# Is the kinetic equation for turbulent gas-particle flows ill posed?

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This paper is about the kinetic equation for gas-particle flows, in particular its well-posedness and realizability and its relationship to the generalized Langevin model (GLM) probability density function (PDF) equation. Previous analyses, e.g. [J.-P. Minier and C. Profeta, Phys. Rev. E **92**, 053020 (2015)], have concluded that this kinetic equation is ill posed, that in particular it has the properties of a backward heat equation, and as a consequence, its solution will in the course of time exhibit finite-time singularities. We show that this conclusion is fundamentally flawed because it ignores the coupling between the phase space variables in the kinetic equation and the time and particle inertia dependence of the phase space diffusion tensor. This contributes an extra *positive* diffusion that always outweighs the *negative* diffusion associated with the dispersion along one of the principal axes of the phase space diffusion tensor. This is confirmed by a numerical evaluation of analytic solutions of these *positive* and *negative* contributions to the particle diffusion coefficient along this principal axis. We also examine other erroneous claims and assumptions made in previous studies that demonstrate the apparent superiority of the GLM PDF approach over the kinetic approach. In so doing, we have drawn attention to the limitations of the GLM approach, which these studies have ignored or not properly considered, to give a more balanced appraisal of the benefits of both PDF approaches.

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## I. INTRODUCTION

The probability density function (PDF) approach has proved very useful in studying the behavior of stochastic systems. Familiar examples of its usage occur in the study of Brownian motion [1] and in the kinetic theory of gases [2]. In more recent times it has been used extensively by Pope and others to model turbulence [3] and turbulence-related phenomena, such as combustion [4] and atmospheric dispersion [5]. This paper is about its application to particle transport in turbulent gas flows, where it has been developed and refined over a number of years by numerous authors. During that time it has been successfully applied to a whole range of turbulent dispersed flow problems involving mixing and dispersion as well as particle collisions and clustering in a particle pair formulation of the approach. It has also formed a fundamental basis for dealing with complex flows in formulating the continuum equations and constitutive relations for the dispersed phase precisely analogous to the way the Maxwell-Boltzmann equation has been used in the kinetic theory. It has become an established technique for studying dispersed flows so much so that the method and its numerous applications are the subject of a recent book [6] and the subject of a chapter in the recent Multiphase Flow Handbook [7].

There are currently two PDF approaches that have been used extensively to describe the transport, mixing, and collisions of small particles in turbulent gas flows. The first approach referred to as the kinetic approach is based on a kinetic equation for the PDF $p(\boldsymbol{x},\boldsymbol{v},t)$ of the particle position $\boldsymbol{x}$ and velocity $\boldsymbol{v}$ at time $t$. This equation is based on a particle equation of motion involving the flow velocity along a particle trajectory derived from a Gaussian stochastic flow field. In the kinetic equation the particles' random motion arising from this stochastic field is manifest as a diffusive flux, which is a linear combination of gradient diffusion in both $\boldsymbol{x}$ and $\boldsymbol{v}$. Transient spatiotemporal structures in the turbulence give rise to an extra force due to clustering and preferential sweeping of particles [8].

In the second PDF approach an equation for the PDF $p(\boldsymbol{x},\boldsymbol{v},\boldsymbol{u},t)$ is constructed, where $\boldsymbol{u}$ is the carrier flow velocity sampled along particle paths. Thus, unlike the kinetic approach, the flow velocity in this approach is retained in the particle phase space and is described by a model evolution equation. In particular, this PDF model is based on a generalized Langevin model (GLM) (see Pope [3]), where the velocity of the underlying carrier flow measured along a particle trajectory is described by a generalized Langevin equation. As such the associated PDF equation is described by a Fokker-Planck equation. This GLM PDF equation has sometimes been inappropriately referred to as the dynamic PDF equation [9], implying that it is a more general PDF approach from which the kinetic equation can in general be derived. However, it is

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important to appreciate the kinetic equation is not a standard Fokker-Planck equation, since it captures the non-Markovian features of the underlying flow velocities.

The problem of closure and the associated realizability and well-posedness of PDF equations are profoundly important in the study of stochastic equations. So despite the successful application of the kinetic equation to a whole range of problems, recent claims in the literature of ill-posedness and realizability of this equation are disturbing and a serious concern. The root cause of this concern is the nonpositive definiteness of the diffusion tensor associated with the phase space diffusion flux. That in particular this tensor has both positive and negative eigenvalues implying that along the principal axis of the diffusion tensor with a *negative* eigenvalue, the particle dispersion exhibits the properties of a backward diffusion equation leading to solutions with finite time singularities. In fact, Minier and Profeta [9], following a detailed analysis of the relative merits of the 2 PDF approaches, have concluded that the kinetic equation is ill posed and therefore an invalid description of disperse two-phase flows (except in the limiting case for particles with large Stokes numbers when the kinetic equation reduces to a Fokker-Planck equation). This raises a number of issues and inconsistencies that we wish to examine and resolve:

(1) The closure of the diffusive terms in the kinetic equation is exact for a Gaussian process for the aerodynamic driving forces in the particle equation of motion. Notwithstanding any *negative* eigenvalues, such dispersion processes are demonstrably forward rather than backward in time with statistical moments that monotonically increase rather than decrease with time. This behavior is reflected in the analytic solutions of the kinetic equation for particle dispersion in shear flows in which the mean shear is linear and the turbulence is statistically homogeneous and stationary (see Hyland *et al.* [10], Swailes and Darbyshire [11]). In these generic flows, there is exact correspondence of the analytical solution with a random walk simulation using a Lagrangian particle tracking approach, solving the individual particle equations of motion in the associated Gaussian random flow field. See, as an example, the illustration in Fig. 1.

(2) In simple generic flows the GLM PDF equation is entirely consistent with the kinetic equation, i.e., the kinetic equation is recoverable from the GLM equations and has exactly the same solution for the same mean flows and statistical correlations for the turbulent velocity $\boldsymbol{u}$ along particle trajectories [12]. They are both compatible with a Gaussian process. The claim of ill-posedness of the kinetic equation would therefore seem to contradict the well-posedness associated with the Fokker-Planck equation of the GLM.

So the first objective of the analysis we present here is to show that despite the non positive definiteness of the phase space diffusion tensor, this does not imply backward diffusion and the existence of finite time singularities, that the kinetic equation is well posed and has realizable solutions that are forward rather than backward in time consistent with a Gaussian process. We shall show that this is intimately related to the non Markovian nature of the kinetic equation, that the time evolution of the phase dispersion tensor from its initial state and the coupling between phase space variables are crucial considerations. In the course of this analysis we will recall the stages of the development of the kinetic equation and the important role played by certain consistency and invariance principles which taken together with the other features determining well-posedness and realizability have not been properly understood or appreciated in previous analyses.

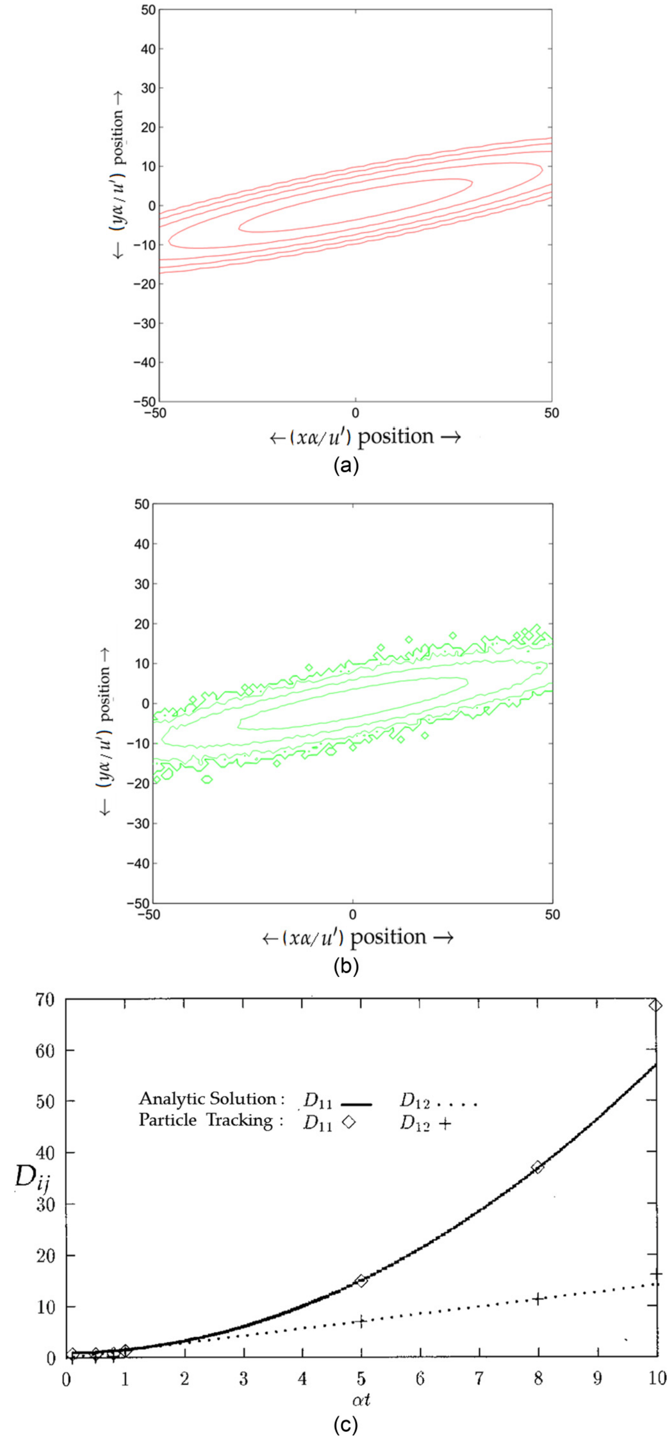


FIG. 1. Dispersion of an instantaneous point source of particles in a simple turbulent shear flow. Comparison of the analytic solution of the kinetic equation for the particle spatial concentration and a random walk simulation based on Stokes drag with a Gaussian process for the aerodynamic driving force. Panels (a) and (b) for the concentration contours are taken from Ref. [11], where the analytic solution is also given. Panel (c) is taken from Ref. [10], where analytic solutions are also given. See also Refs. [7,12].

Previous work has purported to show that the GLM is a more general approach than the kinetic approach. That in particular the kinetic equation can be derived from the GLM, and that the features of transport and mixing in more general non uniform inhomogeneous turbulent flows implicit in the solutions of the kinetic equation are intrinsic to the GLM. So the second objective of this analysis is to examine the basis for this assertion. In the process, we provide a more balanced appraisal of the benefits of both PDF approaches and point out the limitations of the GLM that have been ignored in previous analyses. We regard these limitations to be areas for improvement of the GLM rather than inherent deficiencies. Like all modeling approaches, each of the two approaches considered have their strengths and weaknesses. A categorical dismissal of one in preference to another in previous work would seem misplaced. From a practical point of view this paper is more about how one approach can support the other in solving dispersed flow problems.

## II. ILL-POSED KINETIC PDF EQUATIONS?

In this section, we examine in detail the previous analysis of Minier and Profeta (M&P) [9] that leads to the assertion of ill-posedness of the kinetic equation. For ease of comparison we use the same notation here and throughout the paper. Thus, M&P consider particle phase-space trajectories $\boldsymbol{Z}_p(t) = (\boldsymbol{X}_p(t),\boldsymbol{U}_p(t))$ governed by

$$\dot{\boldsymbol{X}}_p = \boldsymbol{U}_p, \qquad \dot{\boldsymbol{U}}_p = \frac{1}{\tau_p}(\boldsymbol{U}_s - \boldsymbol{U}_p) + \boldsymbol{F}_{\text{ext}}. \tag{1}$$

$\boldsymbol{U}_s(t)$ representing a flow velocity at time $t$ sampled along the trajectory $\boldsymbol{X}_p(t)$, and $\boldsymbol{F}_{\text{ext}}$ an external body force, e.g., gravity. In the kinetic modeling framework, $\boldsymbol{U}_s$ is derived via an underlying flow velocity field $\boldsymbol{u}_f(\boldsymbol{x},t)$, which has both a mean $\langle \boldsymbol{u}_f\rangle$ and fluctuating (zero mean) component $\boldsymbol{u}'_f$. That is $\boldsymbol{U}_s = \boldsymbol{u}_f(\boldsymbol{X}_p(t),t)$. The pdf $p(z,t) = \langle \delta(z - \boldsymbol{Z}_p(t))\rangle$ giving the distribution of $\boldsymbol{Z}_p$ then satisfies the ensemble-averaged Liouville equation:

$$\partial_t p = -\,\partial_{\boldsymbol{x}} \cdot \boldsymbol{v} p - \partial_{\boldsymbol{v}} \cdot \left(\boldsymbol{F}_{\text{ext}} + \frac{1}{\tau_p}(\langle \boldsymbol{u}_f\rangle - \boldsymbol{v})\right)p \\ - \partial_{\boldsymbol{v}} \cdot \left\langle \frac{1}{\tau_p}\boldsymbol{u}'_f \delta(z - \boldsymbol{Z}_p)\right\rangle. \tag{2}$$

A number of works have formulated expressions for the diffusive flux in the final term of Eq. (2), e.g., [13,14]: Modeling $\boldsymbol{u}_f(\boldsymbol{x},t)$ as Gaussian, and treating the particle response time $\tau_p$ as a constant, independent of the particle Reynolds number (i.e., Stokes relaxation), leads to the general form

$$\left\langle \frac{1}{\tau_p}\boldsymbol{u}'_f \delta(z - \boldsymbol{Z}_p)\right\rangle = -(\boldsymbol{\kappa} p + \partial_{\boldsymbol{x}} \cdot \boldsymbol{\lambda} p + \partial_{\boldsymbol{v}} \cdot \boldsymbol{\mu} p),$$

and Eq. (2) can then be written compactly in phase-space notation as

$$\partial_t p = -\partial_z \cdot \boldsymbol{a} p \; + \; \tfrac{1}{2}\partial_z \cdot (\partial_z \cdot \boldsymbol{B} p), \tag{3}$$

where $z = (\boldsymbol{x},\boldsymbol{v})$ refers to the particle position and velocity and

$$\boldsymbol{a} = \left(\boldsymbol{v}, \boldsymbol{F}_{\text{ext}} + \frac{1}{\tau_p}\big(\langle \boldsymbol{u}_f(\boldsymbol{x},t)\rangle - \boldsymbol{v}\big) + \boldsymbol{\kappa}\right), \tag{4}$$

$$\boldsymbol{B} = \left(\begin{array}{c|c} \boldsymbol{0} & \boldsymbol{\lambda} \\ \hline \boldsymbol{\lambda}^{\top} & \boldsymbol{\mu} + \boldsymbol{\mu}^{\top}\end{array}\right), \tag{5}$$

$\boldsymbol{\lambda}$ and $\boldsymbol{\mu}$ are diffusion tensors that define gradient dispersion separately in real space ($\boldsymbol{x}$) and velocity space ($\boldsymbol{v}$), respectively. They are functions of time and depend on the particle response to the carrier flow velocity fluctuations along its trajectory. The specific forms for $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$, and $\boldsymbol{\kappa}$, based on the LHDI closure scheme [15], are

$$\lambda_{ij} = \tau_p^{-2}\int_0^t \langle g_{ki}(t-s)u'_k(\boldsymbol{x},\boldsymbol{v},t|s)u'_j(\boldsymbol{x},t)\rangle ds,$$
$$\mu_{ij} = \tau_p^{-2}\int_0^t \langle \dot{g}_{ki}(t-s)u'_k(\boldsymbol{x},\boldsymbol{v},t|s)u'_j(\boldsymbol{x},t)\rangle ds, \tag{6}$$
$$\kappa_j = \tau_p^{-2}\int_0^t \langle g_{ki}(t-s)u'_k(\boldsymbol{x},\boldsymbol{v},t|s)\partial_{x_i}u'_j(\boldsymbol{x},t)\rangle ds,$$

where the particle response tensor $\boldsymbol{g}(t-s)$ has elements $g_{ki}(t-s)$ corresponding to the displacement at time $t$ in the $i$ direction when $\tau_p u'_f$ is an impulsive force $\delta(t-s)$ applied in the $k$ direction. In general, $\boldsymbol{g}(t-s)$ depends upon the local straining and rotation of the flow. We mention that the response tensor based on the Furutsu-Novikov closure scheme [14] is slightly different in definition (see Ref. [16] for a discussion of the different closure schemes for the kinetic equation). Following the analysis of M&P, we consider the case for dispersion of an instantaneous point source in statistically stationary homogeneous and isotropic turbulence with a zero external force $\boldsymbol{F}_{\text{ext}} = \boldsymbol{0}$, in which case $\boldsymbol{g}(t) = \tau_p(1 - e^{-t/\tau_p})\,\boldsymbol{I}$, and

$$\boldsymbol{\lambda} = \tau_p^{-1}\int_0^t (1 - e^{-s/\tau_p})R(s)ds\; \boldsymbol{I},$$
$$\boldsymbol{\mu} = \tau_p^{-2}\int_0^t e^{-s/\tau_p}R(s)ds\; \boldsymbol{I}, \tag{7}$$
$$\boldsymbol{\kappa} = \boldsymbol{0},$$

where $R(s)$ is the autocorrelation $\frac{1}{3}\langle \boldsymbol{U}'_s(0)\cdot \boldsymbol{U}'_s(s)\rangle$ of the flow velocity fluctuations $\boldsymbol{U}'(s)$ measured along a particle trajectory. Equations (3), (4), and (5) correspond to Eqs. (65), (66), and (67) in Ref. [9]. M&P claim that Eq. (3) is ill posed in the sense that solutions to this can (will) exhibit unphysical behavior except in special or, to use their phrase, "lucky" cases. Specifically, they assert that solutions $p$ of Eq. (3) will exhibit finite-time singularities except for very special initial conditions, for example, with a Gaussian form. Their justification for this claim is based on an analysis centered round the observation that $\boldsymbol{B}$ is not positive-definite but possesses both negative and positive eigenvalues. We show here that their analysis is incorrect.

First, we note that Eq. (3) is *not* a model for the PDF of $\boldsymbol{Z}_p(t)$, but describes precisely how this PDF must evolve. There is an *exact* correspondence between Eq. (3) and the underlying equation of motion Eq. (1). This equivalence, i.e., the formal derivation of Eq. (3) from Eq. (1), is subject only to the requirement that the field $\boldsymbol{u}_f(\boldsymbol{x},t)$ is Gaussian. Then, notwithstanding the nondefiniteness of $\boldsymbol{B}$, Eq. (3) is an exact description of how $p$, as determined by Eq. (1), behaves. Contrary to previous claims [9], no Gaussian (or other) constraint is necessary on the initial distribution $p^0(z)$ of $\boldsymbol{Z}_p(0)$. Thus, should solutions to Eq. (3) exhibit finite-time, or even asymptotic ($t \to \infty$), singularities when $p^0$ is non-Gaussian, then this feature must be inherent in the system determined by Eq. (1). Either this singular behavior is intrinsic to the system, or the analysis upon which M&P base their conclusion is incorrect.

To demonstrate that the nondefiniteness of $\boldsymbol{B}$, coupled with arbitrary initial conditions, does not lead to singular solutions of Eq. (3) we note that the solution to this equation can be written

$$p(t;z) = \int \phi(t;z,z')p^0(z')dz', \tag{8}$$

where $\phi(t;z,z')$ is the fundamental solution satisfying $\phi(0;z,z') = \delta(z - z')$. Now consider the case when $\boldsymbol{U}'_s(t) = \boldsymbol{u}'_f(\boldsymbol{X}_p,t) = \boldsymbol{u}_f(\boldsymbol{X}_p,t) - \langle \boldsymbol{u}_f\rangle(\boldsymbol{X}_p,t)$ is treated *ab initio* as a Gaussian process. The structure of Eq. (3) remains unchanged, except $\boldsymbol{\kappa} \equiv \boldsymbol{0}$ and $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$ are independent of $\boldsymbol{Z}$ (but, crucially, they will still depend on $t$). $\boldsymbol{B}$ still has negative eigenvalues. With $\langle \boldsymbol{u}_f\rangle$ linear in $\boldsymbol{x}$ (and $\boldsymbol{F}_{\text{ext}}$ constant) the form of $\phi$ is well-documented, both in general terms and for a number of specific linear flows [11,12,17]. This solution is Gaussian, and it is straightforward to show that it corresponds exactly, as it must, to the Gaussian form of $\boldsymbol{Z}_p$ determined by Eq. (1). Thus, any singular behavior of the general solution $p$, defined by Eq. (8), can only be a consequence of degeneracy in the Gaussian form of $\phi$, and not the form of an arbitrary initial distribution $p^0$. Again, should such degeneracy exist then it would be symptomatic of behavior determined by Eq. (1), and not some artifact of the nondefiniteness of $\boldsymbol{B}$.

There are several flaws in the analysis upon which M&P base their claim of ill-posedness: To begin, they consider a form of Eq. (3) in which $\boldsymbol{B}$ is taken as independent of time, arguing that this corresponds to stationary isotropic turbulence. This is not correct. $\boldsymbol{B}$ is intrinsically time dependent. This dependence reflects the nonzero time correlations implicit in the turbulent velocity field $\boldsymbol{U}_f$, and the consequent non-Markovian nature of $\boldsymbol{Z}_p$. Moreover, and crucially, $\boldsymbol{B}(0) = \boldsymbol{0}$ unless the initial values $\boldsymbol{U}_p(0)$, $\boldsymbol{U}_s(0)$ are correlated. A detailed analysis of this is given in Ref. [17]. So, even when $\boldsymbol{B} \to \boldsymbol{B}^\infty$ (constant) as $t \to \infty$, it is inappropriate to set $\boldsymbol{B} = \boldsymbol{B}^\infty$ in a formal analysis of the time problem. Indeed, it is straightforward to show that the fundamental solution $\phi$ breaks down for arbitrarily small $t$ when this inappropriate approximation is introduced.

Of course, the nondefiniteness of $\boldsymbol{B}$ is not altered by taking this tensor to be $t$ dependent. The eigensolution-based transformation that M&P introduce can still be invoked. Analogous to Eq. (71) in Ref. [9] we define trajectories $\widetilde{\boldsymbol{Z}}_p(t)$ with components $(\widetilde{Z}_{p1},\widetilde{Z}_{p2})$ in a transformed phase space $\widetilde{z} = (\widetilde{z}_1,\widetilde{z}_2)$ with

$$\widetilde{\boldsymbol{Z}}_p(t) = \mathrm{P}^\top \cdot \boldsymbol{Z}_p(t), \tag{9}$$

where $\mathrm{P}(t)$ is the transformation matrix determined by the (now time dependent) normalized eigenvectors of $\boldsymbol{B}$. Thus, $\mathrm{P}^\top \cdot \mathrm{P} = \mathrm{I}$ and $\mathrm{P}^\top \cdot \boldsymbol{B} \cdot \mathrm{P} = \boldsymbol{\Lambda} = \mathrm{diag}(\omega_i)$, with $\omega_i$ the eigenvalues of $\boldsymbol{B}$. We note that, in applying this to the 2D case considered by the M&P, it is sensible to label the two eigenvalues such that $\omega_1 < 0$, $\omega_2 > 0$ since this gives $\mathrm{P}(0) = \mathrm{I}$. By neglecting the time dependence in $\boldsymbol{B}$ M&P missed this point and chose the opposite ordering (see Eq. (69) in Ref. [9]). Here we take $\omega_1 < 0$.

In using the transform given by Eq. (9), it is important to note that Eq. (1) governing $\boldsymbol{Z}_p(t)$ is not to be interpreted as a stochastic differential equation driven by a white-noise process, and Eq. (3) is not a corresponding Fokker-Planck equation. Clearly, this would be nonsense since $\boldsymbol{B}$ is not positive-definite. It is more transparent (and correct) to note that Eq. (9) implies that the PDF $\widetilde{p}(\widetilde{z},t)$ of $\widetilde{\boldsymbol{Z}}_p(t)$ is related to the PDF $p(z,t)$ of $\boldsymbol{Z}_p(t)$ by $\widetilde{p}\,|J| = p$, where $J = \det[P]$ is the Jacobean of the transform $\widetilde{z} = P^\top \cdot z$. Since $P$ is orthogonal we have $J = 1$. The PDF equation for $\widetilde{p}$ is

$$\partial_t \widetilde{p} = -\partial_{\widetilde{\mathbf{z}}} \cdot \widehat{\boldsymbol{a}}\widetilde{p} \; + \; \tfrac{1}{2}\partial_{\widetilde{\mathbf{z}}} \cdot (\partial_{\widetilde{\mathbf{z}}} \cdot \boldsymbol{\Lambda}\widetilde{p}), \tag{10}$$

where $\widehat{\boldsymbol{a}} = P^\top \cdot \widetilde{\boldsymbol{a}} + R \cdot \widetilde{z}$, $\widetilde{\boldsymbol{a}}(\widetilde{z},t) = \boldsymbol{a}(z,t)$, $R = \dot{P}^\top \cdot P$. This is analogous to Eq. (72) in Ref. [9], except these authors have not included the time dependence in $\boldsymbol{B}$ and so set $\dot{P} = 0$. We note that $R$ represents a rate of rotation matrix, $\mathrm{trace}(R) = 0$. In the 2D model considered, the authors integrate Eq. (10) over $\widetilde{z}_2$ (corresponding to the transformed variable with the positive eigenvalue $\omega_2$) to obtain (compare with Eq. (74) in Ref. [9])

$$\partial_t \widetilde{p}_r = -\partial_{\widetilde{z}_1} \bar{\widehat{\mathrm{a}}}_1 \widetilde{p}_r \; - \; \partial^2_{\widetilde{z}_1} \tfrac{1}{2}|\omega_1|\widetilde{p}_r, \tag{11}$$

where $\widetilde{p}_r$ is the PDF for $\widetilde{Z}_{p1}$ and $\bar{\widehat{\mathrm{a}}}_1 \widetilde{p}_r = \int \widehat{\mathrm{a}}_1 \widetilde{p}\, d\widetilde{z}_2$. Based on the negative diffusion coefficient in Eq. (11), M&P seek to show that this equation and so also Eq. (3) is ill posed. Their argument fails to take into account that the conditional average $\bar{\widehat{\mathrm{a}}}_1$ is a density weighted average, i.e., its value at $z_1$ is dependent upon the distribution of $Z_{p2}(t)$ at $z_1$ which itself can be a function $z_1$. For instance, using a more explicit notation we may write

$$\bar{\widehat{\mathrm{a}}}_1 \equiv \langle \widehat{\mathrm{a}}_1(\widetilde{z}_1,\widetilde{Z}_{p2}(t))\rangle_{\widetilde{z}_1}, \tag{12}$$

where $\langle\cdot\rangle_{\widetilde{z}_1}$ denotes an ensemble average conditioned on $\widetilde{Z}_{p1}(t) = \widetilde{z}_1$. What Eq. (12) illustrates is that only a subset of all trajectories $\widetilde{Z}_{p2}(t)$ contribute to $\bar{\widehat{\mathrm{a}}}_1$, namely those that are also associated with $\widetilde{Z}_{p1}(t) = \widetilde{z}_1$. The term $\bar{\widehat{\mathrm{a}}}_1$ is therefore affected by coupling between $\widetilde{Z}_{p1}(t)$ and $\widetilde{Z}_{p2}(t)$. Indeed, in the case where $\widetilde{Z}_{p1}(t)$ and $\widetilde{Z}_{p2}(t)$ are statistically decoupled, we have

$$\langle \widehat{\mathrm{a}}_1(\widetilde{z}_1,\widetilde{Z}_{p2}(t))\rangle_{\widetilde{z}_1} = \langle \widehat{\mathrm{a}}_1(\widetilde{z}_1,\widetilde{Z}_{p2}(t))\rangle, \tag{13}$$

i.e., all realizations of $\widetilde{Z}_{p2}(t)$ would contribute to $\bar{\widehat{\mathrm{a}}}_1$. In this case, $\bar{\widehat{\mathrm{a}}}_1(z_1)$ is convective as M&P have assumed. However, in general, $\widetilde{Z}_{p1}(t)$ and $\widetilde{Z}_{p2}(t)$ will be statistically coupled, and as a consequence, $\bar{\widehat{\mathrm{a}}}_1$ cannot be treated as an arbitrary convective term. Indeed, as we shall show momentarily, the term $\bar{\widehat{\mathrm{a}}}_1$ is associated with both convective and diffusive fluxes, and its diffusional contribution offsets that associated with the negative eigenvalue.

By failing to appreciate this particular property of $\bar{\widehat{\mathrm{a}}}_1$, M&P [9] have overlooked a fundamental property of the particle dispersion process. That is in the dynamical system described by Eq. (1), the particle position and velocity are not independent. This is reflected in the fixed-frame kinetic Eq. (3) through the term $\partial_x vp$, which couples the spatial and velocity distributions of the particles. In the same way, the distributions of the variables $\widetilde{Z}_{p1}, \widetilde{Z}_{p2}$ are coupled in Eq. (11). The implication of this coupling is that fluctuations in particle velocity give rise to fluctuations in particle position, in addition to the fluctuations in particle position that arise directly from fluctuations in the fluid force $\tau_p^{-1}\boldsymbol{U}_s$. In the moving frame, it is the fluctuations in $\widetilde{Z}_{p2}$ (with the positive eigenvalue, $\omega_2$), via the positive covariance between between $\widetilde{Z}_{p1}$ and $\widetilde{Z}_{p2}$, that overcomes the negative diffusion associated with $\widetilde{Z}_{p1}$ (in the absence of the coupling). We note, for instance, that in Eq. (3), the particle flux $\boldsymbol{v}p$ integrated over all particle velocities is expressible as a net gradient diffusion flux, $\overline{\boldsymbol{v}}p_r$ for which the long term ($t \to \infty$) particle diffusion coefficient $\varepsilon(\infty)$ in statistically stationary, homogeneous, isotropic turbulence is given by

$$\varepsilon(\infty) = \tau_p\{\langle v^2(\infty)\rangle + \lambda(\infty)\}, \tag{14}$$

where $\langle v^2(\infty)\rangle$ is the variance of the particle velocity (which for a Gaussian process is given by $(\tau_p/3)\mathrm{trace}[\boldsymbol{\mu}(\infty)]$; see, e.g., Eqs. (78) and (79) in Ref. [15]), and $\lambda = (1/3)\mathrm{trace}(\boldsymbol{\lambda})$. This simple relationship clearly identifies the two sources of dispersion independently, the first from fluctuations in the particle velocity (the kinetic contribution) and the second term $\lambda(\infty)$ arising from fluctuations in $\tau_p^{-1}\boldsymbol{U}_s$ (the turbulent aerodynamic force contribution). We refer to Ref. [13] for a detailed analysis of how this relationship defines an equation of state for the particle pressure and where $\langle v^2(\infty)\rangle$ and $\lambda(\infty)$ are more correctly identified as the normal components of stress tensors. We refer to Ref. [18] on how a proper treatment of the integrated flux terms in the kinetic equation in inhomogeneous turbulence gives rise to turbophoresis, an important mechanism for particle deposition (in response to the unfounded criticism in Refs. [9,19] that the kinetic equation is inappropriate for modeling particle deposition).

To demonstrate these features in a quantitative way we consider the simple 2D case examined by M&P in which $\langle \boldsymbol{U}\rangle = \boldsymbol{0}$, and $\widetilde{\boldsymbol{Z}}_p(0) = \widetilde{\mathrm{z}}^0$ fixed. Then $\widehat{\mathbf{a}}$ is linear in $\widetilde{\mathrm{z}}$, and $\overline{\widehat{\mathrm{a}}}_1\widetilde{p}_r$ involves $\overline{\widetilde{\mathrm{z}}}_2\widetilde{p}_r = \int \widetilde{\mathrm{z}}_2\widetilde{p}\, d\widetilde{\mathrm{z}}_2$. This can be expressed in terms of convective and gradient diffusive fluxes (see Ref. [14]),

$$\overline{\widetilde{\mathrm{z}}}_2\widetilde{p}_r = \widetilde{m}_2\widetilde{p}_r - \widetilde{\theta}_{21}\partial_{\widetilde{\mathrm{z}}_1}\widetilde{p}_r, \tag{15}$$

where $\widetilde{m}_2, \widetilde{\theta}_{21}$ are components of $\langle\widetilde{\boldsymbol{Z}}_p\rangle = \widetilde{\boldsymbol{m}} = (\widetilde{m}_1, \widetilde{m}_2)$ and $\langle(\widetilde{\boldsymbol{Z}}_p - \widetilde{\boldsymbol{m}})(\widetilde{\boldsymbol{Z}}_p - \widetilde{\boldsymbol{m}})\rangle = \widetilde{\Theta} = (\widetilde{\theta}_{ij})$, satisfying

$$\dot{\widetilde{\boldsymbol{m}}} = \tilde{\boldsymbol{\Gamma}}\cdot\widetilde{\boldsymbol{m}} + \widetilde{\boldsymbol{k}}, \tag{16}$$

$$\dot{\widetilde{\Theta}} = \tilde{\boldsymbol{\Gamma}}\cdot\widetilde{\Theta} + (\tilde{\boldsymbol{\Gamma}}\cdot\widetilde{\Theta})^{\mathrm{T}} + \Lambda, \tag{17}$$

with $\widetilde{\boldsymbol{m}}(0) = \widetilde{\mathrm{z}}^0$, $\widetilde{\Theta}(0) = \boldsymbol{0}$. Here $\tilde{\boldsymbol{\Gamma}} = P^{\mathrm{T}}\cdot\mathbf{A}\cdot P + R$, $\widetilde{\boldsymbol{k}} = P^{\mathrm{T}}\cdot\boldsymbol{k}$ with $\boldsymbol{k} = (\boldsymbol{0}, \boldsymbol{F}_{\mathrm{ext}})$ and $A_{11} = A_{21} = 0, A_{12} = 1, A_{22} = -1/\tau_p^{\mathrm{St}}$. Equations (15), (16), and (17) allow Eq. (11) to be written

$$\partial_t\widetilde{p}_r = -\partial_{\widetilde{\mathrm{z}}_1}\dot{\widetilde{m}}_1\widetilde{p}_r + \partial^2_{\widetilde{\mathrm{z}}_1}\tfrac{1}{2}\dot{\widetilde{\theta}}_{11}\widetilde{p}_r. \tag{18}$$

The net diffusional effect is therefore determined by the particle diffusion coefficient $\widetilde{D}_1(t)$ of the transformed variable $\tilde{z}_1$ (associated with the negative eigenvalue $\omega_1$) and given by

$$\widetilde{D}_1(t) = \tfrac{1}{2}\dot{\widetilde{\theta}}_{11} = (\tilde{\boldsymbol{\Gamma}}\cdot\widetilde{\Theta})_{11} - \tfrac{1}{2}|\omega_1|. \tag{19}$$

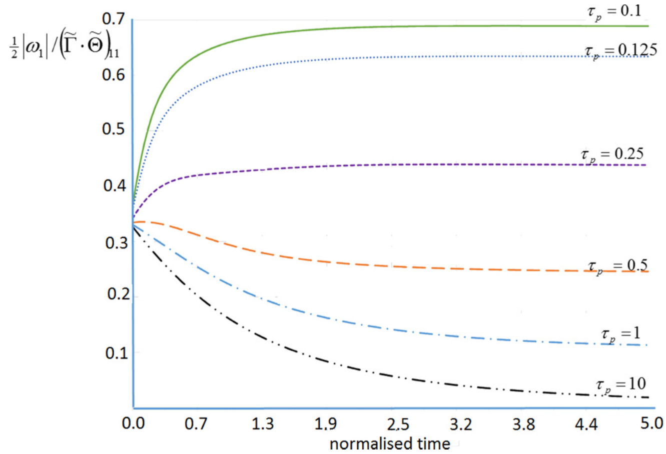


FIG. 2. Plots of $\frac{1}{2}|\omega_1|/(\tilde{\boldsymbol{\Gamma}}\cdot\widetilde{\Theta})_{11}$ Eq. (19) for the ratio of *negative/positive* contributions to the particle diffusion coefficient $\widetilde{D}_1$ of the transformed variable $\widetilde{Z}_{p1}$(with a *negative* eigenvalue) in the moving frame of reference, as a function of time $t$ for a range of values of the particle response time $\tau_p$. Both $t$ and $\tau_p$ are scaled on $T_L$, the Lagrangian integral timescale of the carrier flow measured along a particle trajectory.

This shows how the "antidiffusion" associated with $\omega_1$ is offset by the contribution emerging from the flux $\overline{\widehat{\mathrm{a}}}_1\widetilde{p}_r$ associated with the coupling between $\widetilde{Z}_{p1}$ and $\widetilde{Z}_{p2}$ through their covariance $\widetilde{\theta}_{12}$ in Eq. (19).

Figure 2 demonstrates that $0 \leqslant \frac{1}{2}|\omega_1|/(\tilde{\boldsymbol{\Gamma}}\cdot\widetilde{\Theta})_{11} \leqslant 1$. The plots, which show the time evolution of this ratio for a range of values for $\tau_p$ (with $\mathbf{F}_{\mathrm{ext}} = \mathbf{0}$), were obtained from closed form solutions of Eq. (17). These solutions are constructed by noting that $\widetilde{\boldsymbol{\Theta}} = P^{\mathrm{T}}\cdot\boldsymbol{\Theta}\cdot P$, where the covariances $\boldsymbol{\Theta} = \langle\mathbf{Z}_p\mathbf{Z}_p\rangle$ in the fixed frame are governed by a set of equations analogous to Eq. (17), which can be integrated analytically. We refer to Ref. [13], where analytic solutions are given for $\boldsymbol{\Theta}$ in terms of $\langle\mathrm{U}'_s(0)\mathrm{U}'_s(t)\rangle$ the autocorrelation of the carrier flow velocity fluctuations sampled along particle trajectories. The values of the *negative* to *positive* ratio plotted in Fig. 2 were obtained using an exponential decay $\exp[-t/T_L]$ for this autocorrelation. For completeness we also show in Fig. 3 for a similar range of values of $\tau_p$, the evolution of the particle diffusion coefficient $\widetilde{D}_1(t)$ in the moving frame of reference indicating not only that $\widetilde{D}_1 \geqslant 0$, but also that it reaches an asymptotic limit that is the same for all $\tau_p$. This is is also true of the particle diffusion coefficient $\varepsilon(\infty)$ in the fixed frame of reference, Eq. (14). In particular, in the normalized units used to express the values for $\widetilde{D}_1$ in Fig. 3, $\varepsilon(\infty) = 1$. This result is universally true for a particle equation of motion involving the linear drag form in Eq. (1) for statistically stationary homogeneous isotropic turbulence (see Ref. [20], where it is $T_L$ that depends on $\tau_p$). An evaluation of the asymptotic form of $\langle\mathbf{Z}_p\mathbf{Z}_p\rangle$, which is linear in $t$ in this limit, shows that

$$\begin{aligned}\widetilde{D}_1(\infty) &= 1/(4 - 2\sqrt{2}),\\ \widetilde{D}_2(\infty) &= 1/(4 + 2\sqrt{2}),\end{aligned} \tag{20}$$

and is consistent with the forms for $\widetilde{D}_1(t)$ in Fig. 3 obtained by solving a coupled set of Eq. (17) for $\widetilde{\boldsymbol{\Theta}}$. That the asymptotic result in Eq. (20) agrees with the results in Fig. 3 provides

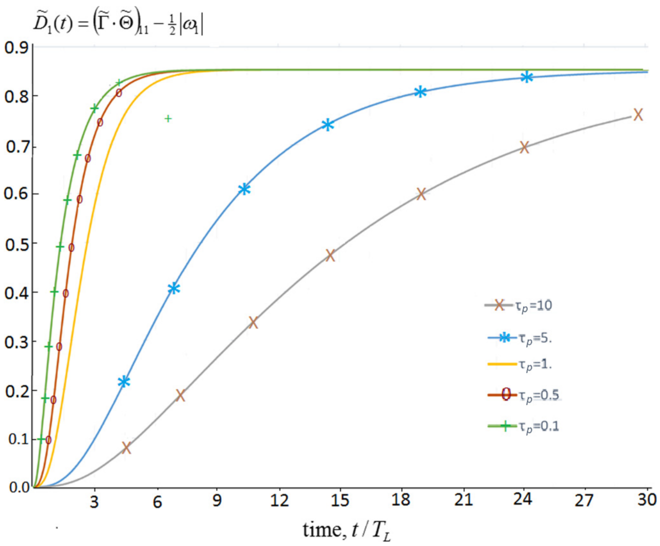


FIG. 3. Evolution of the particle diffusion coefficient $\widetilde{D}_1(t)$ evaluated using Eq. (19) in the moving frame of reference for a range of values of $\tau_p$ (the particle response time normalized by the Lagrangian integral time scale, $T_L$). Time is real time $t$ normalized on $T_L$.

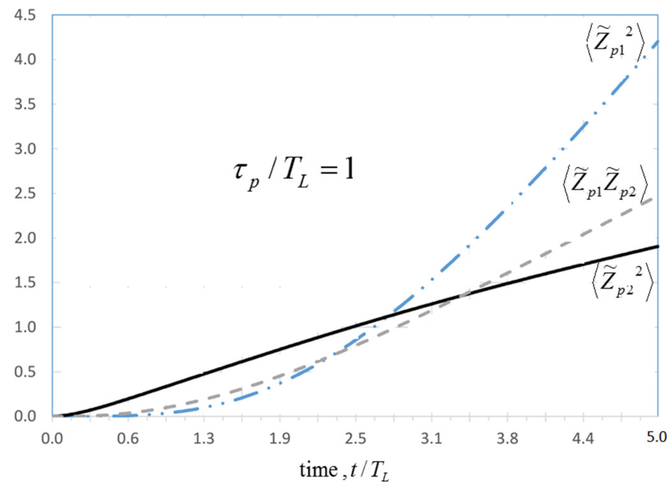


FIG. 4. Moments $\langle \widetilde{Z}_{pi}\widetilde{Z}_{pj}\rangle$ in the moving frame of reference based on the moments $\langle \boldsymbol{Z}_p\boldsymbol{Z}_p\rangle$ for $\tau_p/T_L$ in the fixed frame of reference as solutions of the fixed frame kinetic Eq. (3) or equivalently by evaluating $\langle \boldsymbol{Z}_p\boldsymbol{Z}_p\rangle$ from solutions of the particle equation of motion Eq. (1).

not only a check for the analytic solutions used in Fig. 3, but also a proof that the *positive* contribution to $\widetilde{D}_1(t)$ will always outweigh the *negative* contribution in Eq. (19) (i.e., it applies to all physically acceptable forms of the autocorrelation for $\boldsymbol{U}_s$, and not just the decaying exponential form of $\langle \mathrm{U}_s'(0)\mathrm{U}_s'(t)\rangle$ that we have chosen to obtain our analytical results).

This must be so for two reasons. First, the route involving a solution of the kinetic equation in the fixed frame of reference and the linear relationship between the fixed and transformed variables *always* ensure a realizable Gaussian distribution for the transformed variables. Second, via this route the realizability does not itself explicitly involve or rely in any way on whether one of the eigenvalues $\omega_i < 0$ and any explicit form for $\langle \mathrm{U}_s'(0)\mathrm{U}_s'(t)\rangle$ we might choose, only that the transformation matrix $\boldsymbol{P}$ formed from the normalized eigenvectors of the diffusion matrix exists and is well behaved. However, the second route via Eq. (17) only ensures a realizable Gaussian process if the *positive* contribution to $\widetilde{D}_1(t)$ exceeds the *negative* contribution. But since the two methods of calculating $\widetilde{\Theta}$ are in the end mathematically equivalent to one another, then the *positive* contribution to $\widetilde{D}_1(t)$ must always exceed the *negative* contribution in Eq. (19).

We show the values of the moments $\langle \widetilde{Z}_{pi}\widetilde{Z}_{pj}\rangle$ in Fig. 4 appropriate for the Gaussian function solution of the kinetic equation in the moving frame (see Eq. (87) in Ref. [13]). There is, of course, no hint of a singularity in Fig. 4, all three moments being smoothly varying, monotonically increasing in time, and linear in time for $t/T \gg 1$.

The results also illustrate the now obvious result that, at large times, the two contributions to the diffusional transport are of the same order in $t$. The claim in Ref. [9] that Eq. (11) reduces to the form of a backward heat equation because $\bar{\widetilde{\mathrm{a}}}_1\widetilde{p}_r \to 0$ as $t \to \infty$ is invalid. It fails to acknowledge that $\omega_1\partial_{\widetilde{z}_1}\widetilde{p}_r \to 0$ at the same rate.

Although we have now demonstrated that the transformed kinetic equation is not ill posed, we close this section with some comments on M&P's use of the Feynman-Kac formula (FKF) and the associated arguments in Ref. [9]. In Ref. [9], M&P suggest that Eq. (11) has the structure of a (generalized) backward Kolmogorov equation (BKE), that may be derived from FKF. Noting this, M&P use the FKF to construct the solution to Eq. (11), using the terminal condition $\widetilde{p}_r(\widetilde{\mathrm{z}}_1,T) = \Psi(\widetilde{\mathrm{z}}_1)$, to obtain ($t \in [0,T]$)

$$\widetilde{p}_r(\widetilde{\mathrm{z}}_1,t) = \left\langle \exp\left[\int_t^T \partial_{\widetilde{\mathrm{z}}_1}\bar{\widetilde{\mathrm{a}}}_1(\mathcal{X}(s),s)\,ds\right]\Psi(\mathcal{X}(T))\right\rangle_{\mathcal{X}(t)=\widetilde{\mathrm{z}}_1}, \tag{21}$$

where $\mathcal{X}(s)$ is a stochastic process defined through

$$d\mathcal{X}(s) \equiv \bar{\widetilde{\mathrm{a}}}_1(\mathcal{X}(s),s)ds + \sqrt{|\omega_1(s)|}dW(s), \tag{22}$$

and $W(s)$ is a Wiener process. M&P argue that the solution Eq. (21) implies that only "special" initial ($t = 0$) conditions are permitted when solving Eq. (11), since Eq. (21) specifies

$$\widetilde{p}_r(\widetilde{\mathrm{z}}_1,0) = \left\langle \exp\left[\int_0^T \partial_{\widetilde{\mathrm{z}}_1}\bar{\widetilde{\mathrm{a}}}_1(\mathcal{X}(s),s)\,ds\right]\Psi(\mathcal{X}(T))\right\rangle_{\mathcal{X}(0)=\widetilde{\mathrm{z}}_1}. \tag{23}$$

From this they conclude that since Eq. (21) only applies for the "special initial condition" given by Eq. (23), then Eq. (11) "is an unstable and ill-posed equation." This conclusion is clearly erroneous. Since the FKF employs a terminal condition in solving the PDE, then provided the PDE is well posed as a terminal-value problem, the solution of the PDE at $t = 0$ must of necessity be unique and "special." For a well posed, deterministic PDE, there exists only one solution at $t = 0$ that generates the specified terminal condition at $t = T$; otherwise, solutions to the PDE are not unique!

If Eq. (11) were truly a BKE, then it could indeed be considered ill posed since the BKE is in general ill posed when solved as a time-forward problem (and Eq. (11) is to be solved as a time-forward problem with a prescribed initial condition). However, the important point is that although Eq. (11) superficially appears to have the structure of a BKE, it cannot be considered to be equivalent to a BKE for two reasons. First, as we have already discussed, the term $\widehat{\widetilde{a}}_1$ is not a general convection term, but has a specific form since it is a functional of the solution of the Eq. (11). This is in part a manifestation of the fact that unlike the BKE, Eq. (11) is, in fact, derived from an underlying process that takes place in a higher dimensional space (i.e., the phase-space). Second, Eq. (11) is associated with a non-Markovian process, whereas the BKE corresponds to a Markov process. The implication of this is that Eq. (21) cannot, at least formally, cover the entire solution space of the PDE in Eq. (11), since Eq. (11) admits solutions that correspond to non-Markov trajectories in the space $\widetilde{z}_1$, which Eq. (21) does not account for since it constructs solutions via a conditional expectation over Markov trajectories. Therefore, in the general case, the FKF cannot be used to say anything categorical regarding the solutions to Eq. (11).

## III. KINETIC AND GLM EQUATIONS

It has been claimed in recent studies of PDF methods [9,19] that the kinetic PDF is the marginal of the GLM PDF. This claim is based on analysis that purports to show that the dispersion tensors appearing in a kinetic PDF equation derived from the GLM PDF equation are "strictly identical" to the corresponding tensors emerging directly from the kinetic modeling approach. If this is so the claim of ill-posedness of the kinetic equation contradicts the well-posedness associated with the Fokker-Planck equation of the GLM. Of course, as we have just demonstrated, this claim of ill-posedness is ill founded. Here we consider the validity of the analysis presented in Ref. [9] to demonstrate how the kinetic equation can be derived from the GLM PDF equation.

The analysis is based on the construction of a closure for $\langle \boldsymbol{u}_s \mathcal{P}\rangle$, where $\boldsymbol{u}_s(t;\boldsymbol{x}) = \boldsymbol{U}_s(t) - \langle \boldsymbol{U}_s(t)|(\boldsymbol{X}_p(t) = \boldsymbol{x})\rangle$ and $\mathcal{P}(\boldsymbol{x},\boldsymbol{v},t) = \delta(\boldsymbol{X}_p(t) - \boldsymbol{x})\delta(\boldsymbol{U}_p(t) - \boldsymbol{v})) = \delta(\boldsymbol{Z}_p(t) - \boldsymbol{z})$. We make the simple observation that the ensemble $\langle\cdot\rangle$ to be considered in this closure involves *all* realizations of the system being considered. It is not, nor can it be interpreted as, an average over only those realizations in which the trajectories $\boldsymbol{Z}_p$ satisfy the end-condition $\boldsymbol{Z}_p(t) = \boldsymbol{z}$. Indeed, this is why $\langle \boldsymbol{u}_s \mathcal{P}\rangle = \langle \boldsymbol{u}_s\rangle_{\boldsymbol{z}}\, p(\boldsymbol{z},t)$, where $\langle\cdot\rangle_{\boldsymbol{z}}$ denotes an average based on the subensemble containing only those trajectories satisfying this end-condition. Although self-evident, this point is missed in the closure formulated in Ref. [9]. This closure is constructed by introducing paths $\boldsymbol{\omega}(s) = \boldsymbol{\omega}(s;\boldsymbol{z},t)$ such that $(\boldsymbol{\omega}(t),\dot{\boldsymbol{\omega}}(t)) = \boldsymbol{z}$. These paths are used to partition particle trajectories; for a given path $\boldsymbol{\omega}(\cdot;\boldsymbol{z},t)$, define $\boldsymbol{\Omega}_{\boldsymbol{\omega}} = \{\boldsymbol{Z}_p : \boldsymbol{X}_p(s) = \boldsymbol{\omega}(s;\boldsymbol{z},t)\}$. In Ref. [9] a closure is then considered for the subensemble $\langle \boldsymbol{u}_s \mathcal{P}\rangle^{\boldsymbol{\Omega}_\omega}$ over those trajectories in $\boldsymbol{\Omega}_{\boldsymbol{\omega}}$ (see Eq. (39) in Ref. [9]), and this closure is then integrated over all paths $\boldsymbol{\omega}(\cdot;\boldsymbol{z},t)$. Thus, only trajectories satisfying the specified end-condition $\boldsymbol{Z}_p(t) = \boldsymbol{z}$ have been taken into account. This is wrong. Moreover, the form of the closure for $\langle \boldsymbol{u}_s \mathcal{P}\rangle^{\boldsymbol{\Omega}_\omega}$ questionable. The Furutsu-Novikov formula is invoked; correct application of this should result in a closure framed in terms of the two-time correlation tensor $\mathrm{C}(s,s';\boldsymbol{z},t) = \langle \boldsymbol{u}^{\boldsymbol{\omega}}(s)\boldsymbol{u}^{\boldsymbol{\omega}}(s')\rangle^{\boldsymbol{u}^{\boldsymbol{\omega}}}$ of the process $\boldsymbol{u}^{\boldsymbol{\omega}}(s) = \boldsymbol{u}_s(\boldsymbol{\omega}(s;\boldsymbol{z},t),s)$. However, in Ref. [9] this is conflated with another correlation, namely

$$R(s,\boldsymbol{x};s',\boldsymbol{x}') = \langle \boldsymbol{u}_s(s;\boldsymbol{x},t)\boldsymbol{u}_s(s';\boldsymbol{x}',t')\rangle. \tag{24}$$

Again, this is evidently wrong; C depends on a single phase-space point, $\boldsymbol{z}$, whereas $R$ is defined in terms of two points $\boldsymbol{x}$, $\boldsymbol{x}'$ in configuration space. Not only this, the ensembles over which these two correlation tensors are constructed are different. Finally (and notwithstanding these apparent oversights), even if the resulting forms of the dispersion tensors emerging from the construction given in Ref. [9] were correct, it is incorrect to claim that these tensors are identical to those appearing in the PDF equation of the kinetic model. In the kinetic PDF equation the dispersion tensors are defined in terms of the basic two-point, two-time correlation tensor of the underlying fluctuations in the carrier flow velocity field, that is $\mathcal{R}(\boldsymbol{x},t;\boldsymbol{x}',t') = \langle \boldsymbol{u}'(\boldsymbol{x},t)\boldsymbol{u}'(\boldsymbol{x}',t')\rangle$. This makes no reference to particle trajectories and, therefore, $\mathcal{R}$ cannot be deemed identical to $R$ defined by Eq. (24).

## IV. LIMITATIONS OF THE GLM FOR DISPERSED PARTICLE FLOWS

In the GLM PDF model, the phase-space of the system is extended to include $\boldsymbol{U}_s$, the fluid velocity along the inertial particle trajectory. In this case, the PDF considered is $p(\boldsymbol{z},t) = \langle\delta(\boldsymbol{z} - \boldsymbol{Z}_p(t))\rangle$, but now with $\boldsymbol{Z}_p(t) = (\boldsymbol{X}_p(t),\boldsymbol{U}_p(t),\boldsymbol{U}_s(t))$, and $\boldsymbol{Z}_p(t) \in \boldsymbol{z}$. For this GLM PDF equation, it is then necessary to specify the evolution equation for $\boldsymbol{U}_s(t)$, and by definition

$$\dot{\boldsymbol{U}}_s(t) \equiv \left(\frac{D\boldsymbol{u}_f}{Dt} - (\boldsymbol{u}_f - \boldsymbol{U}_p)\cdot\partial_{\boldsymbol{x}}\boldsymbol{u}_f\right)_{\boldsymbol{x}=\boldsymbol{X}_p(t)}, \tag{25}$$

with $D\boldsymbol{u}_f/Dt$ denoting the fluid acceleration field, and $(\cdot)_{\boldsymbol{x}=\boldsymbol{X}_p(t)}$ denoting that the field variables inside the parenthesis are evaluated at the particle position.

In the PDF equation for $\boldsymbol{Z}_p(t) = (\boldsymbol{X}_p(t),\boldsymbol{U}_p(t),\boldsymbol{U}_s(t))$, the term $\langle\dot{\boldsymbol{U}}_s(t)\rangle_{\boldsymbol{z}}$ appears and is unclosed. In the GLM approach, this term is closed by modeling $\dot{\boldsymbol{U}}_s(t)$ using a (generalized) Langevin equation. Thus, unlike the kinetic equation where assumptions about the *statistics* of the fluid velocities are made, in the GLM approach, assumptions about the *dynamical evolution* of $\boldsymbol{U}_s(t)$ are made. Needless to say, from a fundamental perspective, the use of a Langevin equation in place of Eq. (25) is in principle a strong assumption, since the behavior of $D\boldsymbol{u}_f/Dt$ as governed by the Navier-Stokes equation is vastly more complex than can be described by a simple Langevin equation.

Nevertheless, that the GLM is a model and not a fundamental theory of particle dispersion in turbulent flows is not an issue of critical concern. Like all models it has its advantages as well as its limitations. The kinetic, as well as the GLM PDF equation, invokes approximations in the description of the turbulent flow transporting the particles that are not rigorously justifiable. One important and obvious advantage of the GLM PDF approach is that it includes, in addition to the particle position and velocity variables, a variable for the flow velocity sampled along a particle trajectory. So a solution to the corresponding PDF equation in principle contains more information about the dispersion process than the solution of the kinetic equation. Most notably, Simonin and his coworkers have used this GLM PDF equation to formulate transport equations for the density weighted mean flow velocity $\overline{\boldsymbol{U}}_s$ and the particle-flow covariances and obtained remarkably good agreement with experimental measurement in numerous particle laden flows including jets and vertical channel flows [7,21]. Van Dijk and Swailes [22] solved the GLM PDF equation numerically in the case of particle transport and deposition in a turbulent boundary layer showing the existence of singularities in the near wall particle concentration. Reeks [12] solved this PDF equation for particle dispersion in a simple shear and obtained valuable insights into the influence of the shear on the fluid velocity correlations as well as the dispersion in the streamwise direction which showed a component of contragradient diffusion.

Our aim here is to point out the limitations of the GLM for dispersed gas-particle flows that have been ignored in previous analyses, especially in Ref. [9], to give a more balanced view of its strengths and weaknesses when compared to the kinetic approach. We regard these limitations to be areas for improvement of the model rather than inherent deficiencies. The advantage of models of this sort is that features inherent in more fundamental approaches like the kinetic approach can be included in an *ad hoc* manner.

First, we note that Eq. (25) shows that the process $\dot{\boldsymbol{U}}_s(t)$ is fundamentally connected to the properties of the underlying flow field and, as such, is influenced by the spatiotemporal structure of that field. This is particularly important since it is known, for example, that inertial particles interact with the topology of fluid velocity fields in particular ways, with a preference to accumulate in the strain dominated regions of the flow [8]. Equation (25) captures the way in which the process $\dot{\boldsymbol{U}}_s(t)$ is affected by the properties of the underlying flow. However, in the GLM, $\dot{\boldsymbol{U}}_s(t)$ is modeled using a Langevin equation, and, in consequence, the influence of the spatiotemporal structure of the underlying field on $\dot{\boldsymbol{U}}_s(t)$ is lost. This means then that the GLM cannot properly capture the role of flow structure on inertial particle dynamics in turbulent flows, which is known to be very important in determining the spatial distributions of the particles. In contrast, the kinetic model does capture the role of the spatiotemporal structure of the flow on particle motion. For example, the dispersion tensors $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$, and $\boldsymbol{\kappa}$ capture such effects through their dependence on the two-point, two-time correlation tensor of the fluid velocity field.

A second, related issue, concerns the handling of the term $(\boldsymbol{u}_f - \boldsymbol{U}_p)\cdot\partial_{\boldsymbol{x}}\boldsymbol{u}_f$ in the GLM. The role of this term in Eq. (25) is that it captures how the particle inertia causes the timescale of $\boldsymbol{U}_s(t)$ to deviate from the Lagrangian timescale of the fluid velocity. For example, in the limit $\tau_p \to 0$, one should recover $\dot{\boldsymbol{U}}_s = (D\boldsymbol{u}_f/Dt)_{\boldsymbol{x}=\boldsymbol{X}_p(t)}$, while in the limit $\tau_p \to \infty$ (without body forces), one should recover $\dot{\boldsymbol{U}}_s = (\partial_t \boldsymbol{u}_f)_{\boldsymbol{x}=\boldsymbol{X}_p(t)}$. In the former case, the timescale of $\boldsymbol{U}_s$ is the fluid Lagrangian timescale, whereas in the latter case the timescale of $\boldsymbol{U}_s$ is the fluid Eulerian timescale. With body forces, e.g., gravity, the timescale of $\boldsymbol{U}_s$ for inertial particles would also be affected by the crossing trajectories effect [23].

Conventionally, in the GLM the term $(\boldsymbol{u}_f - \boldsymbol{U}_p)\cdot\partial_{\boldsymbol{x}}\boldsymbol{u}_f$ is either neglected, so that the Langevin model relates to $\dot{\boldsymbol{U}}_s = (D\boldsymbol{u}_f/Dt)_{\boldsymbol{x}=\boldsymbol{X}_p(t)}$, or else its effect is modeled by making the timescale in the Langevin model a function of $\tau_p$. Both approaches are problematic: the first because it neglects the effect of inertia on the timescale, which can be strong, and the second because one then requires an additional model for the timescale of $\boldsymbol{U}_s$ as a function of $\tau_p$. In contrast, in the kinetic model, the role of inertia on $\boldsymbol{U}_s$ is formally accounted for and is an intrinsic part of the model. In particular, it is captured through the dependence of $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$, and $\boldsymbol{\kappa}$ on the correlation tensors of the fluid velocity field evaluated along the inertial particle trajectories.

These issues are related to the fact that the GLM for $\boldsymbol{U}_s$ is constructed in an *ad hoc* manner, in contrast to the case for single-phase turbulence where the GLM for the fluid particle velocity $\boldsymbol{U}_f$ is constructed with reference to the Navier-Stokes equation [3]. In Ref. [24] it was shown how exact (unclosed) transport equations for the statistical moments of $\boldsymbol{U}_s$ may be derived, and it was shown that the GLM model for $\boldsymbol{U}_s$ does not reproduce the closed terms in these transport equations. This shows that even at the one-point level, the GLM for $\boldsymbol{U}_s$ does not have the status of the corresponding GLM for $\boldsymbol{U}_f$ used in single-phase turbulence. However, the transport equations derived in Ref. [24] could be used in future work to improve the GLM for $\boldsymbol{U}_s$, placing it on a more firm foundation.

Another implication of the GLM's use of a Langevin equation to describe $\boldsymbol{U}_s(t)$ is that it cannot accurately describe the Lagrangian properties of the system in the short-time "ballistic" limit. For example, the second-order Lagrangian structure function $\langle\|\boldsymbol{U}_s(t+s) - \boldsymbol{U}_s(t)\|^2\rangle$ should grow as $s^2$ in the limit $s \to 0$, whereas a Langevin equation dictates that it grows as $s$ in the limit $s \to 0$. Interestingly, this very fact has an important bearing on the claim in Ref. [9] of the exact correspondence between the PDF of the kinetic equation and the marginal of the GLM PDF. Even aside from other issues, this claim cannot be correct since the kinetic model gives the correct short-time behavior for $\langle\|\boldsymbol{U}_s(t+s) - \boldsymbol{U}_s(t)\|^2\rangle$ since it allows for the general case where the fluid velocity field is differentiable in time.

In addition to these points, recent criticism of the kinetic equation has failed to appreciate or show any awareness of important consistency and invariance principles that were key guidelines in the construction of the kinetic equation, and highly relevant to the limitations and generality of GLM PDF equations. The first of these is that the kinetic equation should generate the correct equation of state, i.e., the relation between the equilibrium pressure associated with the correlated turbulent motion of the particles and their mass density in homogeneous isotropic statistically stationary turbulence. This can be obtained independently of the kinetic equation by evaluating the Virial for the particle equation of motion (see Sec. II in Ref. [13]). This relates the kinematic pressure $\mathcal{P}$ to the particle diffusion coefficient $\varepsilon$ via the particle response time $\tau_p$, namely $\mathcal{P} = \varepsilon\tau_p^{-1}$.

The second important consideration is that the kinetic equation should satisfy random Galilean transformation (RGT) invariance [15,25,26]. In the development of legitimate closure schemes, invariance to RGT is crucial to account for the transport of small scales of turbulence by the large scales and the $E(k) \sim k^{-5/3}$ spectrum. Specifically, RGT means applying to each realization of the carrier flow a translational velocity, constant in space and time but varying randomly in value from one realization to the next. In Kraichnan's traditional usage of RGT the distribution of velocities is taken to be Gaussian for convenience. Clearly, the internal dynamics should be unaffected by this transformation, and this invariance should be reflected in the equations that describe the average behavior of the resulting system. In the case of the kinetic equation the terms that describe the dispersion due to the aerodynamic driving force and that due to the translational velocity should be separate. When the timescale of $\boldsymbol{U}_s$ is finite (nonzero), RGT cannot be satisfied by a PDF equation with the traditional Fokker-Planck structure. Indeed, RGT invariance implies that the dispersion tensor $\boldsymbol{B}$ in Eq. (3) must have the form given in Eq. (5) [13], which is not compatible with a Fokker-Planck structure for the PDF equation (in which $\boldsymbol{\lambda} \equiv \mathbf{0}$). See Ref. [13] for the form of the dispersion tensor $\boldsymbol{B}$ satisfying RGT invariance for a non-Gaussian process as a cumulant expansion in particle fluid velocity correlations.

A failure to preserve RGT invariance means a failure to reproduce the correct equation of state for the dispersed phase. In the case of the GLM equations it is a failure associated with the short term dispersion prediction of $O(t)$ as opposed to $O(t^2)$. Such a result cannot arbitrarily be changed since the exponentially decaying autocorrelation is a property of the white noise based GLM equation for all time.

This has some bearing on the equivalence of the two approaches, since the kinetic approach does not have this limitation and correctly predicts the short term diffusion. So, whereas in the GLM the form of the particle-flow correlations are calculated and an intrinsic part of the model, in the kinetic equation these are prescribed or calculated using independent knowledge of the statistics of the carrier flow field and a relationship between Eulerian and Lagrangian correlations. As pointed out in Ref. [12] in the case of dispersion in a simple shear flow, if the statistics of the fluid velocity along a particle trajectory are assumed derivable from a Gaussian process and the fluid velocity correlations as a function of time are taken to be the same in either case, then the two approaches are identical, but only then. While in the kinetic equation one is free in principle to choose whatever is physically acceptable for the fluid particle correlation, the problem remains one of calculating carrier flow velocity correlations along particle trajectories, given the underlying Eulerian statistics of the carrier flow velocity field.

### The kinetic equation for nonlinear drag

In closing this section, we wish to address the numerous claims made that the kinetic approach is limited in its application to situations where the drag force is linear in the relative velocity between particle and fluid. This is not correct. We refer in particular to Sec. III in Ref. [15] on the particle motion that specifically deals with the treatment of nonlinear drag and how it is used to evaluate the convective and dispersive terms in the kinetic equation. In particular, the mean and fluctuating aerodynamic driving forces are expressed in terms of the particle mean density weighted particle velocity $\overline{\boldsymbol{v}}(\mathbf{x},t)$ and incorporated into the particle momentum equations by suitably integrating the kinetic equation over all particle velocities. We refer also to Ref. [27], where using the kinetic equation for nonlinear drag, an evaluation is made of the long-term diffusion coefficient for high inertial particles in homogeneous isotropic statistically stationary turbulence.

## V. SUMMARY AND CONCLUSIONS

This paper is about well-posedness and realizability of the kinetic equation and its relationship to the GLM equation for modeling the transport of small particles in turbulent gas flows. Previous analyses [9,19] claim that the kinetic equation is ill posed and therefore *invalid* as a PDF description of dispersed two-phase flows. Specifically, it is asserted that the kinetic equation, as given in Eq. (3), has the properties of a backward heat equation and as a consequence its solutions will in the course of time exhibit finite-time singularities. The justification for this claim is based on an analysis centered around the observation that the phase space diffusion tensor $\boldsymbol{B}$ in Eq. (3) is not positive-definite but possesses both negative and positive eigenvalues. We have examined the validity of assumptions that lead to this conclusion; in particular, the form of the kinetic equation in a moving frame where the PDF $\widetilde{p}(\widetilde{z}_1,\widetilde{z}_2,t)$ gives the distribution of transformed variables $\widetilde{z}_1,\widetilde{z}_2$ relative to the principal axes of $\boldsymbol{B}$ at time $t$ [see Eq. (9)]. Based on the negative diffusion coefficient in the transformed PDF equation, Eq. (11), for the marginal distribution $\widetilde{p}_r(\widetilde{z}_1,t)$, these previous studies have sought to show that this equation [and so also Eq. (3)] is ill posed. However, this analysis assumed that the term $\widehat{\widetilde{\mathrm{a}}}_1$ in Eq. (11) is wholly convective. In fact, it is a density-weighted variable and, because $\widetilde{z}_1$ and $\widetilde{z}_2$ are coupled in phase space, this means that $\widehat{\widetilde{\mathrm{a}}}_1$ has a gradient diffusive component with a *positive* diffusion coefficient which offsets the component in Eq. (11) with a *negative* diffusion coefficient. More particularly, we showed that the solution to the equation considered is Gaussian, with covariances that are the solutions of a set of coupled equations, Eqs. (16) and (17). Based on these solutions, the resultant convection-gradient diffusion equation for $\widetilde{p}_r(\widetilde{z}_1,t)$ is given by Eq. (18) with a diffusion coefficient $\widetilde{D}_1(t)$ given by the sum of the *positive* and *negative* contributions defined in Eq. (19). Using an exponential decaying autocorrelation of the fluid velocity measured along a particle trajectory, we obtained analytic solutions for the *positive* and *negative* components of $\widetilde{D}_1$, which show that the *positive* component *always* outweighs the *negative* component so that $\widetilde{D}_1$ is always *positive*. The corresponding values of $\widetilde{D}_1$ are shown in Fig. 3, which indicates that $\widetilde{D}_1(t)$ approaches an asymptotic value that is independent of the particle response time $\tau_p$, consistent with the derived asymptotic expressions given in Eq. (20). Significantly, we were able to show that this was a general result for all realizable forms for the flow velocity autocorrelation along particle trajectories. As a consequence the kinetic equation is not ill posed.

Finally, in the course of our examination of the analysis of ill-posedness, we pointed out a number of issues with the use of the Feynman-Kac formula (FKF). The application of the FKF to Eq. (11) is problematic because Eq. (11) is not really a backward Kolmogorov equation. Furthermore, the claim that the FKF solution to Eq. (11) implies that the kinetic equation is only solvable for special initial conditions is erroneous. The FKF employs a terminal condition, and therefore there can be only one possible "initial condition," or else solutions to Eq. (11) would not be unique.

Another important issue was the claim made in Ref. [9] that the kinetic equation can be derived from the GLM PDF equation, and that in fact the GLM is a more general approach than the kinetic approach. We showed that this is not the case, that assumptions introduced in averaging processes lead to inappropriate closure approximations that negate this claim.

In the final part of our analysis we sought to give a balanced appraisal of the benefits of both PDF approaches, and in particular to point out limitations of the GLM for gas-particle flows that have been ignored in most studies. We regarded these limitations to be areas for improvement of the GLM rather than inherent deficiencies. As we pointed out, the value of models of this sort is that features inherent in more fundamental approaches can be included in an *ad hoc* manner. We noted that terms fundamental to the modeling (the fluctuating convective strain rate contribution) contain valuable information on the relationships between Lagrangian and Eulerian timescales and the dependence on particle inertia. We suggested how additional features like particle clustering and drift in inhomogeneous turbulent flows, particularly in turbulent boundary layers, might be included in the model to make it more complete. This is one of the ways that the kinetic approach can support the PDF dynamic model by giving specific formulae for these additional features.

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