

# Generalized Diffusion–Advection Schemes and Dispersive Sedimentation: A Fractional Approach<sup>†</sup>

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We consider advection processes following anomalous statistics on an effective transport level, within the framework of fractional kinetic equations. We discuss different realistic situations such as Galilei variant and invariant advection, as well as dispersive sedimentation processes. Exact analytical solutions for the plume and the moments characterizing these transport processes are calculated. We introduce an estimation for the breakthrough of the tracer in the dispersive processes.

## I. Introduction

Advection in a Brownian system can either be modeled through the diffusion–advection equation<sup>1–3</sup>

$$\frac{\partial W}{\partial t} + v \frac{\partial W}{\partial x} = K \frac{\partial^2}{\partial x^2} W(x,t) \quad (1)$$

if the advective drift ( $v$ ) $\partial W/\partial x$  is brought about by an external velocity field  $v$ , or in terms of the monovariate Fokker–Planck equation<sup>3,4</sup>

$$\frac{\partial W}{\partial t} = \left[ -\frac{\partial}{\partial x} \frac{F}{m\eta} + K \frac{\partial^2}{\partial x^2} \right] W(x,t) \quad (2)$$

if the drift is caused by an external, constant force  $F$ , e.g., by gravitation. Equation 2 is often referred to as Smoluchowski equation. Both eqs 1 and 2 are of the same structure, describing the motion of a scalar test (tracer) particle under the influence of a constant drift, and therefore we refer to both as diffusion–advection equations. Thereby,  $K$  is the diffusion coefficient,  $m$  denotes the mass of the tracer particle, and  $\eta$  is the friction constant, a measure for the effective interaction between the diffusing particle and its environment. Both types of diffusion–advection equations define a problem which is Galilei invariant, and the solution is given by

$$W(x,t) = P(x-a^*t,t) \quad (3)$$

where  $a^* = v$  or  $a^* = F/(m\eta)$ , and  $P(x,t)$  is the solution of the diffusion equation

$$\frac{\partial P}{\partial t} = K \frac{\partial^2}{\partial x^2} P(x,t) \quad (4)$$

This is,  $P$  is given by the well-known Gaussian probability density function (pdf)

$$P(x,t) = \frac{1}{\sqrt{4\pi Kt}} \exp\left(-\frac{x^2}{4Kt}\right) \quad (5)$$

for the sharp initial value  $P_0(x) \equiv \lim_{t \rightarrow 0^+} P(x,t) = \delta(x)$ . Note that here and in the following, we consider the one-dimensional case.

Typical realizations of such ideal advection systems, where  $v$  or  $F$  are constants, are encountered in the center section of a Hagen–Poiseuille flow, or in a constant gravity or electric field, two examples highlighting the importance of such models for the spreading of a scalar tracer, e.g., a pollutant, in a groundwater flow system, and its sedimentation therein.<sup>5–9</sup>

In what follows, we consider modifications of eqs 1 and 2 which we believe to be a fair effective description in systems where the existence of porous structures or similar ramified, restricted geometries, leads to an anomalous transport behaviour.

The Brownian nature underlying both eqs 1 and 2 is manifest in the mean-square displacement

$$\langle(\Delta x)^2\rangle \equiv \langle x^2\rangle - \langle x\rangle^2 = 2Kt \quad (6)$$

which increases linearly in time. This is actually an outcome of the central limit theorem.<sup>1,10–12</sup> In the following, we concentrate on such kinds of systems where the existence of broad transport statistics leads to a mean-square displacement of the form<sup>12–14</sup>

$$\langle(\Delta x)^2\rangle \propto t^\alpha, \quad \alpha \neq 1 \quad (7)$$

which may comprise subdiffusion ( $0 < \alpha < 1$ ) and superdiffusion ( $\alpha > 1$ ). Processes called Lévy flights (see below) are even characterized by a diverging mean-square displacement,  $\langle(\Delta x)^2\rangle \rightarrow \infty$  as they occasionally exhibit extremely long displacements in a single jump event.

In what follows, we introduce our concept of modeling anomalous diffusion in terms of fractional equations. The explicit derivation of such equations is briefly reviewed and the concept of fractional models for anomalous diffusion in an external velocity or force field is established. The main emphasis will be put on subdiffusive phenomena.

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**II. Continuous Time Random Walks and Fractional Dynamics: From Amorphous Semiconductors to the Flight of an Albatross**

In their studies of charge carrier transport in a constant electric field in amorphous semiconductors, during the advent years of the photocopying process, Harvey Scher, Elliott Montroll, and collaborators discovered a certain universality in the experimental data obtained for the time dependence of the electrical current which was inconsistent with a Brown/Gaussian description of the charge carrier motion: some sort of anomalous transport process had to be assumed. Their ideas crystallized in the modified random walk picture in which it was assumed that due to the amorphous nature of the semiconductor material, trapping processes occur in which the moving charge carriers get immobilized for a certain time span, the so-called waiting time which is supposed to be drawn from the waiting time pdf. The assumption of a waiting time pdf with a diverging first moment turned out to lead to a remarkably accurate description of all experimentally accessible quantities, including their parametric dependence.<sup>15–17</sup> These studies on the footing of the Montroll–Weiss model<sup>18</sup> led to the success of what is now known as the continuous time random walk (CTRW) model.

Brownian, as well as non-Brownian motion, is described by two pdfs in this CTRW picture, namely the waiting time pdf  $w(t)$  which governs the pausing time spans elapsing between any two jump events, and the jump length distribution  $\lambda(x)$  from which the magnitude of the jumps is drawn. Accordingly, the probability of just having arrived at a certain position  $x$  at time  $t$ ,  $p(x,t)$  obeys the convolution integral equation<sup>19</sup>

$$p(x,t) = \int_0^t dt' \int_{-\infty}^{\infty} dx' p(x',t')w(t-t')\lambda(x-x') + p_0(x)\delta(t) \quad (8)$$

by which this quantity  $p(x,t)$  is related to the pdf of just having arrived in  $x'$  at an earlier time  $t'$ . Note the assumption of instantaneous jumps and the explicit occurrence of the initial value  $p_0(x) \equiv \lim_{t \rightarrow 0^+} p(x,t)$  stating that initially the tracer particles arrive simultaneously at their starting points. The pdf to be at position  $x$  at some time  $t$ ,  $P(x,t)$ , is then related to the cumulative sticking probability (the probability that the particle waits in its site for longer than  $t$ ),

$$\phi(t) = 1 - \int_0^t dt' w(t') \quad (9)$$

in the following way:

$$P(x,t) = \int_0^t dt' p(x,t')\phi(t-t') \quad (10)$$

i.e., a particle is at  $x$  at time  $t$  if it arrived there in a previous time  $t'$  and has not moved since. In Fourier–Laplace space, the relations 8–10 can be recast into the convenient algebraic form<sup>19</sup>

$$P(k,u) = \frac{1 - w(u)}{u} \frac{1}{1 - w(u)\lambda(k)} \quad (11)$$

where  $k$  denotes the wave number and  $u$  the Laplace variable. Three basic classes of diffusion within this multiplicative CTRW picture can be distinguished, according to the finiteness or divergence of the characteristic waiting time

$$T \equiv \int_0^{\infty} tw(t) dt \quad (12)$$

and the jump length variance

$$\Sigma^2 \equiv \int_{-\infty}^{\infty} x^2 \lambda(x) dx \quad (13)$$

(i) If both  $\Sigma^2$  and  $T$  are finite, the process belongs to the domain of attraction of the central limit theorem, and the resulting transport process corresponds to Brownian motion. A possible realization is the choice of a Poissonian waiting time pdf and a Gaussian jump length pdf according to

$$w(t) = \tau^{-1} \exp\{-t/\tau\},$$

$$\lambda(x) = (4\pi\sigma^2)^{-1/2} \exp\{-x^2/(4\sigma^2)\} \quad (14)$$

by means of which, in the long time and long distance limit  $w(u) \sim 1 - u\tau$ ,  $\lambda(k) \sim 1 - \sigma^2 k^2$ , one can by virtue of eq 11 immediately recover the standard diffusion eq 4.

(ii) If  $\Sigma^2$  is kept finite but a waiting time pdf of Lévy stable type is introduced which features the asymptotic long-tail behavior

$$w(t) \sim A_{\alpha} \tau^{-1} (t/\tau)^{-1-\alpha}, \quad 0 < \alpha < 1 \quad (15)$$

the associated dynamical process leaves the basin of attraction of the central limit theorem as  $T \rightarrow \infty$  diverges, and the pdf of this random walk is no longer given by the Gaussian pdf (eq 5). Inserting the Laplace transform of such a waiting time pdf,  $w(u) \sim 1 - (u\tau)^{\alpha}$ , into relation 11 and inverting into the position–time domain, one recovers the fractional diffusion equation (FDE)<sup>14,20–23</sup>

$$P(x,t) - P_0(x) = {}_0D_t^{1-\alpha} K_{\alpha} \frac{\partial^2}{\partial x^2} P(x,t) \quad (16)$$

by means of the generalized integration theorem  $\mathcal{L}\{{}_0D_t^{-\alpha} P(x,t)\} = u^{-\alpha} P(x,u)$  of the Laplace transformation.<sup>24</sup> Equation 16 is written in the integral form. A standard differentiation leads to the differential form of the FDE,

$$\frac{\partial P}{\partial t} = {}_0D_t^{-\alpha} K_{\alpha} \frac{\partial^2}{\partial x^2} P(x,t) \quad (17)$$

Here, the fractional Riemann–Liouville operator  ${}_0D_t^{-\alpha} \equiv d/dt {}_0D_t^{-\alpha}$  is defined by the convolution<sup>24</sup>

$${}_0D_t^{-\alpha} P(x,t) = \frac{1}{\Gamma(\alpha)} \int_0^t dt' \frac{P(x,t')}{(t-t')^{1-\alpha}} \quad (18)$$

From equation 18 it becomes clear that the FDE (eq 17) describes a non-Markovian process with a slowly decaying memory. The generalized diffusion constant  $K_{\alpha}$  is given through  $K_{\alpha} \equiv \sigma^2/\tau^{\alpha}$  and has therefore the fractional dimension  $[K_{\alpha}] \equiv \text{cm}^2 \text{ s}^{-\alpha}$ . Note that the dynamical origin of this generalized diffusion constant has recently been discovered for a trapping system as a time-rescaled version of its Brownian counterpart,  $K$ ,<sup>25,26</sup> see also below.

In terms of Fox’s  $H$ -function the exact analytical solution<sup>14,20</sup>

$$P(x,t) = \frac{1}{\sqrt{4\pi K_{\alpha} t^{\alpha}}} H_{1,2}^{2,0} \left[ \frac{x^2}{4K_{\alpha} t^{\alpha}} \middle| \left(1 - \frac{\alpha}{2}, \alpha\right) \right. \left. \left(0, 1\right), \left(\frac{1}{2}, 1\right) \right] \quad (19)$$

for the pdf  $P(x,t)$  can be found for eqs 16 and 17 which exhibits the asymptotic stretched Gaussian behavior

$$P(x,t) \sim \frac{1}{\sqrt{4\pi K_\alpha t^\alpha}} \sqrt{\frac{1}{2-\alpha} \left(\frac{2}{\alpha}\right)^{(1-\alpha)/(2-\alpha)} \left(\frac{|x|}{\sqrt{K_\alpha t^\alpha}}\right)^{-(1-\alpha)/(2-\alpha)}} \times \exp\left(-\frac{2-\alpha}{2} \left(\frac{\alpha}{2}\right)^{\alpha/(2-\alpha)} \left[\frac{|x|}{K_\alpha t^\alpha}\right]^{1/(1-\alpha/2)}\right) \quad (20)$$

This asymptotic result has the same structure as the solution reported in ref 27 for the continuous time random walk model.

(iii) The third case is related to a finite characteristic waiting time  $T$  in combination with an infinite jump length variance  $\Sigma^2 \rightarrow \infty$  which comes about via a Lévy stable pdf for  $\lambda(x)$ , issuing the long-tail asymptotics<sup>50</sup>

$$\langle x \rangle \sim \beta_\mu \sigma (\sigma |x|)^{-1-\mu}, \quad 1 < \mu < 2 \quad (21)$$

Following along the same steps as for item (ii), one recovers the FDE

$$\frac{\partial P}{\partial t} = K^\mu {}_{-\infty}D_x^\mu P(x,t) \quad (22)$$

with  $K^\mu \equiv \sigma^\mu/\tau$  for this process, where the fractional Weyl or Riesz<sup>51</sup> operator  ${}_{-\infty}D_x^\mu$  is defined through<sup>52</sup>

$${}_{-\infty}D_x^\mu P(x,t) = \frac{t^{1-\mu}}{\Gamma(2-\mu)} \frac{\partial^2}{\partial x^2} \int_{-\infty}^x dx' \frac{P(x',t)}{(x-x')^{\mu-1}} \quad (23)$$

From eq 23 follows the convenient relation

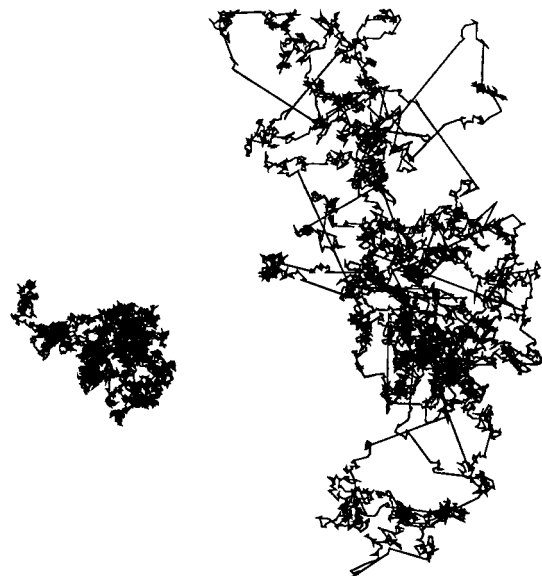
$$\mathcal{F}\{ {}_{-\infty}D_x^\mu P(x,t) \} = -|k|^\mu P(k,t) \quad (24)$$

which generalizes the differentiation theorem of the Fourier transformation. The FDE (eq 22) describes Lévy flights.<sup>22</sup> The latter have a fractal trajectory where local rambling is followed by long sojourns. This taking place on all length scales one encounters the typical clustering shown in Figure 1 where the Lévy flight trajectory is compared to the coillike trajectory determined by an  $\langle x \rangle$  with finite  $\Sigma^2$  which is typical for Brownian and subdiffusive phenomena, items (i) and (ii).

Note that in the case of Lévy flights, the mean-square displacement diverges,  $\langle x^2 \rangle \rightarrow \infty$ ; often, the broadening of the pdf  $P(x,t)$  of the Lévy flight random walk in the course of time is quantified in terms of pseudo mean square displacements.<sup>14</sup> It is worth while mentioning that such Lévy trajectories are assumed to describe qualitatively the search for food of certain living systems such as bacteriae,<sup>28,29</sup> butterflies, or an albatross.<sup>30</sup> The basic idea is that the space is screened much more efficiently for food in the case of a Lévy flight. The efficiency of Lévy flights in spatial screening also explains why they can model the gaze shifts that occur during the visual exploration of an image.<sup>31</sup>

### III. Breaking the Spatial Isotropy: Fractional Diffusion–Advection Equations

The above presented CTRW modeling and the fractional equations derived therefrom correspond to random walk processes where each jump has equal directional probability to go left or right. In those cases where external fields destroy this isotropy, or even the homogeneity of the problem, the concept has to be generalized. For anomalous processes such a generalized concept in terms of fractional equations has been developed for transport in external velocity fields<sup>23,32–36</sup> and in external force fields.<sup>23,25,26,37–42</sup> This straightforward extension demon-



**Figure 1.** Comparison of the trajectories of a Brownian or subdiffusive random walk (left) and a Lévy walk with index  $\mu = 1.5$  (right). Whereas both trajectories are statistically self-similar, the Lévy walk trajectory possesses a fractal dimension, characterizing the island structure of clusters of smaller steps, connected by a long sojourn. Both walks are drawn for the same number of steps (approximately 7000).

strates the advantage of the fractional kinetic equation approach. In what follows, two fractional diffusion–advection equations are presented which are Galilei invariant and variant, respectively. Fractional sedimentation processes are considered in the next section.

#### A. Galilei Invariant Fractional Diffusion–Advection Model.

Galilei invariant processes occur in uniform flow fields such as underground water arteries and denote a stationary state which is described by the Galilei-shifted propagator

$$W(x,t) = P(x - vt, t) \quad (25)$$

The belonging fractional diffusion–advection equation (FDAE) is obtained by observing that the jump statistics in the moving frame are still the same as dealt with in the preceding section, and are given by the jump pdf  $\psi(x,t) = \lambda(x)w(t)$ . In the laboratory frame, the corresponding quantity is thus given by the pdf  $\pi(x,t) = \psi(x - vt, t)$  (i.e., after waiting a time  $t$  the jump length distribution is no longer centered around where the tracer landed but around where it has been dragged to by the fluid), equivalent to the relation  $\pi(k,u) = \psi(k, u + ikv)$  in Fourier–Laplace space. One readily recovers the FDAE<sup>23</sup>

$$\frac{\partial W}{\partial t} + v \frac{\partial W}{\partial x} = {}_0D_t^{1-\alpha} K_\alpha \frac{\partial^2}{\partial x^2} W(x,t) \quad (26)$$

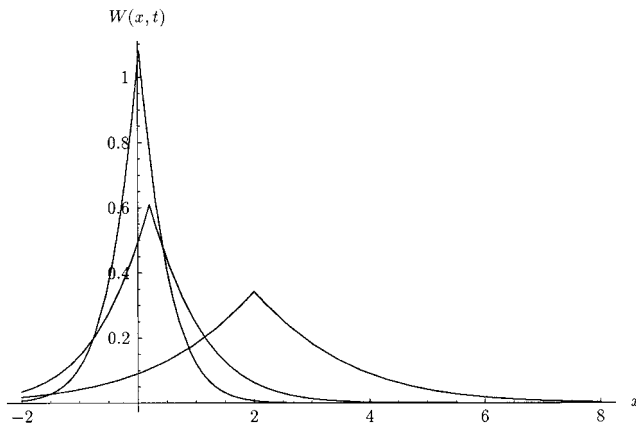
for the subdiffusive case, and the FDAE<sup>23</sup>

$$\frac{\partial W}{\partial t} + v \frac{\partial W}{\partial x} = {}_{-\infty}D_x^\mu K^\mu W(x,t) \quad (27)$$

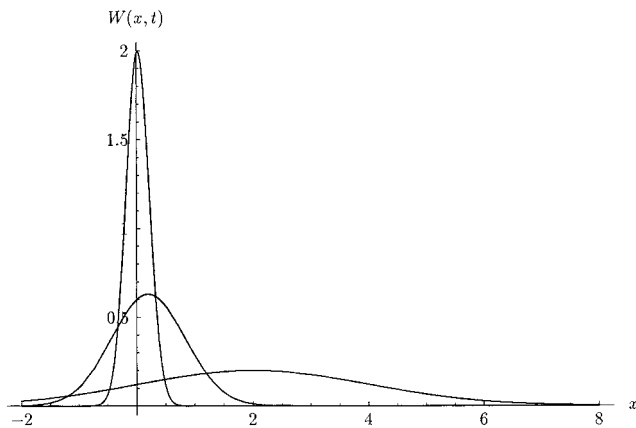
for Lévy flights. In both cases, the Galilei invariance is reflected by the occurrence of the drift term  $(v)\partial W/\partial x$  in eqs 26 and 27. This simple behavior is also manifest in the first moment valid for both cases,

$$\langle x \rangle = vt \quad (28)$$

and in the mean-square displacement for the subdiffusive case,  $\langle (\Delta x)^2 \rangle = 2K_\alpha t^\alpha/\Gamma(1+\alpha)$  which is of the same form as the free contribution, eq 7. Note that the existence of the first



**Figure 2.** Galilei invariant subdiffusive model. The propagator is shown for the dimensionless times  $t = 0.02, 0.2,$  and  $2$ . The propagator is symmetric with respect to its maximum which is translated with velocity  $v = 1$ . The cusps marking the initial condition are distinct in comparison to the Brownian result shown in Figure 3.



**Figure 3.** Galilei invariant Brownian model. The propagator is shown for the dimensionless times  $t = 0.02, 0.2,$  and  $2$ . The propagator is symmetric with respect to its maximum which is translated with velocity  $v = 1$ . The fast decay of the initial condition is mirrored in the smooth peak of the distribution, compare to the subdiffusive result shown in Figure 2.

moment in the Lévy flight case, and therefore a proper velocity, is due to our assumption  $1 < \mu < 2$  in eq 21.

In Figure 2 we plotted the subdiffusive solution  $W(x,t)$  from eqs 25 and 19 for successive times. Note the distinct cusp of the fractional solution, in comparison to the Brownian analogue graphed in Figure 3 for the same sequence of times. For plotting the fractional solution we made use of the properties of the Fox function as listed in the Appendix.

Galilei invariant subdiffusion should occur in the random motion of a small bead immersed in a flowing polymer solution where the dynamic obstacles formed by the polymer chain lead to the slow dispersion of the bead. Similar situations might occur when tracer substances disperse in certain formations of morain or other sediment-enriched flows.

However, the case where the same tracer particles diffuse in a nonuniform velocity field and under the influence of boundary conditions is not as trivial. One instance of this is the situation where the particles diffuse in a Poiseuille–like flow between parallel plates. In ref 33 this situation is looked at making use of eq 17 for the transversal direction and of a Langevin equation for the longitudinal direction. The results show that the leading term in the dispersion of the tracers at long times is a coupling effect of convection and transversal diffusion, thus deserving the name of generalized Taylor dispersion:

$$\langle \Delta x^2 \rangle \sim \overline{v^2} / 2 K_\alpha^{-1} t^{2-\alpha} \quad (29)$$

where  $l$  is the distance between the plates and  $\overline{v^2}$  is typically the square of the mean velocity, assumed nonuniform (for the uniform case the term in eq 29 vanishes and only eq 7 remains, as expected).<sup>33</sup>

**B. Galilei Variant Fractional Diffusion–Advection Equation for Trapped Particles and “Partial Sticking”.** An interesting variation of the Galilei invariant model developed above arises for such cases where so-called “partial sticking” occurs. In such systems the tracer particle gets trapped in the pores or similar obstacles of the substrate off the velocity backbone, but when it eventually jumps, it jumps forward to catch up with the moving fluid, i.e., the average jump length of the particles that were trapped for a time  $\tau$  is  $v\tau$ . A possible realization of partial sticking might occur if the tracer particle leaves the velocity backbone which winds through the substrate, and this particle finds a shortcut along an offstream connection in order to rejoin the backbone somewhere downstream. Such situations might be relevant in ramified aquifers where fine channels exist alongside the main flow. The measured velocity of the partial sticking model,  $\alpha v$ , which is obtained from the first moment and which is related to the backbone only, is consequently smaller than the actual drag velocity,  $v$ . In such a scenario, the Galilei invariance is broken during the waiting times (the pdf for the jumps is actually a Galilei transformation of the pdf in the static case,  $\pi(x,t) = \psi(x - vt,t)$ , while the cumulative sticking probability is not transformed<sup>23</sup>), and the governing FDAE becomes<sup>23</sup>

$$\frac{\partial W}{\partial t} + v \frac{\partial W}{\partial x} = \left[ \frac{\alpha(1-\alpha)}{2} v^2 {}_0D_t^{-1} + K_\alpha {}_0D_t^{1-\alpha} \right] \frac{\partial^2}{\partial x^2} W(x,t) \quad (30)$$

In comparison to eq 26, an additional term occurs which is proportional to the square of the drag velocity,  $v^2$ , and which vanishes in the Brownian limit  $\alpha = 1$ . The difference to the Galilei invariant model is best observed in the temporal evolution of the moments:

$$\langle x \rangle = \alpha vt \quad (31)$$

$$\langle x^2 \rangle = \frac{2K_\alpha}{\Gamma(1+\alpha)} t^\alpha + \frac{\alpha(1+\alpha)}{2} v^2 t^2 \quad (32)$$

$$\langle (\Delta x)^2 \rangle = \frac{2K_\alpha}{\Gamma(1+\alpha)} t^\alpha + \frac{\alpha(1-\alpha)}{2} v^2 t^2 \quad (33)$$

It is interesting to note that the drift grows still linearly with time, but that the effective velocity  $v^* \equiv \alpha v$ , is the drag velocity rescaled by the anomalous diffusion exponent  $\alpha \in (0,1]$ . In the mean-square displacement, a ballistic contribution adds to the free diffusion contribution so that a turnover from the  $\propto t^\alpha$  region to the ballistic behavior  $\propto t^2$  can be observed. This very efficient dispersion can be understood imagining that a trapped particle is much more efficiently separated from an other particle which gets dragged along with velocity  $v$ . We may thus also expect to see enhanced dispersion in other kinds of trapping mechanisms as shown in the next subsection.

**C. Galilei Variant Fractional Diffusion–Advection Equation for Trapped Tracers with Mean Flight-Time.** In this model it is assumed that the tracer particles are trapped in the pores of the substrate in which the transport process takes place during the waiting times but their subsequent jumps are independent of the preceding waiting time. Instead, the jumps

depend on the velocity at their starting points and on an average time-of-flight  $\tau_a$ , which is eventually taken to vanish to be consistent with the instantaneous jump assumption. This model was originally conceived for stationary but space-dependent velocity fields  $v(x)$ .<sup>32,34</sup> In this scheme, the jump pdf is now

$$\pi(x,t;x_0) = \psi(x - v(x_0)\tau_a, t) \quad (34)$$

determining the shape of the laboratory frame jump pdf  $\pi(x,t;x_0)$ . Relation 34 expresses the dilation experienced by the dragged particle in terms of the velocity at prior position  $x_0$  where the motion event started, during the average dilation time  $\tau_a$ . Then, a cleverly introduced limit  $\tau \rightarrow 0$ ,  $\tau_a \rightarrow 0$ , and  $\sigma^2 \rightarrow 0$  leads to the fractional diffusion–advection equation<sup>32,34,35</sup>

$$\frac{\partial W}{\partial t} = {}_0D_t^{1-\alpha} \left[ -A_\alpha \frac{\partial}{\partial x} v(x) + K_\alpha \frac{\partial^2}{\partial x^2} \right] W(x,t) \quad (35)$$

$A_\alpha$  being an advection parameter arising from this derivation and, as argued below, formally related to the advection velocity. Note that eq 35 has the same structure as the fractional Fokker–Planck equation.<sup>40–42</sup> In fact, eq 35 can be thought of as a Brownian process, interrupted by energy conserving trapping events.<sup>25,26</sup> In this multiple trapping scenario, the particle dynamics is governed by the Langevin equation with  $\delta$ -correlated Gaussian noise during the free motion events. Occasionally, the particle gets immobilized in traps which might be related to impurities in a semiconductor, or to pores in a disordered environment. If such trapping periods are governed by a broad waiting time pdf of the form eq 15, and the mean distance between individual traps is of finite measure, the overall dynamics becomes fractional, being controlled by the fractional Klein–Kramers equation whose overdamped limit is the fractional Fokker–Planck equation.<sup>25,26</sup> In the presented advection model this means that adsorption or geometrical trapping in pores confines the particle successively, for self-similar time spans without a characteristic time scale.

Let us investigate eq 35 for a constant velocity. The resulting equation

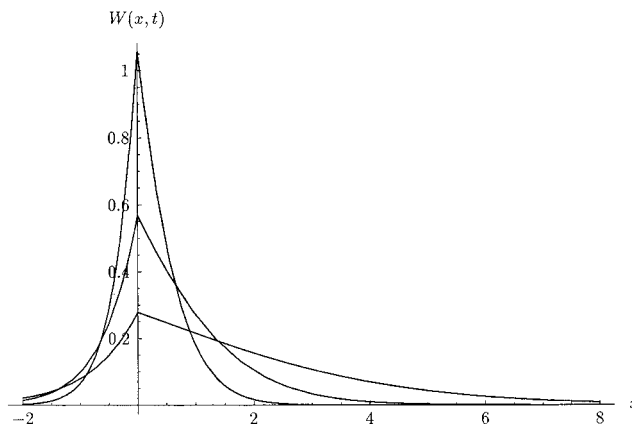
$$\frac{\partial W}{\partial t} = {}_0D_t^{1-\alpha} \left[ -v^* \frac{\partial}{\partial x} + K_\alpha \frac{\partial^2}{\partial x^2} \right] W(x,t) \quad (36)$$

with  $v^* = A_\alpha v$  is not Galilei invariant in the sense of the constant velocity shift  $x' \rightarrow x - vt$ . It can be proved that eq 36 does neither fulfill a generalized Galilei invariance of the form  $x' \rightarrow x - ct^\alpha$  which might be anticipated from the first moment, eq 40 below. This fact becomes obvious regarding the temporal evolution of  $W(x,t)$  from eq 36 displayed in Figure 4. As is known from the continuous time random walk model for broad waiting time distributions,<sup>9,15–17</sup> the initial distribution decays only very slowly in the course of time so that the distribution becomes more and more skewed. This growing asymmetry clearly rules out any kind of generalized Galilei shift of the pdf.

Figure 4 was obtained as follows. Note that the normalized solution of eq 36 in Laplace space fulfils the scaling relation  $W_\alpha(x,u) = u^{\alpha-1} W_1(x, u^\alpha)$  between the fractional solution  $W_\alpha$  and its Brownian counterpart,

$$W_1(x,t) = \frac{1}{\sqrt{4\pi t}} \exp\left(-\frac{(x-t)^2}{4t}\right) \quad (37)$$

for  $v, K \equiv 1$ .<sup>40</sup> In the time domain this corresponds to the transformation<sup>43</sup>



**Figure 4.** Galilei variant subdiffusive model with mean flight time,  $\alpha = 1/2$ . The propagator is shown for the dimensionless times  $t = 0.02$ ,  $0.2$ , and  $2$ . The propagator is asymmetric with respect to its maximum which stays fixed at the origin. The plume stretches more and more into the direction of the velocity.

$$W_\alpha(x,t) = \int_0^\infty ds A(s,t) W_1(x,s) \quad (38)$$

where the kernel  $A(s,t)$  is given through the inverse Laplace transformation

$$A(s,t) = \mathcal{L}^{-1}\{u^{\alpha-1} \exp(-su^\alpha)\} \quad (39)$$

which corresponds to a one-sided Lévy distribution. For  $\alpha = 1/2$ , one finds  $A(s,t) = (\pi t)^{-1/2} \exp(-s^2/(4t))$ . This representation was used in producing Figure 4 by help of the NIntegrate script in Mathematica.<sup>44</sup> This method turns out to be rather fast and robust in convergence. It is superior to the analytical representation in form of a series.<sup>14</sup> Note that the transformation eq 38 guarantees the nonnegativity of the fractional solution.

The moments for the Galilei variant fractional diffusion–advection process can be straightforwardly calculated from eq 36 through the fundamental relation  ${}_0D_t^q t^p = \Gamma(1+p)t^{p-q}/\Gamma(1+p-q)$  of the Riemann–Liouville operator. One obtains

$$\langle x \rangle = \frac{v^* t^\alpha}{\Gamma(1+\alpha)} \quad (40)$$

$$\langle (\Delta x)^2 \rangle = \frac{2K_\alpha t^\alpha}{\Gamma(1+\alpha)} + (v^*)^2 t^{2\alpha} \frac{2\Gamma^2(1+\alpha) - \Gamma(1+2\alpha)}{\Gamma(1+2\alpha)\Gamma^2(1+\alpha)} \quad (41)$$

Consequently, the first moment is sublinear in time and thus mirrors the spatial sticking brought about by the long-tailed waiting time distribution. The second moment features a turnover from an initial  $t^\alpha$  to a  $t^{2\alpha}$  behavior which stems from the much more efficient separation of two particles the one of which is trapped.

*1. Application to a Stratified Random Velocity Field.* A particular interesting situation for groundwater flow research is the question of diffusion of tracer particles in a stratified random velocity field. Matheron and de Marsily<sup>45</sup> were the first to address this issue for the Brownian case. In ref 46 this same situation is further explored in higher dimensions and compared to trapping problems. Recently, diffusion in a stratified random velocity field has also been studied when the tracer particles diffuse according to the non-Galilei invariant scheme that we present in this section.<sup>33</sup> There, eq 31 is solved for a stationary but nonhomogeneous two-dimensional random velocity field given by  $v_y = 0$  and  $\langle v_x \rangle = 0$ ,  $\langle v_x(y)v_x(y') \rangle = \sigma_2 \delta(y - y')$ . The

**TABLE 1. Growth of the Drift in the Various Diffusion–Advection Models<sup>a</sup>**

$t$	$d_1(t)$	$d_2(t)$	$d_3(t)$	$d_4(t)$	$d_5(t)$
1 s	10 cm	5 cm	10 cm	10 cm	10 cm
1 min	6 m	3 m	77 cm	2.2 m	46 cm
1 h	360 m	180 m	600 cm	46 m	2.2 m
1 day	8.6 km	4.3 km	30 m	0.5 km	7.1 m
1 week	60 km	30 km	78 m	2.2 km	15 m
1 year	3200 km	1600 km	561 m	42 km	65 m

<sup>a</sup> The chosen drag velocity is  $v = 10$  cm/s and the anomalous diffusion exponent is  $\alpha = 1/2$ . Here,  $d_1 = vt$ , eq 28, is the Galilei invariant drag with  $v_d = v$ ;  $d_2 = \alpha vt$  is the Galilei variant drag with “partial sticking” (eq 31) where  $v_d = \alpha v$ ;  $d_3 = v^* t^{1/2} / \Gamma(3/2)$  for the Galilei variant model, eq 40, with  $v^* / \Gamma(3/2) = 10$  cm/s<sup>1/2</sup>;  $d_4(t) = \sqrt{A^* t^{3/2}}$  for the Matheron/de Marsily case, eq 42 with  $\alpha = 1$ , for  $A^* = 100$  cm<sup>2</sup>/s<sup>3/2</sup>;  $d_5(t) = \sqrt{A^* t^{3/4}}$  for the fractional Matheron/de Marsily case, eq 42, with the deliberate choice  $A^* = 100$  cm<sup>2</sup>/s<sup>3/4</sup>.

squared mean displacement obtained<sup>33</sup> is

$$\langle \bar{x}^2 \rangle \sim \frac{A^2 \alpha \sigma_2}{\sqrt{K_\alpha}} t^{3\alpha/2} \quad (42)$$

which is slower than the typical behavior of Fickian diffusion in this same situation:<sup>47</sup>  $\langle \bar{x}^2 \rangle \sim t^{3/2}$ . The interest of this result relies on the fact that it is an exact derivation of the dispersion of a tracer particle in a substrate that shows two kinds of structural complexity: random stratification of the velocity field and trapping mechanism in diffusion. Both of these effects are expected to play a role in tracer diffusion in porous aquifers.

**D. Quantitative Analysis of Fractional Diffusion–Advection Processes and Their Measurement.** 1. *Drift Velocity.* A convenient measure for diffusion–advection problems is the drift velocity which can be defined through

$$v_d \equiv \frac{d\langle x \rangle}{dt} \quad (43)$$

and which quantifies the propagation of the peak of the pdf  $W(x,t)$  in a homogeneous velocity field. For the different models presented in this section, the results are the constant drift velocity  $v_d = v$  in the Galilei invariant fractional case, Section IIIA, the constant rescaled drift velocity  $v_d = \alpha v$  in the Galilei variant fractional case with “partial sticking”, the monotonically decreasing “effective” drift velocity  $v_d = v^* t^{\alpha-1} / \Gamma(\alpha)$  in the Galilei variant fractional case with mean flight time, and  $v_d = t^{3/4-1}$  for the stratified velocity field in Section IIIC. In Table 1 we summarize the temporal behavior of the different models, showing that the different transport mechanisms lead to considerably different temporal evolution of the systems.

2. *Flank Velocity.* The propagation of the center of the distribution (the center of mass of the diffusing tracer substance) is not always an adequate measure. Especially when convection is weak, a considerable contribution to the spreading of the tracer which can be observed in breakthrough experiments come from the spreading flanks of the pdf  $W(x,t)$  itself which is characterized by the square root of the mean-square displacement. In addition to the drift velocity (eq 43), we therefore propose the flank velocity

$$v_f \equiv \frac{d}{dt} (\sqrt{\langle (\Delta x)^2 \rangle} + \langle x \rangle) \quad (44)$$

as a measure when a considerable portion (half of the remaining peak concentration) of the tracer arrives at a certain position.

The advance of the flank given through the relation  $(\sqrt{\langle (\Delta x)^2 \rangle} + \langle x \rangle)$  delivers an approximation for the breakthrough of the tracer. The corresponding time needed to reach this point is an estimation for the breakthrough time.

In the velocity advection schemes developed in this section, the flank velocity is given through

$$v_f = \sqrt{\frac{\alpha K_\alpha}{2\Gamma(\alpha)}} t^{\alpha/2-1} + v \sim v \quad (45)$$

for the Galilei invariant fractional model from Section IIIA, through

$$v_f \sim \left( \sqrt{\frac{\alpha(1-\alpha)}{2}} + \alpha \right) v \quad (46)$$

in the long time limit for the Galilei variant model (eq 30) with “partial sticking”, and through

$$v_f \sim (\sqrt{R}\Gamma(1+\alpha) + 1) \frac{v^* t^{\alpha-1}}{\Gamma(\alpha)} \quad (47)$$

for the Galilei variant model (eq 36) with mean flight time where we used

$$R = \frac{2\Gamma^2(1+\alpha) - \Gamma(1+2\alpha)}{\Gamma(1+2\alpha)\Gamma^2(1+\alpha)} \quad (48)$$

Note that now the long time result has a different prefactor than the corresponding result for  $v_d$ .

Accordingly, only the model with mean flight–time shows an ever–decreasing drift or flank velocity whereas the other fractional advection models tend toward a constant value. This corresponds to the scale-free waiting time process involved, i.e., the particle is successively immobilized for comparably long waiting time spans.

#### IV. Dispersive Sedimentation Processes

Subdiffusion in an external force field has recently been formulated in terms of a fractional Fokker–Planck equation (FFPE)<sup>40</sup> which has been derived from random walk principles<sup>41,42</sup> and from a dynamics approach based on a Langevin equation coupled with trapping.<sup>25,26</sup> The FFPE for subdiffusion in the external force field  $F(x) = -\Phi'(x)$  reads<sup>14,25,26,40–42</sup>

$$\frac{\partial W}{\partial t} = {}_0D_t^{1-\alpha} \left[ -\frac{\partial}{\partial x} \frac{F(x)}{m\eta_\alpha} + K_\alpha \frac{\partial^2}{\partial x^2} \right] W(x,t) \quad (49)$$

and is thus very similar to the fractional diffusion–advection eq 35. In eq 49,  $m$  is the mass of the tracer particle,  $\eta_\alpha$  the generalized friction constant, and  $K_\alpha$  denotes the generalized diffusion constant. It has been shown that the generalized Einstein–Stokes relation  $K_\alpha = k_B T / [m\eta_\alpha]$  and the second Einstein relation  $\langle x \rangle_F = 1/2 F \langle x^2 \rangle / k_B T$  between the first moment in the presence of the constant force  $F$  and the force free second moment are fulfilled. Moreover, the generalized coefficients  $K_\alpha$  and  $\eta_\alpha$  have been derived as time-rescaled versions of their Brownian analogues. The stationary solution  $W_{st}(x) \equiv \lim_{t \rightarrow \infty} W(x,t)$  defined through  $\partial W / \partial t = 0$  is given through the Gibbs–Boltzmann form  $W_{st}(x) = N \exp(-\Phi(x) / [k_B T])$ , hence the FFPE (eq 49) is close to thermal equilibrium.<sup>40–42</sup>

For the investigation of the fractional sedimentation process, we assume the gravity field  $F(x) = -mg$  so that the corresponding fractional Fokker–Planck equation reads

$$\frac{\partial W}{\partial t} = {}_0D_t^{1-\alpha} \left[ \frac{\partial}{\partial x} g^* + K_\alpha \frac{\partial^2}{\partial x^2} \right] W(x,t) \quad (50)$$

where  $g^* \equiv g/\eta_\alpha$ . This is equivalent to eq 36 discussed before.

For the case of diffusion–advection, adequate information on the process can be obtained from the first two moments. Sedimentation processes are, for realistic parameters, much slower and therefore it might be desirable to also measure higher order moments. For the dispersive sedimentation FFPE (eq 50) the moments are readily calculated from the Fourier–Laplace transform

$$W(k,u) = \frac{1}{u + ikg^*u^{1-\alpha} + k^2K_\alpha u^{1-\alpha}} \quad (51)$$

the result being

$$\langle x^m \rangle = (-1)^m \sum_{j=0}^M m! \binom{m-j}{j} \frac{(g^*)^{m-2j} K_\alpha^j t^{\alpha(m-j)}}{\Gamma(1 + \alpha(m-j))} \quad (52)$$

where  $M = [m/2]$  denotes the Landau bracket (the integer part) of  $m/2$ .

Note that the Fourier transform of  $W(x,t)$  is given in terms of the Mittag–Leffler function<sup>14,26</sup>

$$E_\alpha(-z) = \sum_{n=0}^{\infty} \frac{(-z)^n}{\Gamma(1 + \alpha n)} \quad (53)$$

through

$$W(k,t) = E_\alpha(-[k^2K_\alpha + ikg^*]t^\alpha) \sim ([k^2K_\alpha + ikg^*]t^\alpha \Gamma(1 - \alpha))^{-1} \quad (54)$$

which expresses the relaxation of a single Fourier mode  $k$ .

An important measure for sedimentation processes is the speed with which the major part of the tracer is setting down. In analogy to the definitions of the drift and the flank velocities in the case of the velocity advection, two definitions are possible from which we mention the analogue of the drift velocity, the conventional setting velocity which is defined through<sup>8</sup>

$$v_s \equiv \frac{d\langle x \rangle}{dt} \quad (55)$$

In the Brownian case,  $v_s = -g/\eta$ . By the definition of  $\eta$ ,  $\eta \equiv 6\pi a\alpha/m$ , in terms of the radius  $a$  and the mass  $m$  of the tracer particle, the friction constant  $\eta$  behaves proportional to the viscosity  $\alpha$  of the embedding fluid. Thus,  $v_s = -mg/[6\pi a\alpha]$  equals the result from the force equilibrium of a particle whose falling velocity equals the Stokes force:  $mg = 6\pi a\alpha v$ .

In the subdiffusive case,  $v_s$  is only an effective velocity, and it is explicitly time dependent:

$$v_s = -\frac{gt^{\alpha-1}}{\eta_\alpha \Gamma(\alpha)} \quad (56)$$

The setting process is illustrated in Table 2 with some rough estimations for the Brownian and the subdiffusive models, the strong difference between the temporal evolution being manifest especially for longer times.

## V. Conclusions

We have demonstrated that the extension of the classical diffusion–advection framework to anomalous diffusion brings

**TABLE 2. Temporal Growth of the Drift in the Brownian and Subdiffusive Sedimentation Models<sup>a</sup>**

$t$	$d_1(t)/\text{cm}$	$d_{1/2}(t)/\text{cm}$
1 s	$4.7 \times 10^{-5}$	$4.7 \times 10^{-5}$
1 h	0.17	0.0028
1 day	4.1	0.014
1 week	28.4	0.04
1 year	1482	0.26

<sup>a</sup> It was assumed that for very short times, here 1 s, the covered distance is the same for each model. An experimental analysis would start with the measurement of the anomalous diffusion exponent  $\alpha$  and the actual transport coefficient  $\eta_\alpha$  from a field measurement, and then extrapolate the behavior. However, even the chosen fictitious numbers demonstrate the huge difference in the sedimentation, surely a very relevant conclusion for environmental considerations.

about a number of different possible scenarios, these being Galilei invariant or variant. We have collected and discussed some existing models, in an integral presentation. For processes with a mean flight time, we have presented novel results, especially concerning the turnover observable in the mean squared displacement. Moreover, we have suggested some quantitative analysis of fractional advection processes. For the first time, sedimentation processes have been considered, and their proximity to the advection process with mean flight time has been shown.

The presented fractional models are oversimplifying real systems where the layer structure features numerous changes in density, structure, and other parameters in dependence of the localization. Nevertheless, even the models under discussion show the importance of the knowledge about the very nature of the transport process, Brownian or anomalous, and its characteristic quantities such as the anomalous diffusion exponent or the generalized friction and diffusion constants. Depending on their magnitude, the resulting transport process might show huge deviations from a classical description in terms of Brownian motion. We believe that our analysis may contribute to the demanding field of the study of the groundwater dynamics and other realizations of advection problems in complex systems.

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## Appendix A: the Fox Function and its Representation in Computable Form

The Fox function<sup>48,49</sup> is defined through the Mellin–Barnes type integral

$$H_{p,q}^{m,n} \left[ z \left| \begin{matrix} (a_1, A_1), (a_2, A_2), \dots, (a_p, A_p) \\ (b_1, B_1), (b_2, B_2), \dots, (b_q, B_q) \end{matrix} \right. \right] = \frac{1}{2\pi i} \int_L ds \chi(s) z^s \quad (A1)$$

with the integral density:

$$\chi(s) = \frac{\Pi_1^m \Gamma(b_j - B_j s) \Pi_1^n \Gamma(1 - a_j + A_j s)}{\Pi_{m+1}^q \Gamma(1 - b_j + B_j s) \Pi_{n+1}^p \Gamma(a_j - A_j s)} \quad (A2)$$

An  $H$ -function can be expressed as a computable series in the form<sup>48,49</sup>

$$H_{p,q}^{m,n}(z) = \sum_{h=1}^m \sum_{v=a}^{\infty} \frac{\prod_{j=1, j \neq h}^m \Gamma(b_j - B_j(b_h + v)/B_h)}{\prod_{j=m+1}^q \Gamma(1 - b_j + B_j(b_h + v)/B_h)} \times \frac{\prod_{j=1}^n \Gamma(1 - a_j + A_j(b_h + v)/B_h)}{\prod_{j=n+1}^p \Gamma(a_j - A_j(b_h + v)/B_h)} \frac{(-1)^v z^{(b_h+v)/B_h}}{v! B_h} \quad (A3)$$

For large argument  $|z| \rightarrow \infty$ , the Fox functions can be expanded as a series over the residues

$$H_{p,q}^{m,n}(z) \sim \sum_{v=0}^{\infty} \text{res}(\chi(s)z^s) \quad (A4)$$

to be taken at the points  $s = (a_j - 1 - v)/A_j$ , for  $j = 1, \dots, n$ .

The series expansion shows a rather slow convergence in numerical evaluations. If  $\alpha$  is a rational number, the Fox function from eq 19 can be reduced to a Meijer  $G$ -function which is implemented in Mathematica.<sup>44</sup> For  $\alpha = 1/2$ , the corresponding result reads

$$W(x,t) = \frac{1}{\sqrt{2\pi^2 K_{1/2} t^{1/2}}} H_{0,2}^{2,0} \left[ \frac{x^2}{8K_{1/2} t^{1/2}} \middle| (0,1), \left(\frac{1}{4}, \frac{1}{2}\right) \right] = \frac{1}{\sqrt{8\pi^3 K_{1/2} t^{1/2}}} G_{0,3}^{3,0} \left[ \left(\frac{x^2}{16K_{1/2} t^{1/2}}\right)^2 \middle| 0, \frac{1}{4}, \frac{1}{2} \right] \quad (A5)$$

by twice using the duplication formula of the Gamma function in the Mellin–Barnes type integral (A1) defining the Fox function.<sup>48,49</sup> The corresponding notation in Mathematica is

$$\frac{1}{\sqrt{8\pi^3 t^{1/2}}} G_{0,3}^{3,0} \left[ \left(\frac{x^2}{16t^{1/2}}\right)^2 \middle| 0, \frac{1}{4}, \frac{1}{2} \right] = 1/(8\pi^{3r(1/2)}) \text{MeijerG}[\{\{ \}, \{ \}, \{0,1/4,1/2\}, \{ \}, \{ \}, x^4/(16^{2r(1/2)})\}] \quad (A6)$$

**References and Notes**

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 (51) Both definitions are, up to a factor of absolute value 1, identical in the one-dimensional case.  
 (52) Note the additional imaginary factor introduced here for convenience.