

Fractional Advection-Diffusion-Asymmetry Equation

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Fractional kinetic equations employ noninteger calculus to model anomalous relaxation and diffusion in many systems. While this approach is well explored, it so far failed to describe an important class of transport in disordered systems. Motivated by work on contaminant spreading in geological formations, we propose and investigate a fractional advection-diffusion equation describing the biased spreading packet. While usual transport is described by diffusion and drift, we find a third term describing symmetry breaking which is omnipresent for transport in disordered systems. Our work is based on continuous time random walks with a finite mean waiting time and a diverging variance, a case that on the one hand is very common and on the other was missing in the kaleidoscope literature of fractional equations. The fractional space derivatives stem from long trapping times, while previously they were interpreted as a consequence of spatial Lévy flights.

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Fractional calculus is an old branch of mathematics that studies noninteger differential operators [1–4]. This method is used extensively to model anomalous diffusion and relaxation in a wide variety of systems [5–8]. To recap, consider the fractional diffusion equation [9,10] for the density of spreading particles $\mathcal{P}(x, t)$,

$$\frac{\partial^\beta \mathcal{P}}{\partial t^\beta} = D_{\beta, \mu} \nabla^\mu \mathcal{P}, \quad (1)$$

where $D_{\beta, \mu}$, with units $\text{m}^\mu/\text{s}^\beta$, is a generalized diffusion constant. The fractional time and space derivatives are convolution operators that more intuitively are defined with their respective Laplace and Fourier transforms (see below). This equation, sometimes called the fractional diffusion-wave equation, reduces to the diffusion equation when $\beta = 1$, $\mu = 2$, and the wave equation for $\beta = 2$, $\mu = 2$. $\mu < 2$ corresponds to long spatial jumps referred to as Lévy flights (LFs), while $\beta < 1$ to long dwelling times between jump events [7]. Originally, this equation was derived using the continuous time random walk (CTRW) model [7,11–16]. More recently, the fractional diffusion equation with $\beta = 1$ was derived for heat transport using models of interacting particles [17,18]. Such fractional kinetic equations are used to describe the time-of-flight experiments of charge carriers in disordered systems where due to trapping $\beta < 1$, $\mu = 2$ [19,20] and anomalous diffusion of cold atoms in optical lattices where the atom-laser interaction induces $\mu < 2$ and $\beta = 1$ [21,22]. Extensions that include external forces are well studied within a framework referred to as the fractional Fokker-Planck equation [23–26] and distributed order fractional equations [27,28]. For an extensive review, see Ref. [7].

Equation (1) exhibits reflection symmetry, and hence, the packet of spreading particles is symmetric around its mean if the initial condition density is localized. In disordered systems with fixed advection, symmetry breaking is found, and Eq. (1) is invalid. Such behavior is found throughout hydrology, for example, for tracer and contaminant spreading in heterogeneous media. For more than two decades, two opposing and competing frameworks were developed in this field. One approach advanced by Benson, Schumer, Meerschaert, and Wheatcraft (BSMW) [29,30] proposed that the mechanism for transport is controlled by nonlocal spatial jumps of the Lévy type [7,31–33]. It was suggested that solute particles may experience long movements in high velocity flow paths, leading to such superdiffusive behavior, possibly in the spirit of LFs in rotational flow [34]. Importantly, since field observations exhibit nonsymmetric shapes of the spreading packet of particles, the microscopic picture introduces a skewed probability density function (PDF) of spatial jump lengths. This approach extensively promoted the use of nonsymmetrical fractional space advection-diffusion equations for LFs; see Ref. [33] for an overview.

The second approach uses what might be considered the opposite strategy. Instead of long nonlocal Lévy jumps in space Berkowitz, Scher and co-workers [35–41] showed that the CTRW framework with a power-law trapping time PDF is the key feature needed to explain the observed data; see also Ref. [16]. Physically, this is the result of long trapping events in geometrically induced dead ends found in strongly disordered porous media. Specifically, based on field experiments and extensive modeling, the trapping time PDF is $\psi(\tau) \sim \tau^{-(1+\beta)}$, and importantly, in many cases $1 < \beta < 2$ [38,39]. Here the mean trapping time is finite, while the variance diverges. In this case, Eq. (1) is certainly

not valid. To see this, consider a CTRW with a finite variance of jump lengths, so $\mu = 2$, and then as mentioned, if we take $\beta \rightarrow 2$, we get the wave equation, which is completely irrelevant for the transport under study. Thus, so far, the Berkowitz-Scher theoretical framework is based on a random walk picture [37] and not on a governing fractional advection-diffusion equation. Both the CTRW and the BSMW frameworks and the experiments in the field agree on one thing: Advection diffusion is anomalous and nonsymmetric [39,42–44] however otherwise these schools promote widely different philosophies.

One goal of this Letter is to promote a better understanding of the meaning of the fractional space derivatives in transport equations. As mentioned in the literature, these are associated with LFs; however, we show here that they are actually related to the long-tailed PDF of trapping times, provided that $1 < \beta < 2$. The mentioned biased CTRW is known to exhibit superdiffusion $\langle (X - \langle X \rangle)^2 \rangle \propto t^{3-\beta}$ [45]; however, this as a stand-alone does not imply a connection to LFs or fractional space kinetic equations. The first important conceptual step toward a unification of LFs and biased CTRW was given by Weeks *et al.* [46,47]. In the presence of bias, an observer in a reference frame moving with the mean speed set by the advection will view the power-law trapping times of the CTRW framework as if the particle is performing large jumps in space. Our challenge is threefold. First we want to extend this idea into a fractional equation showing the role of fluctuations. Second, we wish to develop a mathematical tool capable of dealing with a wide variety of applied problems ranging from calculations of breakthrough curves (see below), effect of time-dependent fields (omnipresent in field experiments), and different boundary conditions by far extending [42–47]. In essence, this framework is the continuum fractional diffusive description of a very large class of random walk processes. Finally, after deriving the fractional equation for the Berkowitz-Scher transport, we will be in the position to compare it to the BSMW LF method.

The fractional advection-diffusion-asymmetry equation (FADAE) investigated in this Letter reads

$$\frac{\partial}{\partial t} \mathcal{P} = D \frac{\partial^2}{\partial x^2} \mathcal{P} - V \frac{\partial}{\partial x} \mathcal{P} + S \frac{\partial^\beta}{\partial (-x)^\beta} \mathcal{P}. \quad (2)$$

The first two terms on the right-hand side of Eq. (2) are the standard diffusion and drift terms, and the last term is the modification we propose. The operator $\partial^\beta / \partial (-x)^\beta$ is a Riemann-Liouville fractional derivative [2,7] of order $1 < \beta < 2$; see the Supplemental Material [48]. The Fourier transform of this operator acting on some test function is $\mathcal{F}[d^\beta g(x) / d(-x)^\beta] = (-ik)^\beta \tilde{g}(k)$, where $\tilde{g}(k)$ is the Fourier transform of $g(x)$. In contrast to the spatial Riemann-Liouville derivatives in Eq. (2), the generalized Laplacian operators in Eq. (1) are symmetric Riesz derivatives [9], where $\mathcal{F}[\nabla^\mu g(x)] = -|k|^\mu \tilde{g}(k)$. Further, in Eq. (2)

we have no fractional time derivatives, and hence, obviously it is very different from the standard fractional diffusion Eq. (1). Here, D describes normal diffusion, V controls the drift, while S is the symmetry breaking parameter. We now explain the meaning of Eq. (2) and its extensions.

When initially $\mathcal{P}(x, t)|_{t=0} = \delta(x)$, namely, the packet of particles is localized on the origin, and when the transport coefficients are time independent and for free boundary conditions, the solution is obtained using Fourier transform. Let $\tilde{\mathcal{P}}(k, t)$ be the Fourier pair of $\mathcal{P}(x, t)$, then Eq. (2) gives

$$\tilde{\mathcal{P}}(k, t) = \exp[-Dk^2 t - ikVt + S(-ik)^\beta t]. \quad (3)$$

Thus, the solution is a convolution of a Gaussian and a nonsymmetric Lévy density [49–53]. These correspond to limit distributions of sums of independent identically distributed random variables described by thin- and fat-tailed densities, respectively. More specifically, we denote $L_\beta(y)$ as the asymmetrical Lévy density whose Fourier transform is $\exp[(-ik)^\beta]$, and hence, $\mathcal{P}(x, t) = L_\beta[x/(St)^\beta](St)^{-\beta} \otimes \exp[-(x - Vt)^2/4Dt] / \sqrt{4\pi Dt}$, where \otimes is the convolution symbol [48,54].

Model.—We treat the problem using the assumption that the particle will wait for some random time τ between two successive jumps. This is exactly the framework of the CTRW that describes a particle performing random independent steps x determined by the PDF $f(x)$, and the waiting time τ distributed according to $\psi(\tau)$ [7,11,43–47]. All the waiting times and the jump lengths are independent. We consider $\psi(\tau) \sim \beta(\tau_0)^\beta \tau^{-1-\beta}$, and as mentioned, $1 < \beta < 2$. The timescale τ_0 together with the finite mean waiting time $\langle \tau \rangle = \int_0^\infty \tau \psi(\tau) d\tau$ is important. The probability of observing N steps at time t is [55,56]

$$Q_i(N) \sim \frac{1}{(t/\bar{\tau})^{1/\beta}} L_\beta \left[\frac{N - t/\langle \tau \rangle}{(t/\bar{\tau})^{1/\beta}} \right] \quad (4)$$

with $\bar{\tau} = \langle \tau \rangle^{1+\beta} / [(\tau_0)^\beta \Gamma(1-\beta)]$. This equation is valid for large times and large N ; for example, the mean number of jumps $\langle N \rangle \sim t/\langle \tau \rangle$ is large. Equation (4) means that Lévy statistics are applicable for the shifted observable $N - \langle N \rangle$. For the jump length distribution $f(x)$, we assume that the mean size of the jumps is a and the variance is σ . For example, in the simulations below, the PDF of jump size is Gaussian

$$f(x) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp \left[-\frac{(x-a)^2}{2\sigma^2} \right]. \quad (5)$$

The parameter a is the bias, and the mean position of the particle after N steps is Na ; hence, on average the packet of particles starting on the origin will be on $at/\langle \tau \rangle$. Clearly, this modeling implies that we do not assume fat-tailed jump

length distributions, unlike the LF picture in the BSMW framework [48].

In the CTRW, the position of the particle after N steps is $X = \sum_{i=1}^N x_i$, and thus, it depends both on the microscopic displacements x_i and the random number of steps N . By conditioning on a specific outcome of N displacements, the PDF of finding the particle at X at time t is $\mathcal{P}_{\text{CTRW}}(X, t) = \sum_{N=0}^{\infty} Q_i(N)P(X|N)$. We are interested in the long time limit since in this limit N is large; hence, we replace $P(X|N)$ with the Gaussian, and similarly replace $Q_i(N)$ with the Lévy distribution Eq. (4). Switching from summation to integration, in the long time limit we find

$$\mathcal{P}_{\text{CTRW}}(X, t) \sim \int_0^{\infty} L_{\beta}\left(\frac{N - t/\langle\tau\rangle}{(t/\bar{t})^{1/\beta}}\right) \frac{\exp\left(-\frac{(X-aN)^2}{2\sigma^2 N}\right)}{\sqrt{2\pi\sigma^2 N}(t/\bar{t})^{1/\beta}} dN. \quad (6)$$

This idea is also known as the subordination of the spatial process X by the temporal process for N and is routinely considered in the literature for $\beta < 1$; see Refs. [20,57]. We already mentioned our intention to derive the spatial derivative usually associated with Lévy spatial jumps using the perfectly Gaussian jump statistics in space, and that is what we do next. In the long time limit, we find

$$\mathcal{P}_{\text{CTRW}} \sim \int_{-\infty}^{\infty} L_{\beta}(y) \frac{\exp\left\{-\frac{[X-at/\langle\tau\rangle-ay(t/\bar{t})^{1/\beta}]^2}{2\sigma^2 t/\langle\tau\rangle}\right\}}{\sqrt{2\pi\sigma^2 t/\langle\tau\rangle}} dy. \quad (7)$$

Technically, this limit is obtained with a change of variables to $\xi = (X - at/\langle\tau\rangle)/a(t/\bar{t})^{1/\beta}$ and ξ is kept fixed while $t \rightarrow \infty$ [48]. We now take the time derivative of the Fourier transform of Eq. (7) and find

$$\begin{aligned} \frac{\partial \tilde{\mathcal{P}}(k, t)}{\partial t} = & -\frac{\sigma^2}{2\langle\tau\rangle} k^2 \tilde{\mathcal{P}}(k, t) - ik \frac{a}{\langle\tau\rangle} \tilde{\mathcal{P}}(k, t) \\ & + (-ik)^{\beta} \frac{a^{\beta}}{t} \tilde{\mathcal{P}}(k, t). \end{aligned} \quad (8)$$

This is the Fourier representation of Eq. (2) when we identify the transport constants

$$D = \frac{\sigma^2}{2\langle\tau\rangle}, \quad V = \frac{a}{\langle\tau\rangle}, \quad S = \frac{a^{\beta}}{t}. \quad (9)$$

The two formulas for D and V are standard relations in the theory of advection diffusion. To summarize, the FADAE (2) describes the biased CTRW process, and this has several consequences which are now discussed.

The importance of bias.—An interesting effect is that in the absence of bias, i.e., $a = 0$, we get $S = 0$; hence, the anomaly is present only when we have advection. Since $S = 0$ implies normal diffusion, in the case of weak advection the solution exhibits nearly normal behavior

even for very long times, an effect crucial for experiments. Further, Eq. (9) shows how the two transport coefficients S and V are generally not independent. To see this, consider linear response theory. Then, we have $a \sim F$ where F is the external force field, and we have $V \sim F$ and $S \sim F^{\beta}$, a prediction that could be tested in experiments.

Packets in two dimensions.—The fact that the asymmetry constant S is bias dependent leads to the following interesting prediction in two dimensions. Imagine the bias is directed in the x direction, then the distortion of the packet of particles is found only along the x axis. In other words, the diffusion in the perpendicular y direction will be perfectly normal. In the Supplemental Material [48], we extend our mathematical treatment of the problem to two dimensions. Here we present this effect graphically in Fig. 1, where the asymmetrical oval-like shape of the spreading packet is clearly visible, with the left tail broader than the right one. Similar experimental observations were reported in Refs. [38,58]. The left tail seen clearly in the figure is due to trapping of particles far lagging behind the mean position of the packet, and this as we showed is modeled with the asymmetry operator $\partial^{\beta}/\partial(-x)^{\beta}$ in Eq. (2). Thus, the physical interpretation of the fractional space derivatives in the FADAE should be made with care, as it does not necessarily mean that the process exhibits LFs.

Temporal variations of the mean velocity $a/\langle\tau\rangle$ are often present in the real world and tested experimentally in Ref. [41]. We explore this issue now using a time-dependant but piecewise constant bias $a(t)$ [41]. Indeed,

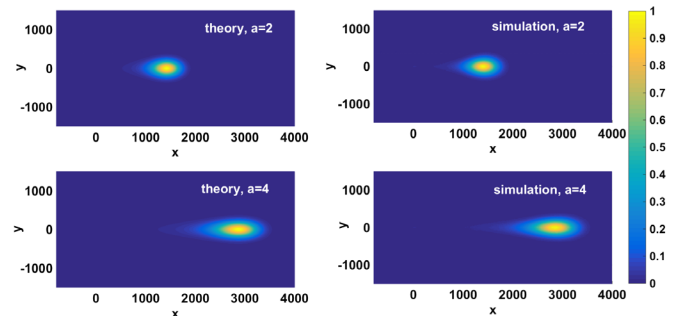


FIG. 1. Packets of particles released from an origin in two dimensions with $\beta = 3/2$, time $t = 200$ where the mean waiting time is $\langle\tau\rangle = 0.3$ and $\tau_0 = 0.1$. The bias is pointing to the x direction, while it is absent along the y axis, and this creates packets distorted in the direction of the field. The symmetry breaking effect is visibly stronger as the bias level is increased. Here we show how simulations of the CTRW process and the analytical solutions of the FADAE nicely match. For theory, we use Eq. (9) which gives $D = 41.7$, $V = a/0.3$, and $S = a^{3/2}/0.44$; the bias a is provided in the figure, while in the y direction $D = 41.7$, and $S = V = 0$. For further details on simulations, see the Supplemental Material [48], for example, a perfect agreement between theory and simulations without fitting for the one-dimensional CTRW.

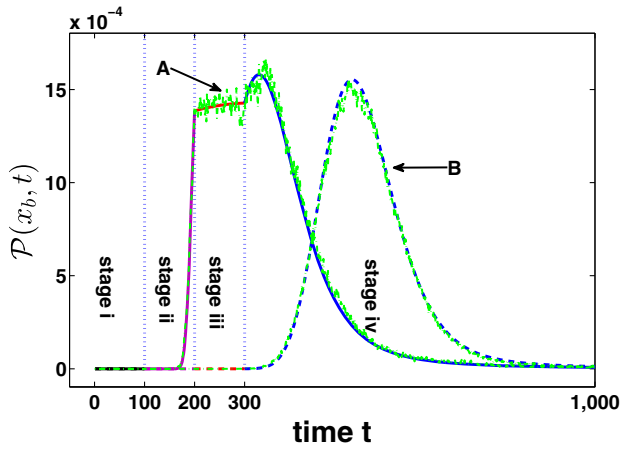


FIG. 2. Particles are released on the origin at time zero and then the density on x_b is recorded versus time. Such breakthrough curves present contaminant spreading from a source (say, the upper part of a stream) to some target on x_b , here $x_b = 1800$. Here we present the solution of the FADAE and compare it with the CTRW simulations with $\beta = 3/2$ without fitting. The bias a is time dependent, and as explained in the text, the dynamics has four stages as indicated in the graph, e.g., stage i, $0 < t < 100$, etc.

in controlled experiments, the velocity V can be modified, and then theoretical predictions can be tested in a nontrivial setting. This example will demonstrate the power of the fractional framework, as it allows for a semianalytical solution of the rather complex behavior and present physical effects related to the magnitude of the bias. We consider four stages of the transport [41]: (i) We use bias $a = 1$, (ii) we then sharply increase a to a value $a = 3.6$, then (iii) we decrease the value then bias to a small number $a = 0.09$, and finally (iv) return to the bias in state (i). All along the second length scale $\sigma = 5$ is fixed. The time lapses of each stage are clearly indicated in Fig. 2, while the derivation of analytical results is left to the Supplemental Material [48]. Note that as we modify the bias a , we are effectively modifying V and S while D remains fixed; see Eq. (9). The essential idea behind the analytical approach is that the final state of each stage serves as an initial condition to the spatial distribution of the next stage. In Fig. 2 (curve A) this analytical method is compared to the numerical solution of the CTRW with $\beta = 3/2$, finding excellent agreement. We also present the case of a constant time-independent $a = 1$ (curve B). The concentration $P(x_b, t)$ at some fixed x_b presented in Fig. 2 is called a breakthrough curve and it is commonly observed in the field of contaminant spreading in hydrology. Figure 2 clearly demonstrates the excellent quantitative agreement between theory and simulation, in a regime of dynamics which is close to real life experiments and far from trivial. Hence, we are confident that our tool, the FADAE, is a useful one.

Lévy flights and the interpretation of experiment.—The CTRW process with long-tailed PDFs is an excellent

model for transport in a wide variety of systems, for example, porous media; hence, the governing FADAE (2) is deeply related to transport in many physical systems [62–7,15,19,43,59]. Still, it is interesting to compare our approach to the fractional model of LFs that reads [29,63]

$$\frac{\partial \mathcal{P}_{\text{LF}}}{\partial t} = -V \frac{\partial \mathcal{P}_{\text{LF}}}{\partial x} + K \left(q \frac{\partial^\mu}{\partial (-x)^\mu} + p \frac{\partial^\mu}{\partial x^\mu} \right) \mathcal{P}_{\text{LF}}. \quad (10)$$

Clearly, this equation is very different from ours; in fact, in some sense it is more general as compared with Eq. (2), as it describes a general class of skewed processes with the phenomenological parameters p and q . In Ref. [31], the authors fit experimental contaminant data and report $V = 0.8$ m/h, $D = 0$, $\mu = 1.51$, $K = 2.8$ m^{1.51}/h, $q = 1$, and $p = 0$ to match the breakthrough curves. Based on this, one may naively interpret the data as stemming from a LF process. However, we realize that these parameters imply, based on our notation Eq. (2), a strong bias in the long time limit of the CTRW. This highlights that the data are consistent with a CTRW with long-tailed trapping times. To summarize, using $p = 0$ in Eq. (10) is consistent with both a LF picture promoted by the BSMW and a CTRW with broad-tailed waiting times.

To distinguish between these two approaches, one needs to analyze the trajectories of the process, not the packet of the spreading particles. More precisely, the CTRW approach and LFs method can give the same predictions for the positional distribution, but the interpretation that a model with fractional space derivatives always implies LFs is wrong. In that sense, we claim that the two competing methods are identical (in some limit relevant to experiments) from the point of view of distributions but the particles trajectories widely differ.

Extensions with subordination.—A key formula is the transformation Eq. (6). It shows how to transform a normal process to an anomalous one, for the case $1 < \beta < 2$, and as mentioned, this idea is called subordination. In Eq. (6), time t is the laboratory time, and N is sometimes referred to as operational time. The idea is simple, N , which is actually the random number of steps in the process, is distributed according to Lévy statistics, as expected from the generalized central limit theorem. We then transform the Gaussian process in the operational time N to the laboratory framework with what we call a Lévy transformation; see Eq. (6). This method can be extended to include cases with different boundary conditions, different spatially dependent force fields, stochastic trajectories, etc., and hence, the mathematical approach we presented is versatile and far more general than what we considered here.

Mean square displacement.— Throughout the manuscript, we focus on the typical fluctuations of the process. The rare events influence the density $\mathcal{P}(x, t)$ in the vicinity of $x \simeq 0$ [44]. Here, Eq. (2) does not work. In that limit, the probability of finding these particles is very small, as

expected for a biased process. In fact, such a cutoff exists also for the diffusion equation, where the telegraph equation can be used to describe far tails of the density of particles. However, in the present case, rare events control the behavior of the mean square displacement, which exhibits superdiffusion. It indicates that Eq. (2) cannot give a valid mean square displacement [43,44]; in this sense, CTRWs are of course very different compared to LFs.

Summary.—The FADAE (2) is controlled by three transport coefficients D , V , and S given in Eq. (9). This framework is valuable in many CTRW systems, ranging from the field of contaminant spreading and geophysics to transport random environments, for example, the quenched trap model [59,64]. What is remarkable is that the long-tailed PDF of trapping times, which for $0 < \beta < 1$ implies a fractional time derivative, is transplanted into a spatial space derivative when $1 < \beta < 2$. And long-tailed PDFs of jump sizes, like in LFs, are not a basic requirement for fractional space operators in transport equations, rather these are related to Lévy statistics applied to the number of jumps in the process. In this sense, we have provided a new physical and widely applicable interpretation of fractional space derivatives, within the context of fractional diffusion. More importantly, we have provided a toolbox with which one may analyze advection diffusion with different boundary conditions (with subordination) and with time-dependent fields.

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