right\}}{\sqrt{2\pi(1-\nu)(\nu x)^{1/(1-\nu)}}} \tag{A7}$$

and as $x\searrow-\infty$,

$$f_c(x;\nu)\approx\frac{1}{\nu}\left[1+\frac{1}{\pi}\sum_{n=1}^{\infty}(-x)^{-n/\nu}\frac{\Gamma(n/\nu)}{\Gamma(n+1)}\sin\frac{\pi n}{\nu}\right]. \tag{A8}$$

Numerical evaluations of these functions (e.g., to produce Fig. 1) were performed by programming the above formulas in the C language.

We note that f can be written in terms of the Fox functions (see, e.g., [14,21]) as follows:

$$f(x;\nu)\equiv\frac{1}{\nu^2 x}H_{1,1}^{1,0}\left[x^{1/\nu}\middle|\begin{matrix}(0,1)\\(0,1/\nu)\end{matrix}\right]\equiv\frac{1}{\nu x}H_{1,1}^{1,0}\left[x\middle|\begin{matrix}(0,\nu)\\(0,1)\end{matrix}\right]$$
$$\equiv\frac{1}{\nu}H_{1,1}^{1,0}\left[x\middle|\begin{matrix}(-\nu,\nu)\\(-1,1)\end{matrix}\right]\equiv H_{1,1}^{1,0}\left[x\middle|\begin{matrix}(1-\nu,\nu)\\(0,1)\end{matrix}\right].$$

We also note that the following form of Eq. (A2) can be helpful: defining $y^2=[1/\nu-1](\nu x)^{1/(1-\nu)}$ leads to

$$f[y(x)]dy\simeq\frac{2}{\sqrt{2\nu\pi}}e^{-y^2}dy,$$

which is similar to the definition of the error function

$$\mathrm{erf}(z)=\frac{2}{\sqrt{\pi}}\int_0^z e^{-y^2}dy.$$

In terms of these two functions, the FPTD and CFPTD solutions as functions of experimental time are written for $0<\beta<1$ as

$$F_{\mathrm{FPTD01}}(t)=\frac{\beta x}{t}f(x;\beta) \tag{A9}$$

and

$$F_{\mathrm{CFPTD01}}(t)=f_c(x;\beta), \tag{A10}$$

where $x\equiv CL/t^{\beta}\equiv(t_{\mathrm{mean\ eff}}/t)^{\beta}$, and for $1<\beta<2$ similarly

$$F_{\mathrm{FPTD12}}(t)=\frac{w}{\beta A}f\left(h;\frac{1}{\beta}\right)=\frac{1}{\beta t_{\mathrm{mean}}b_{\beta,L}^{1/\beta}}f\left(h;\frac{1}{\beta}\right), \tag{A11}$$

and

$$F_{\mathrm{CFPTD12}}(t)=\frac{1}{\beta}f_c\left(h;\frac{1}{\beta}\right), \tag{A12}$$

where $h\equiv(1-t/t_{\mathrm{mean}})/b_{\beta,L}^{1/\beta}$. As noted in [11], $b_{\beta,L}$ should be small enough ($b_{\beta,L}^{1/(1-\beta)}\gg 1$) for these formulas to apply. This condition is required because, strictly speaking, $F_{\mathrm{FPTD12}}(t)$ is normalized to 1 for $-\infty<t<\infty$, which is an artifact, but for sufficiently small $b_{\beta,L}$, the integral from $-\infty$ to 0 is negligible.

The expressions for the spatial profiles were given in Sec. III.

APPENDIX B: DERIVATION OF THE APPARENT DISPERSIVITY

The mean displacement and the variance of the propagator can be calculated from formulas (5) and (10) in Shlesinger [6],

$$\langle l(t)\rangle=\overline{l}\mathcal{L}^{-1}\frac{\psi^*(u)}{u[1-\psi^*(u)]}, \tag{B1}$$

$$\sigma_l^2(t)=\overline{l^2}\frac{\langle l(t)\rangle}{\overline{l}}+2\overline{l}^2\mathcal{L}^{-1}\frac{[\psi^*(u)]^2}{u[1-\psi^*(u)]^2}-\langle l(t)\rangle^2, \tag{B2}$$

where $\overline{l}$ and $\overline{l^2}$ are first and second moments of a single transition length, disregarding the time needed. In the case of no backflow, obviously $\overline{l}\equiv\langle l\rangle$. We thus need to determine

$$\mathcal{L}^{-1}\frac{\psi^*(u)}{u[1-\psi^*(u)]}\equiv\mathcal{L}^{-1}f^*(u)$$

and

$$\mathcal{L}^{-1}\frac{[\psi^*(u)]^2}{u[1-\psi^*(u)]^2}\equiv\mathcal{L}^{-1}g^*(u).$$

It is important to stress that although the leading terms found below do not depend on the exact expression of $\psi^*(u)$, but only on its asymptotic form, the correction terms do depend on it and one must be careful to take this into account. This point was neglected in [6].

First consider the case $0<\beta<1$. As the function $\psi^*(u)=e^{-c_\beta u^\beta}\approx 1-c_\beta u^\beta$ is used in the SCD and FPTD calculations, we also use it here. We denote $x\equiv -c_\beta u^\beta$ and we are interested in the small-x properties of the moments. Thus,

$$\psi^*(u)=1+x+\frac{x^2}{2}+\frac{x^3}{6}+O(x^4),$$

$$1-\psi^*(u)=-x-\frac{x^2}{2}-\frac{x^3}{6}+O(x^4),$$

$$[\psi^*(u)]^2=e^{2x}=1+2x+2x^2+\frac{4x^3}{3}+O(x^4),$$

$$[1-\psi^*(u)]^2=1+[\psi^*(u)]^2-2\psi^*(u)=x^2[1+x+O(x^2)],$$

$$\frac{1}{1-\psi^*(u)}=\frac{1}{-x}\left[1-\frac{x}{2}+O(x^2)\right],$$

$$\frac{1}{[1-\psi^*(u)]^2}=\frac{1}{x^2}\sum_{j=0}^{\infty}[-x+O(x^2)]^j=\frac{1}{x^2}[1-x+O(x^2)].$$

Substituting these expressions into the above formulas for f^* and g^* yields

$$f^*(u)=\frac{1+\frac{x}{2}+O(x^2)}{-xu}\approx\frac{1-\frac{c_\beta u^\beta}{2}}{c_\beta u^{\beta+1}}$$

and

$$g^*(u)=\frac{1+x+O(x^2)}{ux^2}\approx\frac{1-c_\beta u^\beta}{c_\beta^2 u^{2\beta+1}}.$$

(In [6] effectively $\psi^*(u)\equiv 1-c_\beta u^\beta$ was used, which led to similar expressions for f^* and g^* but with the coefficient of the second term in the numerators twice larger.)

After taking the inverse Laplace transform, we obtain

$$\langle l(t)\rangle\approx\overline{l}\left(\frac{t^\beta}{c_\beta\Gamma(\beta+1)}-\frac{1}{2}\right) \tag{B3}$$

and

$$\sigma_l^2(t)\approx\frac{\overline{l}^2t^{2\beta}}{c_\beta^2}\left[\frac{2}{\Gamma(2\beta+1)}-\frac{1}{\Gamma^2(\beta+1)}\right]+\frac{t^\beta(\overline{l^2}-\overline{l}^2)}{c_\beta\Gamma(\beta+1)}$$

$$\approx\left(\langle l(t)\rangle+\frac{\overline{l}}{2}\right)^2\left[\frac{2\Gamma^2(\beta+1)}{\Gamma(2\beta+1)}-1\right]+(\overline{l^2}-\overline{l}^2)\left(\langle l(t)\rangle+\frac{\overline{l}}{2}\right). \tag{B4}$$

We use the natural assumption of the applicability of the derivations, $\langle l(t)\rangle\gg\overline{l}$ to arrive at

$$\frac{\sigma_l^2(t)}{\langle l(t)\rangle^2}\approx\left[\frac{2\Gamma^2(\beta+1)}{\Gamma(2\beta+1)}-1\right]+\frac{\overline{l^2}}{\overline{l}\langle l(t)\rangle}+\frac{2\overline{l}}{\langle l(t)\rangle}\left(\frac{\Gamma^2(\beta+1)}{\Gamma(2\beta+1)}-1\right). \tag{B5}$$

In the case $\overline{l^2}\gg\overline{l}^2$ the last term can be dropped.

Now consider $1<\beta<2$. We denote $\gamma\equiv c_\beta/\overline{t}$ and $\varepsilon\equiv\beta-1$. We could consider

$$\psi^*(u)\equiv e^{-\overline{t}u+\gamma\overline{t}u^\beta}=1-\overline{t}u+\gamma\overline{t}u^\beta+\frac{\overline{t}^2u^2}{2}+O(u^{\beta+1}),$$

but instead we consider the somewhat more general expression $\psi^*(u)=1-\overline{t}u+\gamma\overline{t}u^\beta+\eta\overline{t}u^2+O(u^{\beta+1})$ with η to be specified later. It follows that $1-\psi^*(u)=\overline{t}u[1-\gamma u^\varepsilon-\eta u+O(u^\beta)]$,

$$\frac{1}{1-\psi^*(u)}=\frac{\sum_{j=0}^{\infty}[\gamma u^\varepsilon+\eta u+O(u^\beta)]^j}{\overline{t}u}=\frac{\sum_{j=0}^{\infty}\gamma^j\sum_{r=0}^{j}\binom{j}{r}\left(\frac{\eta}{\gamma}\right)^r u^{r+\varepsilon(j-r)}[1+O(u^\varepsilon)]^r}{\overline{t}u}.$$

Since $j\geq r$ and we are looking only for terms up to u in the numerator (higher order terms will give corrections decreasing with time) then only $r=0$ is relevant, plus the case $j=r=1$, and $O(u^\varepsilon)$ can be dropped. We define a positive integer number $m\equiv[1/\varepsilon]$ [i.e., $m\varepsilon\leq1$ and $(m+1)\varepsilon>1$]. We obtain

$$\frac{1}{1-\psi^*(u)}=\frac{\sum_{j=0}^{m}(\gamma u^\varepsilon)^j+\eta u+o(u)}{\overline{t}u},$$

$$f^*(u)\approx\sum_{j=0}^{m}\frac{\gamma^j}{\overline{t}}u^{\varepsilon j-2}+\left(\frac{\eta}{\overline{t}}-1\right)u^{-1}.$$

Taking the inverse Laplace transform,

$$f(t)\approx\sum_{j=0}^{m}\frac{\gamma^jt^{1-\varepsilon j}}{\overline{t}\Gamma(2-\varepsilon j)}+\left(\frac{\eta}{\overline{t}}-1\right).$$

Again, considering terms up to u in the numerator,

$$\frac{1}{(1-\psi^*(u))^2}=\frac{\left[\sum_{j=0}^{m}(\gamma u^\varepsilon)^j+\eta u\right]^2+o(u)}{\overline{t}^2u^2}=\frac{\sum_{i=0}^{m}\sum_{j=0}^{m}\gamma^{i+j}u^{\varepsilon(i+j)}+2\eta u+o(u)}{\overline{t}^2u^2}=\frac{\sum_{j=0}^{m}(j+1)(\gamma u^\varepsilon)^j+2\eta u+o(u)}{\overline{t}^2u^2}$$

so that

$$g^*(u)=\sum_{j=0}^{m}\frac{(j+1)\gamma^j}{\overline{t}^2}u^{\varepsilon j-3}+\frac{2(\eta-\overline{t})}{\overline{t}^2}u^{-2}+o(u^{-2})$$

and

$$g(t)\approx\sum_{j=0}^{m}\frac{(j+1)\gamma^jt^{2-\varepsilon j}}{\overline{t}^2\Gamma(3-\varepsilon j)}+\frac{2(\eta-\overline{t})}{\overline{t}^2}t.$$

Now we calculate $f^2(t)$ and the lowest power of interest is t,

$$f^2(t)=\sum_{n=0}^{m}\frac{\gamma^nt^{2-\varepsilon n}}{\overline{t}^2}\sum_{i=0}^{n}\frac{1}{\Gamma(2-\varepsilon i)\Gamma[2-\varepsilon(n-i)]}+2\left(\frac{\eta}{\overline{t}}-1\right)\frac{t}{\overline{t}}+o(t).$$

These preliminary calculations lead to

$$\sigma_l^2(t)=\frac{\overline{l}^2}{\overline{t}^2}\sum_{j=0}^{m}\gamma^jt^{2-\varepsilon j}\left[\frac{2(j+1)}{\Gamma(3-\varepsilon j)}-\sum_{n=0}^{j}\frac{1}{\Gamma(2-\varepsilon n)\Gamma(2-\varepsilon(j-n))}\right]+\frac{t}{\overline{t}}\left[\overline{l^2}+\left(\frac{2\eta}{\overline{t}}-2\right)\overline{l}^2\right]. \tag{B6}$$

Note that the term with $j=0$ is zero. To guarantee the positiveness of this expression in all possible cases (including $\gamma=0$ and $\overline{l^2}=\overline{l}^2$) we require that $2\eta/\overline{t}-2\geq-1$, which means $\eta\geq\overline{t}/2$ (so one might conclude that this property will be fulfilled for any function with the asymptotic expansion used above). In the particular case of an exponential form of $\psi^*(u)$ proposed above, $\eta=\overline{t}/2$ and from here on we use this value. Note that using the truncated expansion for $\psi^*(u)$ (equivalent to choosing $\eta=0$) leads to a physically wrong last correction term. As $\beta\searrow1$ ($\varepsilon\searrow0$, gives a δ pulse), all the coefficients of different powers of t greater than 1 [in Eq. (B6)] go to zero, i.e., the dispersivity approaches zero (for no backflow), as is physically correct.

Consider finally $\beta>2$ (thus, effectively, use $\beta=2$). In this case the second temporal moment of $\psi(t)$ exists and we must have $\overline{t^2}/2=(\gamma+\eta)\overline{t}$ [this follows from the small-u expansion of $\psi^*(u)$]. Thus $\gamma=\sigma_t^2/2\overline{t}$, also $m=1$, so that

$$\sigma_l^2(t)\approx\left(\overline{l}^2\frac{\sigma_t^2}{\overline{t}^2}+\sigma_l^2\right)\frac{t}{\overline{t}},$$

where we denoted $\sigma_x^2\equiv\overline{x^2}-\overline{x}^2$.

Calculating the apparent dispersivity leads to

$$D\equiv\frac{1}{2}\frac{d\sigma_l^2(t)}{dt}\simeq\frac{\overline{l}^2 t}{2\overline{t}^2}\sum_{j=0}^{m}(2-\varepsilon j)\gamma^j t^{-\varepsilon j}\left[\frac{2(j+1)}{\Gamma(3-\varepsilon j)}-\sum_{k=0}^{j}\frac{1}{\Gamma(2-\varepsilon k)\Gamma[2-\varepsilon(j-k)]}\right]+\frac{\sigma_l^2}{2\overline{t}}, \tag{B7}$$

$$v\equiv\frac{d\langle l(t)\rangle}{dt}\approx\frac{\overline{l}}{\overline{t}}\left(1+\sum_{j=1}^{m}\frac{\gamma^j t^{-\varepsilon j}}{\Gamma(1-\varepsilon j)}\right). \tag{B8}$$

We see that for ε not very close to zero the velocity reaches its limiting value of $w\equiv\overline{l}/\overline{t}$ fast enough (trivial substitution shows that $\gamma t^{-\varepsilon}\equiv b_\beta\tau^{-\varepsilon}$, which already should be a small number for the validity of the above approximations). For the first two terms

$$\frac{1}{v}=\frac{\overline{t}}{\overline{l}}\left(1-\frac{\gamma t^{-\varepsilon}}{\Gamma(1-\varepsilon)}\right)+O(t^{-2\varepsilon}),$$

and for $\varepsilon<0.5$,

$$\frac{D}{v}\approx\frac{\overline{l}\gamma}{\overline{t}}t^{1-\varepsilon}\left\{\frac{\varepsilon}{\Gamma(2-\varepsilon)}+\gamma t^{-\varepsilon}\left(\frac{1+2\varepsilon}{\Gamma(2-2\varepsilon)}+\frac{\varepsilon^2-1}{\Gamma^2(2-\varepsilon)}\right)\right\},$$

while for $\varepsilon=0.5$,

$$\frac{D}{v}\approx\frac{\overline{l}\gamma}{\overline{t}}\sqrt{\frac{t}{\pi}}+\frac{\overline{l}\gamma^2}{\overline{t}}\left(2-\frac{3}{2\pi}\right)+\frac{\sigma_l^2}{2\overline{l}},$$

and for $\varepsilon>0.5$,

$$\frac{D}{v}\approx\frac{\overline{l}\gamma\varepsilon t^{1-\varepsilon}}{\overline{t}\Gamma(2-\varepsilon)}+\frac{\sigma_l^2}{2\overline{l}}.$$

Considering $\beta\approx1$, we note that the above expressions for $\sigma_l(t)$ (both for $0<\beta<1$ and for $1<\beta<2$) cannot be used simply with $\beta=1$. Inserting $\beta=1$ would lead to $\psi^*(u)=e^{-c_1u}$, so that $\psi(t)$ is a δ function, rather than a function $\psi(t)\sim t^{-2}$ as $t\to\infty$. Unfortunately, to the best of our knowledge, no CTRW solutions exist for $\beta=1$, where $\psi^*(u)\approx1+u\ln u-c_1u$.

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