Reaction-subdiffusion on moving fluids.

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To capture the dynamic behaviors of reaction-subdiffusion in flow fields, in the present paper we analyze a simple monomolecular conversion \(A \rightarrow B\). We derive the corresponding master equations for the distribution of \(A\) and \(B\) particles in continuous time random walks scheme. The new results are then used to obtain the generalizations of advection-diffusion reaction equation, in which the diffusion and advection operators both depend on the reaction rate.

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Reactive transport in flows is an important issue of diffusion theory that has a variety of applications in many topics such as the transport of contaminants in underground water \cite{1}, nuclear waste storage \cite{2}, etc. The macroscopic description of reaction-diffusion in a velocity field is the standard advection-diffusion reaction equation (ADRE) defined in one-dimensional form as:

\[
\frac{\partial C(x,t)}{\partial t} + v \frac{\partial C(x,t)}{\partial x} = K \frac{\partial^2 C(x,t)}{\partial x^2} + f
\]

(1)

where \(C(x,t)\) is the probability density function (PDF) of the particle, \(v\) is constant velocity, \(K\) is diffusion coefficient, and \(f\) denotes the decoupled reaction term.

In recent years the reaction under anomalous diffusion has attracted more and more attention \cite{2}. One of the effective ways to capture anomalous diffusion is continuous time random walk (CTRW) model \cite{3,4,5,6}. By using CTRW Sokolov et al. analyzed reaction-subdiffusion schemes for the monomolecular conversion \(A \rightarrow B\), derived the corresponding kinetic equations for local \(A\) and \(B\) concentrations, and first argued that reaction-subdiffusion equations are not obtained by a trivial change of the diffusion operator for a subdiffusion one \cite{3}. Such nontrivial coupled effect was also found in some other reaction-subdiffusion systems \cite{4,5,6,7}, for instance, on the form of stationary solutions for reaction-diffusion in finite domains (see Ref. \cite{8}).

However, up to now few works have approached the reaction under anomalous diffusion in nonhomogeneous flows \cite{12,14}. Without considering the effect of chemical reactions, Compte \cite{13,15,16} has discussed anomalous diffusion in a nonhomogeneous convection velocity field by applying the CTRW techniques in which the step length distribution function depends on the starting point of the jump, and showed that convection coefficient depends on the waiting time statistics. But to our knowledge, the more basic and interesting problem of how nonhomogeneous velocity field might affect the reaction under anomalous diffusion is still unknown. This is what we shall address in this paper. In what follows we will consider the reaction-subdiffusion process on moving fluid for the reaction \(A \rightarrow B\) in CTRW scheme, then derive the generalizations of advection-diffusion reaction equation and show the interesting coupling relations between the velocity field and the chemical reactions under subdiffusion.

We start by recalling the CTRW model on inhomogeneous flows in one-dimensional case \cite{13,15}. In this model, the jump length \(y\) for the moving particle is dragged along the velocity \(v(x)\) and replaced by \(y/\tau_a v(x)\) where \(\tau_a\) stands for an advection time scale, and \(\tau_a v(x)\) is the mean drag experienced by a particle jumping from the point \(x\). Thus, the particle jumps from \(x\) to \(x+y\) with the jump length PDF \(\lambda(y/\tau_a v(x))\), and then waits at \(x+y\) for time \(t\) drawn from \(\psi(t)\), after which the process is renewed.

We then consider the simplest reaction scheme \(A \rightarrow B\) in this CTRW model. We assume all properties of \(A\) and \(B\) particles are the same and the particles trapped in stagnant regions will react with a relabeling of \(A\) into \(B\) taking place at a rate \(\lambda\). Let \(A(x,t)\) be the PDF of \(A\) particle being in point \(x\) at time \(t\) and \(i^-(x,t)\) the escape rate. By assuming that in the initial distribution all particles have zero resting times, we can find the balance equation for \(A\) particles in a given point:

\[
A(x,t) = A_0(x)\Psi(t)e^{-\alpha t} + \int_{-\infty}^{+\infty} dx' \int_0^t d\tau \int_0^\tau d\tau' i^-(x',\tau')
\]

\[
\lambda(x-x'-\tau_a v(x'))\Psi(t-\tau)e^{-\alpha(t-\tau)}dt'
\]

(2)

A particle being in point \(x\) at time \(t\) is renewed.

where \(A_0(x)\) is the initial state of \(A\) particle, \(\Psi(t)e^{-\alpha t} = (1 - \int_0^t \psi(\tau)d\tau)e^{-\alpha t}\) is the joint survival density of remaining at least at time \(t\) on the spot (without being converted into \(B\)). The density is a sum of outgoing particles from all other points at different times given by the flow, and provided they survived after their arrival till the time \(t\). The first term on the right hand side is just the influence of the initial distribution.

The above equation (2) can be changed to the form

\[
A(x,t) = A_0(x)\Psi(t)e^{-\alpha t} + \int_{-\infty}^{+\infty} dx' \int_0^t i^-(x',\tau')
\]

\[
\phi(x-x',t-t';x')e^{-\alpha(t-t')}dt'
\]

(3)

by using the expression \(\phi(r,\tau;x) = \lambda(r-\tau_a v(x))\Psi(\tau)\).

Fourier transforming \(x \rightarrow k\) and Laplace trans-
forming $t \to u$ of Eq.(3), we obtain

$$A(k, u) = A_0(k)\Psi(u+\alpha) + \int i^-(k', u)\phi(k, u+\alpha; k-k')dk'.$$

(4)

Here, $A_0(k)$ represents the Fourier $x \to k$ transform of the initial condition $A_0(x)$, $\Psi(u+\alpha)$ denote the Laplace transform of joint survival PDF $\Psi(t)e^{-\alpha t}$, $i^-(k, u)$ is the Fourier-Laplace transform of $i^-(x, t)$, and

$$\phi(k, u+\alpha; k-k') = \Psi(u+\alpha)\lambda(k) \int e^{-ik\tau_v(x')}e^{-i(k-k')x'}dx'. (5)$$

To obtain the master equation with respect to $A(x, t)$, we shall give the other balance equation. Noticing that the loss flux is from those particles that were originally at $x$ at $t = 0$ and wait without reacting until time $t$ to leave, and those particles that arrived at an earlier time $t'$ and wait without reacting until time $t$ to leave, we have the second balance equation:

$$i^-(x, t) = A_0(x)\psi(t)e^{-\alpha t} + \int_{t-i}^{t+i}i^-(x', t') \times \lambda(x-x' - \tau_v(x'))\psi(t-t')e^{-\alpha(t-t')}dt'$$

(6)

where $\psi(t)e^{-\alpha t}$ is the non-proper waiting time density for the actually made new step provided the particle survived $\Psi$. By introducing

$$\eta(r, \tau; x) = \lambda(r - \tau_v(x))\psi(\tau)$$

and applying the transform $(x, t) \to (k, u)$ of Eq.(6), we find

$$i^-(k, u) = A_0(k)\psi(u+\alpha) \int i^-(k', u)\eta(k, u+\alpha; k-k')dk'$$

(7)

where the term $\eta(k, u+\alpha; k-k') = \psi(u+\alpha)\lambda(k) \int e^{-ikt\tau_v(x')}e^{-i(k-k')x'}dx'$. We divide Equation (4) by (7) to write

$$i^-(k, u) = \frac{\psi(u+\alpha)}{\Psi(u+\alpha)}A(k, u)$$

(8)

Nothing that $\Psi(u+\alpha) = \frac{1-\psi(u+\alpha)}{u+\alpha}$, we get

$$i^-(k, u) = \Phi_\alpha(u+\alpha)A(k, u)$$

(9)

where $\Phi_\alpha(u+\alpha) = \frac{(u+\alpha)\psi(u+\alpha)}{1-\psi(u+\alpha)}$, which recovers the relation between $A(x, t)$ and $i^-(x, t)$ when the effect of the flow field is not considered in Ref.[3]. Inverting Eq.(9) to the space-time domain $k \to x$, $s \to t$, we obtain

$$i^-(x, t) = \int_0^t \Phi_\alpha(t-t')A(x, t')dt'. (10)$$

Here, the kernel $\Phi_\alpha(t)$ is equal in laplace $t \to u$ space to $\Phi_\alpha(u+\alpha)$. When $\alpha = 0$, it reduces to the usual memory kernel of master equation for CTRW [3,17,18].

We now consider a linear velocity field $v(x) = \omega x$ where $\omega$ is a constant. Then Eq.(4) becomes

$$A(k, u) = \Psi(u+\alpha)A_0(k) + \Psi(u+\alpha)\lambda(k)j(k+v_k, u)$$

(11)

where the symbol $\lambda_k = \tau_\alpha\omega k$. In the limit $\tau_\alpha \to 0$, Eq.(11) gives

$$A(k, u) \simeq \Psi(u+\alpha)A_0(k) + \Psi(u+\alpha)\lambda(k) \times (i^-(k, u) + v_k i^-'(k, u)).$$

(12)

We substitute (9) into (12) and get

$$A(k, u) = \Psi(u+\alpha)A_0(k) + \Psi(u+\alpha)\lambda(k) \times (A(k, u) + v_k A_k^\prime(k, u)).$$

(13)

This simplifies further to the generalized master equation in Fourier-Laplace space for A particles in an $A \to B$ reaction under subdiffusion on linear moving fluid

$$[1-\psi(u+\alpha)\lambda(k)]A(k, u) = v_k \psi(u)\lambda(k)A_k^\prime(k, u) + \Psi(u)$$

(14)

for the CTRW on linear moving fluids in one-dimensional lattice obtained by Compte in Ref.[15].

There is the other way to derive the generalized master equation (14) where the balance condition (3) is replaced by [3]

$$\frac{\partial A(x, t)}{\partial t} = i^+(x, t) - i^-(x, t) - \alpha A(x, t).$$

(16)

Here, $i^+(x, t)$ is the gain flux which can be represented by the loss flux [18]

$$i^+(x, t) = \int_{-\infty}^{+\infty} i^-(x', t)\lambda(x-x' - \tau_v(x'))dx'.$$

(17)

Transforming $(x, t) \to (k, u)$ of (16), one has

$$uA(k, u) - A_0(k) = \frac{\int i^-(k', u)\eta(k, u+\alpha; k-k')dk'}{\psi(u+\alpha)} - i^-(k, u) - \alpha A(k, u)$$

(18)

By using (7) and (18), we can also obtain the relation equation (9). Substitute Eq.(9) into Eq.(18) and assume $\psi(x) = \omega x$, in the limit $\tau_\alpha \to 0$, and we find

$$uA(k, u) - A_0(k) \simeq (\lambda(k)\Phi_\alpha(u) - \Phi_\alpha(u))A(k, u) + \frac{\lambda(k)}{\Phi_0(u)\tau_\alpha A_k^\prime(k, u) - \alpha A(k, u)}$$

(19)

Using $\Phi_\alpha(u+\alpha) = \frac{(u+\alpha)\psi(u+\alpha)}{1-\psi(u+\alpha)}$ and $\Psi(u+\alpha) = \frac{1-\psi(u+\alpha)}{u+\alpha}$, one finally recovers the generalized master
equation (14). This means that the two approaches to derive the generalized master equation for A-particles in reaction-subdiffusion process on moving fluid are equivalent. In what follows we will not distinguish them for

\[
\frac{\partial A(x,t)}{\partial t} + \tau_a \int_{-\infty}^{+\infty} dx' \int_0^t \Phi_\alpha(t-t') \lambda(x-x') \frac{\partial \nu(x')A(x',t')}{\partial x'} dt' = \int_{-\infty}^{+\infty} dx' \int_0^t \Phi_\alpha(t-t') \lambda(x-x') - \delta(x-x')A(x',t') dt' - \alpha A(x,t),
\]

where the reaction rate explicitly affects both the transport term and the advection term. Specifically, we consider a discrete random walk, where the PDF of particles A on site \(x = i\) at time \(t\) is denoted as \(A(i,t)\), and the jump PDF is assumed to be \(\lambda(-1) = \frac{1}{\tau}, \lambda(1) = \frac{1}{\tau}\), meaning that the particle can jump from \(x = i\) to the adjacent grid point, to the right and left directions with the same jump length variance, respectively. Assuming \(\nu(x) = \delta(x)\), substituting \(\Phi_\alpha(u) = \frac{1}{\tau(1-(\frac{1}{\tau}))} (u + \alpha)^{1-\beta}\) into the master equation (19), in the limit of \(\tau \to 0, \sigma \to 0\) and \(\tau_a \to 0\), we obtain

\[
\frac{\partial A(i,t)}{\partial t} = \int_0^t \Phi_\alpha(t-t') \frac{1}{2} A(i-1,t) + \frac{1}{2} A(i+1,t) - A(i,t) dt' - \alpha A(i,t),
\]

obtained by Sokolov in Ref. [3].

We now turn to apply the master equation (19) to derive a ADRE for Gaussian jump length \(\lambda(k) \sim 1 - \frac{\alpha^2 k^2}{2}\) and long-tailed waiting time \(\psi(u) \sim 1 - \Gamma(1-\beta)(\tau u)^\beta\) with \(\tau\) and \(\sigma^2\) being the appropriate time scale and the jump length variance, respectively. Assuming \(\rho_0(x) = \delta(x)\), we then get the generalized ADRE for A-particles in reaction-subdiffusion process on moving fluid are equivalent. In what follows we will not distinguish them for

\[
\frac{\partial A(x,t)}{\partial t} + \tau_a \int_{-\infty}^{+\infty} dx' \int_0^t \Phi_\alpha(t-t') \lambda(x-x') \frac{\partial \nu(x')A(x',t')}{\partial x'} dt' = \int_{-\infty}^{+\infty} dx' \int_0^t \Phi_\alpha(t-t') \lambda(x-x') - \delta(x-x')A(x',t') dt' - \alpha A(x,t),
\]

and becomes a fractional derivative when \(\alpha = 0\). It should be noted that in the generalized ADRE (23) not only diffusion but also advection term depend on the reaction parameter \(\alpha\).

Analogously we shall now derive the generalized ADRE for the B-particles. Let \(B(x,t)\) be the PDF of B particle being in point \(x\) at time \(t\), \(j^+(t)\) be the gain flux and \(j^-(t)\) be the loss flux of particles B at site \(x\) at \(t\). Noting that B-particle that is at (or leaves) site \(x\) at time \(t\) has come there as a B-particle at some prior time or was converted from an A-particle that either was on site \(x\) from the very beginning or arrived there later at \(t' > 0\), and still keeps at (or just leaves) the site \(x\) at time \(t\), we give the following balance equations:

\[
B(x,t) = \int_{-\infty}^{+\infty} dx' \int_0^t j^-(x',t') \phi(x-x',t-t';x') dt' + \int_{-\infty}^{+\infty} dx' \int_0^t j^+(x',t') \phi(x-x',t-t';x') (1 - e^{-\alpha(t-t')} dt' + A_0(x) \psi(t)(1 - e^{-\alpha t}),
\]

and

\[
j^-(x,t) = \int_{-\infty}^{+\infty} dx' \int_0^t j^-(x',t') \eta(x-x',t-t';x') dt' + \int_{-\infty}^{+\infty} dx' \int_0^t j^+(x',t') \eta(x-x',t-t';x') (1 - e^{-\alpha(t-t')} dt' + A_0(x) \psi(t)(1 - e^{-\alpha t}),
\]

with the initial condition \(A_0(x) = \delta(x)\). Here, The integral operator \(\tilde{T}_i(1-\beta,\alpha)f = \tau^\beta \Gamma(1-\beta) \int_0^t \Phi_\alpha(t-t') dt'\) corresponds in time domain to

\[
\tilde{T}_i(1-\beta,\alpha)f = \frac{d}{dt} \int_0^t \frac{e^{-\alpha(t-t')}}{(t-t')^{1-\beta}} f(t') dt' + \alpha \int_0^t \frac{e^{-\alpha(t-t')}}{(t-t')^{1-\beta}} f(t') dt'
\]

and becoming of the two equations (20) to the space-time domain:

\[
\tilde{\Phi}_\alpha(t-t',x) = \frac{\partial}{\partial t} \int_{-\infty}^{+\infty} dx' \int_0^t \Phi_\alpha(t-t') \lambda(x-x') \frac{\partial \nu(x')A(x',t')}{\partial x'} dt' = \int_{-\infty}^{+\infty} dx' \int_0^t \Phi_\alpha(t-t') \lambda(x-x') - \delta(x-x')A(x',t') dt' - \alpha A(x,t),
\]

and becomes a fractional derivative when \(\alpha = 0\). It should be noted that in the generalized ADRE (23) not only diffusion but also advection term depend on the reaction parameter \(\alpha\).

Analogously we shall now derive the generalized ADRE for the B-particles. Let \(B(x,t)\) be the PDF of B particle being in point \(x\) at time \(t\), \(j^+(t)\) be the gain flux and \(j^-(t)\) be the loss flux of particles B at site \(x\) at \(t\). Noting that B-particle that is at (or leaves) site \(x\) at time \(t\) has come there as a B-particle at some prior time or was converted from an A-particle that either was on site \(x\) from the very beginning or arrived there later at \(t' > 0\), and still keeps at (or just leaves) the site \(x\) at time \(t\), we give the following balance equations:

\[
B(x,t) = \int_{-\infty}^{+\infty} dx' \int_0^t j^-(x',t') \phi(x-x',t-t';x') dt' + \int_{-\infty}^{+\infty} dx' \int_0^t j^+(x',t') \phi(x-x',t-t';x') (1 - e^{-\alpha(t-t')} dt' + A_0(x) \psi(t)(1 - e^{-\alpha t}),
\]

and

\[
j^-(x,t) = \int_{-\infty}^{+\infty} dx' \int_0^t j^-(x',t') \eta(x-x',t-t';x') dt' + \int_{-\infty}^{+\infty} dx' \int_0^t j^+(x',t') \eta(x-x',t-t';x') (1 - e^{-\alpha(t-t')} dt' + A_0(x) \psi(t)(1 - e^{-\alpha t}),
\]
tions (25) and (26) yields:

\[ B(k, u) = \int j^-(k', u)\phi(k, u; k - k')dk' + \int i^-(k', u) \times [\phi(k; u; k - k') - \phi(k, u + \alpha; k - k')]dk' + (\Psi(u) - \Psi(u + \alpha))A_0(k), \] (27)

and

\[ j^-(k, u) = \int j^-(k', u)\eta(k, u; k - k')dk' + \int i^-(k', u) \times [\eta(k; u; k - k') - \eta(k, u + \alpha; k - k')]dk' + (\psi(u) - \psi(u + \alpha))A_0(k). \] (28)

Comparing (4), (7), (27) and (28), one has

\[ \frac{A(k, u) + B(k, u)}{\Psi(u)} = \frac{i^-(k, u) + j^-(k, u)}{\psi(u)}, \] (29)

which can be changed to the form:

\[ j^-(k, u) = \Phi_0(u)B(k, u) + (\Phi_0(u) - \Phi_0(\alpha(u)))A_0(k, u). \] (30)

When \( v(x) = 0 \) it is consistent with the result obtained in [3].

If the balance equation (25) is replaced by

\[ \frac{\partial B(x, t)}{\partial t} = \int_{-\infty}^{+\infty} j^-(x', t)\lambda(x - x' - \tau_a v(x'))dx' - j^-(x, t) + \alpha A(x, t), \] (31)

we can also find Eq.(30) by using (7), (18), (28), the transform \((x, t) \rightarrow (k, u)\) of Eq.(31)

\[ B(k, u) = \frac{\int j^-(k', u)\eta(k, u + \alpha; k - k')dk'}{\psi(u + \alpha)} - j^-(k, u) + \alpha A(k, u). \] (32)

and the relation \( \Psi(u) = \frac{1 - \psi(u)}{u} \). It means that the two ways using different balance conditions for B-particles are equivalent, too.

In linear flow \( v(x) = \omega x \) Eq.(32) can be written in the form:

\[ B(k, u) \approx \lambda(k)j^-(k + \omega k, u) - j^-(k, u) + \alpha A(k, u) \] (33)

for small \( \tau_a \). Substitution of (29) into Eq.(33) gives the master equation for the PDF of B-particle in Fourier-Laplace space:

\[ uB(k, u) = \lambda(k)v_kB'_k(k, u)\Phi_0(u) + \lambda(k)v_kA'_k(k, u) + (\Phi_0(u) - \Phi_0(\alpha(u)) + (\lambda(k) - 1)B(k, u)\Phi_0(u) + (\lambda(k) - 1)A(k, u)(\Phi_0(u) - \Phi_0(\alpha(u)) + \alpha A(k, u). \] (34)

Inverting the above equation to the space-time domain, we obtain the master equation in space-time domain:

\[ \frac{\partial B(x, t)}{\partial t} + \tau_a \int_{-\infty}^{+\infty} dx' \int_{0}^{t} \Phi_0(t - t')\lambda(x - x') \frac{\partial \phi(x', \theta)}{\partial \phi} dt' + \alpha A(x, t), \] (35)

For a discrete random walk with \( v(x) = 0 \), and the jump PDF satisfies \( \lambda(-1) = \frac{1}{\tau}, \lambda(1) = \frac{1}{2} \), in the continuum limit, Eq.(35) reduces to Eq.(27) in Ref. [3]. If we substitute the Gaussian jump length \( \lambda(k) \sim 1 - \frac{k_2}{2} \) and power law waiting time PDF \( \psi(u) \sim 1 - \Gamma(1 - \beta) / (\tau u)^{\beta} \) into the master equation (34), and invert \( k \rightarrow x, u \rightarrow t \), in the limit of small \( \tau_a, \tau, \) and \( \alpha \), we then find the generalized master equation for B-particle in the reaction \( A \rightarrow B \) under subdiffusion in linear velocity field

\[ \frac{\partial B(x, t)}{\partial t} + C_\beta \alpha D_t^{1-\beta} B(x, t) \frac{\partial (v(x)A(x, t))}{\partial x} + C_\alpha \cdot 0 D_t^{1-\beta} B(x, t) \frac{\partial (\phi(v(x))B(x, t))}{\partial x} - K_\beta \cdot 0 D_t^{1-\beta} B(x, t) \frac{\partial^2 (A(x, t))}{\partial x^2} + \alpha A(x, t), \] (36)

It should be noted that in (36) both advection and diffusion terms for B-particle couple with the reaction too, since these two terms respectively include not only the advection term \( C_\beta T_l(1 - \beta, \alpha)\frac{\partial (v(x)A(x, t))}{\partial x} \) and diffusion term \( K_\beta T_l(1 - \beta, \alpha) \frac{\partial^2 A(x, t)}{\partial x^2} \) in (23), but also the fractional kinetics memories at previous times for A-particle.

Let \( C(x, t) \) denote the sum of \( A(x, t) \) and \( B(x, t) \). Combining (23) with (36), one has

\[ \frac{\partial C(x, t)}{\partial t} + C_\beta \cdot 0 D_t^{1-\beta} \frac{\partial (v(x)C(x, t))}{\partial x} + C_\alpha \cdot 0 D_t^{1-\beta} \frac{\partial (\phi(v(x))C(x, t))}{\partial x} = K_\beta \cdot 0 D_t^{1-\beta} \frac{\partial^2 C(x, t)}{\partial x^2} \] (37)

which is consistent with the result for subdiffusion in non-reactive liquid in [13, 14]. It is because that the simple reaction we discuss here does not change the sum of the particles in the system.
To sum up we derive the master equations (20) and (35) for the PDF of A and B particles in a simple monomolecular conversion reaction $A \rightarrow B$ taking place at a constant rate $\alpha$ and under subdiffusion in linear flows. As examples, two generalized advection-diffusion reaction equations (23) and (36) are obtained, and the interesting couple relations among diffusion, advection and reaction process are showed. There are problems such as the dynamic behaviors for more complex reaction under subdiffusion in moving fluids are still unknown.

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