Advection–Diffusion in Lagrangian Coordinates

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The advection–diffusion equation can be approximated by a one-dimensional diffusion equation in Lagrangian coordinates along the directions of compression of fluid elements (the stable manifold). This result holds in any number of dimensions, for a velocity field with chaotic trajectories, with an error proportional to the square root of the diffusivity. After some time, the one-dimensional equation becomes invalid, but by that time a large fraction of the scalar variance has decayed.

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I. INTRODUCTION

The importance of transport and mixing processes in the physical sciences cannot be overestimated. Applications abound in the geophysical sciences such as oceanography, atmospheric science, and geology, as well as in astrophysics, chemistry, and general fluid dynamics. Because it spans so many disciplines, a comprehensive understanding of the detailed manner in which mixing proceeds is of paramount importance.

In most physically relevant applications it is the case that the mixing (diffusion) process depends on the stirring (advection) phase to create large gradients that then allow diffusion to act. Without some form of enhanced stirring, diffusion is powerless in the face of weak gradients. The enhancement may be provided by turbulence, but that is not necessary: for even simple laminar flows, fluid trajectories can display sensitivity to initial conditions (chaos) that stretches fluid elements exponentially—a phenomenon termed chaotic advection [1]. This stretching produces the necessary large gradients of the advected quantity, leading to chaotic mixing. Our focus will be on such smooth, chaotic flows.

There are two main results in this paper. First, we show that the advection–diffusion equation in Lagrangian coordinates can be reduced to a one-dimensional diffusion equation along the stable manifold of the flow (the direction along which fluid elements are compressed). The diffusion coefficient of the reduced equation grows exponentially in time, reflecting the enhancement to diffusion due to chaotic trajectories; the reduced equation itself is an approximation valid to exponential accuracy in time. The enhancement to diffusion is not uniform along the stable manifold: the amount of stretching of fluid elements is anticorrelated with the curvature of the stable manifold [2, 3]. The second result is that this approximate, one-dimensional, equation breaks down after a certain time because of a buildup of large gradients along the neglected direction of stretching. By that time, a significant fraction of the scalar variance has decayed, so the one-dimensional equation is often sufficient for describing physical systems.

II. THE REDUCED ADVECTION–DIFFUSION EQUATION

In the present section we give an account of the manner in which the advection–diffusion equation expressed in Lagrangian coordinates can be collapsed to a one-dimensional diffusion equation. The result holds regardless of dimension (two, three, or more as can arise in kinetic equations), and the approximations involved are accurate to order Pe\(^{-1/2}\), where the Péclet number is Pe := \(L v / D\), with \(L\) and \(v\) characteristic spatial and velocity scales, and \(D\) the diffusivity; in many common problems, Pe > 10\(^8\). Though it has been realized for some time that the enhanced diffusion proceeds along the stable (contracting) direction in both smooth [2, 4] and turbulent [5–7] flows, here we make a series of approximations and use a geometrical constraint [2, 3, 8] to rewrite the advection–diffusion equation as an actual one-dimensional diffusion equation along the stable manifold. This is more than mere formalism, because it allows the identification of the role of invariant structures of the flow in the mixing process (Section III). Solving the one-dimensional equation also allows us to show that it must eventually break down (Section IV).

The advection–diffusion equation for a compressible velocity field \(v(x, t)\) on an \(n\)-dimensional bounded domain is

\[
\frac{\partial \phi}{\partial t} + v \cdot \nabla \phi = \frac{1}{\rho} \nabla \cdot (\rho \mathbb{D} \cdot \nabla \phi),
\]

where the scalar field \(\phi(x, t)\) is the concentration of the quantity advected, \(\mathbb{D}(x, t)\) is the diffusivity tensor, and \(\rho\) is the density of the fluid (which satisfies the continuity equation). The advected scalar \(\phi\) can represent for example temperature, salinity, or the concentration of a solute such as helium. The velocity field \(v\) can be obtained by solving the Navier–Stokes equation, or it can be a model system designed to mimic the qualitative features of a realistic flow. The trajectory of a fluid element is given

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by the solution \( x(t, t_0; a) \) to

\[
\frac{\partial x}{\partial t} = v(x,t), \quad x(t = t_0, t_0; a) = a,
\]

so that \( x(t, t_0; a) \) is the coordinate transformation at time \( t \) from the comoving Lagrangian \( (a) \) to stationary Eulerian \( (x) \) coordinates. For smooth velocity fields, the transformation is smooth and invertible.

By construction, in Lagrangian coordinates the advection term drops out, and the advection–diffusion equation (1) takes the form

\[
\frac{\partial \phi_0}{\partial t} \bigg|_a = \frac{1}{\rho_0} \nabla_0 \cdot (\rho_0 \mathbb{D}_0 \cdot \nabla_0 \phi_0),
\]

where 0 subscripts denote Lagrangian quantities: \( \phi_0(a, t) = \phi(x(t, t_0; a), t) \), \( \rho_0(a) = |M| \rho(x(t, t_0; a), t) \), \( \nabla_0 = \partial / \partial a \), and

\[
\mathbb{D}_0 = M^{-1} \cdot D \cdot (M^{-1})^T,
\]

where the tangent map (or Jacobian) \( M \) has components \( M' = \partial x^j / \partial a^i \). The \( a \) subscript on the time derivative in (3) is a reminder that the derivative is taken with \( a \) held fixed, as opposed to the derivative in (1) which has \( x \) fixed.

The covariance of (1) and (3) (i.e., the form of the right-hand side of both equations is the same) suggests the definition of a metric

\[
g := \kappa \mathbb{D}^{-1}
\]

on the tangent space at \( x \), where the dimensional function \( \kappa(a, t) \) is chosen such that \( |g| = 1 \). (For constant \( D \), \( \kappa \) is also constant.) The metric \( g \) is a proper Riemannian metric, since it is symmetric and positive-definite; it induces a metric \( g_0 \) on the tangent space at \( a \),

\[
g_0 = M^T \cdot g \cdot M = \kappa \mathbb{D}^{-1}.
\]

Because it is symmetric and positive-definite, the metric \( g_0 \) can be written in terms of its real-positive eigenvalues \( \Lambda^2 \) and orthonormal eigenvectors \( \mathbf{e}_\sigma \) as

\[
g_0 = \sum_{\sigma=1}^n \Lambda^2_\sigma \mathbf{e}_\sigma \mathbf{e}_\sigma.
\]

The \( \Lambda_\sigma(t, t_0; a) \) are called coefficients of expansion; without loss of generality, they are ordered such that \( \Lambda_1 \geq \Lambda_2 \geq \ldots \geq \Lambda_n \). For a very wide class of flows, including incompressible flows, \( \Lambda_n \) (the smallest coefficient of expansion) is associated with an exponentially contracting direction, so that \( \Lambda_n \ll 1 \). The characteristic directions \( \mathbf{e}_\sigma(t, t_0; a) \) converge exponentially to their time-asymptotic values \( \mathbf{e}_\sigma^\infty(t_0; a) \) \([8, 9]\). The direction of fastest contraction (\( \sigma = n \)) plays a distinguished role in our development, so we emphasize its importance by denoting it by the letter ‘s’ (for “stable”). Thus, \( e_n \) and \( \Lambda_n \) are written \( \mathbf{s} \) and \( \Lambda_s \). Similarly, we will later use the letters ‘u’ (for “unstable”) for the direction of fastest stretching (\( \sigma = 1 \)); for three-dimensional systems, we use ‘m’ (for “median”) when referring to direction \( \sigma = 2 \), which may be stretching, contracting, or neutral.

Replacing \( \mathbb{D}_0 \) by \( \kappa \mathbb{D}_0^{-1} \) using (6), and substituting the diagonal form (7) of the metric into (3), we find

\[
\frac{\partial \phi_0}{\partial t} \bigg|_a = \sum_{\sigma=1}^n \frac{1}{\rho_0} \nabla_0 \cdot (\rho_0 \kappa \Lambda^2_\sigma \mathbf{e}_\sigma \cdot \nabla_0 \phi_0).
\]

Since \( \kappa \sim Pe^{-1} \ll 1 \), for short times the right-hand side of (8) can be neglected completely. However, in a chaotic flow, at least some of the \( \Lambda_\sigma \) achieve exponential behavior after a moderate time and can overcome the small diffusivity. Because the inverse of \( \Lambda_\sigma \) enters (8), the direction of fastest contraction \( \Lambda_s = \Lambda_n \) dominates, and eventually we have \( Pe^{-1} \Lambda_n^{-2} \sim O(1) \). Assuming \( \Lambda_s \sim \exp(\Lambda_s t) \) this occurs roughly at time

\[
t_1 - t_0 \sim \frac{1}{2|\Lambda_s|} \log Pe,
\]

after which we can approximate (8) by

\[
\frac{\partial \phi_0}{\partial t} \bigg|_a = \frac{1}{\rho_0} \nabla_0 \cdot (\rho_0 \kappa \Lambda_s^{-2} \mathbf{s} \cdot \nabla_0 \phi_0) + O(\Lambda_n^{-2}).
\]

The relative error we are making if we neglect the \( O(\Lambda_n^{-2}) \) terms is of order \( (\Lambda_n / \Lambda_s)^{-2} \). Since \( Pe \) is assumed very large, by the time \( \Lambda_n^{-2} \sim Pe^{-1} \ll 1 \). For a 2D time-dependent flow, the error is even smaller, being of order \( (\Lambda_s / \Lambda_n)^{-2} \sim (Pe^{-1} / \Lambda_n^2) \ll Pe^{-1} \). We conclude that in most physically relevant applications the neglect of the higher-order terms in (10) is correct to a very high degree of accuracy. However, in Section IV we will show that this neglect is not justified for long times.

Physically, the leading-order term in (10) reflects the enhanced diffusion due to the presence of a dominant contracting direction: along that direction huge gradients are created by the stretching and folding action of the flow, leading to an exponential increase in the efficiency of diffusion. For all practical purposes the other directions can be ignored, as long as the smallest Lyapunov exponents are nondegenerate, as assumed.

Since the eigenvector \( \mathbf{s} \) converges to its asymptotic value \( \mathbf{s}^\infty \) at a rate \( (\Lambda_s / \Lambda_1)^{-1} \) \([8, 9]\), we may replace \( \mathbf{s}(t, t_0; a) \) in (10) by \( \mathbf{s}^\infty(t_0; a) \). The error committed is proportional to \( \widetilde{\Lambda}_s \sim Pe^{-1/2} \), larger than the order \( Pe^{-1} \) error in (10). The characteristic direction \( \mathbf{s}^\infty(t_0; a) \) can be integrated to yield the stable man-
ifold \( a_s(\hat{s}) \) (or \( \hat{s} \)-line) through a point \( a_0 \),
\[
\frac{\partial a_s(\hat{s})}{\partial \hat{s}} = \hat{s}^{-\infty}(t_0; a_s(\hat{s})), \quad a_s(0) = a_0,
\]
where \( \hat{s} \) denotes the arc length along the stable manifold. The stable manifold is central to our development, and we will reformulate (10) so that it represents a diffusion equation along this manifold.

To achieve this goal two more approximations are needed. It was shown in Ref. [8] that \( \hat{s} \cdot \nabla_0 \log \Lambda_s \) converges exponentially to a time-independent function, at a rate \( \max(\Lambda_s, \Lambda_0/\Lambda_{n-1}) \). This convergence implies that, along an \( \hat{s} \)-line given by \( a_s(\hat{s}) \), we can choose an arbitrary reference point \( a_s(0) = a_0 \) and express the time-dependence of \( \Lambda_s(t, t_0; a_0(\hat{s})) \) as
\[
\Lambda_s(t, t_0; a) = \Lambda_s(t_0; a) \Lambda_n(t, t_0; a_0),
\]
where \( a \) and \( a_0 \) are on the same \( \hat{s} \)-line and \( \Lambda_s \) is a time-independent function. We will discuss the physical significance of the function \( \Lambda_s \), which we call the parallel coefficient of expansion, in Section III. Neglecting the time-dependence of \( \Lambda_s \) does not increase the overall error.

We now rewrite (10) as
\[
\frac{\partial \phi}{\partial t} \bigg|_a \simeq \frac{1}{\rho_0} \nabla_0 \cdot (\rho_0 \hat{\kappa} \hat{s} \cdot \nabla_0 \phi_0),
\]
where we have defined
\[
\hat{\kappa} := \kappa \Lambda_s^{-2}(t, t_0; a_0), \quad \hat{s} := \Lambda_{s}^{-1} \hat{s}^{-\infty}.
\]
In Ref. [8], it was demonstrated that for a chaotic flow in any number of dimensions, \( \hat{s} \) must satisfy the constraint
\[
\text{div}_0 \hat{s} = \Lambda_{s}^{-1}(\text{div}_0 \hat{s}^{-\infty} - \hat{s}^{-\infty} \cdot \nabla_0 \log \Lambda_s) = 0,
\]
where
\[
\text{div}_0 \hat{s} := |g_0|^{-1/2} \nabla_0 \cdot (|g_0|^{1/2} \hat{s})
\]
is the covariant divergence of \( \hat{s} \) [11, p. 49]. Upon using the constraint (15) in (13), we obtain
\[
\frac{\partial \phi}{\partial t} \bigg|_a \simeq \frac{1}{\hat{\rho}} \hat{s} \cdot \nabla_0 \left( \frac{\hat{\rho} \hat{\kappa} \hat{s} \cdot \nabla_0 \phi_0} \right),
\]
where
\[
\hat{\rho} := \frac{|g_0|^{-1/2}}{\rho_0},
\]
and \( \bar{g}_0(a) \) is defined analogously to \( \bar{\Lambda}_s \) in (12),
\[
|g_0(a, t)| = \bar{g}_0(a) |g_0(a_0, t)|.
\]
The decomposition (19) is possible because the derivative of the metric determinant along the \( \hat{s} \)-line converges exponentially to a time-independent value [8]. The factor \( \bar{g}_0 \) in \( \hat{\rho} \) arises from the metric determinant in the covariant divergence (16), and is absent in incompressible flows. Thus, after all the approximations have been made, the overall error is proportional to \( \max(\Lambda_s, (\Lambda_s/\Lambda_{n-1})) \), which scales as \( \rho_0^{-1/2} \).

Finally, we define a derivative along \( \hat{s} \),
\[
\frac{\partial}{\partial \hat{s}} := \hat{s} \cdot \nabla_0 ,
\]
where \( s \) is a new parameter along the \( \hat{s} \)-line that differs from the arc length \( \hat{s} \) because of the weight \( \Lambda_s^{-1} \) in (14). Equation (17) then achieves the simple form
\[
\frac{\partial \phi}{\partial t} \bigg|_a \simeq \frac{1}{\hat{\rho}} \frac{\partial}{\partial \hat{s}} \left( \hat{\rho} \hat{\kappa} \frac{\partial \phi}{\partial \hat{s}} \right).
\]
Equation (21) is a one-dimensional diffusion equation along a line defined by \( a_s(\hat{s}) \). The effective diffusivity \( \hat{\kappa}(a_s(s), t) \) diverges exponentially in time, so that even for a microscopic physical diffusivity \( \kappa \) the gradients along \( \hat{s} \) are eventually smoothed. Note that it is only in terms of the parameter \( s \) that we obtain a one-dimensional diffusion equation; in terms of any other parameter (other than a uniform rescaling of \( s \)), equation (21) contains a “drift” term proportional to \( \nabla_0 \cdot \hat{s}^{-\infty} \).

### III. PHYSICAL INTERPRETATION

We now address the physical meaning of the various constituents of the one-dimensional diffusion equation (21). The essence of the simplification in Section II lies in expressing the advection–diffusion equation in Lagrangian coordinates in terms of a single parameter along a stable manifold: with an appropriate choice of parameter, namely \( s \), the equation collapses to a simple diffusion equation.

There are three main ingredients involved in the one-dimensional equation (21): (i) The exponential part of the coefficient of expansion, \( \Lambda_s(t, t_0; a_0) \); (ii) the stable manifold, \( a_s(\hat{s}) \); and (iii) The parallel coefficient of expansion, \( \bar{\Lambda}_s \). We discuss each in turn.

(i) The exponential part of the coefficient of expansion, \( \Lambda_s(t, t_0; a_0) \), is responsible for the enhanced diffusion coefficient \( \hat{\kappa} \). The variation of the function \( \Lambda_s \) across \( \hat{s} \)-lines along unstable directions is very steep (see Section IV) and is the cause of the poor convergence of Lyapunov exponents [2, 8]. Thus, the diffusion rates on two neighboring \( \hat{s} \)-lines can be quite different in magnitude, though similar in character because \( \Lambda_s(t, t_0; a_0) \) is independent of \( s \) and so gives only a time-dependent overall scaling of the diffusion coefficient in (21).

(ii) The \( \hat{s} \)-line is straightforward to compute \([2, 3]\); it has a fine structure and densely fills an ergodic region. If we treat the stable manifold as a material line to be advected by the flow, then the elements on that line will converge exponentially together as \( t \to \infty \). Thus, if these initial elements carry different concentrations of \( \phi \), the
gradients of $\phi$ will grow exponentially along these trajectories. It is thus natural for the stable manifold to play a central role in the chaotic mixing process. In fact, it is the capacity of the stable manifold to fill an entire chaotic region that allows for a thorough mixing [12].

When evolved backward in time, a small “test parcel” (of a scale comparable to the length at which dissipation acts) will be stretched and folded and will give a filament of fluid that samples the initial data $\phi_0$ [12]. This filament will tend to trace out the stable manifold. This is the Lagrangian analogue of the phenomenon of “asymptotic directionality” invoked to discuss the behavior of a material line as it is evolved forward in time by a flow [13–16].

(iii) The parallel coefficient of expansion $\tilde{\Lambda}_s$, also has deep implications for chaotic mixing. The time-dependent part of $\Lambda$, whose role was explored in (i) above, is uniform on an $\hat{s}$-line. In contrast, the parallel coefficient of expansion $\tilde{\Lambda}_s$ is constant in time (though it depends on $t_0$ for unsteady flows) but varies along the stable manifold. Since the full coefficient of expansion $\Lambda$ gives the rate of contraction of an initial infinitesimal ellipsoid, we must regard $\tilde{\Lambda}_s$ as a deviation from the reference rate of contraction given by $\Lambda_s(t, t_0; a_0)$. Thus, regions of anomalously large $\tilde{\Lambda}_s$ are associated with fluid elements that contract slower than other elements on the same $\hat{s}$-line.

The size of $\tilde{\Lambda}_s$ has an impact on diffusion, because the enhanced diffusion is due to the contraction of fluid elements. The effect is embodied in $\partial / \partial s$: because of (14) and (20), we have $\partial / \partial s = \tilde{\Lambda}_s^{-1} \partial / \partial \hat{s}$, where $\hat{s}$ is the arc length along $\hat{s}$. Hence, in regions of large $\tilde{\Lambda}_s$, gradients of $\phi_0$ are smaller when expressed in the coordinate $s$; the reverse is true for regions of small $\tilde{\Lambda}_s$. A large value of $\tilde{\Lambda}_s$ is thus a hindrance to diffusion.

But what range of values does $\tilde{\Lambda}_s$ achieve? In Fig. 1(a) we plot log $\tilde{\Lambda}_s$ against the arc length $\hat{s}$ on an $\hat{s}$-line. The cellular flow of Solomon and Gollub [17, 18] is used in the computation (with parameter values $A = B = \omega = k = 1$); it is meant to mimic a chain of oscillating convection rolls. The parallel coefficient of expansion $\tilde{\Lambda}_s$ is seen to vary over five orders of magnitude. Thus, there exist many exceptional fluid elements that have very slow rates of contraction.

The important factor, then, is how frequently these large values of $\tilde{\Lambda}_s$ occur along the $\hat{s}$-line. Figure 1(b) shows the probability distribution function (PDF) of log $\tilde{\Lambda}_s$ along two $\hat{s}$-lines: the tail of the distribution is clearly an exponential, decaying as $\exp(-2 \log \tilde{\Lambda}_s) = \tilde{\Lambda}_s^{-2}$. This is surprising because for this flow the distribution of the finite-time Lyapunov exponents themselves has a Gaussian tail, as opposed to a “fat” (exponential) tail. In fact, we have observed the $\tilde{\Lambda}_s^{-2}$ exponential law for the tail of the distribution in several flows, even flows such as the random wave flow [4] (also known as the sine flow) that have no KAM regions. Thus, the anomalously large values of $\tilde{\Lambda}_s$ cannot only be due to sticking to KAM surfaces, though that is certainly a factor. Rather, there are inherent paths in the flow that fluid elements can follow to avoid stretching, and every “final” fluid element contains a mixture of such paths. That the tails of the PDF of log $\tilde{\Lambda}_s$ fall off exponentially rather than as a Gaussian suggests that these exceptional events are important to the long-time decay of scalar variance, in the same manner that the initial alignment of the scalar gradient along the $\hat{s}$-line matters. The statistics of these regions of low stretching is related to the distribution of curvature of material lines [19, 20].

**IV. BREAKDOWN OF THE APPROXIMATION**

We now show that the neglect of the higher-order terms in (10) is not justified for long times, a fact we believe has not been appreciated before. To do that, we compare the magnitude of the terms that were neglected in going from (8) to (10) and show that they become important after a certain time. To simplify the argument, we assume the flow is incompressible, and that the diffusivity $\kappa$ is constant in space and time, so that $\kappa \tilde{t}$ in (14) is constant along the $\hat{s}$-line and depends only on which particular stable manifold we are dealing with [see (i) in Section III]. We label the dependence on the particular manifold under consideration by a parameter $u$, meant to represent displacements along an unstable (stretching) direction $\hat{u}$ of the flow. For an initial condition $\phi_0(s, 0; u) = \exp(iks)$ (we set the initial time $t_0$ to zero), the one-dimensional diffusion equation (21) has solution

$$\phi_0(s, t; u) = \exp \left(iks - k^2 t \mathcal{K}(t; u) \right),$$

where

$$\mathcal{K}(t; u) := \frac{1}{t} \int_0^t \tilde{\kappa}(t'; u) \, dt'$$

is the time-average of $\tilde{\kappa}(t; u)$. From Eq. (14), $\tilde{\kappa}$ is proportional to $\Lambda_s^{-2} \sim \exp(2|\Lambda_s|t)$, so (22) gives a superexponential decay of $\phi_0$.

Consider now the variation of $\phi_0(s, t; u)$ along the unstable direction $\hat{u}$,

$$\partial_u \phi_0 = -k^2 t \partial_u \mathcal{K}(t; u) \phi_0,$$

where $\partial_u := \hat{u} \cdot \nabla_0$. The second derivative of $\phi_0(s, t; u)$ is

$$\partial_u^2 \phi_0 = \left[ \left( k^2 t \partial_u \mathcal{K}(t; u) \right)^2 - k^2 t \partial_u^2 \mathcal{K}(t; u) \right] \phi_0.$$

Keeping only the first term in (25) (the second term does not dominate), we take the ratio of the $\sigma = 1$ (unstable direction) term to the $\sigma = 3$ (stable direction) term in (8),

$$\varepsilon := \frac{|k \Lambda_s^{-2} \left( k^2 t \partial_u \mathcal{K} \right)^2 \phi_0|}{|k \Lambda_s^{-2} k^2 \phi_0|} = \Lambda_s^{-1} \kappa \tilde{t} \partial_u^2 \mathcal{K} | \phi_0|^2.$$


Since $\Lambda_s$ is in the exponential regime, we have that [8]

$$\partial_u \log \Lambda_s = c \Lambda_u ,$$  \hspace{1cm} (28)

where $c$ is a function of space and has only algebraic behavior in time: it neither grows nor decays exponentially in time. The exponential growth in (28) is a reflection of the exponential separation of fluid trajectories in the chaotic flow. Equation (28) applies for almost all initial conditions in essentially all smooth flows and maps (it is sufficient that the velocity field be thrice-differentiable [8]), except in highly exceptional cases such as uniformly stretching systems (e.g., Arnold’s cat map, or flow around a hyperbolic point), for which $\varepsilon \equiv 0$; the superexponential solution (22) then persists forever.

After inserting (27)–(28) in (26), we obtain finally

$$\varepsilon \sim |kc \lambda^{-1} \kappa \Lambda^{-1}|^2 \sim |Pe^{-1} \Lambda^{-1}|^2 .$$  \hspace{1cm} (29)

Assuming that $\Lambda^{-1} \sim \exp(|\lambda_0|t)$, we conclude that $\varepsilon$ becomes $O(1)$ at time

$$t_2 \simeq \frac{1}{|\lambda_0|} \log Pe = 2t_1 ,$$  \hspace{1cm} (30)

where $t_1$ is the time where diffusion sets in, defined by (9).

In three dimensions, we must take into account the $\sigma = 2$ (the ‘m’ or ‘median’ direction) term in (8).

Its role depends on the type of direction it represents. If it is a stretching or neutral direction, then the estimate presented above holds, and diffusion along the $\sigma = 2$ direction becomes important at roughly the same time $t_2$ given by (30). If $\sigma = 2$ is a contracting direction, we must modify the asymptotic behavior (28) to $\partial_m \log \Lambda_s = \bar{c} (\Lambda_s / \Lambda_m)$, as described in [8]. This leads to a time $t'_2 := (1/2|\lambda_m|) \log Pe$, after which the $\sigma = 2$ term in (8) can no longer be neglected. Depending on the magnitude of $|\lambda_m|$, $t'_2$ may come before or after $t_2$.

The breakdown of (21) thus occurs at $\min(t_2, t'_2)$, which we will simply call $t_2$. In two dimensions, we ignore $t'_2$ because there is no median direction.

The overall picture is thus as follows. For time $t < t_1$, $\phi_0$ is approximately constant because the exponential growth of gradients has not yet overcome the small diffusivity. For $t_1 < t < t_2$, Eq. (21) applies and the scalar variance decays superexponentially. For $t > t_2$, the neglect of the $\sigma = 1$ (and $\sigma = 2$ in 3D) terms in (8) is no longer justified, and the one-dimensional diffusion equation (21) must be abandoned: to characterize the advection–diffusion process, the full Eq. (8) must be solved.

The reason that the diffusion along $\mathbf{u}$ cannot be neglected after $t_2$ is that the tiny, exponentially-decaying effective diffusivity associated with $\mathbf{u}$ allows the creation of extremely large gradients of $\phi_0$ along that direction. These gradients are a consequence of the superexponential solution (22). Eventually, the gradients overcome the exponentially-decaying effective diffusivity. In essence, the superexponential solution contains the seeds of its own destruction: the superexponentiality comes from the factor $\mathcal{K}$ in (22), but that factor exhibits extremely rapid
variation along the stretching direction, with no diffusivity to oppose it (until $t \simeq t_2$).

The breakdown of the one-dimensional equation helps to understand a discrepancy between the superexponential decay predicted by that equation and the observed exponential decay of the scalar variance in typical chaotic and turbulent flows. In Ref. [4] a superexponential decay was derived for individual fluid trajectories, and an exponential decay was recovered by averaging over the initial angle between the scalar gradient and the stable direction. Fereday et al. [21] pointed out that the decay of the variance in the (nonuniform) baker’s map is exponential for long times, even though in their case the angle between the scalar gradient and the stable direction is always zero, so that no averaging can be done. Our demonstration of the termination of the superexponential regime after some time shows that there is hope of restoring the more appropriate, exponential decay of the variance, but we have yet to show this because a technique of solution for the Eq. (8) must be found. For the Kraichnan–Kazantsev model of homogeneous, isotropic turbulence (delta-correlated in time), the decay of variance is known to be exponential [5–7], though the predicted rate is too rapid when compared with nonhomogeneous systems [21] which can have slowly-decaying eigenfunctions.

Even though it ceases to be valid after some time, the one-dimensional equation is still relevant, because during the time $t_1 < t < t_2$ a very large fraction of the variance can get dissipated, owing to the very fast decay of the superexponential.

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