Heat and mass transport in nonhomogeneous random velocity fields

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The effective equation describing the transport of passive tracers in nonsolenoidal velocity fields is determined, assuming that the velocity field $\mathbf{U}(\mathbf{r},t)$ is a function of both position $\mathbf{r}$ and time t, albeit remaining locally random. Assuming a strong separation of scales and applying the method of homogenization, we find a Fickian constitutive relation for the coarse-grained particle flux, as the sum of a convective part, $\mathbf{V}_E\overline{c}$, and a diffusive term, $-\mathbf{D}^s\cdot\nabla\overline{c}$, where $\mathbf{V}_E$ is the Eulerian mean tracer velocity, $\overline{c}$ the average particle concentration, and $\mathbf{D}^s$ the effective diffusivity. The latter can be written as $\mathbf{D}^s(\mathbf{r},t)=D_0\mathbf{I}+\mathbf{D}(\mathbf{r},\mathbf{r},t)$, where D_0 is the molecular diffusivity, $\mathbf{I}$ the unit dyadic and $\mathbf{D}(\mathbf{r}_1,\mathbf{r}_2,t)$ the cross diffusion dyadic. Conversely, the Eulerian mean velocity $\mathbf{V}_E(\mathbf{r},t)$ is the sum of the microscale mean tracer velocity $\overline{\mathbf{V}}(\mathbf{r},t)$ and a particle drift velocity, $\mathbf{V}^d(\mathbf{r},t)=-[(\partial/\partial\mathbf{r}_2)\cdot\mathbf{D}^T(\mathbf{r},\mathbf{r}_2,t)]_{\mathbf{r}_2=\mathbf{r}}$, which depends on the nonhomogeneity of the velocity field at the macroscale. The microscale mean particle velocity, in turn, is the sum of the mean fluid velocity and the ballistic tracer velocity, which is due to the local nonuniformity of the concentration field and is therefore structurally different from the tracer drift velocity. In the limit of large Peclet numbers, $\mathbf{D}^s$ coincides with the self-diffusion dyadic, as it measures the local temporal growth of the mean square displacement of a tracer particle from its average position. In this case, the motion of a tracer particle is a random process in the manner of Stratonovich, where the smoothly varying mean tracer velocity equals the microscale mean tracer velocity and the fluctuating term is described through the cross diffusion dyadic $\mathbf{D}(\mathbf{r}_1,\mathbf{r}_2,t)$.

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I. INTRODUCTION

The objective of this paper is to determine the constitutive relation for the heat or mass flux in random velocity fields. Applications can be found in the heat and mass transport in packed beds or in turbulent mixers where the randomness of the velocity field is due, in the first case, to the random distribution of the bed particles and, in the second case, to the turbulent nature of the flow. As is customary in these cases, our primary interest is not the detailed knowledge of the microscale process, but rather its description on a coarse scale, where we expect that it is described through an effective-medium equation and constitutive relation in terms of effective parameters (such as the effective heat and mass diffusivities), which depend on the global characteristics of the microscale velocity field.

Previously, this problem has been studied for solenoidal velocity fields, assuming that the velocity field is locally random and macroscopically both uniform and stationary. In this case, the transport of passive tracers has been shown to be described through a Fickian constitutive relation, as the sum of a convective part, characterized by the average fluid velocity, and a diffusive component, with an effective diffusivity that depends on the microscopic, local characteristic of the flow field. For example, when the randomness of the velocity field is due to turbulence, the average tracer velocity equals the fluid mean velocity, while the effective, or eddy, diffusivity is the time integral (when it is finite) of the fluid velocity autocorrelation function (see Monin and Yaglom [1], and references therein). More recently, this problem was studied also by Biferale *et al.* [2] and by Castiglione *et al.* [3,4], who analyzed standard and anomalous transport in incompressible flow using multiscale techniques. Similar results are found in the related process of convection of a passive tracer by a fluid flowing through a random medium, even if, as mentioned above, in this case the randomness of the flow is provided not by turbulence (in fact, the fluid flow can very well be laminar), but by its interaction with the dispersed particles (see Refs. [5–13]). Another similar case is the transport of Brownian particles convected by a nonhomogeneous laminar flow field, where, again, an effective, so called Taylor-Aris, diffusivity arises as the result of the interaction between the molecular diffusion of the particles and the nonhomogeneity of the flow field (see Refs. [14–17]).

In this work, we will remove the assumptions that the flow field is solenoidal and that it is macroscopically homogeneous and stationary. Nonsolenoidal velocity fields not only refer to compressible fluids, although this is the case that we will consider here; they also apply to suspensions of particles where either particle-particle interactions or particle inertial forces cause the particle velocity to differ from the local fluid velocity. Nonsolenoidal flow fields were studied by Vergassola and Avellaneda [18], who found that in this case the mean tracer velocity differs from the mean fluid velocity by a term that accounts for what they call ballistic transport, due to the nonuniformity of the concentration field at the microscale. An identical phenomenon is observed also in the transport of chemically reacting solute tracers, as shown by Shapiro and Brenner [19] and Mauri [20] (see the following section). More important is our other assumption that the flow field is macroscopically nonhomogeneous and nonstationary, a condition that can be due, for example, to

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the characteristics of the unperturbed flow field, as in the case of time-dependent turbulent shear flow, or to the nonuniform properties of the random flow field, as in the flow through a porous medium with varying porosity, so that we can describe conditions that are more readily found in practical applications

The content of this work is organized as follows. After formulating the problem and establishing the basic scaling (see Sec. II), in Secs. III and IV we describe the method of solution and derive the effective constitutive equation. The case of large Peclet number is studied in detail in Sec. V, while, eventually, in Sec. VI, the main results of this work are summarized and discussed.

II. GOVERNING EQUATION AND SCALING

Consider the convection of Brownian tracers in a velocity flow field $\mathbf{U}(\mathbf{r},t)$. Neglecting inertia and all interactions among the particles, the tracer molar concentration $c(\mathbf{r},t)$ at location $\mathbf{r}$ and time t satisfies the following convection-diffusion equation:

$$\frac{\partial c}{\partial t}+\nabla\cdot(\mathbf{U}c)-D_0\nabla^2 c=0, \tag{1}$$

where D_0 is the tracer molecular diffusivity, to be solved with a given initial condition,

$$c(\mathbf{r},0)=c_0(\mathbf{r}). \tag{2}$$

The velocity field $\mathbf{U}(\mathbf{r},t)$ is nonsolenoidal (i.e., the fluid is compressible), with known statistical properties. In particular, it has the mean value

$$\langle \mathbf{U}(\mathbf{r},t)\rangle_0=\overline{\mathbf{U}}(\mathbf{r},t)=U_0\overline{\mathbf{u}}(\mathbf{r},t) \tag{3}$$

and Lagrangian velocity autocorrelation function

$$\langle \tilde{\mathbf{U}}(\mathbf{r},t)\tilde{\mathbf{U}}(\mathbf{r}+\tilde{\mathbf{r}},t+\tilde{t})\rangle_0=U_0^2\mathbf{f}(\mathbf{r},t,\tilde{t}), \tag{4}$$

where U_0 denotes a typical value of the fluid velocity, the brackets indicate ensemble average, $\tilde{\mathbf{U}}=\mathbf{U}-\overline{\mathbf{U}}$ is the velocity fluctuation, while $\mathbf{r}+\tilde{\mathbf{r}}$ is the position, at time $t+\tilde{t}$, of the fluid particle which, at time t, is located at $\mathbf{r}$. Equations (3) and (4) indicate that the velocity field is not stationary in time, nor homogeneous in space. Clearly, although the Lagrangian velocity autocorrelation can be extracted from numerical simulations of fluid flows in turbulent mixers or in packed beds, a direct experimental measurement of this function is not feasible, in general. However, it should be remarked that, since the Eulerian and Lagrangian probability distribution functions of the velocity fluctuations are related to one another (see Ref. [21]), it is possible to determine the Lagrangian velocity autocorrelation function from its Eulerian counterpart, which is more easily measurable.

Rewriting the governing equation (1) as

$$\frac{\partial c}{\partial t}+\mathbf{U}\cdot\nabla c+rc=D_0\nabla^2 c, \tag{5}$$

where $r=\nabla\cdot\mathbf{U}$, we see that fluid compressibility plays the role of a first-order chemical reaction. Therefore, when the random velocity field is stationary in time and homogeneous in space, mass transport in compressible flow fields is a particular case of the transport of chemically reacting tracers (see Shapiro and Brenner [19], Mauri [20], Edwards *et al.* [22]).

Here, we assume that the mean velocity field has characteristic length L and characteristic time $\tau_L=L^2/D_0$, while the velocity autocorrelation function decays over a length scale $\ell=\epsilon L$ and time scale $\tau_\ell=\ell^2/D_0=\epsilon^2\tau_L$, with $\epsilon\ll 1$. Therefore, the geometry of the problem is characterized by two length scales, ℓ and L, and two time scales, τ_ℓ and τ_L, indicating the typical correlation length and time of the velocity field at the microscale and macroscale, respectively. In the following, we will assume that $\mathrm{Pe}=O(1)$, where $\mathrm{Pe}=\ell U_0/D_0$ is the microscale Peclet number. This assumption means that convection and diffusion balance each other at the microscale and therefore convection dominates diffusion at the macroscale, as $\mathrm{Pe}_L=LU_0/D_0=O(1/\epsilon)$.

Clearly, this problem, in principle, could be solved exactly, provided that the velocity field were known. In reality, the velocity field is known only statistically and, in addition, we are not interested in the detailed knowledge of the microscale process, but rather in its description on a coarse scale. In fact, the macroscopic effective equations describing the motion of the test particle will be determined using a multiple-scale perturbation analysis otherwise denoted as method of homogenization (Bensoussan *et al.* [6], Sanchez-Palencia [23]). The main idea is that the effective equation is expected to arise naturally from Eq. (1) through a regular perturbation analysis in terms of the small parameter $\epsilon=\ell/L$. Accordingly, each quantity can be represented separately in terms of a microscale coordinate $\tilde{\mathbf{r}}$, with $|\tilde{\mathbf{r}}|=O(\ell)$ and of a macroscale position vector $\mathbf{r}$, with $|\mathbf{r}|=O(L)=O(\ell/\epsilon)$. In particular, the gradient operator ∇ can be expanded in terms of ϵ as

$$\ell\nabla=\frac{\partial}{\partial\mathbf{x}}+\epsilon\frac{\partial}{\partial\mathbf{X}}, \tag{6}$$

where $\mathbf{X}=\mathbf{r}/L$ and $\mathbf{x}=\tilde{\mathbf{r}}/\ell$ are nondimensional macroscale and microscale variables, respectively.

A similar scaling applies also to time, obtaining,

$$\frac{\ell^2}{D_0}\frac{\partial}{\partial t}=\frac{\partial}{\partial\theta}+\epsilon\frac{\partial}{\partial\Theta_1}+\epsilon^2\frac{\partial}{\partial\Theta_2}, \tag{7}$$

where Θ_2, Θ_1, and θ measure time at the macroscale, mesoscale, and microscale, respectively.

In the following, we will assume that any function $f(\mathbf{x},\theta)$ is ergodic, that is, its (microscale) volume and time integral coincides with its ensemble average. This assumption derives from the fact that the fluctuations of the velocity field result from a very large number of interactions and therefore, according to the central limit theorem, they must be locally random, i.e., homogeneous in space and stationary in time at the microscale (see comments in Ref. [24]). In the different, albeit related, context of suspension flow, Marchioro and Acrivos [25] explicitly demonstrated that, although the system of equations describing the particle motions is deterministic, the particle displacements present a Gaussian distribution and therefore constitute a random process.

Substituting Eqs. (6) and (7) into Eq. (1), we obtain

$$\epsilon^2 \frac{\partial c}{\partial \Theta_2} + \epsilon \frac{\partial c}{\partial \Theta_1} + \frac{\partial c}{\partial \theta} + \mathrm{Pe}\left[\frac{\partial}{\partial \mathbf{x}} \cdot (\mathbf{u}c) + \epsilon \frac{\partial}{\partial \mathbf{X}} \cdot (\mathbf{u}c)\right] = \left(\frac{\partial}{\partial \mathbf{x}} + \epsilon \frac{\partial}{\partial \mathbf{X}}\right)^2 c, \tag{8}$$

where $\mathbf{u}(\mathbf{X},\mathbf{x},\Theta,\theta) = \mathbf{U}/U_0$ represents the nondimensional fluid velocity, with $\Theta = (\Theta_1, \Theta_2)$. This governing equation is subjected to the initial condition (2),

$$\lim_{t \to 0} c(\epsilon, \mathbf{X}, \mathbf{x}, t) = c_0(\mathbf{X}), \tag{9}$$

where we have assumed that the initial concentration c_0 is a smooth function of position. Generalization to the case where c_0 depends on $\mathbf{x}$, and therefore on ϵ, does not add anything to our physical understanding of the phenomenon.

In the following, we will apply a multiple scale analysis to derive the effective equations satisfied by the system. The key idea here is to expand the probability distribution as the following power series:

$$c(\epsilon, \mathbf{X}, \mathbf{x}, \Theta, \theta) = \sum_{n=0} \epsilon^n c^{(n)}(\mathbf{X}, \mathbf{x}, \Theta, \theta), \tag{10}$$

substitute it into the governing equation (8) and then collect equal powers of ϵ.

When, as in this case, some of the leading-order problems turn out to be ill posed, this multiple scale technique has to be applied with special care. Similar problems have been encountered in the past, and were solved either by "subtracting" the dominant term from the governing equation and "summing" it back at the end (Rubinstein and Mauri [11]), "warping" time scales and length scales (Mauri [20]), or expanding the independent variables in time as well as in space (see Mauri [13], Mazzino [26], and references therein). Here, as indicated in the expression (7), we adopt the last of these methods.

III. MULTIPLE SCALE ANALYSIS

A. Leading-order terms

At leading, $O(1)$ order, substituting Eq. (10) into (8), we obtain

$$\frac{\partial c^{(0)}}{\partial \theta} + \mathrm{Pe}\frac{\partial}{\partial \mathbf{x}} \cdot (\mathbf{u}c^{(0)}) - \frac{\partial^2 c^{(0)}}{\partial \mathbf{x}^2} = 0, \tag{11}$$

with initial condition (9),

$$\lim_{\Theta, \theta \to 0} c^{(0)}(\mathbf{X}, \mathbf{x}, \Theta, \theta) = c_0(\mathbf{X}). \tag{12}$$

The solution of Eq. (11) is trivially,

$$c^{(0)}(\mathbf{X}, \mathbf{x}, \Theta, \theta) = A_0(\mathbf{X}, \mathbf{x}, \Theta, \theta)\overline{c}(\mathbf{X}, \Theta), \tag{13}$$

with $\overline{c}(\mathbf{X}, 0) = c_0(\mathbf{X})$, while $A_0(\mathbf{X}, \mathbf{x}, \Theta, \theta)$, representing the microscale probability function, can be determined solving Eq. (11), with initial condition $A_0(\mathbf{X}, \mathbf{x}, \Theta, \theta = 0) = 1$. For solenoidal flow fields, this problem admits the trivial solution $A_0 = 1$. Note that A_0 is normalized as

$$\langle A_0(\mathbf{X}, \mathbf{x}, \Theta, \theta) \rangle = 1, \tag{14}$$

where the bracket indicates microscale averaging both in space and in time for any function $f(\mathbf{X}, \mathbf{x}, \Theta, \theta)$, i.e.,

$$\langle f \rangle(\mathbf{X}, \Theta) = \int \int f(\mathbf{X}, \mathbf{x}, \Theta, \theta) d\mathbf{x} d\theta. \tag{15}$$

In addition, using the normalization condition of c, together with the following additional constraints, which will arise spontaneously later,

$$\langle c^{(n \neq 0)}(\mathbf{X}, \mathbf{x}, \Theta, \theta) \rangle = 0, \tag{16}$$

we find the normalization condition

$$\langle c(\mathbf{X}, \mathbf{x}, \Theta, \theta) \rangle = \langle c^{(0)}(\mathbf{X}, \mathbf{x}, \Theta, \theta) \rangle = \overline{c}(\mathbf{X}, \Theta). \tag{17}$$

Although these additional assumptions are not necessary to the development of our analysis, they do remove the degree of arbitrariness in the definition (10) of $c^{(i)}$ and greatly simplify our results. In any case, although these conditions are not unique, any other condition that we may choose in place of Eq. (17) would lead, eventually, to the same final results (Mauri, 1991).

B. First-order terms

At first, $O(\epsilon)$ order, we obtain

$$\frac{\partial c^{(1)}}{\partial \theta} + \mathrm{Pe}\frac{\partial}{\partial \mathbf{x}} \cdot (\mathbf{u}c^{(1)}) - \frac{\partial^2 c^{(1)}}{\partial \mathbf{x}^2} = -\frac{\partial c^{(0)}}{\partial \Theta_1} - \mathrm{Pe}\frac{\partial}{\partial \mathbf{X}} \cdot (\mathbf{u}c^{(0)}) + 2\frac{\partial}{\partial \mathbf{x}} \cdot \frac{\partial c^{(0)}}{\partial \mathbf{X}}, \tag{18}$$

subjected to the initial condition (9),

$$\lim_{\Theta, \theta \to 0} c^{(1)}(\mathbf{X}, \mathbf{x}, \Theta, \theta) = 0. \tag{19}$$

Now, apply the solvability condition to Eq. (18), taking the volume and time integral over the $\mathbf{x}$ and θ variables and considering that the left-hand side does not contribute because the volume integral of the gradient of any locally random function is identically zero. Finally, we obtain,

$$\frac{\partial \overline{c}}{\partial \Theta_1} = -\mathrm{Pe}\frac{\partial}{\partial \mathbf{X}} \cdot (\mathbf{v}_0 \overline{c}), \tag{20}$$

where

$$\mathbf{v}_0(\mathbf{X}, \Theta) = \langle \mathbf{u}(\mathbf{X}, \mathbf{x}, \Theta, \theta) A_0(\mathbf{X}, \mathbf{x}, \Theta, \theta) \rangle \tag{21}$$

is the leading-order mean tracer velocity, that is, the velocity that the suspended particles (with the given particle distribution A_0) would have if they moved with the same speed as the fluid's.

The main conclusion that we draw from Eq. (21) is that the fluid compressibility determines a nonuniform concentration field at the microscale, which, in turn, causes the leading-order mean tracer velocity $\mathbf{v}_0$ to differ from the mean fluid velocity $\overline{\mathbf{u}}$, defined as

$$\overline{\mathbf{u}}(\mathbf{X},\Theta)=\langle \mathbf{u}(\mathbf{X},\mathbf{x},\Theta,\theta)\rangle. \tag{22}$$

If fact, Eq. (21) can be rewritten as

$$\mathbf{v}_0=\overline{\mathbf{u}}+\mathbf{v}_0^b, \tag{23}$$

where

$$\mathbf{v}_0^b(\mathbf{X},\Theta)=\langle \mathbf{u}(\mathbf{X},\mathbf{x},\Theta,\theta)[A_0(\mathbf{X},\mathbf{x},\Theta,\theta)-1]\rangle \tag{24}$$

coincides with the ballistic velocity, as is defined by Vergassola and Avellaneda [18]. This result is identical to the leading-order term obtained by Shapiro and Brenner [19] and Mauri [20], who studied the transport of chemically reacting tracers, with the reaction speed taking the place of the fluid velocity divergence [see Eq. (5) and related comments].

Now let us consider Eq. (18), assuming that its solution can be expressed as

$$c^{(1)}(\mathbf{X},\mathbf{x},\Theta,\theta)=A_1(\mathbf{X},\mathbf{x},\Theta,\theta)\overline{c}(\mathbf{X},\Theta) - \frac{\partial}{\partial \mathbf{X}}\cdot[\mathbf{B}(\mathbf{X},\mathbf{x},\Theta,\theta)\overline{c}(\mathbf{X},\Theta)]. \tag{25}$$

Substituting Eq. (20) and (25) into Eq. (18), we see that the function $A_1(\mathbf{X},\mathbf{x},\Theta,\theta)$ and the vector function $\mathbf{B}(\mathbf{X},\mathbf{x},\Theta,\theta)$ satisfy the following problems:

$$\frac{\partial A_1}{\partial\theta}+\mathrm{Pe}\frac{\partial}{\partial\mathbf{x}}\cdot(\mathbf{u}A_1)-\frac{\partial^2 A_1}{\partial\mathbf{x}^2}=-\frac{\partial A_0}{\partial\Theta_1}-\mathrm{Pe}\frac{\partial}{\partial\mathbf{x}}\cdot\left(\mathbf{B}\cdot\frac{\partial}{\partial\mathbf{X}}\mathbf{u}\right) - \mathrm{Pe}\mathbf{v}_0\cdot\frac{\partial A_0}{\partial\mathbf{X}}, \tag{26}$$

and

$$\frac{\partial\mathbf{B}}{\partial\theta}+\mathrm{Pe}\frac{\partial}{\partial\mathbf{x}}\cdot(\mathbf{u}\mathbf{B})-\frac{\partial^2\mathbf{B}}{\partial\mathbf{x}^2}=\mathrm{Pe}\tilde{\mathbf{u}}A_0-2\frac{\partial A_0}{\partial\mathbf{x}}, \tag{27}$$

where $\tilde{\mathbf{u}}(\mathbf{X},\mathbf{x},\Theta,\theta)=\mathbf{u}(\mathbf{X},\mathbf{x},\Theta,\theta)-\mathbf{v}_0(\mathbf{X},\Theta)$ is the velocity fluctuation. These equations must be solved with initial conditions

$$A_1(\mathbf{X},\mathbf{x},\Theta,\theta=0)=0,\quad \mathbf{B}(\mathbf{X},\mathbf{x},\Theta,\theta=0)=\mathbf{0}, \tag{28}$$

and imposing the normalization condition (17), i.e.,

$$\langle A_1(\mathbf{X},\mathbf{x},\Theta,\theta)\rangle=0;\quad \langle\mathbf{B}(\mathbf{X},\mathbf{x},\Theta,\theta)\rangle=\mathbf{0}. \tag{29}$$

C. Second-order terms

Now we proceed to the second, $O(\epsilon^2)$ order, obtaining

$$\frac{\partial c^{(2)}}{\partial\theta}+\mathrm{Pe}\frac{\partial}{\partial\mathbf{x}}\cdot(\mathbf{u}c^{(2)})-\frac{\partial^2 c^{(2)}}{\partial\mathbf{x}^2} = -\frac{\partial c^{(0)}}{\partial\Theta_2}-\frac{\partial c^{(1)}}{\partial\Theta_1}-\mathrm{Pe}\frac{\partial}{\partial\mathbf{X}}\cdot(\mathbf{u}c^{(1)})+2\frac{\partial}{\partial\mathbf{X}}\cdot\frac{\partial c^{(1)}}{\partial\mathbf{x}} + \frac{\partial^2 c^{(0)}}{\partial X^2}, \tag{30}$$

subjected to the initial condition (9),

$$\lim_{\Theta,\theta\to 0} c^{(2)}(\mathbf{X},\mathbf{x},\Theta,\theta)=0. \tag{31}$$

Applying the solvability condition to this equation, i.e., integrating over the $\mathbf{x}$ and θ variables, we obtain

$$\frac{\partial\overline{c}}{\partial\Theta_2}=-\mathrm{Pe}\frac{\partial}{\partial\mathbf{X}}\cdot\tilde{\mathbf{j}}, \tag{32}$$

where

$$\tilde{\mathbf{j}}(\mathbf{X},\Theta)=-\frac{1}{Pe}\frac{\partial\overline{c}}{\partial\mathbf{X}}(\mathbf{X},\Theta)+\langle\mathbf{u}c^{(1)}\rangle(\mathbf{X},\Theta) \tag{33}$$

is the fluctuation-induced particle flux. Substituting Eq. (25) into Eq. (33) and rearranging, we can express the particle flux as follows:

$$\tilde{\mathbf{j}}=(\mathbf{v}_1^b+\mathbf{v}^d)\overline{c}-\mathbf{d}^s\cdot\frac{\partial\overline{c}}{\partial\mathbf{X}}. \tag{34}$$

Here, $\mathbf{v}_1^b$ is the velocity that the suspended particles, with the given particle distribution A_1, would have if they moved with the same speed as the fluid's,

$$\mathbf{v}_1^b(\mathbf{X},\Theta)=\langle\mathbf{u}A_1\rangle. \tag{35}$$

Also, $\mathbf{v}^d$ and $\mathbf{d}^s$ are fluctuation-induced tracer drift velocity and diffusivity, respectively, which, considering the normalization condition (29), can be written as follows:

$$\mathbf{v}^d=-\left\langle\mathbf{u}\left(\frac{\partial}{\partial\mathbf{X}}\cdot\mathbf{B}\right)\right\rangle=-\left\langle\tilde{\mathbf{u}}\left(\frac{\partial}{\partial\mathbf{X}}\cdot\mathbf{B}\right)\right\rangle, \tag{36}$$

and

$$\mathbf{d}^s=\frac{1}{Pe}\mathbf{I}+\langle\mathbf{u}\cdot\mathbf{B}\rangle=\frac{1}{Pe}\mathbf{I}+\langle\tilde{\mathbf{u}}\mathbf{B}\rangle, \tag{37}$$

where $\mathbf{I}$ denotes the unit dyadic.

IV. THE EFFECTIVE EQUATION

Substituting Eqs. (20) and (32)–(37) into Eq. (7), we obtain the Fokker-Planck equation,

$$\frac{L^2}{D_0}\frac{\partial \overline{c}}{\partial t}+\mathrm{Pe}\frac{\partial}{\partial \mathbf{X}}\cdot\overline{\mathbf{j}}=0, \tag{38}$$

with the following constitutive relation for the coarse-grained non-dimensional particle flux $\overline{\mathbf{j}}(\mathbf{X},\Theta)$,

$$\overline{\mathbf{j}}=-\frac{1}{Pe}\frac{\partial\overline{c}}{\partial\mathbf{X}}+\left\langle \mathbf{u}\left(\frac{1}{\epsilon}c^{(0)}+c^{(1)}\right)\right\rangle=\frac{1}{\epsilon}\mathbf{v}_0\overline{c}+\tilde{\mathbf{j}}. \tag{39}$$

Therefore we obtain

$$\overline{\mathbf{j}}=\mathbf{v}_E\overline{c}-\mathbf{d}^s\cdot\frac{\partial\overline{c}}{\partial\mathbf{X}}, \tag{40}$$

where $\mathbf{v}_E$ is the Eulerian mean particle velocity,

$$\mathbf{v}_E=\frac{1}{\epsilon}\overline{\mathbf{v}}+\mathbf{v}^d, \tag{41}$$

denoting the sum between the fluctuation-induced particle drift velocity $\mathbf{v}^d$ [see Eq. (36)] and the microscale mean tracer velocity $\overline{\mathbf{v}}$, with

$$\overline{\mathbf{v}}=\mathbf{v}_0+\epsilon\mathbf{v}_1=\langle\mathbf{u}(A_0+\epsilon A_1)\rangle. \tag{42}$$

The microscale mean tracer velocity can also be written as

$$\overline{\mathbf{v}}=\overline{\mathbf{u}}+\mathbf{v}^b, \tag{43}$$

where $\mathbf{v}^b=\mathbf{v}_0^b+\epsilon\mathbf{v}_1^b$ is the ballistic tracer velocity (Vergassola and Avellaneda [18]), expressing the difference between the microscale mean tracer velocity and the mean fluid velocity, due to the nonhomogeneous concentration field at the microscale.

The particle drift velocity $\mathbf{v}^d$ of Eq. (36) can be rewritten as

$$\mathbf{v}^d(\mathbf{X},\Theta)=-\left[\frac{\partial}{\partial\mathbf{X}_2}\cdot\mathbf{d}^T(\mathbf{X},\mathbf{X}_2,\Theta)\right]_{\mathbf{X}_2=\mathbf{X}}, \tag{44}$$

with $(d^T)_{ij}=d_{ji}$. Here, $\mathbf{d}(\mathbf{X}_1,\mathbf{X}_2,\Theta)$ is the following cross-diffusion dyadic:

$$\mathbf{d}(\mathbf{X}_1,\mathbf{X}_2,\Theta)=\langle\tilde{\mathbf{u}}(\mathbf{X}_1,\mathbf{x},\Theta,\theta)\mathbf{B}(\mathbf{X}_2,\mathbf{x},\Theta,\theta)\rangle, \tag{45}$$

with

$$\mathbf{d}^s(\mathbf{X},\Theta)=\frac{1}{\mathrm{Pe}}\mathbf{I}+\mathbf{d}(\mathbf{X},\mathbf{X},\Theta). \tag{46}$$

Note that, from its definition (44), the drift velocity $\mathbf{v}^d$ is due to macroscale nonhomogeneities and therefore it is structurally different from the ballistic velocity $\mathbf{v}^b$, which depends on microscale nonhomogeneities instead.

The constitutive relation (40) can also be written as

$$\overline{\mathbf{j}}=\mathbf{v}_L\overline{c}-\frac{\partial}{\partial\mathbf{X}}\cdot(\mathbf{d}^s\overline{c}), \tag{47}$$

where $\mathbf{v}_L$ is the Lagrangian mean particle velocity, which is related to its Eulerian counterpart through the relation $\mathbf{v}_L=\mathbf{v}_E+\boldsymbol{\nabla}\cdot\mathbf{d}^s$. Therefore, considering that

$$\frac{d}{dx}D(x,x)=\left[\frac{\partial}{\partial y}D(y,x)\right]_{y=x}+\left[\frac{\partial}{\partial z}D(x,z)\right]_{z=x}, \tag{48}$$

we can express the Lagrangian mean particle velocity as

$$\mathbf{v}_L(\mathbf{X},\Theta)=\frac{1}{\epsilon}\overline{\mathbf{v}}(\mathbf{X},\Theta)+\left[\frac{\partial}{\partial\mathbf{X}_1}\cdot\mathbf{d}(\mathbf{X}_1,\mathbf{X},\Theta)\right]_{\mathbf{X}_1=\mathbf{X}}. \tag{49}$$

In particular, when the flow field is solenoidal,

$$\frac{\partial}{\partial\mathbf{X}}\cdot\mathbf{u}=\frac{\partial}{\partial\mathbf{x}}\cdot\mathbf{u}=0, \tag{50}$$

then, from Eqs. (11), (26), and (27) we obtain

$$A_0=1,\quad A_1=0,\quad \frac{\partial}{\partial\mathbf{X}}\cdot\mathbf{B}(\mathbf{X},\mathbf{x},\Theta,\theta)=0. \tag{51}$$

As a consequence, we see that for solenoidal flow fields the constitutive equation presents three important features:

(a) The microscale mean particle velocity $\overline{\mathbf{v}}$ coincides with the mean fluid velocity $\overline{\mathbf{u}}$ [cf. Eqs. (21), (35), and (42)] and therefore the ballistic velocity vanishes, i.e., $\mathbf{v}^b=\mathbf{0}$.

(b) From its definition (36), the particle drift velocity vanishes identically, i.e., $\mathbf{v}^d=\mathbf{0}$.

(c) From its definition (37), the diffusivity has zero divergence, i.e., $\boldsymbol{\nabla}\cdot\mathbf{d}^s=\mathbf{0}$. In addition, as we will see in the following section, $\mathbf{d}^s$ is equal to one half the long-time growth rate of the mean square displacement of the tracer particle from its average position and therefore coincides with the self-diffusivity.

Consequently, the constitutive relation for the flux of passive tracers in solenoidal flow fields reduces to

$$\overline{\mathbf{j}}=\frac{1}{\epsilon}\overline{\mathbf{u}}-\mathbf{d}^s\cdot\boldsymbol{\nabla}\overline{c}. \tag{52}$$

Rewriting our results in dimensional form, we may conclude that the coarse-grained concentration field $\overline{c}(\mathbf{r},t)$ of Brownian tracers convected by and diffusing in a nonhomogeneous and nonstationary fluid flow field $\mathbf{U}(\mathbf{r},t)$ satisfies the Fokker-Planck equation,

$$\frac{\partial\overline{c}}{\partial t}+\boldsymbol{\nabla}\cdot\mathbf{J}=0, \tag{53}$$

with the following constitutive relation for the coarse-grained dimensional particle flux $\mathbf{J}(\mathbf{r},t)$:

$$\mathbf{J}=\mathbf{V}_E\overline{c}-\mathbf{D}^s\cdot\boldsymbol{\nabla}\overline{c}. \tag{54}$$

Here $\mathbf{V}_E=\epsilon U_0\mathbf{v}_E$ is the Eulerian mean particle velocity,

$$\mathbf{V}_E(\mathbf{r},t)=\overline{\mathbf{V}}(\mathbf{r},t)-\left[\frac{\partial}{\partial \mathbf{r}_2}\cdot \mathbf{D}^T(\mathbf{r},\mathbf{r}_2,t)\right]_{\mathbf{r}_2=\mathbf{r}}, \tag{55}$$

where $\overline{\mathbf{V}}=U_0\overline{\mathbf{v}}$ is the microscale mean particle velocity, $\mathbf{D}(\mathbf{r}_1,\mathbf{r}_2,t)=(U_0\ell)\mathbf{d}(\mathbf{X}_1,\mathbf{X}_2,\Theta)$ is the cross diffusion dyadic (45), while the diffusivity $\mathbf{D}^s(\mathbf{r},t)=(U_0\ell)\mathbf{d}^s(\mathbf{X},\Theta)$ is defined as

$$\mathbf{D}^s(\mathbf{r},t)=D_0\mathbf{d}^*=D_0\mathbf{I}+\mathbf{D}(\mathbf{r},\mathbf{r},t). \tag{56}$$

V. LARGE PECLET NUMBER LIMIT

In this section, for the sake of brevity, the dependence on the macrovariables $\mathbf{X}$ and Θ is not explicitly indicated, although it is always implicitly assumed. When $\mathrm{Pe}\gg 1$, the solution of Eq. (27) for the $\mathbf{B}$ field can be written as

$$\mathbf{B}(\mathbf{z},\theta')=\langle G(\mathbf{z}-\mathbf{x},\theta'-\theta,)\tilde{\mathbf{u}}(\mathbf{x},\theta)A_0(\mathbf{x},\theta)\rangle, \tag{57}$$

where, Here $G(\mathbf{z}-\mathbf{x},\theta'-\theta)$ is the propagator of Eq. (27) for large Peclet number, satisfying the following equation:

$$\frac{\partial G}{\partial \theta}+\mathrm{Pe}\frac{\partial}{\partial \mathbf{x}}\cdot(\mathbf{u}G)=\mathrm{Pe}\,\delta(\mathbf{z}-\mathbf{x})\,\delta(\theta'-\theta), \tag{58}$$

therefore denoting the probability that a tracer, which at time θ is located at $\mathbf{x}$, is found at $\mathbf{z}$ at time $\theta'>\theta$. Note that, since the process is locally random (i.e., stationary in time and homogeneous in space), G depends on the differences $\mathbf{z}-\mathbf{x}$ and $\theta'-\theta$. Substituting Eq. (57) into Eq. (37) for $\mathrm{Pe}\gg 1$, we see that the diffusivity tensor can be expressed as the following time integral:

$$\mathbf{d}^s=\int_0^\infty \mathbf{f}(\theta)d\theta. \tag{59}$$

Here $\mathbf{f}(\theta'-\theta)$ is the average covariance of the velocity fluctuations at the points occupied by the tracer (and fluid as well) particle at the times θ and $\theta'>\theta$ (see, for example, Monin and Yaglom [1], Mauri [13]),

$$\mathbf{f}(\theta'-\theta)=\int \langle\tilde{\mathbf{u}}(\mathbf{z},\theta')G(\mathbf{z}-\mathbf{x},\theta'-\theta,)\tilde{\mathbf{u}}(\mathbf{x},\theta)\rangle_0 d\mathbf{z}. \tag{60}$$

Here the brackets denote the average of any quantity $h(\mathbf{x},\theta)$ over the local $\mathbf{x}$ and θ variables, weighted upon the probability to find a tracer particle at that location and at that time, i.e.,

$$\langle h\rangle_0=\langle hA_0\rangle=\int\int h(\mathbf{x},\theta)A_0(\mathbf{x},\theta)d\mathbf{x}d\theta, \tag{61}$$

and therefore it coincides with the ensemble average, due to the assumption that the process is locally ergodic. Equations (58)–(60) show clearly that for large Pe, as both G and $\mathbf{u}$ are $O(1)$ quantities, the effective diffusivity is proportional to V_0, i.e., to Pe. At this point, consider that the product

$$A_0^{(2)}(\mathbf{x},\mathbf{z},\theta,\theta')=G(\mathbf{z}-\mathbf{x},\theta'-\theta,)A_0(\mathbf{x},\theta) \tag{62}$$

is the leading-order joint microscale probability that the tracer particle is located at position $\mathbf{x}$ at time θ and at $\mathbf{z}$ at time θ'. Consequently, Eq. (60) can be written as

$$\mathbf{f}(\theta'-\theta)=\int\int \tilde{\mathbf{u}}(\mathbf{x},\theta)\tilde{\mathbf{u}}(\mathbf{z},\theta')A_0^{(2)}(\mathbf{x},\mathbf{z},\theta,\theta')d\mathbf{x}d\mathbf{x}'d\theta, \tag{63}$$

which is identical to the Lagrangian velocity autocorrelation function defined in Eq. (4),

$$\mathbf{f}(\theta'-\theta)=\langle\tilde{\mathbf{u}}(\mathbf{x},\theta)\tilde{\mathbf{u}}(\mathbf{z},\theta')\rangle_0, \tag{64}$$

where $\mathbf{z}$ is the position of a tracer (and fluid) particle at time θ', provided that at time $\theta<\theta'$ it is located at $\mathbf{x}$. Clearly, the identification of $\mathbf{f}$ with the Lagrangian velocity autocorrelation function defined in Eq. (4) holds only when $\tilde{\theta}$ in Eq. (4) is a microscale time.

Now, since at the microscale our process is stationary, we can apply the microscale reversibility property (otherwise called principle of detailed balance),

$$\mathbf{f}(\mathbf{X},\Theta,\theta)=\mathbf{f}(\mathbf{X},\Theta,-\theta) \tag{65}$$

to obtain Onsager's relation (see de Groot and Mazur [27]), showing that both $\mathbf{f}$ and $\mathbf{d}^s$ are symmetric tensors. Again, note that the Lagrangian autocorrelation function depends on the difference $\theta-\theta'$, due to the assumption that the process is stationary (and homogeneous) at the microscale. Now, denote,

$$\tilde{\mathbf{z}}(\mathbf{x},\theta)=\int_0^\theta \tilde{\mathbf{u}}[\mathbf{z}(\theta'),\theta']d\theta', \tag{66}$$

as the difference between the position of the test particle at time θ and its average position (i.e., the position that the test particle would have, had it moved with the average velocity $\overline{\mathbf{v}}_0$), with $\mathbf{z}(\theta=0)=\mathbf{x}$ and $\tilde{\mathbf{z}}(\theta=0)=\mathbf{0}$. Therefore, considering that

$$\tilde{\mathbf{u}}(\mathbf{x},\theta)=\frac{d\tilde{\mathbf{z}}}{d\theta}(\mathbf{x},\theta), \tag{67}$$

substituting Eq. (66) into Eqs. (59), and (64), we obtain

$$\mathbf{d}^s=\lim_{\theta\to\infty}\left\langle\frac{d\tilde{\mathbf{z}}}{d\theta}\tilde{\mathbf{z}}\right\rangle_0=\lim_{\theta\to\infty}\frac{1}{2}\frac{d}{dt}\langle\tilde{\mathbf{z}}\cdot\tilde{\mathbf{z}}\rangle_0, \tag{68}$$

where the last equality stems from the fact that the dyadic $\mathbf{d}^s$ is symmetric [and, in any case, its antisymmetric part does not play any role in the Fokker-Planck equation (38)]. Equation (68) reveals that $\mathbf{d}^s$ equals one half the long-time growth rate of the mean square displacement of a tracer (and fluid) particle from its mean position and hence, by definition, it coincides with the particle (and fluid) self-diffusivity dyadic.

Note that when the flow field is solenoidal, the solution of the advection equation (58) is

$$G(\mathbf{z}-\mathbf{x},\theta'-\theta)=\delta(\mathbf{z}-\mathbf{x})\,\delta(\theta'-\theta). \tag{69}$$

Substituting Eq. (69) into Eq. (57), with $A_0=1$, we obtain

$$\mathbf{B}(\mathbf{x},\theta)=\int_0^{\theta}\tilde{\mathbf{u}}(\mathbf{z},\theta')\,d\theta'=\tilde{\mathbf{z}}(\theta), \tag{70}$$

and since $\mathbf{d}^s=\langle\tilde{\mathbf{u}}\cdot\mathbf{B}\rangle$, we obtain again Eq. (68).

Now we can include in this treatment the effect of a Brownian movement since, for Pe$\gg1$, it is superimposed to and independent of the advection of the flow field. In fact, denoting by $d\mathbf{z}_B/d\theta$ the Brownian velocity, that is, the difference between the instantaneous tracer velocity, $d\mathbf{z}/d\theta$, and the fluid velocity $\mathbf{u}$, we know that (Kubo *et al.* [28])

$$\left\langle\frac{d\mathbf{z}_B}{d\theta}(\mathbf{x},\theta)G(\mathbf{x}-\mathbf{x}',\theta-\theta')\frac{d\mathbf{z}_B}{d\theta}(\mathbf{x}',\theta')\right\rangle_0 =\mathbf{I}\delta(\mathbf{x}-\mathbf{x}')\,\delta(\theta-\theta'). \tag{71}$$

Therefore, considering that the Brownian motion is uncorrelated with the velocity field (and its fluctuations, as well), we see that Eq. (59) is still valid, provided that the Lagrangian correlation function (64) is referred to the tracer velocity fluctuations $d\tilde{\mathbf{z}}/d\theta$, instead of the fluid velocity fluctuations $\tilde{\mathbf{u}}$, i.e.,

$$\mathbf{f}(\theta-\theta')=\left\langle\frac{d\tilde{\mathbf{z}}}{d\theta}(\mathbf{x},\theta)\frac{d\tilde{\mathbf{z}}}{d\theta}(\mathbf{z},\theta')\right\rangle_0. \tag{72}$$

Therefore, we obtain again Eq. (68), showing that, when the Brownian movement is uncorrelated with the velocity field, the effective diffusivity can still be interpreted as one half the long-time growth rate of the mean square displacement of a tracer from its mean position.

Tracer transport as a random process

Here we intend to show that, in agreement with Stratonovich (see Kubo *et al.* [28]), the same Fokker-Planck equations (53)–(56) could be obtained, in the limit Pe$\gg1$, assuming that the trajectory $\mathbf{r}(t)$ of any tracer particle is a random variable satisfying the following generalized nonlinear Langevin equation:

$$\frac{d\mathbf{r}}{dt}(\mathbf{r},t)=\bar{\mathbf{V}}(\mathbf{r},t)+\tilde{\mathbf{V}}(\mathbf{r},t). \tag{73}$$

Here $\bar{\mathbf{V}}(\mathbf{r},t)$ is a smoothly varying mean tracer velocity, while $\tilde{\mathbf{V}}(\mathbf{r},t)$ is its random component, which is described through the following white stochastic process:

$$\langle\tilde{\mathbf{V}}(\mathbf{r},t)\rangle_0=\mathbf{0}, \tag{74}$$

$$\langle\tilde{\mathbf{V}}(\mathbf{r}_1,t)\tilde{\mathbf{V}}(\mathbf{r}_2,t+\Delta t)\rangle_0=2\mathbf{D}^T(\mathbf{r}_1,\mathbf{r}_2,t)\,\delta(\Delta t), \tag{75}$$

with the angular brackets indicating ensemble averaging and $\mathbf{D}(\mathbf{r}_1,\mathbf{r}_2,t)$ denoting (by definition) the time integral of the Lagrangian velocity cross correlation dyadic. In fact, integrating Eq. (73) for a short time interval Δt, we obtain

$$\Delta\mathbf{r}=\mathbf{r}(t+\Delta t)-\mathbf{r}(t) =\bar{\mathbf{V}}(\mathbf{r},t)\Delta t+\int_t^{t+\Delta t}\tilde{\mathbf{V}}[\mathbf{r}(t_1),t_1]dt_1+o(\Delta t), \tag{76}$$

where we have considered that $\bar{\mathbf{V}}$ changes in time much slower than $\tilde{\mathbf{V}}$. Now expand

$$\tilde{\mathbf{V}}[\mathbf{r}(t_1),t_1]=\tilde{\mathbf{V}}[\mathbf{r}(t),t_1]+\Delta\mathbf{r}_1\cdot\frac{\partial}{\partial\mathbf{r}}\tilde{\mathbf{V}}[\mathbf{r}(t),t_1]+o(\Delta t), \tag{77}$$

where

$$\Delta\mathbf{r}_1=\mathbf{r}(t_1)-\mathbf{r}(t)=\int_t^{t_1}\tilde{\mathbf{V}}[\mathbf{r}(t),t_2]dt_2. \tag{78}$$

Consequently, we can define the mean Lagrangian particle velocity $\mathbf{V}_L$, and the particle self-diffusivity $\mathbf{D}^s$ as

$$\mathbf{V}_L(\mathbf{r},t)=\lim_{\Delta t\to0}\frac{\langle\Delta\mathbf{r}\rangle}{\Delta t}=\bar{\mathbf{V}}(\mathbf{r},t)+\left[\frac{\partial}{\partial\mathbf{r}_1}\cdot\mathbf{D}(\mathbf{r}_1,\mathbf{r},t)\right]_{\mathbf{r}_1=\mathbf{r}} \tag{79}$$

and

$$\mathbf{D}^s(\mathbf{r},t)=\frac{1}{2}\lim_{\Delta t\to0}\frac{\langle(\Delta\mathbf{r})^2\rangle}{\Delta t}=\mathbf{D}(\mathbf{r},\mathbf{r},t). \tag{80}$$

These quantities constitute the convective and diffusive parts of the particle flux appearing in the Fokker-Planck equation (53), with

$$\mathbf{J}=\mathbf{V}_L\bar{c}-\boldsymbol{\nabla}\cdot(\mathbf{D}^s\bar{c}), \tag{81}$$

where the Lagrangian mean particle velocity is related to its Eulerian counterpart, $\mathbf{V}_E$, through the relation [cf. Eqs. (47)–(49)]

$$\mathbf{V}_L=\mathbf{V}_E+\boldsymbol{\nabla}\cdot\mathbf{D}^s. \tag{82}$$

Therefore, we may conclude that in the limit Pe$\gg1$ the convection-diffusion equation (53) is equivalent to the Stratonovich nonlinear stochastic process (73)–(75). In particular, it means that (a) the smoothly varying mean tracer velocity $\bar{\mathbf{V}}$ appearing in the random process is the microscale mean tracer velocity (which in turn is the sum of the mean fluid velocity and the ballistic tracer velocity); (b) the effective diffusivity $\mathbf{D}^s$ appearing in the Fokker-Planck equation is a self-diffusion dyadic, as it equals the time derivative of the Lagrangian velocity autocorrelation function. An identical conclusion was reached recently [29] for the constitutive relations of the volumetric flux of a suspension of rigid particles immersed in a viscous fluid.

VI. CONCLUSIONS AND COMMENTS

The main result of this work is that the coarse-grained concentration $\bar{c}$ of Brownian tracers convected and diffusing in a nonhomogeneous, nonstationary fluid flow field satisfies the Fokker-Planck equation [cf. Eqs. (53)–(56)],

$$\frac{\partial \overline{c}}{\partial t}+\nabla\cdot\mathbf{J}=0, \tag{83}$$

with the following constitutive relation for the coarse-grained dimensional particle flux $\mathbf{J}$:

$$\mathbf{J}=\mathbf{V}_E\overline{c}-\mathbf{D}^s\cdot\nabla\overline{c}. \tag{84}$$

Here $\mathbf{D}^s$ is the effective diffusivity dyadic, defined as

$$\mathbf{D}^s(\mathbf{r},t)=D_0\mathbf{I}+\mathbf{D}(\mathbf{r},\mathbf{r},t), \tag{85}$$

where $\mathbf{D}=D_0\mathbf{d}$ indicates the cross-diffusivity tensor (45).

The Eulerian mean tracer velocity $\mathbf{V}_E$ appearing in the convective term of the constitutive relation (84) is equal to the sum of the microscale mean tracer velocity $\overline{\mathbf{V}}(\mathbf{r},t)$ and the tracer drift velocity $\mathbf{V}^d$,

$$\mathbf{V}_E(\mathbf{r},t)=\overline{\mathbf{V}}(\mathbf{r},t)+\mathbf{V}^d(\mathbf{r},t). \tag{86}$$

In turn, the microscale mean tracer velocity is the sum of the mean fluid velocity and the ballistic tracer velocity, $\overline{\mathbf{V}}=\overline{\mathbf{U}}+\mathbf{V}^b$, the latter accounting for the nonuniform tracer concentration at the microscale. Conversely, the tracer drift velocity is equal to the spatial derivative of the cross-diffusivity tensor $\mathbf{D}$, i.e.,

$$\mathbf{V}^d(\mathbf{r},t)=-\left[\frac{\partial}{\partial\mathbf{r}_2}\cdot\mathbf{D}^T(\mathbf{r},\mathbf{r}_2,t)\right]_{\mathbf{r}_2=\mathbf{r}}. \tag{87}$$

In the limit of large Peclet numbers, $\mathbf{D}^s$ is also equal to the time integral of the Lagrangian velocity autocorrelation dyadic (75),

$$\mathbf{D}^s(\mathbf{r},t)=\int_0^\infty\langle\tilde{\mathbf{V}}(\mathbf{r}+\mathbf{r}',t+t')\tilde{\mathbf{V}}(\mathbf{r},t)\rangle_0 dt', \tag{88}$$

where the brackets indicate ensemble average, $\tilde{\mathbf{V}}=\mathbf{V}-\overline{\mathbf{V}}$ is the tracer velocity fluctuation around the microscale mean tracer velocity, while $\mathbf{r}+\mathbf{r}'$ is the position of the tracer at time $t+t'$, assuming that at time t it is located at $\mathbf{r}$. Therefore, the tracer drift velocity can be written in the manner of Stratonovich as follows:

$$\mathbf{V}^d(\mathbf{r},t)=-\int_0^\infty\langle\tilde{\mathbf{V}}(\mathbf{r}+\mathbf{r}',t+t')[\nabla\cdot\tilde{\mathbf{V}}(\mathbf{r},t)]\rangle_0 dt'. \tag{89}$$

We may conclude that the flux of passive tracers can be expressed through a Fickian constitutive relation, characterized by an effective diffusivity and an Eulerian mean tracer velocity, the latter being equal to the sum of the mean fluid velocity, the ballistic tracer velocity, and the particle drift velocity. As we see from its definition (42)–(43), the ballistic velocity is nonzero whenever the concentration field is nonuniform at the microscale, irrespective of whether the flow field is homogeneous at the macroscale. On the contrary, the drift velocity takes into account the coupling between the nonsolenoidal character of the flow field and its nonhomogeneity at the macroscale. Therefore, as it vanishes whenever the flow field is homogeneous at the macroscale, the tracer drift velocity is structurally different from the ballistic tracer velocity. So, for example, in the flow of a compressible fluid through a random but statistically uniform porous medium, we find a nonzero ballistic tracer velocity and a drift velocity that is identically zero. An identical problem was studied by Shapiro and Brenner [19], who considered chemically reacting solute particles dispersed in a fluid flowing through a porous medium.

Finally, we should note that, as we have mentioned in the Introduction, the nonsolenoidal character of the flow field is not necessarily due to the fluid compressibility. In fact, particle-particle interactions and particle inertial forces can cause the particle velocity to differ from the local fluid velocity, so that the particle velocity field can be nonsolenoidal even when the fluid is incompressible. Therefore, the results of this work can find wider applications than the transport of passive tracers in compressible fluid flow. For example, the volumetric flux of concentrated viscous suspensions has been recently derived by Mauri [29], finding results that are very similar to those obtained here. In particular, even in that case we see that the motion of each suspended particle is a random process satisfying a nonlinear Langevin equation in the manner of Stratonovich, where the fluctuating term is described through the cross-diffusivity tensor.

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