A connection between the Lagrangian stochastic–convective and cumulant expansion approaches for describing solute transport in heterogeneous porous media

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The equation describing the ensemble-average solute concentration in a heterogeneous porous media can be developed from the Lagrangian (stochastic–convective) approach and from a method that uses a renormalized cumulant expansion. These two approaches are compared for the case of steady flow, and it is shown that they are related. The cumulant expansion approach can be interpreted as a series expansion of the convolution path integral that defines the ensemble-average concentration in the Lagrangian approach. The two methods can be used independently to develop the classical form for the convection–dispersion equation, and are shown to lead to identical transport equations under certain simplifying assumptions. In the development of such transport equations, the cumulant expansion does not require a priori the assumption of any particular distribution for the Lagrangian displacements or velocity field, and does not require one to approximate trajectories with their ensemble-average. In order to obtain a second-order equation, the cumulant expansion method does require truncation of a series, but this truncation is done rationally by the development of a constraint in terms of parameters of the transport field. This constraint is less demanding than requiring that the distribution for the Lagrangian displacements be strictly Gaussian, and it indicates under what velocity field conditions a second-order transport equation is a reasonable approximation. © 1998 Elsevier Science Limited. All rights reserved

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1 INTRODUCTION

Stochastic methods have become indispensable to the development of solute transport equations in heterogeneous porous media. This is due largely to the realization that the variations in the velocity field in the subsurface systems dominate the spreading of solutes as they are transported, and by the fact that the important scales of heterogeneity in the conductivity field (and, hence, the velocity field) cannot be sufficiently resolved by measurement to provide a fully deterministic representation. A variety of stochastic methods have been developed to describe the spreading of solutes from both the Lagrangian and Eulerian perspective. Much of the fundamental work in this area has been reviewed by Dagan13,14, Gelhar27, Cushman11. In particular, the work of Dagan13,14 provides a comprehensive review that describes the development of stochastic transport equations from both of the Eulerian and Lagrangian perspectives.

The Lagrangian approach to stochastic transport was developed in application to turbulent diffusion by Taylor52, and introduced for nonreactive transport in porous media by Dagan and Bresler17 and Simmons49. This approach has been widely applied to subsurface hydrology, and examples of such applications can be found in the work of Dagan15, Neuman and Zhang44, Cushman and Ginn40, Rajaram and
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Gelhar, Cvetkovic and Dagan, Zhang, Neuman, Zhang, Kavvas and Karakas, and Cushman and Hu. In particular, we are interested in the

Examples of these approaches can be found in Winter et al., Kabala and Sposito, Neuman, Zhang, Kavvas and Karakas, and Cushman and Hu. In particular, we are interested in the approach of Kabala and Sposito and more recently Kavvas and Karakas; in both works a cumulant expansion has been adopted for development of an ensemble-average expression for the solute concentration. The cumulant expansion described in this paper is renormalized by the ensemble-average velocity field, and this will be shown to have some advantageous properties.

Direct comparisons among the results for various stochastic methods are often difficult because of inherent differences the techniques that are used. When such comparisons are available, they can be helpful in understanding and interpreting the results that are obtained. The purpose of this paper is to compare the Lagrangian stochastic–convective and renormalized cumulant expansion methods. The Lagrangian stochastic–convective solution for the ensemble-average concentration is expressed as a convolution of the Lagrangian displacement probability density function (pdf) and the initial condition function. The pdf that arises must be determined by measurement, or by making assumptions about the statistics of the Lagrangian displacements. If, under certain simplifying assumptions, the pdf is assumed to be Gaussian, then the conventional convection–dispersion equation can be recovered. We show that the cumulant expansion approach can be considered to be a series expansion of the convolution integral that arises from the Lagrangian stochastic–convective approach. The utility of the cumulant expansion is that it represents the solution in terms of the cumulants of the velocity field directly, without the assumption of a particular form for the pdf. Additionally, a constraint can be developed to indicate under what conditions the expansion can be rationally truncated at second order to yield a closed-form solute transport equation. When this constraint is met, the cumulant expansion also leads to the conventional convection–dispersion equation. This constraint is less restrictive than the requirement that the velocity field be strictly Gaussian, and has the additional advantage of indicating under what conditions the velocity field is close enough to Gaussian such that a second-order truncation reasonably approximates the governing transport equation.

The paper is organized as follows. In Section 2 the problem of pure convection in a random velocity field is set up, and solutions are obtained by the Lagrangian stochastic–convective method and by the cumulant expansion approach. In this section it is shown that the expectation over Lagrangian trajectories leads to a convolution path integral formulation for the Lagrangian stochastic–convective method, and that the renormalized cumulant expansion can be interpreted as a series expansion of this integral. In Section 3 the conventional convection–dispersion equation is developed starting from the results for each of the methods that were developed in Section 2. Here, it is shown that under certain limiting assumptions the two approaches lead to results that are similar in form, and the approximations required for each of the approaches are examined and compared. In Section 4 these results are summarized. Although the importance of local dispersion under certain conditions has been well established, in the interest of simplicity of notation and clarity in the developments, local dispersion is neglected. Neglecting local dispersion is not at all required for use of the renormalized cumulant expansion method (cf. Fox, Kabala and Sposito, Kavvas and Karakas); it is neglected here only because it would complicate the comparison of these two methods without adding much to the meaning. Interested readers can find developments that include the effects of local dispersion from both the Lagrangian stochastic–convective perspective (e.g., Dagan) and the cumulant expansion perspective. A brief discussion of how inclusion of the local dispersion tensor alters the results obtained in this work is provided in Section 4.

2 COMPARISON OF THE METHODS

Although the stochastic–convective approach can be cast in terms of an Eulerian travel time perspective, we will be adopting a Lagrangian approach here. This approach has been described by Simmons and Dagan, and we will derive it from a slightly different perspective. To begin with, we consider the problem of solute transport in a steady flow field. We do not assume that the porosity or water content are necessarily constant; therefore, the velocity field is not in general divergence-free (i.e., \( \nabla \cdot v(x) \neq 0 \)). We are interested mainly in the macroscopic spreading due to the velocity field variations, and as discussed above we will neglect local-scale dispersion for this example (cf. Cvetkovic and Dagan, Simmons, Simmons, Simmons, Simmons). It is assumed that the solute initial distribution is known and that there are no solute fluxes on the boundaries so that the system may be treated as an initial value problem (cf. Dagan). The conservation equations that apply for describing the solute transport and the steady flow of fluid at the Darcy scale can be determined by an appropriate upscaling technique. These conservation equations apply either to either saturated (where \( \theta \) represents the porosity) or unsaturated (where \( \theta \) represents the water content) conditions, and are listed here as

\[
\frac{\partial (\theta c)}{\partial t} + \nabla \cdot (\theta vc) = 0
\]
Connection between Lagrangian and cumulant expansion approaches

Once leads to the solution

Eqn (4) represents an initial value problem, and integrating this system takes the form

\[
x(t) = x(t_0) + \int_{t_0}^t dr \, v[x(r)]
\]  

(5b)

Eqn (5a) is the mathematical statement that the solute concentration does not change in time if one adopts the coordinate system defined by the trajectories, and eqn (5b) defines the trajectories of the velocity field. In the developments that follow, we will make use of the definition

\[
L[x(t)] = \int_{t_0}^t dr \, v[x(r)]
\]  

(6)

where \(L[x(t)]\) represents the displacement trajectory between the initial location \(x(t_0)\) and the location at time \(t\) given by \(x(t)\) (Fig. 1). If we adopt \(x(t)\) as the independent variable, eqn (5) can be solved by specifying the location \(x(t)\), that is fixed at the time and space coordinates for which we want a solution. These coordinates can be specified as an ordered pair \((x, t)\), allowing us to express eqn (5) more compactly as

\[
c(x, t) = f(x - L[x(t)])
\]  

(7a)

\[
x(t_0) = x - L[x(t)]
\]  

(7b)

Some interpretation may be helpful at this point. Eqn (7) can be used to determine the solution of the initial value problem posed by eqn (5) as follows. First, we fix a point in space and time \((x, t)\) for which we want a solution. The velocity field trajectory (the path at particle would follow from its initial location \(x(t_0)\) at time \(t_0\), such that it arrives at the location \(x\) at time \(t\) is traced backwards to the initial time (eqn (7b)). Then, \(c(x, t)\) is given by the initial concentration at this traced back location, as indicated by eqn (7a) (Fig. 1). As a simple example, consider transport in one-dimension with a constant velocity given by \(v\). Let the initial time be set such that \(t_0 = 0\), and the initial location set at \(x = 0\). Then, the displacement expressed by eqn (6) is given by \(L[x,t] = vt\), and the traced-back location is given by \(x(t_0) = x - vt\) for all time. The solution for the concentration specified by eqn (7a) is given by the simple relation \(c(x, t) = f(x - vt)\). Keeping this example in mind will help in interpreting the developments that follow.

To find the expression that governs the ensemble-average concentration, we take the expectation of eqn (7a) to yield

\[
\langle c(x, t) \rangle = \langle f(x - L[x(t)]) \rangle
\]  

(8)

where \(\langle \ldots \rangle\) represents ensemble expectation. Eqn (8) can be interpreted as the average of a Lagrangian quantity, where the average is conducted over the members of the ensemble of velocity fields. Each member of the ensemble represents a different realization for the trajectory \(- L[x(t)]\) that starts from the point located at \(x\) at time \(t\) and traces back to the initial location at time \(t_0\) as indicated in Fig. 2; in the material that follows we will refer to this quantity as the ‘backward’ trajectory or path. The ensemble-average given in eqn (8) represents an average over all possible backward paths to the initial time. Eqn (8) can be re-expressed in the...
form of a path integral or functional integral (Bogoliubov and Shirkov\(^4\), ch. 8; Feynman\(^20\), Feynman and Hibbs\(^21\), ch. 12). In the notation of Feynman this is

\[
\langle c(x, t) \rangle = \int_{\Omega} f(x - L[x(t)]) p(L[x(t)]) \, dL[x(t)]
\]

(9)

where \(\Omega\) represents the space of all possible paths between the deterministic location \(x\) at time \(t\), and the random initial location \(x(t_0)\) at time \(t_0\). The quantity \(p(L[x(t)]) \, dL[x(t)]\) represents the (differential) probability measure associated with a particular trajectory, subject to the normalization condition

\[
\int_{\Omega} p(L[x(t)]) \, dL[x(t)] = 1
\]

(10)

Although not explicitly indicated, the probability functional \(p(L[x(t)])\) may in general also depend upon the location \(x\) (fixed at time \(t\)) from which the trajectory is traced backward. The main difficulty in the use of such functional integrals is that ultimately one needs to be able to describe in what sense the backward trajectories are measurable so that the probability measure can be defined. If the particle trajectories are composed of independent Gaussian displacements, Feynman’s formulation leads immediately to the Wiener measure\(^2\), and for many analyses this approximation will be appropriate. Technical details aside, the formalism of integration in functional spaces has been used in hydromechanics for some time (e.g., Hopf\(^32\)), so we will adopt the perspective of Feynman and Hibbs\(^21\) that “if the problem is not Gaussian, it can at least be formulated and studied using path integrals.”

The path integral in eqn (9) is defined in terms of the trajectories, and according to eqn (5) the concentration along these trajectories is constant. This implies that between any two points fixed at \(x\) at time \(t\) and \(x(t_0)\) at time \(t_0\), it is only the total displacement (rather than the particular path) that influences the integral in eqn (9)
2.2 Renormalized cumulant expansion approach

The cumulant expansion method, pioneered by Kubo\textsuperscript{37}, has been used in the study of multiplicative stochastic processes by physics community for some time\textsuperscript{25–28,53–55,58}. This approach has been applied to subsurface hydrology by Sposito and Barry\textsuperscript{31}, Kabala and Sposito\textsuperscript{33,34}, and Kavvas and Karakas\textsuperscript{36}. Here our objective is to use the cumulant expansion in combination with the definition of an operator exponential to derive a differential equation for transport directly from the ensemble-average solution to the transport problem given by eqn (8).

We begin by noting that in eqn (8) the coordinates of the initial condition function $f(x)$ are translated backwards along trajectories of the velocity field by the amount $-L[x(t)]$. Eqn (8) can be rewritten in terms of an operator exponential whose effect is to translate the function it operates on along trajectories of the velocity field, and takes the form\textsuperscript{56}

$$
 f(x - L[x(t)]) = \exp \left( - \int_{t_0}^{t} dr \, v(x) \cdot \nabla \right) f(x) \quad (14)
$$

The series that appears in the second line of eqn (14) is a Lie series\textsuperscript{26,48}, and the differential operators involved operate on all terms to the right of them. The definition provided by eqn (14) technically requires that a Taylor series for $f(x)$ exists (i.e., $f(x)$ must be analytic). However, we can find analytic functions that approximate most of the discontinuous initial conditions that would be of interest (e.g., a step pulse) as closely as we would like, this presents no real limitation. Eqn (14) can be proved in a straightforward manner by integrating both sides of eqn (3) to determine an implicit solution, and then iterating this solution. Note that in eqn (14) it is the Eulerian velocity that appears in the exponential; the Lie series automatically generates the trajectories from the Eulerian velocity field and its derivatives. One can think of the exponential in eqn (14) as the product of many infinitesimal displacement operators; these displacements generate the Lagrangian particle path in terms of only the Eulerian velocity field. An illustrative example showing how such a trajectory is generated for finite time intervals is provided in Appendix A.

Combining eqn (8) and eqn (14), one can now express the ensemble-average concentration as the expectation of an operator acting on the initial condition

$$
 \langle c(x, t) \rangle = \left( \exp \left( - \int_{t_0}^{t} dr \, v(x) \cdot \nabla \right) \right) f(x) \quad (15)
$$

Here $f(x)$ has been removed from the expectation operator because it is assumed to be a deterministic function. It is clear from eqn (14) how eqn (15) could be expanded in terms of moments by simply applying the expectation operator to each of the terms in the series. However, such an expansion would include terms proportional to powers of $t$ and lead to time secularities; that is, any finite number of terms of the expansion will provide a good approximation to the series for only sufficiently small times. To rectify this problem, we consider a renormalized cumulant expansion that expands eqn (15) in terms of powers of the Lagrangian correlation time of the velocity field perturbations\textsuperscript{32–35}. The advantage of such an expansion is that if the correlation time is small enough, the series is well approximated by the first few terms for all time.

As a first step towards developing a renormalized cumulant expansion, we decompose the velocity field as the sum of a deterministic component and a random component

$$
 v(x) = v_s(x) + \tilde{v}(x) \quad (16)
$$

where $v_s(x)$ is a deterministic velocity field, and $\tilde{v}(x)$ represents a perturbation from the deterministic field. Although $v_s(x)$ can be taken as any deterministic function that proves to be useful, it is often convenient to take it to be the ensemble-average velocity. In this case, eqn (16) becomes

$$
 v(x) = \langle v(x) \rangle + \tilde{v}(x) \quad (17)
$$

Eqn (15) can now be written in terms of this decomposition

$$
 \langle c(x, t) \rangle = \left( \exp \left( - \int_{t_0}^{t} dr [ \langle v(x) \rangle \cdot \nabla + \tilde{v}(x) \cdot \nabla ] \right) \right) f(x) \quad (18)
$$

The operator exponential of the sum can be expressed as the product of exponentials. Because in eqn (18) the operators do not in general commute, the exponential is somewhat more complicated than if only scalar functions were involved. However, there is a theorem that allows one to express the exponential of a sum of operators as the product of time-ordered exponentials\textsuperscript{19,40,41}. In Appendix B we provide a simple development that proves this theorem for continuous velocity fields, and the following identity results (cf. Feynman\textsuperscript{19}, Masani\textsuperscript{40,41}, van Kampen\textsuperscript{55})

$$
 \exp \left[ - (t - t_o) \langle v(x) \rangle \cdot \nabla \right] - \exp \left[ - (t - t_o) \tilde{v}(x) \cdot \nabla \right] = \exp \left[ - (t - t_o) \tilde{v}(x) \cdot \nabla \right] \exp \left[ - \int_{t_0}^{t} dr \exp \left[ (t - t_o) \tilde{v}(x) \cdot \nabla \right] f(x) \cdot \nabla \right] - \exp \left[ - (t - t_o) \langle v(x) \rangle \cdot \nabla \right] \quad (19)
$$

Here, \textsuperscript{T} indicates that the exponential is time-ordered\textsuperscript{19,24,37,53}. In this case, the ordering symbol indicates that operators with the earliest time index (i.e., $\tau$ closest to $t_o$) occur to the far right in positional notation, whereas the operators with the latest time index are to the far left. The time-ordered exponential can be expanded in the same manner as an ordinary exponential, with the prescription that each of the terms in the expansion must be ordered as dictated by the ordering symbol.
To simplify the notation, in the subsequent developments we set
\[
U^{(i)}_+(t, \tau) = \exp([t - \tau]T_+(x) V) \tag{20a}
\]
\[
U^{(i)}_-(t, \tau) = \exp(-[t - \tau]T_-(x) V) \tag{20b}
\]
where the explicit dependence on space has been suppressed. These operators have the effect of shifting the functions or operators that they act upon along the trajectories of the ensemble-average velocity (either forward or backward depending on which of the two operators acts) and are often referred to as propagators.\(^2,56\) It is important to note that although \(V(x)\) is the Eulerian velocity field perturbation, the action of the propagator \(U^{(i)}_\pm\) is to shift \(V(x)\) forward along the trajectory formed by the ensemble-average velocity field, and this will ultimately render it Lagrangian.

With the identity provided by eqn (19) and the definitions provided by eqn (20a) and eqn (20b), the solution given in eqn (18) can be rewritten in the form
\[
\begin{align*}
\langle x(t, \tau) \rangle &= U^{(0)}_+(t, \tau) \int d\tilde{\gamma}(x) \left[ \frac{\exp[-\int d\tilde{\gamma}^{(0)}_+(t, \tau) \tilde{V}(x)]}{\det(\tilde{V}(x))} \right]
\end{align*}
\]
\[
\nabla U^{(0)}_+(t, \tau) / f(x) \tag{21}
\]
A similar expression has been derived by Kubo and Hashitsume\(^39\) for a Brownian motion model of atomic spins. Eqn (21) has essentially the same form as eqn (15); the difference is that eqn (21) is renormalized in the sense that the ensemble mean velocity field action has been removed from the random velocity field. The only random quantities left in eqn (21) are the velocity perturbations from the ensemble-mean velocity field. Much like eqn (15), an expansion of the exponential followed by the application of the averaging operator at this point will lead to an expansion that has secular terms. This problem can be rectified by considering an expansion in terms of time-ordered cumulants. A cumulant expansion can be considered a rearrangement of the terms in a moment expansion that improves the series convergence. The expansion has been studied extensively in the physics literature, and we can define the time-ordered cumulant expansion\(^24,37,53\) by the following expression
\[
\begin{align*}
\langle x(t, \tau) \rangle &= U^{(0)}_+(t, \tau) \int d\tilde{\gamma}(x) \left[ \frac{\exp[-\int d\tilde{\gamma}^{(0)}_+(t, \tau) \tilde{V}(x)]}{\det(\tilde{V}(x))} \right]
\end{align*}
\]
\[
\nabla U^{(0)}_+(t, \tau) / f(x) + \int d\tilde{\gamma} d\tilde{\xi} U^{(0)}_+(t, \tau \eta) \tilde{V}(x) \nabla U^{(0)}_+(t, \tau \eta) + \cdots \tag{22}
\]
where \(\langle \cdot \cdot \rangle\) defines the cumulant operator. The relationship between the cumulants and the moments can be found by expanding the right-hand sides of eqn (21) and eqn (22) and equating terms of the same power. The general relationship between time-ordered cumulants and moments for all orders have been presented by van Kampen\(^5\) and Fox\(^24\); here we list only the first two such relations. First, let
\[
B(t, \tau) = U^{(0)}_+(t, \tau) \tilde{V}(x); \nabla U^{(0)}_+(t, \tau) \tag{23}
\]
where the dependence on space has been suppressed. Then the first two cumulants can be expressed in terms of moments by
\[
\begin{align*}
\langle B(t, \tau) \rangle &= \langle B(t, \tau) \rangle \tag{24a}
\end{align*}
\]
\[
\begin{align*}
\langle B(t, \tau) B(t, \eta) \rangle &= \langle B(t, \tau) B(t, \eta) \rangle 
\end{align*}
\]
\[
- \langle B(t, \tau) B(t, \eta) \rangle \tag{25b}
\]
Noting that for our choice of decomposition given in eqn (17), we must have that \(\langle \tilde{V}(x) \rangle = 0\), so that eqn (22) can be rewritten
\[
\begin{align*}
\langle x(t, \tau) \rangle &= U^{(0)}_+(t, \tau) \int d\tilde{\gamma}(x) \left[ \frac{\exp[-\int d\tilde{\gamma}^{(0)}_+(t, \tau) \tilde{V}(x)]}{\det(\tilde{V}(x))} \right]
\end{align*}
\]
\[
\nabla U^{(0)}_+(t, \tau) / f(x) + \int d\tilde{\gamma} d\tilde{\xi} U^{(0)}_+(t, \tau \eta) \tilde{V}(x) \nabla U^{(0)}_+(t, \tau \eta) + \cdots \tag{26}
\]
Eqn (12) and eqn (26) were both developed directly from eqn (8), and must therefore represent the same solution. However, in eqn (26) we obtain an expansion in terms of the velocity perturbations rather than a convolution over the displacement pdf. Note that to this point no assumptions regarding the statistics of the underlying velocity field have been imposed; therefore, this expression applies equally well to stationary and nonstationary velocity fields.

### 3 DEVELOPMENT OF THE CONVENTIONAL CONVECTION–DISPERSION EQUATION

The discussion above suggests that the stochastic–convective result (eqn (12)) and the renormalized cumulant expansion result (eqn (26)) are equivalent. In eqn (12) one must know the Lagrangian displacement pdf a priori to solve for the ensemble-average concentration. In eqn (26) the solution for the ensemble-average concentration is expressed not by the pdf, but by an infinite series of cumulants of the convection operator perturbations. To make the connection between the two solutions more explicit, we will derive the classical differential equation for transport (convective–dispersion equation) in a saturated porous media for steady-state flow conditions from both perspectives. We will begin from eqn (8) for the stochastic–convective approach, and from eqn (26) for the cumulant expansion approach to show that, under the appropriate conditions, the same transport equation can be derived from either starting point. This will allow us to directly compare the results, and carefully examine the assumptions that are imposed in both approaches. The development starting from eqn (8) will parallel the developments of Dagan\(^3,15\), and will include
the assumptions that are made in that development. The first such assumption is that the porosity (θ) of eqn (2) is treated as a constant, and this implies that θ = 0.

3.1 Lagrangian stochastic–convective development

Some of the techniques used above in the development of the renormalized cumulant expansion can also be employed to derive the transport equation from the conventional Lagrangian perspective. We begin with eqn (8), and express the translation of the initial condition in terms of a Taylor series in the Lagrangian displacements L[x(t)]. The result is

\[ c(x, t) = f(x) - (L[x(t)]) \cdot \nabla f(x) + \frac{1}{2!} [L[x(t)][L[x(t)]]] \cdot \nabla^2 f(x) + \ldots \]

(27)

where LL·∇ is Gibbs notation for the contraction

\[ L_i L_j \frac{\partial}{\partial x_i} \frac{\partial}{\partial x_j} \]

Eqn (27) can be expressed in a notation similar to that used in eqn (15)

\[ \frac{\partial c(x, t)}{\partial t} = \left\{ \exp \left[ -L[x(t)] \cdot \nabla \right] \right\} f(x) + \frac{1}{2!} \left\{ \exp \left[ -L[x(t)] \cdot \nabla \right] \right\} \cdot \nabla^2 f(x) + \ldots \]

(28)

However, there is a crucial difference between the previous representation given in eqn (15) and the representation given in eqn (28). In eqn (15) it is the Eulerian velocity that appears in the exponential; the trajectories are generated automatically by the group property of the convection operator. In that formulation, the exponential is expanded in a Lie series where the operator acts iteratively upon itself. In eqn (28) the motion along the trajectory is accounted for directly by the use of a Lagrangian velocity function. Therefore, one can expand in a conventional Taylor series using the (presumably known) displacements as done in eqn (27).

Like eqn (15), eqn (28) can be expanded in terms of cumulants. The cumulants are defined by

\[ c(x, t) = \exp \left\{ -L[x(t)] \cdot \nabla \right\} f(x) + \frac{1}{2!} \left\{ \exp \left[ -L[x(t)] \cdot \nabla \right] \right\} \cdot \nabla^2 f(x) + \ldots \]

(29)

Because there is no time-ordering and the displacements freely commute with themselves, the cumulants defined in eqn (29) are related to the moments in a simpler manner than for the time-ordered case. The first two cumulants are listed here as

\[ \langle L[x(t)]\rangle_i = \langle L[x(t)] \rangle_j \]

\[ \langle L[x(t)][L[x(t)]\rangle_i = \langle L[x(t)]\rangle_j - \langle L[x(t)]\rangle_i \langle L[x(t)]\rangle_j \]

(30a)

(30b)

Eqn (29) is an expression involving derivatives of infinite order; similar such results have been obtained previously by Simmons and Zhang. Dagan states that if dis-placements specified by L[x(t)] are Gaussian, then the expansion given in eqn (29) truncates identically at second order. In fact, if L[x(t)] is a Gaussian random variable the series in eqn (29) does truncate at second order, and we avoid the difficulties associated with discarding potentially secular terms. However, it is difficult to determine under what conditions L[x(t)] can be exactly Gaussian. Weinstock has suggested that the Lagrangian displacements can never be exactly Gaussian for finite times. This does not preclude the distribution from being near Gaussian in some sense (as it must approach Gaussian for large times as dictated by the central limit theorem), but in the usual approach to this problem there are no guidelines to indicate the conditions such that the Gaussian approximation is valid. With these limitations in mind, we will keep with the assumption made by Dagan and require that L[x(t)] is Gaussian and spatially stationary. This simplifies eqn (29) to

\[ \frac{\partial c(x, t)}{\partial t} = \left\{ \exp \left[ -L[x(t)] \cdot \nabla \right] \right\} f(x) + \frac{1}{2!} \left\{ \exp \left[ -L[x(t)] \cdot \nabla \right] \right\} \cdot \nabla^2 f(x) 
\]

(31)

Substituting the definition of the displacements given by eqn (6) into eqn (31), and using the definition of the second cumulant given by eqn (30) and the velocity decomposition given by eqn (17), one obtains after some algebra

\[ \frac{\partial c(x, t)}{\partial t} = -v_x \cdot \nabla \int_v \exp \left[ \hat{v}(x(t)) \cdot \nabla \right] \hat{v}(x(t)) : \nabla c(x, t) \]

(32)

where \( v_x = \tilde{v}(x(t)) \) and represents the (constant) ensemble-average velocity. Eqn (32) is not useful in its present form because it still contains the unknown trajectory realizations x(t). At this point, we make a final assumption that the displacements L[x(t)] defined in eqn (6) can be approximated by replacing the unknown trajectory x(t) by the ensemble-average trajectory \( \langle x(t) \rangle \),

\[ \langle x(t) \rangle = x(t_o) + \int_{t_o}^t \exp \left[ \hat{v}(x(t)) \cdot \nabla \right] \hat{v}(x(t)) \cdot \nabla \]

(33)

As pointed out by Dagan, this approximation is similar to the first-order terms of an iterative scheme for homogenous turbulence suggested by Phythian. However, Neuman and Zhang have criticized the use of this lowest-order approximation, and no restrictions that define the conditions such that this lowest-order approximation is valid appear to be available. Again, to be consistent with previous studies, we adopt this assumption with full realization of its limitations. Eqn (32) can then be expressed in the form

\[ \frac{\partial c(x, t)}{\partial t} = v_x \cdot \nabla c(x, t) + \int_{t_o}^t \exp \left[ \hat{v}(x(t)) \cdot \nabla \right] \hat{v}(x(t)) \cdot \nabla c(x, t) \]

(34)
and this result is identical to the result of Dagan\textsuperscript{13,15}. This takes the form of the conventional convection–dispersion equation by setting
\[ D(t) = \int_{t_o}^{t} dt' \mathcal{V}(\mathbf{x}(t_o) + (t - t_o)\mathbf{v}_o) \mathcal{V}(\mathbf{x}(t_o) + (t - t_o)\mathbf{v}_o) \]
(35)
yielding the transport equation
\[ \frac{\partial \mathcal{C}(\mathbf{x}, t)}{\partial t} = -\mathbf{v}_o \nabla \mathcal{C}(\mathbf{x}, t) + D(t) : \nabla \nabla \mathcal{C}(\mathbf{x}, t) \]
(36)
To be absolutely clear about the assumptions involved in this development, we recall that they are (1) the porosity can be treated as a constant, (2) the Eulerian velocity field is steady, (3) the displacements \( \mathbf{L}_f(x(t)) \) are Gaussian, (4) the Lagrangian velocity field is spatially stationary, and (5) we can replace \( \mathbf{x}(t) \) by its lowest-order approximation \( \langle \mathbf{x}(t) \rangle \) given by eqn (33).

3.2 Renormalized cumulant expansion development

Our goal is now to develop an expression similar to eqn (36) by the cumulant expansion approach rather than the Lagrangian stochastic–convective one. We will begin with eqn (26) as the starting point, and again adopt the assumptions that the porosity is constant and the velocity field is spatially stationary. Under these conditions, the meaning of the operators involved in eqn (26) becomes more transparent. When spatial stationarity holds, the operators \( U_f^{(0)} \) and \( U_f^{(1)} \) are independent of location, and they freely commute with the gradient operator. Recall that the action of the operator \( U_f^{(0)} \) is to shift the spatial coordinate of the functions on which it operates forward along the trajectory of the ensemble-average velocity field\textsuperscript{38,45}. It is easy to show that under the conditions of spatial stationarity, we have the relationships
\[ U_f^{(0)}(x_o, \tau) \mathcal{V}(\mathbf{x}) \nabla U_f^{(0)}(x_o, \tau) = \frac{1}{\tau} \left[ \mathcal{V}(\mathbf{x} + (\tau - t_o)\mathbf{v}_o) \nabla - \mathcal{V}(\mathbf{x} + (\tau - t_o)\mathbf{v}_o) \mathcal{V} \right] \]
(37)
where here \( \mathbf{v}_o \) is the constant ensemble-average velocity.

Using the shifting property of the propagators, we can work out the effects of these operators in the exponential on the right-hand side of eqn (26). Although eqn (26) is somewhat complicated, by starting at the right-hand side and applying each of the propagators inside the brackets to everything to the right of them, we quickly arrive at the simplified expression
\[ \langle \mathcal{C}(\mathbf{x}, t) \rangle = U_f^{(0)}(t, t_o) \mathcal{C}(\mathbf{x}, t_o) \mathcal{V} \exp \left[ \int_{t_o}^{t} \! \! dr \int_{t_o}^{r} \! \! dq \right] \times \langle \mathcal{V}(\mathbf{x} + (\tau - t_o)\mathbf{v}_o) \mathcal{V} \rangle \]
\[ \times \mathcal{V}(\mathbf{x} + (\eta - t_o)\mathbf{v}_o) \mathcal{V} + \ldots \] \( f(\mathbf{x}) \)
(38)
Note that even though the velocity that appears in eqn (26) is the Eulerian field, the propagators induce a Lagrangian representation by transforming the coordinate system to the Lagrangian one.

Eqn (38) has some advantages over the expansion in moments (that can be generated immediately from eqn (15)), or the Lagrangian expansion given in eqn (29). Due to the so-called cluster property of cumulants\textsuperscript{24,37}, a cumulant becomes negligible (relative to its maximum value) as soon as any two terms in a particular cumulant are separated by more than the Lagrangian correlation time \( \tau_r \). This is useful for making order-of-magnitude estimates for the terms that appear in eqn (38), and van Kampen\textsuperscript{54,55} uses this property to develop the estimate \( \tau_r^{-1} \mathcal{O}(\mathbf{v}(\mathbf{x}) \nabla) \) for the \( n \)-th order term in the cumulant expansion. We can use these order-of-magnitude estimates to develop a constraint that indicates under what conditions a second-order truncation of eqn (38) is appropriate. For example, if we require that the third-order and higher terms in eqn (38) be negligible relative to the second-order terms, we can impose the restriction\textsuperscript{54,55}
\[ \mathcal{O}[^{\tau_r} \mathcal{O}(\mathbf{v}(\mathbf{x}) \nabla)] \ll 1 \]
(39)
and this leads to a constraint of the form
\[ \mathcal{O}[^{\tau_r} \mathcal{O}(\mathbf{v}(\mathbf{x}))] \ll 1 \]
(40)
where \( I \) is the identity operator. Realizing that in eqn (38) all operations are ultimately on the initial condition \( f(\mathbf{x}) \), one can further refine this constraint to be
\[ \mathcal{O} \left[ \frac{Z_f}{L_f} \right] \ll 1 \]
(41)
where \( v \) is a characteristic measure of the velocity perturbations and \( L_f \) represents a characteristic length for changes in the initial condition field. Here we have estimated the gradient of \( f(\mathbf{x}) \) as \( \mathcal{O}(\Delta f/L_f) \), and assumed that at most \( \partial f/\partial f(\mathbf{x}) \) in the development of the inequality (eqn (41)). The dimensionless group on the left-hand side of the inequality (eqn (41)) is conventionally known as the Kubo number\textsuperscript{38,39}. Similar such constraints have been developed for stochastic matrix processes by Fox\textsuperscript{25}, and in the analysis of turbulent dispersion by Weinstock\textsuperscript{56}. Under the condition that the constraint given by eqn (41) is met, then only the first two cumulants in eqn (38) are non-negligible, and the solution may be approximated to second order by the expression
\[ \langle \mathcal{C}(\mathbf{x}, t) \rangle = U_f^{(0)}(t, t_o) \mathcal{C}(\mathbf{x}, t_o) \mathcal{V} \exp \left[ \int_{t_o}^{t} \! \! dr \int_{t_o}^{r} \! \! dq \right] \times \langle \mathcal{V}(\mathbf{x} + (\tau - t_o)\mathbf{v}_o) \mathcal{V} \rangle \mathcal{V}(\mathbf{x} + (\eta - t_o)\mathbf{v}_o) \mathcal{V} + \ldots \]
(42)
To get a differential equation for transport, we take the derivatives of both sides of eqn (42). Because this step is algebraically complicated, the details of the development
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4 SUMMARY AND CONCLUSIONS

The stochastic–convective and cumulant expansion methods are clearly connected, as can be seen from the developments above. For the conditions described here, the cumulant expansion can be interpreted as an expansion of the convolution integral that appears in eqn (9). By the developments of Section 3, it is clear that under certain limiting conditions one can obtain identical solute transport equations directly from the stochastic–convective equation for the ensemble-average concentration, or from the equivalent cumulant expansion.

One might ask the question as to why the cumulant expansion is needed at all. If the desired end-result of our efforts is to derive an ensemble-average transport equation as done in Section 3, then the renormalized cumulant expansion provides some advantages.

(1) The renormalized cumulant expansion is not limited to certain distributions for the trajectory displacements or Lagrangian velocities. The development in terms of a renormalized cumulant expansion is applicable regardless of the distributions involved as long as the expansion is convergent. More useful is the case when a correlation time constraint for the perturbations (given by the inequality eqn (41)) is valid, indicating that the cumulant expansion can be truncated at second order. Note that this is less restrictive than assuming that the pdf of the Lagrangian velocities is strictly Gaussian; this assumption indicates only that the pdf can be well approximated by the first two cumulants of the velocity field provided certain physical constraints are met. Therefore, the assumption of a Gaussian velocity field is replaced by a restriction that suggests under what conditions a second-order solute transport equation will be a reasonable approximation. Higher-order equations for the ensemble-average concentration can be developed by retaining more terms in the truncation of the cumulant expansion, although the utility of such higher-order expansions needs to be investigated further.

(2) The renormalized cumulant expansion does not require the assumption that the trajectories $\mathbf{x}(t)$ be replaced by their ensemble-average $\langle \mathbf{x}(t) \rangle$. In fact, a similar (but not identical) renormalization occurs automatically, and results from the disentangling theorem given by eqn (19). Thus, the relevant Lagrangian cumulants are expressed in terms of the trajectories that arise from the $\mathbf{v}$-ensemble average velocity field. For spatially stationary velocity fields the induced trajectories take a particularly simple form as indicated by eqn (37). If one specifies particular values for $\mathbf{x}$ such that $\mathbf{x}=\mathbf{x}(t_0)+(t-t_0)\mathbf{v}_0$, the resulting transport eqn (43) can be identical to that of Dagan\textsuperscript{15}.
Although not discussed here, the renormalized cumulant expansion allows a development of the ensemble-average transport equations from Darcy-scale equations that do not neglect local dispersion (cf. Kavvas and Karakas\(^{36}\)). Dagan\(^{13,15}\) also presents a Lagrangian development that allows for local dispersion (treated as a Brownian motion component of the velocity field), and develops a convection–dispersion equation that includes this local dispersion term. Under the same assumptions that are applied to Dagan’s analysis, it is possible to show that a convection–dispersion equation of identical form can be garnered from a cumulant expansion approach\(^{36}\). In this case, the dispersion tensor given by eqn (44) is modified only by the addition of a local diffusion tensor.

For reactive flows that are spatially inhomogeneous, the stochastic–convective approach leads to a solution that involves potentially complicated path integrations, as discussed in Section 2. Because the renormalized cumulant expansion approach is a series expansion of these integrals, it results in solutions that are tractable. A publication that details how the cumulant expansion is carried out for reactive transport is presently in process.

Finally, we point out that there are very few restrictions that have been imposed on the development of the transport eqn (26) by the cumulant expansion method. Consequently, eqn (26) should apply equally well to spatially nonstationary velocity fields as is does to the case of the stationary velocity field examined in Section 3. Additionally, in nonstationary fields a shear effect may arise that modifies the ensemble-average velocity field, and this effect is captured by the cumulant expansion approach (as described in Appendix C). Although the cumulant expansion method has been applied to more complex nonstationary and non-steady flow fields\(^{3,36}\), it still represents a challenging area for continued research.

**NOMENCLATURE**

- \(c(x,t)\): solute concentration (mol/m\(^3\))
- \(D(t)\): effective dispersion tensor as defined by eqn (35) or eqn (44) (m\(^2\)/s)
- \(f(x)\): initial solute concentration distribution function (mol/m\(^3\))
- \(L_f\): characteristic length-scale associated with the concentration field (m)
- \(L(x(t))\): displacement trajectory, defined by eqn (6) (m)
- \(p(L[x(t)])\): probability functional for trajectories, as defined in eqn (9)
- \(p(y|x)\): probability function for displacements, defined by eqn (13)
- \(\tau_p\): time ordering symbol, as defined in eqn (19)
- \(U_{\text{forward}}(t,p)\): forward propagator or translation operator, defined by eqn (20a)
- \(U_{\text{backward}}(t,p)\): backward propagator or translation operator, defined by eqn (20b)
- \(v(x)\): Darcy-scale fluid pore velocity (m/s)
- \(\langle v(x) \rangle\): ensemble-average fluid pore velocity (m/s)
- \(v_x\): constant ensemble-average fluid pore velocity (m/s)
- \(v_c\): characteristic measure of fluid pore velocity perturbations, defined in the inequality (eqn (41)) (m/s)
- \(x(t)\): trajectory of the steady velocity field, defined by eqn (5) (m)
- \(y\): net displacement vector, defined as in Fig. 1
- \(\theta(x)\): porosity (saturated systems) or water content (unsaturated systems)
- \(\tau_{LC}\): Lagrangian correlation time for the velocity field (s)

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APPENDIX A TRAJECTORIES GENERATED FROM THE OPERATOR EXPONENTIAL

In the mathematics and physics literature, integrals of the form of eqn (14) have been studied for some time (starting with the work of Volterra), and are referred to as product integrals. The example that follows illustrates how the Lagrangian trajectories are generated by the exponential of the Eulerian velocity field, and is meant to be motivational rather than mathematically rigorous. We begin by discretizing the integral in eqn (14) into a sum over small time intervals \( D_t \). Then, the familiar property of exponentials allows us to represent this sum as a product of the form

\[
\exp\left( \int_0^t dt \mathbf{v}(x) \cdot \mathbf{r} \right) = \lim_{N \to \infty} \prod_{i=1}^N \exp\left[ -\Delta t_i \mathbf{v}(x) \cdot \mathbf{r} \right]
\]

\[\text{(A1)}\]

Now, for purely illustrative purposes, consider the case where only two such discrete displacements occur, as illustrated in Fig. 4. Then, taking the time intervals to be equal and using the properties of such operator exponentials, we have

\[
\exp[ -\Delta t \mathbf{v}(x) \cdot \mathbf{r} ] \exp[ -\Delta t \mathbf{v}(x) \cdot \mathbf{r} ] = \exp[ -\Delta t \mathbf{v}(x) \cdot \mathbf{r} ]
\]

\[\text{(A2)}\]

so that the action of these two exponentials is in fact to trace backwards to the initial condition. This representation becomes exact as \( \Delta t \) approaches zero. Note that the translating action of the two operator exponentials is easy to prove by a simple Taylor series expansion. The important point is that it is only the Eulerian velocity field that appears in the exponential in eqn (A2), and that the trajectories of the velocity field are generated automatically by this exponential form.

APPENDIX B RENORMALIZED CUMULANT EXPANSION FORM

We begin with the equations of transport given by eqn (3) and eqn (3)

\[
\frac{\partial c(x, t)}{\partial t} = -\nabla \cdot (\mathbf{v}(x) c(x, t)) \tag{B1a}
\]

Initial condition: \( c(x, t_0) = f(x) \) \tag{B1b}

Making the decomposition

\[
\mathbf{v}(x) = (\mathbf{v}(x)) + \tilde{\mathbf{v}}(x)
\]

\[\text{(B2)}\]

and using the definition of the operator exponential, eqns (B1) can be integrated directly to yield

\[
c(x, t) = \exp[ -t \mathbf{v}(x) \cdot \mathbf{r} ] c(x, t_0) \tag{B3}
\]

Now, if the velocity field that appears in Eq. (B1a) were spatially stationary, one could adopt a new coordinate system that moves with the velocity of the ensemble-average flow field. One can still adopt a transformation of variables that accomplishes much the same thing for more general flow fields. This is accomplished by passage to the so-called interaction representation, which is defined by

\[
c(x, t) = \exp[ -(t - t_0) \mathbf{v}(x) \cdot \mathbf{r} ] c(x, t_0) \tag{B4}
\]
Taking time derivatives of both sides of eqn (B4) yields the result
\[
\frac{\partial c(x,t)}{\partial t} = - (v(x)) \cdot \nabla \exp[-(t-t_o)v(x)] \cdot \nabla u(x,t)
+ \exp[-(t-t_o)v(x)] \frac{\partial u(x,t)}{\partial t}.
\] (B5)

Substituting this result into the left-hand side of Eq. (B1a) along with the decomposition given by eqn (B2) yields some algebra
\[
\frac{\partial u(x,t)}{\partial t} = - \exp[(t-t_o)v(x)] \cdot \nabla \tilde{v}(x) \cdot \nabla
\times \exp[-(t-t_o)v(x)] \cdot \nabla u(x,t).
\] (B6)

There is one important complication that arises in defining the transformed equation given by (B6). The coefficient of \(u(x,t)\) on the right-hand side of eqn (B6) is now a function of time, and is in general it represents a non-commuting operator\(^{35}\). Eqn (B6) can be integrated by the use of a time-ordered exponential\(^{19,24,37,53}\), which can be thought of as the operator version of an integrating factor. Integrating and returning to the original variable \(c\) (by inverting eqn (B4)) results in
\[
c(x,t) = \exp[-(t-t_o)v(x)] \cdot \nabla \tilde{v}(x) \cdot \nabla \exp[-(t-t_o)v(x)] \cdot \nabla f(x)
\] (B7)

where \(\tilde{v}(x)\) indicates that the exponential is time-ordered\(^{19,24,37,53}\). The ordering symbol indicates that operators with the earliest time index (i.e., \(t\) closest to \(t_o\)) occur to the far right in positional notation, whereas the operators with the latest time index are to the far left. Comparing the solutions given by eqn (B3) and eqn (B7), one finds a (somewhat simplified) version of the disentangling theorem proposed by Feynman\(^{19}\) and Masani\(^{40,41}\).
\[
\exp[-(t-t_o)v(x)] \cdot \nabla \tilde{v}(x) \cdot \nabla \exp[-(t-t_o)v(x)] \cdot \nabla\]
\[
\exp\left\{- \int_{t_o}^{t} \mathrm{d}r \exp\left\{ (t-r_o)v(x) \cdot \nabla \tilde{v}(x) \cdot \nabla\right\}\right\}
\] (B8)

This is eqn (19) in the main body of the paper.

**APPENDIX C DEVELOPMENT OF THE DIFFERENTIAL EQUATION**

By design we are interested in obtaining the differential equation for transport of the ensemble-average concentration. However, eqn (42) (repeated here for convenience)
\[
\langle c(x,t) \rangle = U_{\omega}^{(0)}(t,t_o) \exp \left\{ \int_{t_o}^{t} \mathrm{d}\eta \left[ \tilde{f}(x + (t-t_o)v) \cdot \nabla \right] \cdot \nabla f(x) \right\}
\] (C1)

is not in differential form. To obtain the differential equation, we begin by noting the time derivative of \(U_{\omega}^{(0)}\) acting on a function \(g(x,t)\) is given by a straightforward application of the chain rule
\[
\frac{\partial}{\partial t} U_{\omega}^{(0)}(t_o,t) g(x,t) = \frac{\partial}{\partial t} \exp\left\{ - (t-t_o)v(x) \cdot \nabla \right\} g(x,t)
\]
\[
= - (v(x)) \cdot \nabla \exp\left\{ - (t-t_o)v(x) \cdot \nabla \right\}
\times g(x,t) + \exp\left\{ - (t-t_o)v(x) \cdot \nabla \right\}
\times \frac{\partial g(x,t)}{\partial t}
\]
\[
= - (v(x)) \cdot \nabla U_{\omega}^{(0)}(t_o,t) g(x,t)
\]
\[
+ U_{\omega}^{(0)}(t_o,t) \frac{\partial g(x,t)}{\partial t}.
\] (C2)

Taking the time derivative of both sides of eqn (C1), and use of the chain rule given in eqn (C2) leads to
\[
\frac{\partial \langle c(x,t) \rangle}{\partial t} = - (v(x)) \cdot \nabla U_{\omega}^{(0)}(t,t_o)
\]
\[
\times \exp\left\{ \int_{t_o}^{t} \mathrm{d}\eta \left[ \tilde{f}(x + (t-t_o)v) \cdot \nabla \right] \cdot \nabla f(x) \right\}
\]
\[
\times \exp\left\{ \int_{t_o}^{t} \mathrm{d}r \exp\left\{ (t-r_o)v(x) \cdot \nabla \tilde{v}(x) \cdot \nabla\right\}\right\}
\]
\[
\times \frac{\partial g(x,t)}{\partial t}
\]
\[
\left\{ \tilde{f}(x + (t-t_o)v) \cdot \nabla \tilde{f}(x + (t-t_o)v) \cdot \nabla f(x) \right\}
\] (C3)

Comparison of the first term on the right-hand side of eqn (C3) with eqn (C1) indicates that this term is simply \(- (v(x)) \cdot \nabla \tilde{c}(c(x,t))\). For the second term, we insert the identity
\[
U_{\omega}^{(0)}(t_o,t) U_{\omega}^{(0)}(t,t_o) = I
\] (C4)

before the last exponential on the right-hand side of eqn (C3) and again use eqn (C1) to extract the ensemble-average
concentration. Incorporating the results of these two simplifications yields the expression

$$\frac{\partial \tilde{c}(\mathbf{x}, t)}{\partial t} = -\langle \nabla \tilde{c}(\mathbf{x}, t) \rangle$$

$$+ U^{(o)}(t, t_o) \left[ \int_{t_o}^t d\tau \langle \tilde{v}[\mathbf{x} + (t - \tau)\mathbf{v}_o] \cdot \nabla \tilde{c}(\mathbf{x}, t) \rangle \right]$$

$$\cdot \tilde{v}[\mathbf{x} + (\eta - t_o)\mathbf{v}_o], \nabla$$

$$U^{(o)}(t, t_o) \langle \tilde{c}(\mathbf{x}, t) \rangle$$

(C5)

Finally, applying the propagators $U^{(o)}$ and $U^{(0)}$, we get the local equation

$$\frac{\partial \tilde{c}(\mathbf{x}, t)}{\partial t} = -\langle \nabla \tilde{c}(\mathbf{x}, t) \rangle$$

$$+ \left[ \int_{t_o}^t d\tau \langle \tilde{v}(\mathbf{x}) \cdot \nabla \tilde{v}[\mathbf{x} - (t - \eta)\mathbf{v}_o] \cdot \nabla \rangle \right] \langle \tilde{c}(\mathbf{x}, t) \rangle$$

(C6)

This result is identical to the result of Kavvas and Karakas for the case of a spatially stationary velocity field and a negligible the local dispersion coefficient. Note that because the gradient operators act on all terms to the right, the term in brackets on the right-hand side of eqn (C6), by the chain rule will generate two terms. The first of these terms is of the form $\langle \tilde{v}(\mathbf{x}) \cdot (\nabla \tilde{v}[\mathbf{x} - (t - \eta)\mathbf{v}_o]) \cdot \nabla \rangle \langle \tilde{c}(\mathbf{x}, t) \rangle$ and represents a shear effect that modifies the ensemble-average velocity field. Such terms are not unique to this study (cf. Weinstock, Appendix C; Fox, eqn (42)), and for the case of incompressible flow with a spatially stationary Lagrangian covariance function this terms is identically zero. To be consistent with the work of Dagan, we will assume that the Lagrangian covariance function is spatially stationary. The second term that arises from the application of the chain rule is of the form of the conventional Green-Kubo macrodispersion tensor, and leads to the definition of the classical convection–dispersion equation

$$\frac{\partial \tilde{c}(\mathbf{x}, t)}{\partial t} = -\mathbf{v}_s \cdot \nabla \tilde{c}(\mathbf{x}, t) + \left[ \int_{t_o}^t d\tau \langle \tilde{v}(\mathbf{x}) \cdot (\nabla \tilde{v}[\mathbf{x} - (t - \eta)\mathbf{v}_o]) \cdot \nabla \rangle \right] \langle \tilde{c}(\mathbf{x}, t) \rangle$$

(C7)

This is eqn (43) of the main body of the paper.