Drift-free kinetic equations for turbulent dispersion

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The dispersion of passive scalars and inertial particles in a turbulent flow can be described in terms of probability density functions (PDFs) defining the statistical distribution of relevant scalar or particle variables. The construction of transport equations governing the evolution of such PDFs has been the subject of numerous studies, and various authors have presented formulations for this type of equation, usually referred to as a kinetic equation. In the literature it is often stated, and widely assumed, that these PDF kinetic equation formulations are equivalent. In this paper it is shown that this is not the case, and the significance of differences among the various forms is considered. In particular, consideration is given to which form of equation is most appropriate for modeling dispersion in inhomogeneous turbulence and most consistent with the underlying particle equation of motion. In this regard the PDF equations for inertial particles are considered in the limit of zero particle Stokes number and assessed against the fully mixed (zero-drift) condition for fluid points. A long-standing question regarding the validity of kinetic equations in the fluid-point limit is answered; it is demonstrated formally that one version of the kinetic equation (derived using the Furutsu-Novikov method) provides a model that satisfies this zero-drift condition exactly in both homogeneous and inhomogeneous systems. In contrast, other forms of the kinetic equation do not satisfy this limit or apply only in a limited regime.

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I. INTRODUCTION

The study of scalar and particle dispersion in turbulent flows is a field of great interest, not only because of the need for detailed understanding in industrial and environmental applications (particle transport in pipelines, formation of water droplets in clouds, dispersion of radioactive aerosols in nuclear fission reactors, combustion of fuel droplets in engines, etc.) but also because there remain many theoretical challenges and unanswered questions in the field.

There are two distinct but closely related areas of interest: the dispersion of fluid points (i.e., particles with $\mathrm{St} = 0$, where St is the particle Stokes number) and the dispersion of inertial particles. Both present challenges, but the dispersion of inertial particles is more complex since, by virtue of their inertia, the particles do not follow the flow exactly. This leads to some important phenomena. For example, studies have shown that inertial particles suspended in homogeneous turbulence are not, as might be expected, uniformly mixed by the turbulence but tend to cluster in high-strain, low-vorticity regions of the flow (e.g., [1]). Then again, in inhomogeneous turbulence such as a boundary layer, inertial particles are distributed nonuniformly, both instantaneously and on average (e.g., [2]). This is in contrast to the distribution of fluid points, which, if initially uniform, will remain so for all times (if the flow is incompressible). The strong nonuniformity of inertial particle distributions in a turbulent boundary layer occurs even when body forces such as gravity are absent and is a consequence of two competing mechanisms: turbophoretical drift and preferential sampling of the flow by particles (e.g., [3–5]). Developing models capable of accurately predicting such phenomena is nontrivial and a subject of continuing research. In this context kinetic equations play an important role. These equations, which capture the dynamics of a system via a probability density function (PDF) for a phase-space distribution, can be used as the basis for constructing continuum equations that describe the transport of the mean-field statistics of the particles (or fluid points) (i.e., the transport of the moments of the PDF).

PDF kinetic equations and their associated continuum equations have been shown to successfully predict the dispersion statistics of particles suspended in homogeneous flows (e.g., [6]). However, for inhomogeneous systems the predictions are only, at best, in adequate agreement with equivalent simulation data (e.g., [7]) Turbulence inhomogeneity presents a major challenge in the construction of appropriate PDF equations.

In this work PDF equations are considered for both scalar dispersion as well as for inertial particle transport: In the case of passive scalars the PDF of interest, $\rho(\boldsymbol{x},t)$, will define, in essence, the distribution at time t of the position $\boldsymbol{x}^f(t)$ of a marked fluid point, governed by the equation of motion

$$\frac{d}{dt}x_i^f = u_i(\boldsymbol{x}^f,t). \tag{1}$$

The field $\boldsymbol{u}(\boldsymbol{x},t)$ is to be interpreted as a stochastic model for a turbulent, incompressible fluid flow, exhibiting correlations in both space and time. This flow field may be homogeneous or inhomogeneous. While it is possible to include molecular diffusion by the addition of a further delta-correlated in time contribution to the field $\boldsymbol{u}$ this will not be considered here.

By extension, for inertial particles, the PDF of interest, $p(\boldsymbol{x},\boldsymbol{v},t)$, will define the joint distribution of the position $\boldsymbol{x}^p(t)$ and velocity $\boldsymbol{v}^p(t) = \dot{\boldsymbol{x}}^p(t)$ of a pointlike particle. The equation of motion defining the trajectories $\boldsymbol{x}^p$ will be problem specific.

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A generic form for this equation is considered here, namely,

$$\frac{d^2}{dt^2}x_i^p = F_i(\boldsymbol{x}^p,\boldsymbol{v}^p,t) + f_i(\boldsymbol{x}^p,t). \tag{2}$$

In this equation, which embraces a wide range of models, the stochastic acceleration experienced by a particle with phase-space position $(\boldsymbol{x},\boldsymbol{v})$ at time t has been decomposed into a mean deterministic component $\boldsymbol{F}$ and a zero-mean stochastic term $\boldsymbol{f}$. Clearly both $\boldsymbol{F}$ and $\boldsymbol{f}$ will depend in some way upon the underlying fluid velocity field $\boldsymbol{u}$. The precise nature of this dependence is not of concern here, since the analysis to be presented is of a general nature.

Consideration is given first to the case of inertial particles and the formulation of transport equations for p. This leads naturally to the limiting case of fluid points obtained as $\tau_p \to 0$, where τ_p is some particle response (relaxation) time, characterizing particle inertia. We consider various forms of the PDF equation for p and the consequent $\tau_p \to 0$ forms for ρ. The aim is to resolve some long-standing questions concerning the equivalence (or otherwise) of these different forms and the adherence of the models to the fundamental physical constraint of zero drift. This constraint, which relates to the preservation of a fully mixed state of fluid points in incompressible flow, is discussed in detail later in the paper.

For a single realization of $\boldsymbol{f}$ (and corresponding trajectory $\boldsymbol{x}^p$) we define the fine-grain PDF

$$\mathcal{P}(\boldsymbol{x},\boldsymbol{v},t) = \delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\delta(\boldsymbol{v}^p(t)-\boldsymbol{v}). \tag{3}$$

Then $p = \langle\mathcal{P}\rangle$ and, corresponding to Eq. (2), the evolution of this PDF is governed by

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial x_i}[pv_i] - \frac{\partial}{\partial v_i}[pF_i] - \frac{\partial}{\partial v_i}\langle\mathcal{P}f_i\rangle. \tag{4}$$

Here $\langle\cdot\rangle$ denotes an ensemble average over all realizations of not only the field $\boldsymbol{f}$ but also the initial conditions $\boldsymbol{x}^p(0)$, $\boldsymbol{v}^p(0)$. While (4) represent an exact equation for p it is of little value in this form since the average $\langle\mathcal{P}\boldsymbol{f}\rangle$, referred to as the phase-space diffusion flux, requires closure. The challenge lies in the formulation of closed-form expressions for $\langle\mathcal{P}\boldsymbol{f}\rangle$ that properly take into account both spatial and temporal correlations of $\boldsymbol{u}$ (and by extension $\boldsymbol{f}$) and also any inhomogeneity inherent in this flow field.

A number of authors have addressed this issue and the next section summarizes key results from three different approaches to this closure problem. It is often stated (e.g., [6,8,9]) and widely assumed that, although the methods used in these approaches are distinct, the resulting representations for $\langle\mathcal{P}\boldsymbol{f}\rangle$ are equivalent. One of the purposes of this paper is to draw attention to the fact that this is not so and to assess the implications of the differences. A critical test of the validity of any closure for $\langle\mathcal{P}\boldsymbol{f}\rangle$ is provided by considering the limiting form of the closure for fluid-point dispersion. This limiting form must be consistent with the zero-drift (fully mixed) condition for fluid points. Models which fail to satisfy this physical criterion are said to possess spurious drift, and this defect indicates that the model is not strictly consistent with the underlying dynamics of the particle equation of motion and the turbulent flow field. This issue is addressed in Sec. III. This presents the main results of the paper, showing that only one form of closure can be considered truly consistent with this fully mixed condition. This then also establishes an appropriate form of the passive-scalar PDF equation for $\rho(\boldsymbol{x},t)$.

II. EXPRESSIONS FOR $\langle\mathcal{P}\boldsymbol{f}\rangle$

A range of strategies for closing the flux $\langle\mathcal{P}\boldsymbol{f}\rangle$ can be found in the literature. The methodologies underpinning these approaches can be divided into three distinct categories: (a) Furutsu-Novikov-based methods [6,9–14], (b) Lagrangian history direct interaction (LHDI) methods [15], and (c) van Kampen (VK) operator representation methods [8,16].

In this section results from each of these three methods are presented and analyzed. This highlights that fact that these approaches do not lead to the same result, even though it is often claimed that they do (e.g., [6,8,9]).

A. The Furutsu-Novikov approach

A formula developed independently by Furutsu [17] and Novikov [18] can be used to reformulate correlations of the type $\langle\mathcal{P}\boldsymbol{f}\rangle$. (See [19] for an extensive discussion of the application of this approach.) The method is quite general but the result takes a particularly simple (and *exact*) form when $\boldsymbol{f}$ is a Gaussian field. The result, based on that given in Ref. [12], is

$$\langle\mathcal{P}f_i\rangle = -\left(\frac{\partial}{\partial x_k}p\lambda_{ki} + \frac{\partial}{\partial v_k}p\mu_{ki} - p\kappa_i\right), \tag{5}$$

where the dispersion tensors $\boldsymbol{\lambda}(\boldsymbol{x},\boldsymbol{v},t)$, $\boldsymbol{\mu}(\boldsymbol{x},\boldsymbol{v},t)$, and $\boldsymbol{\kappa}(\boldsymbol{x},\boldsymbol{v},t)$, are given by

$$\lambda_{ki} = \int_0^t \langle\Gamma_{kj}(t;t')R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t)\rangle_{\boldsymbol{x},\boldsymbol{v}}dt', \tag{6}$$

$$\mu_{ki} = \int_0^t \langle\dot{\Gamma}_{kj}(t;t')R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t)\rangle_{\boldsymbol{x},\boldsymbol{v}}dt', \tag{7}$$

$$\kappa_i = \int_0^t \left\langle\Gamma_{kj}(t;t')\frac{\partial}{\partial x_k}R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t)\right\rangle_{\boldsymbol{x},\boldsymbol{v}}dt', \tag{8}$$

where $\langle\cdot\rangle_{\boldsymbol{x},\boldsymbol{v}}$ denotes a conditioned ensemble average in which only values of $\boldsymbol{\Gamma}$ and $\boldsymbol{R}$ evaluated along particle trajectories satisfying $\boldsymbol{x}^p(t)=\boldsymbol{x},\boldsymbol{v}^p(t)=\boldsymbol{v}$ contribute to the average. In these expressions $\boldsymbol{R}$ denotes the Eulerian two-point, two-time correlation tensor for the field $\boldsymbol{f}$, that is,

$$R_{ji}(\boldsymbol{x}',t';\boldsymbol{x},t) = \langle f_j(\boldsymbol{x}',t')f_i(\boldsymbol{x},t)\rangle. \tag{9}$$

Further, in Eq. (8), the gradient operator $\partial/\partial x_k$ acts on the second spatial variable of $\boldsymbol{R}$, that is,

$$\frac{\partial}{\partial x_k}R_{ji}(\boldsymbol{x}',t';\boldsymbol{x},t) = \left\langle f_j(\boldsymbol{x}',t')\frac{\partial}{\partial x_k}f_i(\boldsymbol{x},t)\right\rangle. \tag{10}$$

The response tensor $\boldsymbol{\Gamma}$ in Eqs. (6)–(8) is a functional derivative,

$$\Gamma_{kj}(t;t') = \frac{\delta x_k^p(t)}{\delta f_j(\boldsymbol{x}^p(t'),t')dt'}. \tag{11}$$

This describes the effect of a perturbation in the field $\boldsymbol{f}$ at the particle position at time t' upon the position of the particle at time t. The evolution of $\boldsymbol{\Gamma}$ with respect to t (with $t \geqslant t'$) is governed by Refs. [19,20]

$$\frac{d^2}{dt^2}\Gamma_{kj} = \frac{\partial F_k}{\partial v_i}\frac{d}{dt}\Gamma_{ij} + \left(\frac{\partial F_k}{\partial x_i} + \frac{\partial f_k}{\partial x_i}\right)\Gamma_{ij}, \tag{12}$$

with "initial" conditions $\Gamma_{ij}(t';t') = 0$, $\dot{\Gamma}_{ij}(t';t') = \delta_{ij}$. In Eq. (12) the derivatives of $\boldsymbol{F}$ and $\boldsymbol{f}$ are evaluated on the particle phase-space trajectory $(\boldsymbol{x}^p(t),\boldsymbol{v}^p(t))$.

Note that the representation of $\langle \mathcal{P}\boldsymbol{f}\rangle$ provided by Eq. (5) does not, by itself, constitute a complete closure of this flux since the integrands in Eqs. (6)–(8) still contain unclosed conditional averages involving particle trajectories $\boldsymbol{x}^p(t')$. Some second closure step is necessary to model these trajectories. Only then is the PDF equation properly closed. This second modeling step is not the subject of this paper. Here attention is restricted to the more fundamental question concerning significance of differences resulting from the different basic approaches. In passing it is noted that a new methodology for closing the conditional averages in Eqs. (6)–(8) appropriate for particle dispersion in strongly inhomogeneous turbulence typical of wall-bounded flows has recently been developed [21].

Several authors have made use of the Furutsu-Novikov (FN) approach to close $\langle \mathcal{P}\boldsymbol{f}\rangle$; see, in particular, Hyland *et al.* [6], Derevich and Zaichik [10], Zaichik [9], and Swailes and Darbyshire [12]. However, there appear to be a number of errors present in these works. In fact we would claim that none of these present a strictly correct form: Both [6] and [12] cite an incorrect form of Eq. (12), omitting the spatial gradient term $\partial f_k/\partial x_i$. The implications of this omission are discussed later in Sec. III, where it will be seen that this gradient term plays a pivotal role in ensuring the recovery of the fully mixed, zero-drift condition in the fluid-point limit. The contribution from the fluctuating acceleration gradient is implicit in the derivation presented by Zaichik [9] (see also [10,19]), although this contribution is subsequently neglected in the analysis (and some later papers, e.g., [14]) where approximations for $\boldsymbol{\Gamma}$ appropriate for quasihomogeneous flows are constructed.

Another important distinction to be made between the forms of the dispersions tensors $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$, and $\boldsymbol{\kappa}$ given here and those cited by Hyland *et al.* [6] and Zaichik [9] lies in the definition of $\boldsymbol{R}$: Instead of the deterministic form given here by Eq. (9) both Hyland and Zaichik replace this with the stochastic form

$$\mathcal{R}_{ji}(\boldsymbol{x}',t';\boldsymbol{x},t) = f_j(\boldsymbol{x}',t')f_i(\boldsymbol{x},t). \tag{13}$$

Clearly (9) and (13) are intrinsically different. The form given by Eq. (13) is more in keeping the the closure obtained via the LHDI approach discussed below, but in the context of the FN approach this form would appear to be incorrect. The outline of the FN approach presented in the Appendix emphasizes this point, and a simple illustration of the significance of replacing $\boldsymbol{R}$ by $\boldsymbol{\mathcal{R}}$ is discussed in Ref. [22].

For homogeneous systems the introduction of $\boldsymbol{\mathcal{R}}$ in place of $\boldsymbol{R}$ is not critical since Corrsin's hypothesis [23] can be invoked to justify the claim that the two forms lead to equivalent definitions of the dispersion coefficients. However, for inhomogeneous systems this seems doubtful; the use of Eq. (13) will generate averages in which only those realizations of $\boldsymbol{f}$ leading to the end condition $(\boldsymbol{x}^p(t),\boldsymbol{v}^p(t)) = (\boldsymbol{x},\boldsymbol{v})$ will contribute. In contrast, using (9) there is an implicit contribution from all realizations of $\boldsymbol{f}$, and it is the full Eulerian statistics of this field that are sampled along particle trajectories. Consequently, in inhomogeneous systems, there is likely to be a bias induced by using (13).

A further relation between the dispersion tensors, as introduced in the work by Zaichik, is also important to note. In Ref. [9] (and in later papers such as [7,14,24] by Zaichik and co-workers) it is stated (or at least implied) that

$$\kappa_i \equiv \frac{\partial}{\partial x_k}\lambda_{ki}. \tag{14}$$

At first sight this might appear reasonable since, by comparing (6) and (8), this amounts to interchanging the order of averaging and differentiation in Eq. (8). However, this fails to take into account the fact that the averages are conditional on the end point $\boldsymbol{x}$ so that this interchange is not valid. Formally, $\kappa_i - \frac{\partial}{\partial x_k}\lambda_{ki}$ can be written as

$$-\int_0^t \int_{\boldsymbol{x}'} R_{ji}(\boldsymbol{x}',t';\boldsymbol{x},t)\frac{\partial}{\partial x_k}Q_{kj}(\boldsymbol{x}',t';\boldsymbol{x},\boldsymbol{v},t)\,d\boldsymbol{x}'dt', \tag{15}$$

where

$$Q_{kj}(\boldsymbol{x}',t';\boldsymbol{x},\boldsymbol{v},t) = \langle \Gamma_{kj}(t;t')\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')\rangle_{\boldsymbol{x},\boldsymbol{v}}, \tag{16}$$

and there is no reason to assume that, in general, the derivative of $\boldsymbol{Q}$ with respect to $\boldsymbol{x}$ [the end-point condition on the average in Eq. (16)] is identically zero. The relation given by Eq. (14) can only be considered appropriate for fully homogeneous systems. A conclusive demonstration that (14) does not hold in general can be found in a recent paper [21], where the dispersion tensors were computed explicitly from the simulation of particle trajectories in an inhomogeneous flow. This shows that there is an important contribution to the particle mass flux modeled by the term $\boldsymbol{\kappa} - \nabla\cdot\boldsymbol{\lambda}$. If relation (14) is assumed then this flux will not be taken into account. This issue is intimately related to the concept of spurious drift and the realization of the fully mixed condition for fluid points. This is addressed in Sec. III.

Finally, in Ref. [6] an approximation is made which results in the trajectories $\boldsymbol{x}^p(t')$ introduced in the definition of the dispersion tensors being treated as deterministic (i.e., independent of $\boldsymbol{f}$; see pages 6182 and 6185 in Ref. [6]). This approximation is said to be invoked to allow the evaluation of the spatial integral that appears in the FN derivation. As shown in the Appendix this is both incorrect and unnecessary. Furthermore, it is claimed in Ref. [6] that a similar approximation is necessary in the LHDI formulation of the dispersion tensors (see page 6182 in Ref. [6]). This is not the case. The LHDI approach is considered next.

B. The LHDI approach

In Ref. [15] Reeks derived an expression for $\langle \mathcal{P}\boldsymbol{f}\rangle$ using the method of Lagrangian history direct interaction. The framework used in LHDI is fundamentally different to that using the FN approach. The FN closure is based upon the statistical properties of the underlying turbulent carrier phase in an Eulerian framework, and the closure is exact when the Eulerian field $\boldsymbol{f}$ is Gaussian. In contrast, the LHDI closure is developed based upon the statistical properties of $\boldsymbol{f}$ evaluated along particle trajectories, i.e., $\boldsymbol{f}(\boldsymbol{x}^p(s),s)$, the statistics of which are a subset of those of the field $\boldsymbol{f}$. In the LHDI approach it is the process $\boldsymbol{\phi}(s) \equiv \boldsymbol{f}(\boldsymbol{x}^p(s),s)$ which is considered to be (or approximated as) Gaussian.

The LHDI result for the phase-space diffusion flux, as given in Ref. [15], is

$$\langle \mathcal{P} f_i \rangle = -\left(\frac{\partial}{\partial x_k} p L_{ki} + \frac{\partial}{\partial v_k} p M_{ki} - p K_i \right), \tag{17}$$

with tensors $\boldsymbol{L}(\boldsymbol{x},\boldsymbol{v},t)$, $\boldsymbol{M}(\boldsymbol{x},\boldsymbol{v},t)$, and $\boldsymbol{K}(\boldsymbol{x},\boldsymbol{v},t)$ given by

$$L_{ki} = \int_0^t \langle G_{kj}(t;t')\, f_j(\boldsymbol{x}^p(t'),t')\, f_i(\boldsymbol{x},t) \rangle_{x,v} dt', \tag{18}$$

$$M_{ki} = \int_0^t \langle \dot{G}_{kj}(t;t')\, f_j((\boldsymbol{x}^p(t'),t')\, f_i(\boldsymbol{x},t) \rangle_{x,v} dt', \tag{19}$$

$$K_i = \int_0^t \left\langle G_{kj}(t;t')\, f_j(\boldsymbol{x}^p(t'),t')\, \frac{\partial f_i}{\partial x_k}(\boldsymbol{x},t) \right\rangle_{x,v} dt'. \tag{20}$$

There are two major differences between these forms and those obtained from the FN approach, Eqs. (6)–(8): The first of these differences, as already indicated, lies in the inclusion of the stochastic $\boldsymbol{\mathcal{R}}$, as given by Eq. (13), rather than the deterministic $\boldsymbol{R}$ given by Eq. (9). The second difference is between the LHDI tensor $\boldsymbol{G}$ and the FN response tensor $\boldsymbol{\Gamma}$. The evolution of $\boldsymbol{G}$ is given by [compare with (12)]

$$\frac{d^2}{dt^2} G_{kj} = \frac{\partial F_k}{\partial v_i} \frac{d}{dt} G_{ij} + \frac{\partial F_k}{\partial x_i} G_{ij}, \tag{21}$$

with $G_{kj}(t';t') = 0$, $\dot{G}_{kj}(t';t') = \delta_{kj}$. The absence of spatial gradients $\partial f_k/\partial x_i$ in Eq. (21) is a consequence of the fact that it is a process, $\boldsymbol{\phi}$, rather than a field, $\boldsymbol{f}$, that forms the stochastic input to the model. In Sec. III both the FN and the LHDI formulations are considered in the context of spurious drift. That the FN formulation is drift free while the LHDI formulation is not will be seen to stem from this fundamental difference between $\boldsymbol{\Gamma}$ and $\boldsymbol{G}$.

Reeks, aware that his original LHDI formulation suffered from this drift, developed an alternative "Lagrangian-based" kinetic equation [25,26]. As presented in these references the approach presupposes the existence of a particle velocity field $\boldsymbol{V}(\boldsymbol{x},t)$ such that $\dot{\boldsymbol{x}}^p = \boldsymbol{V}(\boldsymbol{x}^p,t)$. Although this approach resolves the drift problem in a formal sense, from a practical perspective the introduction of a particle velocity field $\boldsymbol{V}$ presents its own problems; this field, unlike $\boldsymbol{u}$, is not a natural model input, and an explicit representation of $\boldsymbol{V}$ (except in the limit $\tau_p \to 0$) is not simple. Indeed, in general, this field need not be unique. For these reasons this formulation is not considered further here. A more natural comparator for the FN and LHDI formulations is that obtained using the operator representation technique introduced by van Kampen. This is considered next.

C. The VK approach

In Ref. [16] Pozorski and Minier derived a PDF kinetic equation using an operator representation technique developed by van Kampen (see, for example, [27,28]). Mashayek and Pandya [8] recognized that the result given in Ref. [16] was incorrect (a fact confirmed by the authors of Ref. [16]) and gave a modified version of the kinetic equation obtained using the VK approach. The method, as presented in Ref. [8], is based on Eq. (4) written in the form

$$\frac{\partial}{\partial t} p = A^0 p + \alpha \langle A^1 \mathcal{P} \rangle, \tag{22}$$

where the operators A^0 and A^1 are defined as

$$A^0[\,\cdot\,] = -\frac{\partial}{\partial x_i}[v_i \cdot] - \frac{\partial}{\partial v_i}[\mathrm{F}_i \cdot], \tag{23}$$

$$\alpha A^1[\,\cdot\,] = -\frac{\partial}{\partial v_i}[f_i \cdot]. \tag{24}$$

The VK method is then used to provide a closure for $\alpha\langle A^1 \mathcal{P}\rangle$. The resulting representation is, in general, only appropriate for $\alpha\tau_c \ll 1$, where τ_c is the correlation time for A^1. It would appear, therefore, that the VK result is not exact but only an approximation valid in a small-parameter regime (and it is not clear that such a condition is satisfied in general for a turbulent flow). Based on this approach the closure of the phase-space dispersion flux can be written as (using a notation that is in keeping with the other closure representations)

$$\langle \mathcal{P} f_i \rangle = -\left(\mathcal{L}_{ik} \frac{\partial}{\partial x_k} p + \mathcal{M}_{ik} \frac{\partial}{\partial v_k} p - \mathcal{K}_i p \right) \tag{25}$$

(see Eq. (4.125) in Ref. [8]), where

$$\mathcal{L}_{ik} = \int_0^t \langle \mathcal{G}_{kj}(t;t') f_j(\boldsymbol{\mathcal{X}}(t'),t') f_i(\boldsymbol{x},t) \rangle dt', \tag{26}$$

$$\mathcal{M}_{ik} = \int_0^t \langle \dot{\mathcal{G}}_{kj}(t;t') f_j(\boldsymbol{\mathcal{X}}(t'),t') f_i(\boldsymbol{x},t) \rangle dt'. \tag{27}$$

(The expression for $\mathcal{K}_i$ and the definition of $\boldsymbol{\mathcal{X}}$ are given below.)

While there is an evident similarity of form between (26), (27) and (18), (19) there are significant differences. Most obviously, the dispersion tensors $\boldsymbol{\mathcal{L}}$ and $\boldsymbol{\mathcal{M}}$ in Eq. (25) appear to the left of the respective partial differential operators. In both the FN and LHDI formulations, Eqs. (5) and (17), the dispersion tensors emerge naturally from the analysis to the right of the operators (which therefore act on the phase-space dependence of these tensors). Moreover, other aspects of the VK formulation that distinguish this from the LHDI form preclude a straightforward reordering of terms in Eq. (25) to place the tensors to the right of the derivatives. Specifically, $\boldsymbol{\mathcal{X}}$ is not the same as $\boldsymbol{x}^p$, and $\mathcal{G}$ is not the same as $\boldsymbol{G}$ or $\boldsymbol{\Gamma}$.

First, $\boldsymbol{\mathcal{X}}$ (unlike $\boldsymbol{x}^p$) is a deterministic trajectory, defined as the solution to

$$\frac{d^2}{dt'^2}\mathcal{X}_i(t') = \frac{d}{dt'}\mathcal{V}_i(t') = F_i(\boldsymbol{\mathcal{X}}(t'),\boldsymbol{\mathcal{V}}(t'),t'), \tag{28}$$

subject to end ($t' = t$) conditions $\boldsymbol{\mathcal{X}}(t) = \boldsymbol{x}$, $\dot{\boldsymbol{\mathcal{X}}}(t) = \boldsymbol{v}$. Therefore $\boldsymbol{\mathcal{X}}(t')$ should be considered an abbreviation for a more precise notation $\boldsymbol{\mathcal{X}}(\boldsymbol{x},\boldsymbol{v},t|t')$. This notation also assists in specifying $\mathcal{G}$ precisely; if we define

$$\mathcal{D}_{mn}(\boldsymbol{y},\boldsymbol{w},s|t) = \frac{\partial \mathcal{X}_m}{\partial v_n}(\boldsymbol{y},\boldsymbol{w},s|t), \tag{29}$$

then

$$\mathcal{G}_{mn}(t;t') = \mathcal{D}_{mn}(\boldsymbol{\mathcal{X}}(\boldsymbol{x},\boldsymbol{v},t|t'),\boldsymbol{\mathcal{V}}(\boldsymbol{x},\boldsymbol{v},t|t'),t'|t). \tag{30}$$

Loosely speaking, $\mathcal{G}$ describes the rate of change of $\boldsymbol{\mathcal{X}}(\boldsymbol{x},\boldsymbol{v},t|t)$ with respect to the rate of change of $\boldsymbol{\mathcal{V}}(\boldsymbol{x},\boldsymbol{v},t\ |t')$. What this shows is that $\mathcal{G}$ (unlike $\boldsymbol{\Gamma}$ and $\boldsymbol{G}$) has an intrinsic dependence on the end condition (phase-space coordinates) $(\boldsymbol{x},\boldsymbol{v})$. Strictly speaking it would be more precise to write $\mathcal{G}(\boldsymbol{x},\boldsymbol{v},t;t')$. It is this dependence of $\mathcal{G}$ (and $\boldsymbol{\mathcal{X}}$) on the coordinates $(\boldsymbol{x},\boldsymbol{v})$ that precludes a simple rearrangement of the VK form Eq. (25)

to place the dispersion tensors on the right-hand side of the differential operators. Definition (30) also makes it clear that, whereas both $\boldsymbol{\Gamma}$ and $\boldsymbol{G}$ are governed by simple ordinary differential equations, Eqs. (12) and (21), there is no equivalent (simple) characterization for $\mathcal{G}$.

The form of $\mathcal{K}$ in Eq. (25) is

$$\mathcal{K}_i = -\int_0^t \langle \psi(\boldsymbol{x},\boldsymbol{v},t|t') f_i(\boldsymbol{x},t)\rangle dt', \tag{31}$$

where

$$\psi(\boldsymbol{x},\boldsymbol{v},t|t') = \mathcal{G}_{kj}(\boldsymbol{x},\boldsymbol{v},t;t')\frac{\partial}{\partial x_k} f_j(\boldsymbol{x},\boldsymbol{v},t|t') + \dot{\mathcal{G}}_{kj}(\boldsymbol{x},\boldsymbol{v},t;t')\frac{\partial}{\partial v_k} f_j(\boldsymbol{x},\boldsymbol{v},t|t'), \tag{32}$$

with

$$f_j(\boldsymbol{x},\boldsymbol{v},t|t') \equiv f_j(\boldsymbol{\mathcal{X}}(\boldsymbol{x},\boldsymbol{v},t|t'),t'). \tag{33}$$

As a final point note that the ensemble averages in Eqs. (26), (27), and (31) do not (unlike the corresponding LHDI and FN formulations) involve a conditionality on the realizations of $\boldsymbol{f}$ contributing to these ensembles; all realizations of $\boldsymbol{f}$ are involved. This again draws attention to the marked difference between the VK formulation and the other two.

In summary, it has been demonstrated that the PDF kinetic equations obtained using the FN, LHDI, and VK closures are not equivalent but differ in some fundamental aspects. Although the FN and LHDI formulations are not equivalent they may both be "correct," in the sense that each may be exact within its respective Gaussian framework. On the other hand, the VK result may be appropriate when $\alpha\tau_c \ll 1$. A natural question therefore is: Which of the formulations is most suitable for modeling particle dispersion in inhomogeneous flow?

One way in which this question can be addressed is with regard to the issue of spurious drift. For an initially uniform distribution of fluid points in an inhomogeneous, incompressible flow, the distribution of the fluid points necessarily remains spatially uniform for all times. Models which fail to satisfy this physical criterion are said to posses spurious drift, and such a defect indicates that the model is not strictly consistent with the underlying particle dynamics and turbulent flow field.

In the next section it will be shown that in order for a kinetic equation to be free from spurious drift, the closure for the phase-space flux must satisfy certain conditions in the limit of fluid points. All three closures are examined in this limit.

III. PDF EQUATIONS IN THE LIMIT $\tau_p \to 0$

The aim is to assess the closure methodologies in the context of the fully mixed condition for fluid points, especially in the case of inhomogeneous flows. There are two questions that must addressed: The first concerns whether or not a closure methodology is consistent with the fully mixed condition when applied directly to the case of fluid-point dynamics. If one assumes that it is consistent then the second, subsidiary question concerns whether or not the closure, as formulated for inertial particles, recovers the limiting form for fluid points as $\tau_p \to 0$.

To make precise what is meant by the fully mixed condition, and to answer these questions, consider fluid points transported in a velocity field $\boldsymbol{u}(\boldsymbol{x},t)$, with trajectories $\boldsymbol{x}^f(t)$ defined by Eq. (1). As an initial condition set $\boldsymbol{x}^f(0) = \boldsymbol{x}^0$, with $\boldsymbol{x}^0$ a random variable with some PDF $\varphi^0(\boldsymbol{x})$. Then, treating the field $\boldsymbol{u}$ as stochastic (and independent of $\boldsymbol{x}^0$), we define the following PDFs:

$$\varrho(\boldsymbol{x},t) = \delta(\boldsymbol{x}^f(t) - \boldsymbol{x}), \tag{34}$$

$$\varphi(\boldsymbol{x},t) = \langle \varrho(\boldsymbol{x},t)\rangle_{\boldsymbol{u}}^{x^0}, \tag{35}$$

$$\rho(\boldsymbol{x},t) = \langle \varrho(\boldsymbol{x},t)\rangle = \langle \varphi(\boldsymbol{x},t)\rangle^{\boldsymbol{u}}. \tag{36}$$

Here $\langle\cdot\rangle_{\boldsymbol{u}}^{x^0}$ denotes an ensemble average over all realizations of $\boldsymbol{x}^0$ for a given, single realization of the flow field $\boldsymbol{u}$, and $\langle\cdot\rangle^{\boldsymbol{u}}$ denotes an ensemble average over all $\boldsymbol{u}$. The decomposition $\langle\cdot\rangle = \langle\langle\cdot\rangle_{\boldsymbol{u}}^{x^0}\rangle^{\boldsymbol{u}}$ is used later in the analysis. From Eq. (34)

$$\frac{\partial}{\partial t}\varrho(\boldsymbol{x},t) = -\frac{\partial}{\partial x_i}[\varrho(\boldsymbol{x},t)]u_i(\boldsymbol{x}^f(t),t). \tag{37}$$

Averaging Eq. (37) over all realizations of $\boldsymbol{x}^0$ (for a single realization of $\boldsymbol{u}$) gives

$$\frac{\partial}{\partial t}\varphi(\boldsymbol{x},t) = -\frac{\partial}{\partial x_i}[\varphi(\boldsymbol{x},t)u_i(\boldsymbol{x},t)]. \tag{38}$$

The solution to Eq. (38) with initial condition $\varphi(\boldsymbol{x},0) = \varphi^0(\boldsymbol{x})$ can be expressed as

$$\varphi(\boldsymbol{x},t) = \varphi^0(\boldsymbol{x}^f(\boldsymbol{x},t|0)) \times \exp\left[-\int_0^t \frac{\partial u_i}{\partial x_i}(\boldsymbol{x}^f(\boldsymbol{x},t|t'),t')dt'\right], \tag{39}$$

where $\boldsymbol{x}^f(\boldsymbol{x},t|t')$ denotes the fluid-point trajectory $\boldsymbol{x}^f(t')$ which satisfies $\boldsymbol{x}^f(t) = \boldsymbol{x}$. Now suppose that the fluid points are are fully mixed (uniformly distributed) at $t = 0$, so φ^0 is independent of position. Suppose, further, that the velocity field is incompressible. Then, from Eq. (39), it follows that $\varphi(\boldsymbol{x},t) \equiv \varphi^0$ for all $\boldsymbol{x},t$. Hence also $\rho(\boldsymbol{x},t) \equiv \varphi^0$. The fluid points remain fully mixed. Note that this result holds even when the field $\boldsymbol{u}$ is inhomogeneous, and it is this condition that must be respected by the PDF equation for $\rho(\boldsymbol{x},t)$. This equation follows from averaging Eq. (38) over all realizations of $\boldsymbol{u}$. With $\boldsymbol{u}$ decomposed into mean and fluctuating fields, $\langle\boldsymbol{u}\rangle$ and $\boldsymbol{u}' = \boldsymbol{u} - \langle\boldsymbol{u}\rangle$, respectively, this averaging gives [analogous to Eq. (4)]

$$\frac{\partial}{\partial t}\rho = -\frac{\partial}{\partial x_i}\rho\langle u_i\rangle - \frac{\partial}{\partial x_i}\langle\varrho u_i'\rangle. \tag{40}$$

It is necessary to formulate a closure for the flux $\langle\varrho\boldsymbol{u}'\rangle$. Note that if the system is fully mixed then

$$\langle\varrho u_i'\rangle = \left\langle\langle\varrho u_i'\rangle_{\boldsymbol{u}}^{x^0}\right\rangle^{\boldsymbol{u}} = \langle\varphi u_i'\rangle^{\boldsymbol{u}} = \varphi^0\langle u_i'\rangle^{\boldsymbol{u}} = 0. \tag{41}$$

Thus, any closure for $\langle\varrho\boldsymbol{u}'\rangle$ must be shown to be consistent with result (41) when applied to a fully mixed system of fluid points. This, in turn, will ensure that the resulting solution of the PDF equation (40) will preserve this fully mixed state. The FN closure is considered first. As will be seen, this closure does respect the fully mixed condition. The analysis involved in demonstrating this fact then, in turn, draws attention to deficiencies in the LHDI and VK formulations.

A. FN drift analysis

Applying the FN methodology to $\langle \varrho \boldsymbol{u}' \rangle$ leads to the representation [29]

$$\langle \varrho u_i' \rangle = \rho \kappa_i^f - \frac{\partial}{\partial x_k} \rho \lambda_{ki}^f, \tag{42}$$

where $\boldsymbol{\lambda}^f(\boldsymbol{x},t)$ and $\boldsymbol{\kappa}^f(\boldsymbol{x},t)$ are given by [compare with Eqs. (6) and (8)]

$$\lambda_{ki}^f = \int_0^t \left\langle \Gamma_{kj}^f(t;t') R_{ji}^f(\boldsymbol{x}^f(t'),t';\boldsymbol{x},t) \right\rangle_{\boldsymbol{x}} dt', \tag{43}$$

$$\kappa_i^f = \int_0^t \left\langle \Gamma_{kj}^f(t;t') \frac{\partial}{\partial x_k} R_{ji}^f(\boldsymbol{x}^f(t'),t';\boldsymbol{x},t) \right\rangle_{\boldsymbol{x}} dt', \tag{44}$$

with

$$R_{ji}^f(\boldsymbol{x}',t';\boldsymbol{x},t) = \langle u_j'(\boldsymbol{x}',t')\, u_i'(\boldsymbol{x},t) \rangle \tag{45}$$

and

$$\Gamma_{kj}^f(t;t') = \frac{\delta x_k^f(t)}{\delta u_j'(\boldsymbol{x}^f(t'),t')dt'}. \tag{46}$$

Note that if the flow is incompressible ($\partial u_j/\partial x_j = 0$) then

$$\frac{\partial}{\partial x_j'} R_{ji}(\boldsymbol{x}',t';\boldsymbol{x},t) = 0. \tag{47}$$

The FN methodology underpinning this representation for $\langle \varrho \boldsymbol{u}' \rangle$ is such that the result should be exact provided only that the field $\boldsymbol{u}'$ is Gaussian. This field can be inhomogeneous, and the result should still be exact. The fully mixed condition provides a stringent test of this statement. To establish that the FN formulation is consistent with the fully mixed condition first note the closure expression given by Eq. (42) will satisfy (41) in the fully mixed case provided that the corresponding forms of $\boldsymbol{\lambda}^f$ and $\boldsymbol{\kappa}^f$ satisfy

$$\kappa_i^f - \frac{\partial}{\partial x_k} \lambda_{ki}^f = 0, \qquad \text{all } i. \tag{48}$$

The challenge, then, is to demonstrate directly that the expressions for $\boldsymbol{\lambda}^f$ and $\boldsymbol{\kappa}^f$ given by Eqs. (43) and (44) satisfy (48) in the case of fully mixed points in an incompressible flow. That this is the case is far from obvious and requires a formal demonstration. Such a demonstration, which is nontrivial, is given below.

An important point is that relation (48) need not hold for a passive scalar that is not fully mixed. In the case of scalar dispersal from localized sources the left-hand side of Eq. (48) will contribute to the scalar flux. In contradiction to this, the relation (14) introduced by Zaichik *et al.* [7,9,24] would imply that (48) was invariably recovered as $\tau_p \to 0$, irrespective of whether the initial distribution of points was uniform. Not only is the relation given by Eq. (14) invalid for inertial particles, it cannot, in general, be considered a reliable asymptotic approximation.

For a given realization of the field $\boldsymbol{u}$ let $\boldsymbol{x}^f(\boldsymbol{y},t|s)$ denote a fluid-point trajectory $\boldsymbol{x}^f(s)$ satisfying $\boldsymbol{x}^f(t) = \boldsymbol{y}$. There is no restriction or presumption here on the sign of $t - s$. For an incompressible flow field this trajectory will be unique. The key to establishing that the fully mixed condition is preserved by the FN formulation is the corresponding Jacobian tensor $\boldsymbol{J}(\boldsymbol{y},t,s)$ defined by

$$J_{mn}(\boldsymbol{y},t,s) = \frac{\partial}{\partial y_n} x_m^f(\boldsymbol{y},t|s). \tag{49}$$

The Jacobian tensor $\boldsymbol{J}$ possesses an inverse relation with the response tensor $\boldsymbol{\Gamma}^f$. To see this, consider the evolution of $\boldsymbol{J}$ with respect to s,

$$\frac{\partial}{\partial s} J_{mn}(\boldsymbol{y},t,s) = \frac{\partial u_m}{\partial x_k}(\boldsymbol{x}^f(\boldsymbol{y},t|s),s) J_{kn}(\boldsymbol{y},t,s). \tag{50}$$

By noting that $J_{ij}(\boldsymbol{y},t,t) = \delta_{ij}$ (all $\boldsymbol{y},t$), it follows that

$$\boldsymbol{J}(\boldsymbol{y},t,s) = \exp\left(\int_t^s \nabla \boldsymbol{u}(\boldsymbol{x}^f(\boldsymbol{y},t|s'),s')ds' \right), \tag{51}$$

where $(\nabla \boldsymbol{u})_{mk} = \partial u_m/\partial x_k$. Now consider the response tensor $\boldsymbol{\Gamma}^f(t;t')$ given by Eq. (46). The evolution of this with respect to t ($t \geqslant t'$) is given by Refs. [19,20]

$$\frac{\partial}{\partial t} \Gamma_{kj}^f(t;t') = \frac{\partial u_k}{\partial x_i}(\boldsymbol{x}^f(t),t) \Gamma_{ij}^f(t;t') \tag{52}$$

with $\Gamma_{kj}^f(t';t') = \delta_{kj}$. Thus

$$\boldsymbol{\Gamma}^f(t;t') = \exp\left(\int_{t'}^t \nabla \boldsymbol{u}(\boldsymbol{x}^f(t''),t'')dt'' \right). \tag{53}$$

If the trajectory $\boldsymbol{x}^f$ is given by $\boldsymbol{x}^f(t'') = \boldsymbol{x}^f(\boldsymbol{y},t|t'')$, then $\boldsymbol{\Gamma}^f(t;t')$ becomes dependent on $(\boldsymbol{y},t)$. We write

$$\boldsymbol{H}(\boldsymbol{y},t,t') = \exp\left(\int_{t'}^t \nabla \boldsymbol{u}(\boldsymbol{x}^f(\boldsymbol{y},t|t''),t'')dt'' \right). \tag{54}$$

Comparing Eqs. (54) and (51) indicates that

$$\boldsymbol{H}(\boldsymbol{y},t,t') = \boldsymbol{J}^{-1}(\boldsymbol{y},t,t'). \tag{55}$$

This inverse relation between the Jacobian and response tensors is central to the following drift analysis. From Eq. (49) it also follows that

$$\boldsymbol{J}(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t',t) = \boldsymbol{J}^{-1}(\boldsymbol{y},t,t'). \tag{56}$$

Now consider the left-hand side of Eq. (48) with $\boldsymbol{\lambda}^f$ and $\boldsymbol{\kappa}^f$ given by Eqs. (43) and (44):

$$\kappa_i^f - \frac{\partial}{\partial x_k} \lambda_{ki}^f = \int_0^t \left\langle \Gamma_{kj}^f(t;t') \frac{\partial}{\partial x_k} R_{ji}^f(\boldsymbol{x}^f(t'),t';\boldsymbol{x},t) \right\rangle_{\boldsymbol{x}} - \frac{\partial}{\partial x_k} \left\langle \Gamma_{kj}^f(t;t') R_{ji}^f(\boldsymbol{x}^f(t'),t';\boldsymbol{x},t) \right\rangle_{\boldsymbol{x}} dt'. \tag{57}$$

In the first term of the integrand in Eq. (57) the operator $\partial/\partial x_k$ acts only on the second spatial input of $\boldsymbol{R}^f$. In the second term the differentiation is with respect to both this dependence in $\boldsymbol{R}^f$ and also the conditionality $\boldsymbol{x}^f(t) = \boldsymbol{x}$ on the average.

Consequently, Eq. (57) can be written as

$$\kappa_i^f - \frac{\partial}{\partial x_k}\lambda_{ki}^f = -\int_0^t \frac{\partial}{\partial y_k}\big\langle \Gamma_{kj}^f(t;t')R_{ji}^f(\boldsymbol{x}^f(t'),t';\boldsymbol{x},t)\big\rangle_{\boldsymbol{y}} dt'\big|_{y=x}, \tag{58}$$

where $\partial/\partial y_k$ is now used instead of $\partial/\partial x_k$ to emphasize that this differentiation is now with respect to the condition $\boldsymbol{x}^f(t) = \boldsymbol{y}$ and not the $\boldsymbol{x}$ dependence in $\boldsymbol{R}$.

A sufficient condition to ensure that Eq. (48) is satisfied is therefore

$$\frac{\partial}{\partial y_k}\big\langle \Gamma_{kj}^f(t;t')R_{ji}^f(\boldsymbol{x}^f(t'),t';\boldsymbol{x},t)\big\rangle_{\boldsymbol{y}} = 0, \text{ all } i. \tag{59}$$

A simple manipulation of weighted averages shows that any conditional average of the form $\langle\cdot\rangle_{\boldsymbol{y}}$ can be decomposed in the form

$$\langle\cdot\rangle_{\boldsymbol{y}} = \frac{1}{\rho(\boldsymbol{y},t)}\big\langle \varphi(\boldsymbol{y},t)\langle\cdot\rangle_{\boldsymbol{y},\boldsymbol{u}}^{\boldsymbol{x}^0}\big\rangle^{\boldsymbol{u}}. \tag{60}$$

The outer average is over all realizations of the field $\boldsymbol{u}$, while the inner average is over all realizations of the initial condition $\boldsymbol{x}^0$ (for a given realization of $\boldsymbol{u}$) such that the resulting trajectories $\boldsymbol{x}^f(s)$ satisfy $\boldsymbol{x}^f(t) = \boldsymbol{y}$. If $\boldsymbol{u}$ is incompressible then there is a single trajectory contributing to this inner average, namely, $\boldsymbol{x}^f(\boldsymbol{y},t|s)$. By making use of this decomposition on Eq. (59) it follows that, for a fully mixed system, it is sufficient to show

$$\frac{\partial}{\partial y_k}\big[H_{kj}(\boldsymbol{y},t,t')R_{ji}^f(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t';\boldsymbol{x},t)\big] = 0, \text{ all } i. \tag{61}$$

The left-hand side of Eq. (61) can be written as

$$\underbrace{H_{kj}(\boldsymbol{y},t,t')\left[\frac{\partial}{\partial y_k}R_{ji}^f(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t';\boldsymbol{x},t)\right]}_{\mathbf{1}} + \underbrace{\left[\frac{\partial}{\partial y_k}H_{kj}(\boldsymbol{y},t,t')\right]R_{ji}^f(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t';\boldsymbol{x},t)}_{\mathbf{2}}. \tag{62}$$

It is therefore sufficient to show that, in Eq. (62), both terms **1** and **2** are identically zero. Consider term **1**:

$$\begin{aligned} H_{kj}(\boldsymbol{y},t,t')&\left[\frac{\partial}{\partial y_k}R_{ji}^f(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t';\boldsymbol{x},t)\right] \\ &= H_{kj}(\boldsymbol{y},t,t')\frac{\partial}{\partial y_k}x_n^f(\boldsymbol{y},t|t')\frac{\partial}{\partial x_n'}R_{ji}^f(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t';\boldsymbol{x},t) \\ &= H_{kj}(\boldsymbol{y},t,t')J_{nk}(\boldsymbol{y},t,t')\frac{\partial}{\partial x_n'}R_{ji}^f(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t';\boldsymbol{x},t) \\ &= \frac{\partial}{\partial x_j'}R_{ji}^f(\boldsymbol{x}^f(\boldsymbol{y},t|t'),t';\boldsymbol{x},t) \equiv 0. \end{aligned} \tag{63}$$

The last line follows by virtue of the inverse relation between $\boldsymbol{H}$ and $\boldsymbol{J}$, Eq. (55), and the incompressibility condition, Eq. (47).

It remains, therefore, to show that term **2** in Eq. (62) is identically zero, that is,

$$\frac{\partial}{\partial y_k}H_{kj}(\boldsymbol{y},t,t') \equiv 0, \qquad \text{all } j. \tag{64}$$

This can be demonstrated by using (55) again. For incompressible flow det$[\boldsymbol{J}] = 1$ and so, in three dimensions, the inverse of $\boldsymbol{J}$ is given by

$$\boldsymbol{J}^{-1} = \begin{pmatrix} +(J_{22}J_{33}-J_{23}J_{32}) & -(J_{12}J_{33}-J_{13}J_{32}) & +(J_{12}J_{23}-J_{13}J_{22}) \\ -(J_{21}J_{33}-J_{23}J_{31}) & +(J_{11}J_{33}-J_{13}J_{31}) & -(J_{11}J_{23}-J_{13}J_{21}) \\ +(J_{21}J_{32}-J_{22}J_{31}) & -(J_{11}J_{32}-J_{12}J_{31}) & +(J_{11}J_{22}-J_{12}J_{21}) \end{pmatrix}. \tag{65}$$

Therefore, with $j = 1$,

$$\begin{aligned}\frac{\partial}{\partial y_k}H_{k1} = &+\frac{\partial}{\partial y_1}(J_{22}J_{33}-J_{23}J_{32}) - \frac{\partial}{\partial y_2}(J_{21}J_{33}-J_{23}J_{31}) \\ &+\frac{\partial}{\partial y_3}(J_{21}J_{32}-J_{22}J_{31}),\end{aligned} \tag{66}$$

and similarly for the $j = 2,3$ components of $\partial H_{kj}/\partial y_k$. Result (64) then follows by making use of the fact that

$$\frac{\partial}{\partial y_k}J_{ij} = \frac{\partial}{\partial y_k}\frac{\partial}{\partial y_j}x_i^f = \frac{\partial}{\partial y_j}\frac{\partial}{\partial y_k}x_i^f = \frac{\partial}{\partial y_j}J_{ik}. \tag{67}$$

The conclusion, therefore, is that the FN formulation of the kinetic equation for fluid-point dispersion preserves the fully mixed condition in both homogeneous and inhomogeneous flows, and so it is free of artificial drift.

It remains to demonstrate that the FN closure for inertial particles reduces asymptotically to the corresponding fluid-point form in the limit of $\tau_p \to 0$. To simplify the demonstration of this result assume, *a priori*, that the dependence of $\boldsymbol{f}$ on $\boldsymbol{u}'$ is linear. This avoids the need to introduce any linearization and small-τ_p order analysis, and it ensures that $\boldsymbol{f}$ and $\boldsymbol{u}'$ can both be treated consistently as Gaussian. This case is realized by the simple Stokes drag model for the particle equation of motion: $\ddot{\boldsymbol{x}}^p = \tau_p^{-1}(\boldsymbol{u}(\boldsymbol{x}^p,t) - \dot{\boldsymbol{x}}^p)$. We write $\boldsymbol{f} = \tau_p^{-1}\boldsymbol{u}'$, so that

$$\tau_p\langle\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})f_i\rangle = \langle\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})u_i'\rangle. \tag{68}$$

In the limit $\tau_p \to 0$ the right-hand side of Eq. (68) becomes $\langle\varrho u_i'\rangle$ with the fluid-point closure given by Eq. (42). To see that this limit is recovered independently from the left-hand side of Eq. (68) using the closure for inertial particles given by Eq. (5) note that, from this equation,

$$\tau_p\langle\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})f_i\rangle = \tau_p\int_{\boldsymbol{v}}\langle\mathcal{P}f_i\rangle d\boldsymbol{v} = \tau_p\left(\rho_p\overline{\kappa}_i - \frac{\partial}{\partial x_k}\rho_p\overline{\lambda}_{ki}\right). \tag{69}$$

Here the PDF ρ_p gives the particle spatial distribution,

$$\rho_p(\boldsymbol{x},t) = \langle\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\rangle = \int_{\boldsymbol{v}} p(\boldsymbol{x},\boldsymbol{v},t)d\boldsymbol{v}, \tag{70}$$

and

$$\overline{\lambda}_{ki} = \frac{1}{\rho_p}\int_{\boldsymbol{v}} p\lambda_{ki}d\boldsymbol{v} = \int_0^t\langle\Gamma_{kj}(t;t')R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t)\rangle_{\boldsymbol{x}}dt', \tag{71}$$

$$\overline{\kappa}_i = \frac{1}{\rho_p}\int_{\boldsymbol{v}} p\kappa_i d\boldsymbol{v} = \int_0^t\left\langle\Gamma_{kj}(t;t')\frac{\partial}{\partial x_k}R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t)\right\rangle_{\boldsymbol{x}}dt'. \tag{72}$$

Clearly $\rho_p = \langle\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\rangle \to \langle\delta(\boldsymbol{x}^f(t)-\boldsymbol{x})\rangle = \rho$ as $\tau_p\to 0$. It follows that the closure for inertial particles given by Eq. (69) will reduce to the fluid-point closure for $\langle\varrho\boldsymbol{u}'\rangle$ given by Eq. (42) in the limit $\tau_p\to 0$ provided

$$\lim_{\tau_p\to 0}\tau_p\overline{\boldsymbol{\lambda}} = \boldsymbol{\lambda}^f, \qquad \lim_{\tau_p\to 0}\tau_p\overline{\boldsymbol{\kappa}} = \boldsymbol{\kappa}^f. \tag{73}$$

That these conditions are satisfied follows from consideration of the relations among $\boldsymbol{R}$, $\boldsymbol{\Gamma}$, and $\boldsymbol{R}^f$, $\boldsymbol{\Gamma}^f$. Namely, comparing (45) and (46) to Eqs. (9) and (11) with $\boldsymbol{f} = \tau_p^{-1},\boldsymbol{u}'$, one has

$$R_{ji} = \tau_p^{-2}R_{ji}^f \quad\text{and}\quad \tau_p^{-1}\Gamma_{kj}\to\Gamma_{kj}^f. \tag{74}$$

B. LHDI drift analysis

The preceding analysis for the FN formulation can be applied in a similar manner to the LHDI closure. This leads to the fully mixed requirement

$$K_i^f - \frac{\partial}{\partial x_k}L_{ki}^f = 0, \qquad \text{all } i, \tag{75}$$

where now

$$K_i^f - \frac{\partial}{\partial x_k}L_{ki}^f = -\int_0^t\frac{\partial}{\partial y_k}\big\langle G_{kj}^f(t;t')u_j'(\boldsymbol{x}^f(t'),t')u_i'(\boldsymbol{x},t)\big\rangle_{\boldsymbol{y}}dt'\big|_{\boldsymbol{y}=\boldsymbol{x}}. \tag{76}$$

The crucial difference lies in the distinction between $\boldsymbol{\Gamma}^f$ and $\boldsymbol{G}^f$: Whereas the definition of $\boldsymbol{\Gamma}^f$, Eq. (52), involves gradients of the instantaneous field $\boldsymbol{u}$, the definition of $\boldsymbol{G}^f$ replaces this with the mean field $\langle\boldsymbol{u}\rangle$ [analogous to the difference between (11) and (21)]. Thus, instead of $\boldsymbol{H}$ defined by Eq. (54), the subsequent analysis will introduce

$$\widetilde{\boldsymbol{H}}(\boldsymbol{y},t,t') = \exp\left(\int_{t'}^t\nabla\langle\boldsymbol{u}\rangle(\boldsymbol{x}^f(\boldsymbol{y},t|t''),t'')dt''\right). \tag{77}$$

The significance of this difference is that $\widetilde{\boldsymbol{H}}\neq\boldsymbol{J}^{-1}$ [compare with (55)] so that, in general, the integrand in Eq. (76) will not be identically zero in the fully mixed case. The conclusion, therefore, is that the LHDI methodology does not guarantee a drift-free closure for $\langle\mathcal{P}\boldsymbol{f}\rangle$, but it is only strictly valid for the homogeneous case, and with $\langle\boldsymbol{u}\rangle\equiv\boldsymbol{0}$, so that $\boldsymbol{G}^f\equiv\boldsymbol{I}$.

C. VK drift analysis

Applying the VK methodology to $\langle\varrho\boldsymbol{u}'\rangle$ leads to the representation

$$\langle\varrho u_i'\rangle = \rho\mathcal{K}_i^f - \mathcal{L}_{ik}^f\frac{\partial}{\partial x_k}\rho, \tag{78}$$

where [compare with Eqs. (26) and (31)]

$$\mathcal{L}_{ik}^f = \int_0^t\big\langle\mathcal{G}_{kj}^f(\boldsymbol{x},t;t')u_j'(\boldsymbol{\mathcal{X}}^f(\boldsymbol{x},t|t'),t')u_i'(\boldsymbol{x},t)\big\rangle dt', \tag{79}$$

$$\mathcal{K}_i^f = -\int_0^t\langle\psi^f(\boldsymbol{x},t|t')u_i'(\boldsymbol{x},t)\rangle dt'. \tag{80}$$

Here $\boldsymbol{\mathcal{X}}^f(\boldsymbol{x},t|t')$ satisfies

$$\frac{d}{dt'}\mathcal{X}_i^f = \langle u_i\rangle(\boldsymbol{\mathcal{X}}^f,t'), \quad \boldsymbol{\mathcal{X}}^f(\boldsymbol{x},t|t) = \boldsymbol{x}, \tag{81}$$

and $\mathcal{G}^f(\boldsymbol{x},t;t')$ is defined by

$$\mathcal{G}_{mn}^f(\boldsymbol{x},t;t') = \mathcal{D}_{mn}^f(\boldsymbol{\mathcal{X}}^f(\boldsymbol{x},t|t'),t'|t), \tag{82}$$

with

$$\mathcal{D}_{mn}^f(\boldsymbol{y},s|t) = \frac{\partial\mathcal{X}_m^f}{\partial x_n}(\boldsymbol{y},s|t). \tag{83}$$

In Eq. (80)

$$\psi^f(\boldsymbol{x},t|t') = \mathcal{G}_{kj}^f(\boldsymbol{x},t;t')\frac{\partial}{\partial x_k}u_j'(\boldsymbol{x},t|t'), \tag{84}$$

with

$$u_j'(\boldsymbol{x},t|t')\equiv u_j'(\boldsymbol{\mathcal{X}}^f(\boldsymbol{x},t|t'),t'). \tag{85}$$

Consideration of Eq. (78) shows that, in contrast to conditions (48) and (75) for the FN and LHDI formulations, the VK formulation requires $\boldsymbol{\mathcal{K}}^f = \boldsymbol{0}$ to be satisfied for a fully mixed system.

Comparing (83) with Eq. (49) shows that $\boldsymbol{\mathcal{D}}^f$ is the Jacobian associated with trajectories $\boldsymbol{\mathcal{X}}^f$ defined by the mean flow field $\langle\boldsymbol{u}\rangle$. Therefore, comparing (82) with Eqs. (55) and (56), one has

$$\mathcal{G}^f(\boldsymbol{x},t;t') = (\boldsymbol{\mathcal{D}}^f)^{-1}(\boldsymbol{x},t|t'). \tag{86}$$

It follows that $\psi^f = 0$, and so also $\boldsymbol{\mathcal{K}}^f = \boldsymbol{0}$, provided only that the flow is incompressible. This result holds irrespective of whether or not the system is fully mixed. It would therefore seem that the VK formulation fails to capture a scalar flux contribution associated with nonuniform distributions in inhomogeneous, incompressible flow.

There is a further issue with the VK formulation. Consider the dispersion of an initially nonuniform distribution of fluid points in incompressible isotropic turbulence. In such a system the fluid points would disperse throughout the flow field until their distribution $\rho(\boldsymbol{x},t)$ became spatially uniform, and the diffusion coefficient describing this dispersion process would be related to the Lagrangian integral time scale of the flow. In the VK formulation the diffusion coefficient for fluid points is given by Eq. (79), in which the correlations of $\boldsymbol{u}'$ are determined along the deterministic trajectory $\boldsymbol{\mathcal{X}}^f(\boldsymbol{x},t|t')$. For isotropic turbulence $\langle \boldsymbol{u}(\boldsymbol{x},t)\rangle = \boldsymbol{0}$, and consequently from Eq. (81) we have $\boldsymbol{\mathcal{X}}^f(\boldsymbol{x},t|t') = \boldsymbol{x}$. The implication of this is that for fluid-point dispersion in isotropic turbulence the VK diffusion coefficient contains correlations of $\boldsymbol{u}'$ evaluated at a fixed point in space, $\boldsymbol{x}$, so that the dispersion rate predicted would be proportional to the *Eulerian* integral time scale of $\boldsymbol{u}'$ rather than the *Lagrangian* integral time scale. This is clearly incorrect and points to a serious defect in the dispersion tensors resulting from the VK formulation of the PDF equation.

IV. CONCLUSIONS

The analysis presented in this paper has highlighted important differences among three PDF kinetic equations used to model the distribution of passive scalars or inertial particles dispersing in a turbulent flow. The three forms of the kinetic equation originate from three distinct methods (FN, LHDI, and VK) used to construct a closed expression for a turbulence induced mass (or number) density flux. The three closure expressions exhibit superficial similarities, and in the literature it is often claimed that the corresponding kinetic equations are essentially equivalent. It has been shown that this is not the case and that differences among the models have an important bearing on the efficacy of these when applied to inhomogeneous flows.

An important contribution of the work is the formal demonstration that one form of kinetic equation, obtained using the FN closure, does not exhibit the defect of spurious drift and will preserve the fully mixed state of fluid points transported in an incompressible, inhomogeneous turbulent flow. Moreover, this analysis explains why the kinetic equation obtained from the LHDI closure fails to satisfy this condition and is only strictly valid for homogeneous systems. The third form of the kinetic equation, obtained from the VK closure, is formally consistent with the fully mixed condition but fails to account for a nonzero scalar flux contribution associated with nonuniform scalar distributions in inhomogeneous, incompressible systems. Further, this VK closure is based on an expansion in terms of a small parameter reflecting the magnitude of turbulence intensity relative to a rate scale for the decorrelation of turbulent flow velocities. This approximation is manifest in the formulation by the presence of mean (as opposed to stochastic) particle trajectories in the definition of the VK dispersion tensors. While this affords a significant simplification to the closure problem—removing conditional averages within the dispersion tensors—the validity of such an approximation in strongly inhomogeneous flows is still a moot point.

However, even for the simpler case of dispersion in isotropic turbulence there seems to be a serious issue with the VK formulation. The VK diffusion coefficient for fluid-point dispersion in isotropic turbulence suggests that the dispersion rate should be proportional to the Eulerian integral time scale, rather than the Lagrangian integral time scale. This is clearly incorrect and arises precisely because the VK dispersion tensors contain correlations of the fluid velocity field evaluated along mean trajectories. Therefore the question of the validity of the VK closure relates not only to inhomogeneous flows but also to homogeneous, isotropic flows.

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APPENDIX: FN FLUX CLOSURE

This Appendix outlines the Furutsu-Novikov (FN) derivation of the flux closure. For a Gaussian field $\boldsymbol{f}$ the FN formula gives the exact result [12,19]

$$\langle \mathcal{P} f_i\rangle = \int_0^t \int_{\boldsymbol{x}'} R_{ij}(\boldsymbol{x},t;\boldsymbol{x}',t')\left\langle \frac{\delta \mathcal{P}}{\delta f_j(\boldsymbol{x}',t')d\boldsymbol{x}'dt'}\right\rangle d\boldsymbol{x}'dt'. \tag{A1}$$

The functional derivative

$$\frac{\delta \mathcal{P}}{\delta f_j(\boldsymbol{x}',t')d\boldsymbol{x}'dt'} \tag{A2}$$

describes how a perturbation in $\boldsymbol{f}$ at $(\boldsymbol{x}',t')$ will affect the fine-grain, phase-space PDF $\mathcal{P}$ of the particle at time t. This derivative can be rewritten [6,9,10,12] as

$$\begin{aligned}\frac{\delta \mathcal{P}}{\delta f_j(\boldsymbol{x}',t')d\boldsymbol{x}'dt'} &= -\frac{\partial \mathcal{P}}{\partial x_k}\Gamma_{kj}(t;t')\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')\\ &\quad -\frac{\partial \mathcal{P}}{\partial v_k}\dot{\Gamma}_{kj}(t;t')\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}'),\end{aligned} \tag{A3}$$

where

$$\Gamma_{kj}(t;t') = \frac{\delta x_k^p(t)}{\delta f_j(\boldsymbol{x}^p(t'),t')dt'}, \tag{A4}$$

and $\dot{\boldsymbol{\Gamma}} = \frac{d}{dt}\boldsymbol{\Gamma}$. Therefore, noting that the averaging and integration operations in Eq. (A1) commute, we have

$$\begin{aligned}\langle \mathcal{P} f_i\rangle &= -\int_0^t \left\langle \int_{\boldsymbol{x}'} R_{ij}(\boldsymbol{x},t;\boldsymbol{x}',t')\right.\\ &\quad\left.\times\left(\frac{\partial \mathcal{P}}{\partial x_k}\Gamma_{kj}+\frac{\partial \mathcal{P}}{\partial v_k}\dot{\Gamma}_{kj}\right)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')d\boldsymbol{x}'\right\rangle dt'\\ &= -\int_0^t\left\langle R_{ij}(\boldsymbol{x},t;\boldsymbol{x}^p(t'),t')\left(\frac{\partial \mathcal{P}}{\partial x_k}\Gamma_{kj}+\frac{\partial \mathcal{P}}{\partial v_k}\dot{\Gamma}_{kj}\right)\right\rangle dt'.\end{aligned} \tag{A5}$$

This can be rearranged further (by noting that $\boldsymbol{R}$ is a function of $\boldsymbol{x}$ but that $\boldsymbol{\Gamma}$ is not) as

$$\begin{aligned}\langle \mathcal{P} f_i\rangle &= -\int_0^t \frac{\partial}{\partial x_k}\langle \mathcal{P}\Gamma_{kj}R_{ij}(\boldsymbol{x},t;\boldsymbol{x}^p(t'),t')\rangle\\ &\quad -\left\langle \mathcal{P}\Gamma_{kj}\frac{\partial}{\partial x_k}R_{ij}(\boldsymbol{x},t;\boldsymbol{x}^p(t'),t')\right\rangle\\ &\quad +\frac{\partial}{\partial v_k}\langle \mathcal{P}\dot{\Gamma}_{kj}R_{ij}(\boldsymbol{x},t;\boldsymbol{x}^p(t'),t')\rangle dt'.\end{aligned} \tag{A6}$$

Finally, by making use of the basic result [30]

$$\langle \mathcal{P}(\boldsymbol{x},\boldsymbol{v},t)\cdot\rangle = p(\boldsymbol{x},\boldsymbol{v},t)\,\langle\cdot\rangle_{\boldsymbol{x},\boldsymbol{v}} \tag{A7}$$

and the fact that $R_{ij}(\boldsymbol{x},t;\boldsymbol{x}',t') = R_{ji}(\boldsymbol{x}',t';\boldsymbol{x},t)$, the closure becomes

$$\langle \mathcal{P} f_i \rangle = -\frac{\partial}{\partial x_k}\left[p \int_0^t \langle \Gamma_{kj} R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t)\rangle_{\boldsymbol{x},\boldsymbol{v}} dt' \right] - \frac{\partial}{\partial v_k}\left[p \int_0^t \langle \dot{\Gamma}_{kj} R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t)\rangle_{\boldsymbol{x},\boldsymbol{v}} dt' \right] + p \int_0^t \left\langle \Gamma_{kj} \frac{\partial}{\partial x_k} R_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x},t) \right\rangle_{\boldsymbol{x},\boldsymbol{v}} dt', \quad \text{(A8)}$$

which is the result given by Eqs. (5)–(8).

Two important points should be noted about this derivation and result. First, and contrary to the assertion made in Ref. [6], it is unnecessary (and incorrect) to replace the stochastic particle trajectories $\boldsymbol{x}^p$ appearing in the ensemble averages with deterministic trajectories $\boldsymbol{X}$ governed by $\ddot{\boldsymbol{X}} = \boldsymbol{F}(\boldsymbol{X},\dot{\boldsymbol{X}},t)$. Second, it is the correlation tensor $\boldsymbol{R}$ that must be used in Eq. (A1) (and the subsequent conditional averages) and not the nonaveraged form $\boldsymbol{f}(\boldsymbol{x},t)\boldsymbol{f}(\boldsymbol{x}',t')$.

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