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Diffusion and Brownian motion in Lagrangian coordinates

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In this paper we consider the convection-diffusion problem of a passive scalar in Lagrangian coordinates, i.e., in a coordinate system fixed on fluid particles. Both the convection-diffusion partial differential equation and the Langevin equation are expressed in Lagrangian coordinates and are shown to be equivalent for uniform, isotropic diffusion. The Lagrangian diffusivity is proportional to the square of the relative change of surface area and is related to the Eulerian diffusivity through the deformation gradient tensor. Associated with the initial value problem, we relate the Eulerian to the Lagrangian effective diffusivities (net spreading), validate the relation for the case of linear flow fields, and infer a relation for general flow fields. Associated with the boundary value problem, if the scalar transport problem possesses a time-independent solution in Lagrangian coordinates and the boundary conditions are prescribed on a material surface/interface, then the net mass transport is proportional to the diffusion coefficient. This can be also shown to be true for large Péclet number and time-periodic flow fields, i.e., closed pathlines. This agrees with results for heat transfer at high Péclet numbers across closed streamlines. © *2007 American Institute of Physics*.
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I. INTRODUCTION

Molecular diffusion is a fundamental physical process that must be properly accounted for in the analysis of engineering and natural flows. When a substance is present in a moving fluid, it will be transported by the fluid (convection). If molecular diffusion is present, molecules of the substance can move in and out of fluid particles and their location will differ from that of the fluid particles due to random/Brownian motion. For example, dye and smoke are often used as flow tracers and are assumed to "mark" the fluid particles. However, due to molecular diffusion, this marking may be transferred to different fluid particles over time and thus the flow tracing becomes questionable.[1] The flux associated with molecular diffusion is generally assumed to be proportional to the substance concentration gradient and the constant of proportionality is identified as the mass diffusivity, D. Incorporating this basic assumption (gradient transport) to a conservation law in the Eulerian frame leads to the governing equation where the convection and diffusion terms are distinctly identified, i.e., the convection-diffusion equation. The latter simplifies mathematically to the Laplacian for the case of isotropic uniform diffusion. Similar arguments hold for heat transfer.

Einstein[2] was able to obtain the diffusion equation considering the probability density function (pdf) for the position of a Brownian particle. This was later identified as the first term in the Kramers-Moyal expansion of the Chapman-Kolmogorov equation, which is the principal way in which the Fokker-Planck equation enters into physics. Langevin,[3] considering the dynamics of an individual particle and its interaction with nearby particles, formulated the process in an "infinitely more simple" manner by introducing a stochastic force in the ordinary differential equation governing the position of an individual particle. This force is the aggregate interaction force of the particle with other nearby particles. Concerning the initial value problem, i.e., a particle convected/diffusing in an infinite field, the equivalence between the two approaches can be demonstrated through the use of Ito's calculus.[4] Their equivalence ceases when boundary conditions are present.

In this study, we are interested in characterizing convective-diffusive passive scalar (e.g., substance concentration) transport and finding relations that characterize the net spreading or, more "mathematically," the covariance matrix of the position of a "random" (Brownian) particle. It is important to note that the diffusivity tensor $\mathbf{D}$ identifies the local rate of spreading,[5] whereas we are interested in the *effective diffusivity*, or *dispersivity*, which characterizes the macroscopic spreading. A common objective of relevant analyses has been to develop expressions for a dispersion coefficient that accounts for the effect of convection, as reviewed by Isichenco.[6] In general, the term "effective diffusivity" is associated with the initial value problem and is related to the mean-square displacement of a Brownian particle. When referring to the Dirichlet boundary value problem, particle statistics are not applicable, and the term effective diffusivity is used to characterize the net mass transport.

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In the following sections we briefly review the various analyses present in the literature that lead to an effective diffusivity. For the sake of clarity, we have classified the approaches in terms of the mathematical formulation of the problem. Which approach is preferable will depend on the formulation of the specific problem, however, results obtained through different methods should be consistent. For example, for the case of linear flow fields (Sec. IV A 1), the effective diffusivities obtained through the Lagrangian and Eulerian formulations, though not equal, can be related through the deformation gradient tensor.

A. Eulerian coordinates

1. Initial value problems

Foister and Van de Ven[7] consider the Eulerian initial value problem through both the convective diffusion equation for the time-dependent probability density function and the Langevin equations, and demonstrate the equivalence of the two approaches for the case of linear flow fields.

2. Boundary value problems

In contrast to the initial value problem, the presence of boundaries and discontinuities leads to boundary layers that must be considered and matched to the bulk (homogeneous) solution.[8] Referring to the Eulerian convection-diffusion equation, the series expansion of the velocity field about the location of the interface suggests that convection, has a profound effect on the structure of the boundary layer and, consequently, on the net mass transport. The net mass transport is of $\mathcal{O}(\mathrm{Pe}^{-1/2})$ in the case of convection-dominated diffusion,[9] and of $\mathcal{O}(\mathrm{Pe}^{-1/3})$ in the case of shear flow,[10–12] where Pe is the Péclet number which is the ratio of diffusion to convection time scales.

Another elementary boundary value problem is the steady-state heat conduction through a material with multiple constituents in which the thermal diffusivity varies periodically in space. Obtaining a mathematical solution that includes such variations can be very difficult. A simpler alternative is to develop equations that effectively smooth out the substructure variations and replace the heterogeneous diffusivity with an "effective" diffusivity that is homogeneous at large scales, i.e., a homogeneous solution. A naive approach is to define the classical average (arithmetic mean) of the heterogeneous diffusivity as the "homogeneous effective" diffusivity, however, the relation between the two diffusivities is elusive. As shown in Hinch,[13] the effective diffusivity for the one-dimensional heat diffusion with space-dependent diffusivity, is the harmonic average (harmonic mean) of the heterogeneous diffusivity.

3. Initial value problems with multiple length scales

A special class of problems are initial value problems with two length scales: the "long" length scale and the "short" length scale.

In the case of flow through a domain with spatially periodic structure, a domain decomposition technique, the theory of moments, introduced by Taylor[14] and developed by Aris,[15] Horn,[16] and Brenner,[17] is effective and widely used in diffusion problems in porous media. The formulation can address boundary value problems with Neumann boundary conditions specifying the normal derivative of the function on a surface being zero (no flux, impermeable). Because of the impermeable walls, a (Brownian) particle cannot escape from the domain, hence particle statistics (mean, variance) are applicable. In this technique, the average interstitial velocity and the effective dispersivity tensor can be obtained by taking spatial moments of the convection-diffusion equation governing the probability density function of a Brownian particle.

Another technique to obtain the effective diffusivity is to treat the problem by multiple scales analysis in time and space considering an initially slowly varying scalar field.[18] The effect of eddies and boundary layers is incorporated through the multiple scales in space without explicitly identifying boundary conditions. The solvability condition of the second-order problem leads to an expression for the effective diffusivity.

B. Lagrangian coordinates

In Lagrangian (fluid particle) coordinates, if the flow is time periodic the time average of the resulting diffusivity tensor, which is related to the velocity gradient tensor, was referred to as the effective Lagrangian diffusivity.[20] If the scalar field has a time-independent solution then the net mass transport, associated with the boundary value problem, is proportional[19] to the diffusion coefficient D, or more precisely to the inverse of the Péclet number $(1/\mathrm{Pe})$. The approach requires that fluid particles return to their original position after one period (i.e., closed pathlines), the Péclet number is large, and the Dirichlet boundary conditions are prescribed on a material surface/interface. An example satisfying the above conditions is mass transport across the interface of a spherical bubble undergoing volume oscillations in an incompressible liquid.[21,22]

A propagating wave is another example of a mass transfer problem with a dynamic interface. In general, the treatment of the diffusion problem associated with a dynamic interface would lead to a nonlinear, nonorthogonal transformation between Eulerian and Lagrangian coordinates. However, when pathlines coincide with streamlines, i.e., the velocity field can be expressed by a time-independent streamfunction (ψ) and velocity potential (ϕ) , a variance of the von Mises transformation leads to a conformal transformation between the (x,y) and (ϕ,ψ) variables.[23] This was exploited by Witting[24] and Szeri[25] to consider the effect of capillary waves on the enhancement of transport across a wavy interface. In all these examples, the effective diffusivity is proportional to the diffusion coefficient (D). This agrees with results for heat transfer at high Péclet numbers across closed streamlines.[26] The closed streamline pattern inhibits the coupling between convection and diffusion; hence, heat/mass can only escape by diffusing slowly across closed streamlines (stream surfaces) on the large scale.

C. Scope and outline

The objective of the present analysis is to obtain results for the diffusivity and effective diffusivity in Lagrangian coordinates, first considered by Press and Rybicki.[20] We consider both the initial value problem and the boundary value problem. For the latter, the main results are presented in Sec. IV B 1. For the former, we address both the convection-diffusion equation for the time-dependent probability density function and the Langevin equation, and demonstrate their equivalence (Sec. II B). We explicitly determine the effective Lagrangian diffusivity for the case of linear flow fields and we show how the Lagrangian results are related to the Eulerian results and, in particular, to the results obtained by Foister and Van de Ven[7] (Sec. IV A 1). The advantage of the Lagrangian formulation is that the effect of convection is incorporated in the Lagrangian diffusion through the deformation gradient tensor; hence, we are able to infer an effective diffusivity for general flow fields (Sec. IV A 2).

In Sec. II, we present the Eulerian and Lagrangian formulations of the transport equations. In Secs. III and IV, the Lagrangian diffusivity and the effective diffusivity are presented, respectively, and compared. Conclusions are given in Sec. V.

II. ANALYSIS

The physical problem we consider is that of a moving fluid (carrier substance) in which there is a dilute suspension of spherical colloidal particles (contaminant substance). Thus, the suspended particles are larger than the molecules of the carrier fluid but sufficiently small to exhibit Brownian motion. An example is dust or smoke in air. The carrier fluid flow is incompressible. We consider fluid particles to be material elements which can carry and exchange the colloidal particles.

A. Transport in Eulerian coordinates

The transport equation describing the convection and diffusion of a passive scalar, f, is[27]

$$\frac{\partial f}{\partial t}+\mathbf{u}(\mathbf{x},t)\cdot\nabla f=\nabla\cdot\mathbf{D}\cdot\nabla f, \tag{1}$$

where $\mathbf{D}$ is the molecular diffusivity tensor which is symmetric. We assume that the vector field $\mathbf{u}$ is solenoidal (i.e., the flow field is incompressible $\nabla\cdot\mathbf{u}=0$). In index notation, the equation takes the form

$$\frac{\partial f}{\partial t}+u^i(x,t)\frac{\partial f}{\partial x^i}=\frac{\partial}{\partial x^i}\left\{D^{ij}\frac{\partial f}{\partial x^j}\right\}, \tag{2}$$

in Cartesian coordinates.

In the context of Brownian motion, f represents the probability density function of the location of a colloidal particle. Given that the position of the particle is known at the initial time, i.e., the particle is at the origin $\mathbf{x}=\mathbf{0}$, the stochastic process has an initial condition

$$f(t=0)=\delta(\mathbf{x})$$

[where $\delta(\mathbf{x})$ represents the Dirac function] and the homogeneous boundary condition

$$f(\mathbf{x}\to\infty)=0.$$

This formulation is identified as the initial value problem and there is no time-independent asymptotic solution associated with this problem.

If explicit boundary conditions are defined over the boundaries, this is identified as a boundary value problem, and one would expect a time-independent steady-state solution, as the diffusivity tensor is time independent.

1. Langevin equation

The initial value problem is equivalent to an Ito stochastic differential equation (SDE), known as the Langevin equation.[4] The high friction limit (diffusion limit) of the Langevin equation is as follows:[28]

$$\mathrm{d}\mathbf{x}=\mathbf{u}(\mathbf{x},t)\mathrm{d}t+\mathbf{B}\cdot\mathrm{d}\mathbf{W}, \tag{3}$$

where $\mathbf{u}(\mathbf{x},t)$ represents the underlying (carrier) fluid velocity, $\mathbf{W}$ are independent Wiener processes, and the matrix $\mathbf{B}$ is related to the diffusivity tensor through

$$\mathbf{B}=(2\mathbf{D})^{1/2}.$$

It is important to point out that the Langevin equation (3) and the diffusion equation (1) might not be equivalent for any arbitrary tensor $\mathbf{D}(\mathbf{x},t)$, unless the advective velocity is modified by a drift term equal to the divergence of the diffusivity tensor.[29] This is beyond the scope of this work, where we are mostly interested in uniform, isotropic diffusion, i.e., $\mathbf{D}=D\mathbf{I}$, where D is the diffusion coefficient and $\mathbf{I}$ the identity matrix.

B. Transport in Lagrangian coordinates

The problem can be recast in Lagrangian (carrier fluid) coordinates

$$\boldsymbol{\xi}=\boldsymbol{\xi}(\mathbf{x},\tilde{t})\text{ or }\xi_i=\xi_i(x_1,x_2,x_3,\tilde{t})$$

as follows:[27] We let $x_1(\boldsymbol{\xi},t)$, $x_2(\boldsymbol{\xi},t)$, $x_3(\boldsymbol{\xi},t)$ be the location at time t of a fluid particle that was located at (ξ_1,ξ_2,ξ_3) at time $t=0$. Hence, the coordinates (ξ_1,ξ_2,ξ_3) identify a fluid particle. The governing equations for (x_1,x_2,x_3) are

$$\mathbf{u}(x_1,x_2,x_3,t)=\frac{\mathrm{d}\mathbf{x}}{\mathrm{d}t}=\left(\frac{\partial\mathbf{x}}{\mathrm{d}\tilde{t}}(\xi_1,\xi_2,\xi_3,t)\right), \tag{4}$$

with initial conditions ($t=0$), $\mathbf{x}=\boldsymbol{\xi}$. In order to distinguish between time in Eulerian and Lagrangian coordinates a tilde has been used, i.e., $\tilde{t}$. Integration of (4) gives the path of a fluid particle. Alternatively, note that $\boldsymbol{\xi}(\mathbf{y},s)$ identifies the particle passing through the point $\mathbf{y}$ in space, at time $t=s$; for example, $\boldsymbol{\xi}(\mathbf{0},0\leq t\leq s)$ identifies the particles that pass from the origin within the time-frame $0\leq t\leq s$.

The convection-diffusion equations (1) and (2) in Lagrangian coordinates take the form[20,27,30]

$$\frac{\partial f}{\partial \tilde{t}} = \frac{\partial}{\partial \xi^i}\left\{\underbrace{\frac{\partial \xi^i}{\partial x^k}\frac{\partial \xi^j}{\partial x^l}D^{kl}_{\text{lab}}}_{D^{kl}}\frac{\partial f}{\partial \xi^j}\right\}, \tag{5}$$

where D^{kl}_{lab} denotes the components of the diffusivity tensor in Cartesian coordinates (orthonormal laboratory basis) while the term D^{kl} (in underbrace) is the contravariant component of the diffusivity tensor. Note that the convection term has been absorbed in view of the identity

$$\frac{\partial}{\partial \tilde{t}} = \frac{\partial}{\partial t} + \mathbf{u}\cdot\nabla_{\mathbf{x}},$$

i.e., the contravariant component of the diffusivity tensor, the "Lagrangian diffusivity," incorporates the effect of convection.

The diffusion equation can be written in vector notation employing the deformation gradient tensor $\mathbf{F}$ (Ref. 31)

$$\mathbf{F} = \nabla_{\xi}\mathbf{x} = \frac{\partial \mathbf{x}}{\partial \boldsymbol{\xi}}$$

and that the contravariant components of a tensor can be obtained through[32] the transformation $D^{ij}=F^{-1}D_{\text{lab}}F^{-T}$,

$$\frac{\partial f}{\partial \tilde{t}} = \nabla_{\xi}\cdot\{\mathbf{F}^{-1}\cdot\mathbf{D}\cdot\mathbf{F}^{-T}\}\cdot\nabla_{\xi}f, \tag{6}$$

where the superscript $(-T)$ denotes inverse transpose.

In the case of uniform, isotropic diffusion ($\mathbf{D}=D\mathbf{I}$ in the Cartesian system), the right-hand side (RHS) of Eqs. (5) and (6) simplifies to

$$\frac{\partial f}{\partial \tilde{t}} = D\frac{\partial}{\partial \xi^i}\left\{g^{ij}\frac{\partial f}{\partial \xi^j}\right\} \equiv D\nabla_{\xi}\cdot\mathbf{C}^{-1}\cdot\nabla_{\xi}f, \tag{7}$$

where g^{ij} is the contravariant metric tensor. The tensor $\mathbf{C}^{-1}$ is the inverse of the Green (Cauchy-Green) tensor $\mathbf{C}$, which is related to the deformation gradient tensor $\mathbf{F}$ through $\mathbf{C}=\mathbf{F}^T\cdot\mathbf{F}$ (Refs. 31 and 33).

1. Langevin equation

The results of the previous section [Eq. (5)] raise the issue of expressing the Langevin equation (3) in Lagrangian coordinates. We proceed by considering a differential displacement,

$$\mathrm{d}x^i(\xi^j,t) = \frac{\partial x^i}{\partial \xi^j}\mathrm{d}\xi^j + \frac{\partial x^i}{\partial t}\mathrm{d}t, \tag{8}$$

where the first term on the RHS represents the displacement associated with a change in fluid particle and the second term represents the displacement associated with a change in time. In view of Eq. (4) and the Langevin equation (3), (8) can be further manipulated to

$$\mathrm{d}x^i(\xi^j,t) = \frac{\partial x^i}{\partial \xi^j}\mathrm{d}\xi^j + u^i\,\mathrm{d}t = u^i\,\mathrm{d}t + B^i_j\,\mathrm{d}W^j.$$

The equation leads to the Langevin equation in Lagrangian coordinates

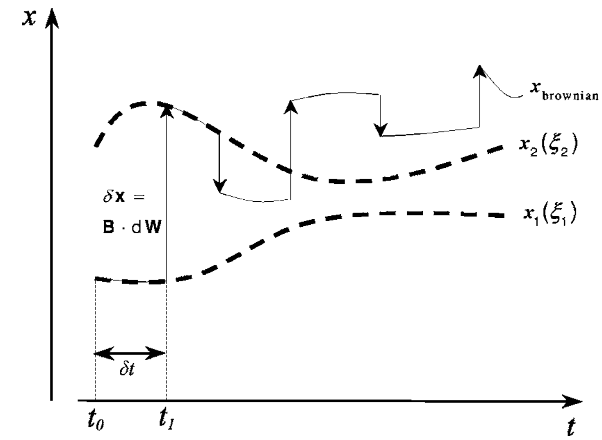

FIG. 1. A Brownian particle (thin line) jumping across fluid particles (thick lines).

$$\mathrm{d}\xi^i = \frac{\partial \xi^j}{\partial x^i}B^i_k\mathrm{d}W^k. \tag{9}$$

A physical interpretation of this equation can be obtained as follows, referring to Fig. 1. Consider two fluid particles, $\boldsymbol{\xi}_1$ and $\boldsymbol{\xi}_2$. At time t_1 the Brownian particle $\mathbf{x}_b$ takes a step $\delta\mathbf{x}$ equal to $\mathbf{B}\cdot\mathrm{d}\mathbf{W}$. The small displacement vector $\delta\mathbf{x}$ can be expressed as

$$\delta\mathbf{x} = \mathbf{x}(\boldsymbol{\xi}_2) - \mathbf{x}(\boldsymbol{\xi}_1) = \mathbf{x}(\boldsymbol{\xi}+\delta\boldsymbol{\xi}) - \mathbf{x}(\boldsymbol{\xi}) = \mathbf{F}\cdot\delta\boldsymbol{\xi},$$

which leads to the vector form of Eq. (9),

$$\mathrm{d}\boldsymbol{\xi} = \mathbf{F}^{-1}\cdot\mathbf{B}\cdot\mathrm{d}\mathbf{W}. \tag{10}$$

The Langevin equation in Lagrangian coordinates can be identified as the convected derivative[27] of the Lagrangian coordinate. This is identically zero for the case that there is no diffusion.

For isotropic, uniform diffusion the equivalence between (9) and (5) can be obtained as follows. The SDE (9) is equivalent to the Fokker-Planck equation,[34]

$$\frac{\partial f}{\partial \tilde{t}} = \frac{B^2}{2}[g^{ij}f]_{,ij},$$

where the subscript $,ij$ denotes covariant differentiation (covariant differentiation reduces to the partial derivative in Cartesian coordinates). In view of the identity $g^{ij}_{,i}=0$, the above equation simplifies to

$$\frac{\partial f}{\partial \tilde{t}} = D[g^{ij}f_{,i}]_{,j}$$

and finally to[27]

$$\frac{\partial f}{\partial \tilde{t}} = D\frac{\partial}{\partial \xi^i}\left\{g^{ij}\frac{\partial f}{\partial \xi^j}\right\}.$$

Similar to the Eulerian formulation, the equivalence between the two formulations, Fokker-Planck and Langevin, is limited to the initial value problem.

Furthermore, in the Lagrangian coordinate frame, the spatial variability of the diffusivity tensor, manifested through the contravariant metric g^{ij}, might suggest a drift velocity[29] in the Langevin equations (9) and (10). However, as mentioned in the proof earlier, the covariant divergence of the metric tensor g^{ij} is equal to zero, i.e., $\nabla \cdot \mathbf{C}^{-1} = g^{ij}_{,i} = 0$.

III. LAGRANGIAN DIFFUSIVITY

In view of the diffusion equation in Lagrangian coordinates (6), it is tempting to identify the tensor $\mathbf{D}^L = \mathbf{F}^{-1} \cdot \mathbf{D} \cdot \mathbf{F}^{-T}$ as the Lagrangian diffusivity tensor.[20] To reveal the physical significance of the tensor, consider the case of uniform isotropic diffusion, where the term simplifies to the contravariant metric tensor [see Eqs. (7)], which is the inverse of the Green or Cauchy-Green tensor. Physically this tensor gives the square of the relative change of an element of surface area,[31]

$$\hat{\mathbf{n}}' \cdot \mathbf{C}^{-1} \cdot \hat{\mathbf{n}}' = \frac{\mathrm{d}\mathbf{S} \cdot \mathrm{d}\mathbf{S}}{\mathrm{d}\mathbf{S}' \cdot \mathrm{d}\mathbf{S}'},$$

where $\mathrm{d}\mathbf{S}'$ is the initial area of a surface element (undeformed state) and $\hat{\mathbf{n}}'$ the normal to the interface in the un-deformed state.

In the context of finite deformation theory the Cauchy-Green[33] tensor $\mathbf{C}$ is related to the Green-Lagrange strain tensor $\mathbf{E}$ through

$$\mathbf{E} = \tfrac{1}{2}(\mathbf{C} - \mathbf{I}).$$

Hence, for small deformations, the inverse of the Cauchy-Green tensor $\mathbf{C}^{-1}$ is related to the infinitesimal strain tensor $\boldsymbol{\varepsilon}$ through

$$\mathbf{C}^{-1} \approx \mathbf{I} - 2\boldsymbol{\varepsilon}.$$

A. Scalar flux in Lagrangian coordinates

The mass flux across an element of a surface area is given by

$$\frac{\mathrm{d}\,\delta m}{\mathrm{d}t} = D\,\nabla f \cdot \mathrm{d}\mathbf{S}, \tag{11}$$

where $\mathrm{d}\mathbf{S}$ is associated with an element of the surface/interface. In Lagrangian coordinates, and in view of incompressibility, this can be expressed as[32]

$$\frac{\mathrm{d}\,\delta m}{\mathrm{d}t} = D\frac{\partial f}{\partial \xi^j}\mathbf{g}^j \cdot n_i' \mathbf{g}^i\, \mathrm{d}S' = D g^{ij} \frac{\partial f}{\partial \xi^j} n_i'\, \mathrm{d}S'.$$

Hence, in agreement with the diffusion equation (5), the appearance of the factor g^{ij} suggests that the rate of mass transport from an element of a material interface is related to the square of the relative change of the surface area. More precisely, if a material interface is a scalar isosurface then the following simplifications are applicable, $\partial f / \partial \xi^j = \alpha n_j'$, because the normal vector points in the contravariant direction. For this simple case, the mass flux is given by

$$\frac{\mathrm{d}\ \ \delta m / \delta S'}{\mathrm{d}t} = D\alpha n_i' g^{ij} n_j' = D\alpha(\hat{\mathbf{n}}' \cdot \mathbf{C}^{-1} \cdot \hat{\mathbf{n}}'), \tag{12}$$

where the term in parentheses is precisely the definition of the relative local area change squared.

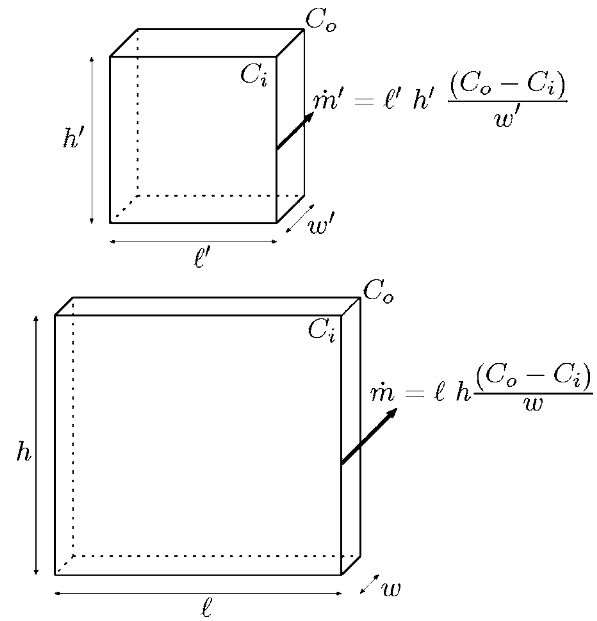

FIG. 2. A simple model that demonstrates the physical interpretation of the convection-enhanced diffusion process. The inner surface is at uniform concentration C_i and the outer surface at a uniform concentration C_o. The surface expands rapidly such that diffusion has no time to occur. This would lead to a steeper concentration gradient which, along with the increase of the surface area $(\ell h / \ell' h')$, would lead to a twofold increase of mass transfer rates.

In Fig. 2, we illustrate through a simple argument the relation of the mass transport through a material interface to the relative change of its area. An initial volume element with dimensions h', ℓ', w' expands to the new dimensions h, ℓ, w. The expansion is fast enough such that diffusive mass transfer has no time to occur. The ratio between the mass transport rates is equal to

$$\frac{\dot{m}}{\dot{m}'} = \frac{\ell h \frac{\Delta C}{w}}{\ell' h' \frac{\Delta C}{w'}} = \frac{\ell h w'}{\ell' h' w} = \left(\frac{w'}{w}\right)^2,$$

where $\ell h w = \ell' h' w'$ as a result of incompressibility. The quantity $w'/w = \ell h / \ell' h'$ is the relative change in surface area.

Physically, the effect of a dynamic interface on the diffusion process is twofold: an increase/decrease of the surface area and a steepening/smoothing of the concentration gradient. The latter is a consequence of the movement of fluid particles that are close to the interface in order to satisfy incompressibility (volume conservation), i.e., particles move closer to an expanding interface resulting in steeper concentration gradients; the opposite holds for a contracting interface.

B. The Lagrangian covariance tensor $\langle \xi \cdot \xi^T \rangle$

The Eulerian pdf $f(\mathbf{x},t)$ and the Lagrangian pdf $\phi(\boldsymbol{\xi},t)$ are related through $\phi(\boldsymbol{\xi},t) \equiv |F| f(\mathbf{x},t)$ (Refs. 35 and 36), hence they are equivalent in view of incompressibility, i.e., $\phi(\boldsymbol{\xi},t) \equiv f(\mathbf{x}(\boldsymbol{\xi},t),t)$. Furthermore, without loss of generality we can assume that the reference particle is at the origin of the coordinate system, hence its mean $\langle \boldsymbol{\xi} \rangle = \langle \mathbf{x} \rangle = 0$. Consequently, the second moment tensor, covariance tensor, correlation tensor, and mean-square displacement tensor can be used interchangeably.

The diffusion tensor $\mathbf{D}$ gives the local rate of the mean-square displacement at the point occupied by the particle,[5] i.e.,

$$\mathbf{D} = \frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\langle \mathbf{x}\cdot\mathbf{x}^T\rangle = \lim_{f(\mathbf{x},t)\to\delta(\mathbf{x})}\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\int \mathbf{x}\cdot\mathbf{x}^T f(\mathbf{x},t)\mathrm{d}\mathbf{x}.$$

Similarly, the Lagrangian diffusivity tensor can be defined as

$$\mathbf{D}^L = \frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\langle \boldsymbol{\xi}\cdot\boldsymbol{\xi}^T\rangle = \lim_{f(\boldsymbol{\xi},t)\to\delta(\boldsymbol{\xi})}\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\int \boldsymbol{\xi}\cdot\boldsymbol{\xi}^T \phi(\boldsymbol{\xi},t)\mathrm{d}\boldsymbol{\xi}.$$

The mean-squared Lagrangian displacement tensor, $\langle\boldsymbol{\xi}\cdot\boldsymbol{\xi}^T\rangle$, can be obtained, employing the Ito rules,[4] to Eq. (10),

$$\mathrm{d}\langle\boldsymbol{\xi}\cdot\boldsymbol{\xi}^T\rangle = B^2\langle\mathbf{F}^{-1}\cdot\mathbf{F}^{-T}\rangle\mathrm{d}t, \tag{13}$$

where angle brackets denote ensemble average. When there is no motion the deformation gradient tensor $\mathbf{F}=\mathbf{I}$ and above expression simplifies to the classical result $\langle\boldsymbol{\xi}\cdot\boldsymbol{\xi}^T\rangle=\langle\mathbf{x}\cdot\mathbf{x}^T\rangle = B^2t\mathbf{I}=2Dt\mathbf{I}$. We can also "claim" that a physical interpretation to the equation of the mean-square displacement [Eqs. (13) and (19)] is that it is related to the net mass transport across a material isosurface [Eq. (12)].

A differential equation for $\mathbf{C}^{-1}=\mathbf{F}^{-1}\cdot\mathbf{F}^{-T}$ can be obtained through the matrix differential equation relating $\mathbf{F}$ with the velocity gradient tensor $\nabla\mathbf{u}$ (Refs. 30 and 33),

$$\frac{\mathrm{d}\mathbf{F}}{\mathrm{d}t} = \nabla\mathbf{u}\cdot\mathbf{F}, \tag{14}$$

with the initial condition $\mathbf{F}(t=0)=\mathbf{I}$. Using the properties of the inverse and transpose of matrix differential equations we obtain

$$\frac{\mathrm{d}\mathbf{C}^{-1}}{\mathrm{d}t} = -2\mathbf{F}^{-1}\cdot\mathbf{S}\cdot\mathbf{F}^{-T},$$

where $\mathbf{S}$ is the rate of deformation tensor defined as

$$\mathbf{S} = \frac{1}{2}(\nabla\mathbf{u}+\nabla\mathbf{u}^T).$$

IV. EFFECTIVE DIFFUSIVITY

The classical result by Einstein states that the second moment of the position of a Brownian particle, $\langle x^2\rangle$, is related to the diffusion coefficient through[4] $\langle x^2\rangle=2Dt$. When there is no mean displacement for the particle, the mean-squared displacement tensor $\langle\mathbf{x}\cdot\mathbf{x}^{\mathbf{T}}\rangle$ identifies the dispersion of the Brownian particle through diffusive-convective coupling and its time derivative

$$\mathbf{D}^* = \lim_{t\to\infty}\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\langle\mathbf{x}\cdot\mathbf{x}^T\rangle = \lim_{t\to\infty}\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\int\mathbf{x}\cdot\mathbf{x}^T f(\mathbf{x},t)\mathrm{d}\mathbf{x}$$

is termed the dispersivity[17,37] and can be identified as an effective diffusion coefficient. In a similar manner the effective Lagrangian diffusivity can be defined,

$$\mathbf{D}^{L*} = \lim_{t\to\infty}\frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\langle\boldsymbol{\xi}\cdot\boldsymbol{\xi}^T\rangle = \lim_{t\to\infty}\tfrac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\int\boldsymbol{\xi}\cdot\boldsymbol{\xi}^T\phi(\boldsymbol{\xi},t)\mathrm{d}\boldsymbol{\xi}.$$

Clearly, the definition of the effective diffusivities does not allow the presence of general boundary conditions as the particle under consideration must remain in the domain. However, no flux (impermeable) boundary conditions are allowed as they do not invalidate the above definitions.

A. Lagrangian versus Eulerian effective diffusivities

As mentioned earlier, in view of incompressibility, the Lagrangian and Eulerian pdfs are equivalent. However, what would be more significant is to relate the spatial moments between the two coordinate systems. In order to proceed we take some particular point as the origin of the coordinate system (identifying a fluid particle) and relate the two coordinate systems using a Taylor series[38] about the origin ($\boldsymbol{\xi}=0$),

$$\mathbf{x}(\boldsymbol{\xi},t) = \mathbf{x}(t)+\mathbf{F}(t)\cdot\boldsymbol{\xi}+\tfrac{1}{2}\boldsymbol{\xi}^T\cdot\nabla\mathbf{F}(t)\cdot\boldsymbol{\xi}+\text{h.o.t.} \tag{15}$$

Higher-order terms can be omitted in the series expansion as they would contribute higher-order moments in the ensemble average of $\langle\mathbf{x}\cdot\mathbf{x}^T\rangle$ (moments higher than the second would be small as we can assume that displacements would have a Gaussian distribution). In addition, we anticipate that $\boldsymbol{\xi}$ would be proportional to $\sqrt{D}$ [Eq. (10)], hence for small diffusivity (D) higher-order moments are insignificant as they are proportional to higher powers of D. Using the above expansion [Eq. (15)], in the next section we compare the Lagrangian and Eulerian effective diffusivities for linear shear fields.

1. Diffusion in linear shear fields

Two-dimensional linear shear fields can be expressed[7] as

$$\mathbf{u}^T = (u_x,u_y) = (Gy,\alpha Gx),$$

where G is the shear rate and the parameter α may range from -1 to $+1$. The velocity gradient tensor is the constant matrix

$$\nabla\mathbf{u} = \begin{pmatrix} 0 & G \\ \alpha G & 0 \end{pmatrix}$$

and the deformation gradient tensor, $\mathbf{F}$, can be obtained by employing the (matrix) differential [Eq. (14)]

$$\mathbf{F} = e^{\nabla\mathbf{u}t}.$$

Hence, the contravariant metric tensor, g^{ij}, can be expressed in terms of the velocity gradient tensor,

$$g^{ij} = \mathbf{F}^{-1}\cdot(\mathbf{F}^{-1})^T = e^{-(\nabla\mathbf{u})t}\cdot e^{-(\nabla\mathbf{u}^T)t}, \tag{16}$$

which is independent of $\boldsymbol{\xi}$.

When we substitute the result for the metric tensor [Eq. (16)] in the covariance [Eq. (13)] we obtain the final result for the mean-squared displacement tensor in Lagrangian coordinates

$$\langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle = 2D \int e^{-(\nabla \mathbf{u})t} \cdot e^{-(\nabla \mathbf{u}^T)t} \mathrm{d}t.$$

In particular, for the three special cases by Foister *et al.*[7] we obtain

- Pure rotation $(\alpha = -1)$

$$\langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle = \begin{pmatrix} 2Dt & 0 \\ 0 & 2Dt \end{pmatrix};$$

- Simple shear $(\alpha = 0)$

$$\langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle = \begin{pmatrix} 2Dt\left(1 + \frac{1}{3}(Gt)^2\right) & -DGt^2 \\ -DGt^2 & 2Dt \end{pmatrix};$$

- Pure elongation $(\alpha = 1)$

$$\langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle = \begin{pmatrix} \dfrac{D}{G}\sinh(2Gt) & \dfrac{D}{G}(1 - \cosh(2Gt)) \\ \dfrac{D}{G}(1 - \cosh(2Gt)) & \dfrac{D}{G}\sinh(2Gt) \end{pmatrix}.$$

We should point out that since the deformation gradient tensor depends only on time, there is no clear distinction between the Lagrangian diffusivity and effective Lagrangian diffusivity. The important point is, however, that, unlike Eulerian coordinates, in Lagrangian coordinates the effective diffusivity is identified explicitly. A physical explanation of the effect of convection on the mean-square displacement (in Lagrangian coordinates) of a Brownian particle can be provided using the arguments of Sec. III A. When a Brownian particle takes a step along the "compressing" direction, it would end up further away due to the crowding up of the fluid particles in this direction. The contrary holds in the "stretching" direction.

The agreement between these results and the results obtained by Foister *et al.*[7] (Eulerian coordinates) can be verified through the use of Eq. (17). For the linear shear fields considered, $\nabla \mathbf{F} = 0$, along with its higher derivatives, and an exact expression, can be obtained for the covariance tensor in Eulerian coordinates in terms of the Lagrangian covariance matrix

$$\begin{aligned} \langle \mathbf{x} \cdot \mathbf{x}^T \rangle &= \langle [\mathbf{x}(t) + \mathbf{F}(t) \cdot \boldsymbol{\xi}] \cdot [\mathbf{x}^T(t) + \boldsymbol{\xi}^T \cdot \mathbf{F}^T(t)] \rangle \\ &= \mathbf{F}(t) \cdot \langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle \cdot \mathbf{F}^T(t). \end{aligned} \tag{17}$$

The last expression is obtained through the simplifications $\mathbf{x}(t) = 0$ and $\langle \boldsymbol{\xi} \rangle = 0$ and justifies the equivalence between the Lagrangian effective diffusivity (present results) and the Eulerian effective diffusivity obtained by Foister and Van De Ven.[7]

2. Turbulent flow fields

For the case of no diffusion, i.e., $D = 0$, the solution for $\boldsymbol{\xi}$ obtained from Eq. (9) is the trivial solution $\boldsymbol{\xi} = 0$, hence, its moments are identically equal to zero. If, however, the velocity field is turbulent, i.e., the velocity $\mathbf{u}$ is a non-delta-correlated, stationary, random variable, the equations relating the Eulerian and Lagrangian coordinates (4) should be interpreted in the Stratonovitch sense,[4] and the mean-square displacement is obtained through

$$\overline{\mathbf{x} \cdot \mathbf{x}^T} = \overline{\mathbf{x}(t) \cdot \mathbf{x}^T(t)},$$

where the overbar denotes an average over the turbulent ensemble[39] and characterizes turbulent dispersion.[40] In view of the expression above and Eq. (17), it is tempting to infer that the two dominant terms characterizing the turbulent scalar transport are

$$\langle \overline{\mathbf{x} \cdot \mathbf{x}^T} \rangle \approx \overline{\mathbf{x}(t) \cdot \mathbf{x}^T(t)} + \overline{\mathbf{F}(t) \cdot \langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle \cdot \mathbf{F}^T(t)} + \text{h.o.t.},$$

where $\langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle$ is given in Eq. (13). One can assume that the Brownian motion and the macroscopic motion are statistically uncorrelated.[39]

B. Effective Lagrangian diffusivity at large Péclet and time-periodic flow fields

The Péclet number $\mathrm{Pe} = l^2/(t_c D)$ is the ratio of diffusion to convection time scales, where l and t_c are the characteristic length and time scales, respectively. In Sec. IV A 1, it was shown that for the homogeneous initial value problem associated with linear shear fields, both the effective Lagrangian and Eulerian diffusivities are proportional to the diffusivity D (more precisely inversely proportional to the Péclet number).

In what follows, we will present asymptotic results for the effective diffusivity at large Péclet for both the initial and boundary value problem. We will restrict our analysis to the convection-diffusion process associated with time-periodic flow fields, i.e., fluid particles with closed trajectories.

1. Convection-diffusion equation

In the limit of small diffusion, i.e., large Péclet number, and time-periodic flow field the diffusion problem can be addressed using multiple scales in time.[13,22] We expect that convection will dominate the process, hence a fluid particle will retain its scalar property while diffusion will evolve in the slow time scale $\lambda = \tilde{t}/\mathrm{Pe}$. The elimination of secular behavior in the first-order problem leads to an average equation governing the zeroth-order problem,

$$\frac{\partial f^0}{\partial t} = \frac{1}{\mathrm{Pe}} \frac{\partial}{\partial \xi^i} \left\{ \overline{g^{ij}(\boldsymbol{\xi}, t)} \frac{\partial f^0}{\partial \xi^j} \right\}, \tag{18}$$

where the overbar denotes time average, which requires that fluid particles return to their original position after one period. Press and Rybicki[20] argue that, in general, the analysis is valid provided the motion of the fluid has no mean component of secular drift.

Here, we should clearly distinguish between the boundary value problem and the initial value problem. Associated with the boundary value problem, in view of the fact that the coefficients are not time dependent, there is a time-independent solution for f^0 provided that the Dirichlet boundary conditions are compatible with the Lagrangian formulation, i.e., the boundary conditions should be prescribed on a material interface. Based on this formulation, we can conclude that the net mass flux is proportional to Pe^{-1}, a result in agreement with other studies dealing with convective transport at high Péclet numbers both across closed pathlines[21,22] and streamlines.[26] The rationale here is that the transport process is diffusion controlled because the closed pathline pattern inhibits the coupling between convection and diffusion; the mass escapes by diffusing slowly across closed pathlines (material surfaces).

Regarding the initial value problem the effective diffusivity may not be proportional to Pe^{-1} except of course if the Lagrangian diffusivity is a function only of time.[20] This is clarified further in the next section.

2. Langevin equation

The initial value problem is the fundamental solution of the diffusion equation for an impulse at time $t=0$. Because the formulation of this problem is equivalent to the Langevin equation (3), the mean-square displacement can be also obtained through

$$\frac{\mathrm{d}\langle \boldsymbol{\xi} \cdot \boldsymbol{\xi}^T \rangle}{\mathrm{d}t} = B^2 \langle \mathbf{F}^{-1} \cdot (\mathbf{F}^{-1})^T \rangle \equiv 2D \langle g^{ij}(\boldsymbol{\xi},t) \rangle. \tag{19}$$

The requirement of closed pathlines implies that the tensor g^{ij} is periodic in time. In the limit of small diffusion, i.e., $D \ll 1$, the associated average system is[41]

$$\frac{\mathrm{d}\langle \xi_i \xi_j \rangle}{\mathrm{d}t} = 2D \langle \overline{g^{ij}(\boldsymbol{\xi},t)} \rangle, \tag{20}$$

which renders the RHS of the equation independent of time giving, locally, the average square relative increase/decrease of the surface area. If $g^{ij}(\boldsymbol{\xi},t)$ is independent of $\boldsymbol{\xi}$ clearly the effective diffusivity is proportional to the diffusivity[20] (D). This agrees with the averaged form of the Lagrangian convection-diffusion equation (18) and the conclusions of the previous section.

V. CONCLUSIONS

We have examined the general problem of convection diffusion of a passive scalar in Lagrangian coordinates, i.e., in a coordinate system defined with respect to fluid particles. We have also developed a description of Brownian motion of spherical colloidal particles in Lagrangian coordinates for the case of dilute suspensions. More precisely, we have obtained the "diffusion limit," high-friction limit, of the Langevin description of the diffusion process in Lagrangian coordinates and show that this is equivalent to the passive scalar transport equation in Lagrangian coordinates. The resulting diffusivity tensor is defined as the Lagrangian diffusivity which, for uniform, isotropic diffusion is proportional to the contravariant metric tensor. This tensor gives the square of the relative change of an element of surface area (inverse of Green tensor) and characterizes the mass transport rate across an isoscalar material surface/interface. The two diffusivity tensors, Eulerian and Lagrangian, are related through the deformation gradient tensor.

We have distinguished the physical meaning between the Lagrangian diffusivity and the *effective* Lagrangian diffusivity. Similar to diffusion in Eulerian coordinates, the former identifies the *local* rate of spreading whereas the latter characterizes the macroscopic spreading. In general, when dealing with the initial value problem, the effective diffusivity is obtained through the statistics of a Brownian particle whereas when referring to the Dirichlet boundary value problem, the effective diffusivity characterizes the net mass transport.

Associated with the initial value problem, we determine that the effective diffusivities in Eulerian and Lagrangian coordinate systems can be related through the deformation gradient tensor to a first approximation. We validate this for the case of linear flow fields and infer a relation for turbulent flow fields. In general, the effective diffusivity associated with the initial value problem is not proportional to the diffusion coefficient except, for example, for the case of linear flow fields. The results can be justified using both the convection-diffusion equation and the Langevin equation.

Associated with the boundary value problem, if the scalar transport problem possesses a time-independent solution in Lagrangian coordinates, then the net mass transport is proportional to the diffusion coefficient. For large Péclet number and time-periodic flow fields, i.e., fluid particles return to their original position (closed pathlines), we formulate an equivalent, autonomous problem in Lagrangian coordinates through multiple-scales analysis. Hence, the net mass flux across a material surface is proportional to the diffusion coefficient because there is a time-independent solution associated with the averaged convection-diffusion equation. However, the boundary conditions should be consistent with the Lagrangian formulation, i.e., they should be prescribed on a material surface/interface.

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