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Derivation of a pdf kinetic equation for the transport of particles in turbulent flows

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Abstract. A transport equation for the particle phase space density (probability density function (pdf) kinetic equation) is derived for the motion of a dilute suspension of particles in a turbulent flow. The underlying particle equation of motion is based upon a Langevin equation but with a non-white noise driving force derived from an Eulerian aerodynamic force field whose statistics are assumed known. Specifically both the particle position and velocity are considered to be functionals of the driving force and an application of a more general form of the Furutsu–Novikov theorem leads to closed expressions for the phase space diffusion current (i.e. the net force due to the turbulence acting on the particles per unit volume of phase space). In the case of a Gaussian random driving force the closed expressions reduce to a simple Boussinesq form in gradients of the pdf with respect to particle velocity and position. As a practical application solutions of the equation are compared with results obtained from particle tracking in a developing simple shear generated by large eddy simulation.

1. Introduction

Probability density function (pdf) equations have proved to be of value in understanding the behaviour of stochastic systems. Obvious examples of their usage occur in the study of Brownian motion [1] and in the kinetic theory of gases [2]. In more recent times they have been used extensively by Pope and others to model both turbulence [3] and turbulent related phenomena such as combustion [4] and atmospheric dispersion [5]. In all these cases the independent variables are the phase space variables of the system and the pdf equation describes the transport of the average phase space density in terms of those variables in phase space: the solution of the equation is the pdf that the system will be in any particular state as it evolves randomly in time from a given initial distribution. One of us in a series of papers [6–8] has used this approach to obtain the so-called continuum equations for a two-fluid model of dilute particle laden flows. In this particular case the pdf referred to is a function of both the particle velocity and position at any given time. As in kinetic theory, the continuum equations for the particle (dispersed) phase were obtained by integrating the pdf equation, multiplied by a suitable power of the velocity, over all particle velocities at a particular location in space. Henceforth, we will refer to the pdf equation describing the transport of particles in phase space as a pdf kinetic equation to emphasize the close link with kinetic theory.

This so-called pdf kinetic equation was derived by averaging the Liouville equation for the instantaneous particle phase space density over all realizations of the turbulent aerodynamic

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force field that acts upon the particle at a particular instant and location in phase space. Assuming that this force is separable into a steady resistive force (dependent upon the particle velocity) and a fluctuating driving force (independent of particle velocity), a crucial feature of the derivation is the closure approximation for the phase space 'diffusion' current $\boldsymbol{j}$ representing the contribution to the pdf equation from the aerodynamic driving force. This was based on an application of Kraichnan's Lagrangian history direct interaction (LHDI) approximation [9, 10] which gives [7]:

$$\boldsymbol{j} = -\left(\frac{\partial}{\partial \boldsymbol{v}} \cdot \boldsymbol{\mu} + \frac{\partial}{\partial \boldsymbol{x}} \cdot \boldsymbol{\lambda} + \boldsymbol{\gamma}\right) P(\boldsymbol{x}, \boldsymbol{v}, t) \tag{1}$$

where $P(\boldsymbol{x}, \boldsymbol{v}, t)$ is the probability density for a particle with position $\boldsymbol{x}$ and velocity $\boldsymbol{v}$ at time t and $\boldsymbol{\mu}$, $\boldsymbol{\lambda}$ and $\boldsymbol{\gamma}$ are dispersion tensors dependent upon the distribution in displacements in velocity and position about $(\boldsymbol{x}, \boldsymbol{v})$ in times of order of the timescale of the fluctuating aerodynamic driving force along a particle trajectory containing $(\boldsymbol{x}, \boldsymbol{v})$ at time t. In more precise terms, $\boldsymbol{j}/P(\boldsymbol{x}, \boldsymbol{v}, t)$ is the net aerodynamic driving force (due to the turbulence) per unit mass conditional upon a particle released into the flow field at some time $t' < t$ and being at $\boldsymbol{x}$, $\boldsymbol{v}$ at time t.

Further, $\boldsymbol{\gamma}$ is a body force dependent upon inhomogeneities in the turbulence of the carrier flow and $\boldsymbol{\mu}$ and $\boldsymbol{\lambda}$ are such that in the limit of particles with long response times compared with the fluid's $\boldsymbol{j} \to -(\partial/\partial \boldsymbol{v}) \cdot \boldsymbol{\mu} P$ (see [6] for details); that is the pdf equation takes on the same form as the classical Fokker–Planck equation of Brownian motion. In other words a Markov process (in which the fluctuating aerodynamic driving force is equivalent to white noise) is a special case of the random motion considered here. Indeed not only is the closure approximation in (1) appropriate for all particle response times, it is an exact closure when the fluctuating aerodynamic driving force is a Gaussian random process. This is a particularly important result since it ensures realizability of P under a certain set of non-trivial circumstances. The particular form of $\boldsymbol{j}$ (involving gradients in *both* velocity and position) was shown to be consistent with random Galilean transformation invariance (RGT) [6, 11]. Indeed using this principle Reeks [8] was able to construct the form for $\boldsymbol{j}$ appropriate for homogeneous flows. However, to tackle the problem of non-uniform flows the more general-purpose procedure of LHDI was used since it was known to satisfy RGT invariance [10].

Although LHDI is a powerful technique, it involves an elaborate procedure not easy to apply to even the simplest of systems. As an illustration, for this particular system it is first necessary to transform to a new phase for which the phase space density is constant along a particle trajectory. Particle trajectories in phase space are then used to define generalized vectors and response functions. Thus $\boldsymbol{f}(\boldsymbol{x}, \boldsymbol{v}, t|s)$ refers to the value of $\boldsymbol{f}$ measured at time s (the measuring time) along a particle trajectory containing $\boldsymbol{x}$, $\boldsymbol{v}$ at time t (the labelling time) and the response function $\hat{G}(\boldsymbol{x}, \boldsymbol{v}, t|s; \boldsymbol{x}', \boldsymbol{v}', t'|s')$ is the instantaneous phase space density, in a particular realization of the flow, measured at time s at some location along a particle trajectory containing $(\boldsymbol{x}, \boldsymbol{v})$ at time t arising from an instantaneous point source at time s' located by the second trajectory $(\boldsymbol{x}', \boldsymbol{v}', t'|s')$. The response function of interest is a particular case of this generalized response function when $t = s$ and $t' = s'$. An expression for the associated phase space diffusion current in (1) is recovered only after a closed expression for the equivalent term in the averaged Liouville equation for $\langle\hat{G}\rangle$ is obtained and transformed back to the original phase space. The reason for this circuitous route is that in the so-called primitive expansion for $\hat{G}(\boldsymbol{x}, \boldsymbol{v}, t|s; \boldsymbol{x}', \boldsymbol{v}', t'|s')$ intermediate labelling times can be replaced by t without changing the expansion. This together with the fact that $\langle\hat{G}\rangle$ is independent of the measuring time leads to a simplification of the closure approximation in this transformed phase space, in which memory and particle relaxation effects are contained in integrals over particle trajectories in phase space independent of $\langle \hat{G} \rangle$ and the closure approximation depends upon local gradients of $\langle \hat{G} \rangle$.

Most of this procedure, fortunately, can be avoided using a more direct approach which, in addition, permits the consideration of non-Gaussian driving forces and offers the potential of dealing with more complex flows involving, for example, thermal and mass coupling between the phases.

This method is a functional approach similar to that adopted in fluid turbulence [12]. In this approach, the particle position and velocity are considered functionals of the random aerodynamic driving force. By averaging the phase space density function over all realizable states of the fluctuating aerodynamic driving force (with their associated probabilities), one again arrives at the averaged form of Liouville's equation for the pdf but containing one unknown, the phase space diffusion current. An expression for this can be found in terms of the pdf by employing results from functional calculus. This expression involves the statistics of the random aerodynamic driving force (which are assumed known) and a new unknown which can be considered as the transition probability. Rather than explicitly specifying this transition probability, but instead using an approximation based on the particle equations of motion, the resultant expression is found to be identical to that derived from the LHDI procedure. However, this method is less complicated and, arguably, more mathematically rigorous than the LHDI approach.

The analysis presented here makes direct use of the Furutsu–Novikov–Donsker theorem [13–15]. However, the same results can be obtained using an analysis explicitly involving the characteristic functional of the random flow field in time and its functional derivatives as is the approach used in [6] for the dispersion of particles in a homogeneous turbulent flow field. We note that this latter approach has similarities to that used in the propagation of waves in random media [16] and to that presented in [17].

The format of this paper is as follows. In section 2, the phase space density function is defined and Liouville's equation for the pdf is derived from the Langevin equation of motion. As mentioned above, this contains an unknown term: the phase space diffusion current. In order to derive a closed form for this term, the statistics of the random aerodynamic driving force must be specified. In section 3, the statistics are assumed to be Gaussian in nature and an expression for the phase space diffusion current for inhomogeneous flows is obtained. This method is then extended to consider the situation of initial correlation effects in section 4. As a practical application of the above theory, in section 5 results obtained from the pdf kinetic equation are compared with those previously obtained for dispersion in a developing simple shear using large eddy simulation (LES).

2. The Liouville equation

This study is only concerned with the mechanical effect of the carrier flow on the particulate phase: there is no thermal coupling or mass exchange. For convenience, it is supposed that the dispersed phase is mono-disperse and that it is also dilute so that particle–particle collisions can be ignored. It is further assumed that the suspended particles are sufficiently large so that Brownian motion may be ignored: thus the particle motion is driven exclusively by aerodynamic forces which are assumed to depend upon the relative velocity between the particle and local carrier flow.

The Langevin equation we consider is

$$\frac{\mathrm{d}x_i^p}{\mathrm{d}t} = v_i^p \qquad (2)$$

$$\frac{\mathrm{d}v_i^p}{\mathrm{d}t} = -\beta_{ij}v_j^p + F_i + f_i \tag{3}$$

where $\boldsymbol{x}^p(t)$ and $\boldsymbol{v}^p(t)$ are the position and velocity of the particle respectively, $\boldsymbol{\beta}(\boldsymbol{x}^p(t), t)$ is a 'dissipative' tensor, the components of whose inverse, β_{ij}^{-1}, are the particle response times to changes in the velocity in the i direction due to changes in the flow velocity in the j direction. Further, $\boldsymbol{F}(\boldsymbol{x}^p(t), t)$ represents the external forces, or the mean aerodynamic driving force, or both, which act on the particle, with $\boldsymbol{f}(\boldsymbol{x}^p(t), t)$ being the fluctuating part of the aerodynamic driving force. At the moment nothing will be assumed about the statistics of $\boldsymbol{f}$ except that it has zero mean.

In general, the mean aerodynamic driving force is a function of the relative velocity between the particle and the carrier flow, in which case $\boldsymbol{\beta}$ is a function of the particle mean velocity, and hence particle position, and is an average over the local carrier flow velocity. However, at low particle Reynolds numbers, $\boldsymbol{\beta}$ is a tensor whose components are constants of the motion. An example of this situation is Stokes drag acting in a dilute suspension of particles: in this case $\boldsymbol{F}$ and $\boldsymbol{f}$ can be written respectively as

$$F = \boldsymbol{\beta}\cdot\bar{\boldsymbol{u}} + \boldsymbol{F}^e \qquad \boldsymbol{f} = \boldsymbol{\beta}\cdot\boldsymbol{u}'$$

where $\bar{\boldsymbol{u}}(\boldsymbol{x}, t)$ and $\boldsymbol{u}'(\boldsymbol{x}, t)$ are the mean and fluctuating components of the carrier flow and $\boldsymbol{F}^e(\boldsymbol{x}, t)$ represents any external forces present such as gravity. The exact forms of $\boldsymbol{\beta}$ and the mean aerodynamic driving force are to be found in Reeks [7] and though the exact form of these need not concern us here, it should be noted that neither depend explicitly on $\boldsymbol{f}$.

Following Pope [4] the 'fine grained' phase space density function, $W(\boldsymbol{x}, \boldsymbol{v}, t)$, that the position field, $\boldsymbol{x}^p(t)$, and the velocity field, $\boldsymbol{v}^p(t)$, of the particle take the particular set of values $\boldsymbol{x}$ and $\boldsymbol{v}$ at time t respectively, in any one realization of the flow (i.e. for a given $\boldsymbol{f}$), is defined by

$$W(\boldsymbol{x}, \boldsymbol{v}, t) = \delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - \boldsymbol{v}). \tag{4}$$

The pdf itself is then defined by

$$P(\boldsymbol{x}, \boldsymbol{v}, t) = \langle W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = \langle \delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - \boldsymbol{v})\rangle$$

where averaging is over all realizable states of the random aerodynamic driving force with their appropriate probabilities (see, for example, [18]); a more rigorous treatment of averaging in terms of functionals is given by McComb [19]. Differentiating both sides of (4) with respect to t and using the chain rule gives

$$\begin{aligned}
\frac{\partial W}{\partial t} = \frac{\partial x_i^p}{\partial t}\frac{\partial}{\partial x_i^p}[\delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - \boldsymbol{v})] + \frac{\partial v_i^p}{\partial t}\frac{\partial}{\partial v_i^p}[\delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - v)] \\
= -\frac{\partial x_i^p}{\partial t}\frac{\partial}{\partial x_i}[\delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - \boldsymbol{v})] \\
-\frac{\partial v_i^p}{\partial t}\frac{\partial}{\partial v_i}[\delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - \boldsymbol{v})] \\
= -\frac{\partial}{\partial x_i}\left[\frac{\partial x_i^p}{\partial t}\delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - \boldsymbol{v})\right] \\
-\frac{\partial}{\partial v_i}\left[\frac{\partial v_i^p}{\partial t}\delta(\boldsymbol{x}^p(t) - \boldsymbol{x})\delta(\boldsymbol{v}^p(t) - \boldsymbol{v})\right]
\end{aligned} \tag{5}$$

where in deriving the second line use has been made of the identity

$$\frac{\partial}{\partial x}f(x - y) = -\frac{\partial}{\partial y}f(x - y). \tag{6}$$

Also, (5) follows from the previous line by noting that $\partial x_i^p/\partial t$ is not a function of $\boldsymbol{x}$ and $\partial v_i^p/\partial t$ is not a function of $\boldsymbol{v}$ as can be seen from (2) and (3). Substituting (2) and (3) into (5) gives

$$\begin{aligned}\frac{\partial W}{\partial t} = -\frac{\partial}{\partial x_i}[v_i^p\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\delta(\boldsymbol{v}^p(t)-\boldsymbol{v})] - \frac{\partial}{\partial v_i}[\{-\beta_{ij}(\boldsymbol{x}^p(t),t)v_j^p \\ +F_i(\boldsymbol{x}^p(t),t) + f_i(\boldsymbol{x}^p(t),t)\}\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\delta(\boldsymbol{v}^p(t)-\boldsymbol{v})].\end{aligned}$$

This equation is now averaged over all realizations, to yield

$$\begin{aligned}\frac{\partial P}{\partial t} &= -\frac{\partial}{\partial x_i}\langle v_i^p\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\delta(\boldsymbol{v}^p(t)-\boldsymbol{v})\rangle - \frac{\partial}{\partial v_i}\langle\{-\beta_{ij}(\boldsymbol{x}^p(t),t)v_j^p + F_i(\boldsymbol{x}^p(t),t) \\ &\quad + f_i(\boldsymbol{x}^p(t),t)\}\delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\delta(\boldsymbol{v}^p(t)-\boldsymbol{v})\rangle \\ &= -\frac{\partial}{\partial x_i}(v_iP) + \frac{\partial}{\partial v_i}(\beta_{ij}(\boldsymbol{x},t)v_jP) \\ &\quad -\frac{\partial}{\partial v_i}(F_i(\boldsymbol{x},t)P) - \frac{\partial}{\partial v_i}\langle f_i(\boldsymbol{x},t)W(\boldsymbol{x},\boldsymbol{v},t)\rangle. \end{aligned} \quad (7)$$

This is a partial differential equation for $P(\boldsymbol{x},\boldsymbol{v},t)$ and is a form of Liouville's equation. However, it contains one unknown term, $\langle f_i(\boldsymbol{x},t)W(\boldsymbol{x},\boldsymbol{v},t)\rangle$, the phase space diffusion current. Thus, to 'close' the equation for P, an expression for this unknown term has to be found.

3. Closure for Gaussian random force fields

If the fluctuating aerodynamic driving force, $\boldsymbol{f}$ is Gaussian then a result from functional calculus can be used. This result is called the *Furutsu–Novikov–Donsker* formula and seems to have been derived independently by Furutsu [13], Novikov [14] and Donsker [15]. It is given below in the form presented in the last paper. In what follows, use is made of functionals and functional derivatives and a brief explanation of these are given in appendix A.

Furutsu–Novikov–Donsker formula. Let $f_i(s)$ be arbitrary Gaussian random functions with zero mean and with a correlation tensor

$$\langle f_i(s)f_k(s')\rangle = F_{ik}(s,s') \quad (8)$$

where s is the aggregate of arguments on which the random function depends. Then if $R[\boldsymbol{f}]$ is any functional of $\boldsymbol{f}$,

$$\langle f_i(s)R[\boldsymbol{f}]\rangle = \int F_{ik}(s,s')\left\langle\frac{\delta R[\boldsymbol{f}]}{\delta f_k(s')\,\mathrm{d}s'}\right\rangle \mathrm{d}s'$$

where the integral extends over the region in which the functions are defined.

This result is well known and so a derivation will not be presented (see, for example, [13, 15, 20]). In particular, (8) gives the result, that for $\boldsymbol{f}(\boldsymbol{x},t)$, a random Gaussian field of zero mean,

$$\langle f_i(\boldsymbol{x},t)R[\boldsymbol{f}]\rangle = \iint \langle f_i(\boldsymbol{x},t)f_j(\boldsymbol{x}',t')\rangle\left\langle\frac{\delta R[\boldsymbol{f}(\boldsymbol{x},t)]}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'}\right\rangle \mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'. \quad (9)$$

From (7), it can be seen that the functional R we wish to consider is

$$R[\boldsymbol{f}] = W(\boldsymbol{x},\boldsymbol{v},t) = \delta(\boldsymbol{x}^p(t)-\boldsymbol{x})\delta(\boldsymbol{v}^p(t)-\boldsymbol{v}) \quad (10)$$

with both $\boldsymbol{x}^p$ and $\boldsymbol{v}^p$ being themselves functionals of $\boldsymbol{f}$ as can be seen from the equation of motion. Thus in order to close the equation for P, the functional derivative of W with respect to $\boldsymbol{f}$ has to be found. This is

$$\begin{aligned}\frac{\delta W}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} &= \frac{\delta}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'}[\delta(\boldsymbol{x}^p-\boldsymbol{x})\delta(\boldsymbol{v}^p-\boldsymbol{v})]\\ &= \frac{\delta x_k^p(t)}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'}\frac{\partial}{\partial x_k^p}[\delta(\boldsymbol{x}^p-\boldsymbol{x})\delta(\boldsymbol{v}^p-\boldsymbol{v})]\\ &\quad+\frac{\delta v_k^p(t)}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'}\frac{\partial}{\partial v_k^p}[\delta(\boldsymbol{x}^p-\boldsymbol{x})\delta(\boldsymbol{v}^p-\boldsymbol{v})]\\ &= -\left[\frac{\partial}{\partial x_k}\frac{\delta x_k^p(t)}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'}+\frac{\partial}{\partial v_k}\frac{\delta v_k^p(t)}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}x'\,\mathrm{d}t'}\right]W(\boldsymbol{x},\boldsymbol{v},t) \end{aligned}\tag{11}$$

where in deriving the last line use has been made of (6) and also that the functional derivatives of $\boldsymbol{x}^p$ and $\boldsymbol{v}^p$ with respect to $\boldsymbol{f}(\boldsymbol{x}',t')$ are independent of $\boldsymbol{x}$ and $\boldsymbol{v}$, respectively. In order to proceed, these functional derivatives now have to be evaluated. This is carried out in appendix B, with the resultant expressions given by

$$\frac{\delta x_i^p(t)}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} = G_{ji}(\boldsymbol{x}',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')$$

$$\frac{\delta v_i^p(t)}{\delta f_j(\boldsymbol{x}',t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} = \frac{\mathrm{d}}{\mathrm{d}t}G_{ji}(\boldsymbol{x}',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')$$

where the generalized response functions, $\boldsymbol{G}$, are defined by

$$G_{ji}(\boldsymbol{x}^p(t'),t';\boldsymbol{x}^p(t),t) = \frac{\delta x_i^p(t)}{\delta f_j(\boldsymbol{x}^p(t'),t')\,\mathrm{d}t'}\tag{12}$$

and which satisfy

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}G_{ji}+\beta_{in}\frac{\mathrm{d}}{\mathrm{d}t}G_{jn}+G_{jk}\frac{\partial\beta_{in}}{\partial x_k^p}\frac{\mathrm{d}x_n^p}{\mathrm{d}t}-G_{jk}\frac{\partial F_i}{\partial x_k^p} = \delta_{ji}\delta(t-t').\tag{13}$$

The closed expression for the phase space diffusion current is found to be (see appendix B for details)

$$\langle f_i(\boldsymbol{x},t)W(\boldsymbol{x},\boldsymbol{v},t)\rangle = -\left[\frac{\partial}{\partial x_j}\lambda_{ji}+\frac{\partial}{\partial v_j}\mu_{ji}+\gamma_i\right]P(\boldsymbol{x},\boldsymbol{v},t)\tag{14}$$

where

$$\lambda_{ji} = \int_0^t \mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_k(\boldsymbol{x},\boldsymbol{v},t|t')\rangle G_{kj}(t'|t)\tag{15}$$

$$\mu_{ji} = \int_0^t \mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_k(\boldsymbol{x},\boldsymbol{v},t|t')\rangle \frac{\mathrm{d}}{\mathrm{d}t}G_{kj}(t'|t)\tag{16}$$

$$\gamma_i = -\int_0^t \mathrm{d}t'\left\langle\frac{\partial f_i(\boldsymbol{x},t)}{\partial x_j}f_k(\boldsymbol{x},\boldsymbol{v},t|t')\right\rangle G_{kj}(t'|t).\tag{17}$$

In (15)–(17), $\boldsymbol{f}(\boldsymbol{x},\boldsymbol{v},t|t')$ represents the value of $\boldsymbol{f}$ measured at time t' along a particle trajectory that passes through $\boldsymbol{x}$, $\boldsymbol{v}$ at time t. Also, $\boldsymbol{G}(t'|t)$ is shorthand for $\boldsymbol{G}(\boldsymbol{x}',t';\boldsymbol{x},t)$ which satisfies (13) but with $\boldsymbol{x}^p$ replaced by $\boldsymbol{x}$. The final form of the pdf kinetic equation for inhomogeneous flow is thus

$$\frac{\partial P}{\partial t}+v_i\frac{\partial P}{\partial x_i}-\frac{\partial}{\partial v_i}(\beta_{ij}v_jP)+\frac{\partial}{\partial v_i}(F_iP) = \frac{\partial}{\partial v_i}\left[\frac{\partial}{\partial v_j}(\mu_{ji}P)+\frac{\partial}{\partial x_j}(\lambda_{ji}P)+\gamma_iP\right].$$

This is the same as found by Reeks [7] using the LHDI approximation. Further, if $\boldsymbol{f}$ is a white noise process, the above reduces to the classical Fokker–Planck equation of Brownian motion (see the discussion at the end of appendix B.1).

4. Initial conditions

Throughout the preceding sections it has been tacitly assumed that there is no initial correlation between the phase space density W and the random aerodynamic driving force, $\boldsymbol{f}$, i.e.

$$\langle f_i(\boldsymbol{x}_0, 0) W(\boldsymbol{x}_0, \boldsymbol{v}_0, 0)\rangle = 0$$

where $\boldsymbol{x}_0$ and $\boldsymbol{v}_0$ are the particle's initial position and velocity at $t = 0$. In this section we show how to take into account any initial correlation.

The starting point is (2) and (3), or

$$\frac{\mathrm{d}^2 x_i^p}{\mathrm{d}t^2} + \beta_{ij}(\boldsymbol{x}^p, t)\frac{\mathrm{d}x_j^p}{\mathrm{d}t} - F_i(\boldsymbol{x}^p, t) = f_i(\boldsymbol{x}^p, t) \tag{18}$$

but with the initial conditions

$$x_i^p(0) = b_i[\boldsymbol{f}(\boldsymbol{x}^p(0), 0)] \qquad \dot{x}_i^p(0) = a_i[\boldsymbol{f}(\boldsymbol{x}^p(0), 0)]$$

where $\boldsymbol{b}$ and $\boldsymbol{a}$ are arbitrary functionals of the initial random aerodynamic driving force $\boldsymbol{f}(\boldsymbol{x}^p(0), 0)$. To solve the above, we use the notion of 'extending' a differential operator (see [21] for an excellent discussion). That is, if we define the operator L to be the right-hand side of (18), then the extended definition of L is

$$L[\boldsymbol{x}^p] = f_i(\boldsymbol{x}^p, t) + a_i\delta(t) + b_i\delta'(t)$$

where $\delta(t)$ is the Dirac delta function and $\delta'(t)$ is its derivative. Thus the generalized response functions, $G_{ji}(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t)$ as defined by (12), satisfy,

$$M[G_{ji}(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t)] = \delta_{ij}\delta(t - t') + A_{ji}\delta(t) + B_{ji}\delta'(t) \tag{19}$$

where

$$M[G_{ji}] = \frac{\mathrm{d}^2}{\mathrm{d}t^2}G_{ji} + \beta_{ik}\frac{\mathrm{d}}{\mathrm{d}t}G_{jk} + \frac{\mathrm{d}x_k^p}{\mathrm{d}t}\frac{\partial\beta_{ik}}{\partial x_m^p}G_{jm} - \frac{\partial F_i}{\partial x_k^p}G_{jk}$$

and

$$A_{ji} = \frac{\delta a_i[\boldsymbol{f}(\boldsymbol{x}^p(0), 0)]}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'} \qquad B_{ji} = \frac{\delta b_i[\boldsymbol{f}(\boldsymbol{x}^p(0), 0)]}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'}. \tag{20}$$

To proceed, G_{ji} is split into two components $G_{ji} = G_{ji}^1 + G_{ji}^2$ such that

$$M[G_{fi}^1(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t)] = \delta_{ij}\delta(t - t') \tag{21}$$

$$M[G_{ji}^2(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t)] = A_{ji}\delta(t) + B_{ji}\delta'(t). \tag{22}$$

From (21) it can be seen that

$$M[G_{ji}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t)] = \delta_{ij}\delta(t)$$

and

$$M\left[\frac{\partial G_{ji}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t)}{\partial t'}\right] = -\delta_{ij}\delta'(t).$$

Hence

$$M\left[A_{jm}G_{mi}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t) - B_{jm}\frac{\partial G_{mi}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t)}{\partial t'}\right] = A_{ji}\delta(t) + B_{ji}\delta'(t)$$

and comparing this equation with (22) it can be seen that

$$G_{ji}^2 = A_{jm}G_{mi}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t) - B_{jm}\frac{\partial G_{mi}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t)}{\partial t'}. \tag{23}$$

Once (21) has been solved for G_{ji}^1, we use (23) to obtain G_{ji}^2, and hence the solution G_{ji} of (19). The phase space diffusion current is then found by substituting this G_{ji} for that used in appendix B, to arrive at similar equations to (14)–(17).

5. Comparison with LES data

Recent LES results have been obtained for particle dispersal in a homogeneous developing shear flow [22]. In this section results obtained from the pdf kinetic equation will be compared with the LES data for three particle sizes. The results obtained from the LES were for the particle Reynolds stresses, $\overline{v_i'v_j'}$, where $\boldsymbol{v}' = \boldsymbol{v} - \bar{\boldsymbol{v}}$ is the fluctuating particle velocity and $\bar{\boldsymbol{v}}$ is the mean particle velocity. These are obtained from the pdf kinetic equation, by deriving, and then solving, a transport equation for the particle Reynolds stresses, though other techniques can also be used [23]. This transport equation is obtained by rewriting the pdf kinetic equation in terms of $\boldsymbol{v}'$, then multiplying the resultant equation by $\frac{1}{2}mv_i'v_j'$ and then integrating it over all velocities [7, 24]. For the case considered here, the transport equation is found to be†

$$\frac{\mathrm{d}}{\mathrm{d}t}\overline{v_i'v_j'} = -2\beta\overline{v_i'v_j'} - \frac{\partial\bar{v}_i}{\partial x_m}(\overline{v_j'v_m'} + \lambda_{mj}) - \frac{\partial\bar{v}_j}{\partial x_m}(\overline{v_i'v_m'} + \lambda_{mi}) + \mu_{ij} + \mu_{ji} \tag{24}$$

where we have set $\beta_{ij} = \beta\delta_{ij}$ with β^{-1} being the particle response time.

The LES data involves fluid statistics at various times: these include the fluid Reynolds stresses, $\overline{\boldsymbol{u}'\boldsymbol{u}'}$, the turbulent kinetic energy, k, and turbulent dissipation rate, ϵ; they are used to obtain expressions for the quantities $\langle \boldsymbol{ff}\rangle$ and τ which appear in the dispersion tensors $\boldsymbol{\lambda}$ and $\boldsymbol{\mu}$ given in appendix C. By assuming that only Stokes drag is acting on the particle,

$$\langle \boldsymbol{ff}\rangle = \beta^2\overline{\boldsymbol{u}'\boldsymbol{u}'}. \tag{25}$$

An expression is also required for τ and this is chosen to be

$$\tau = 0.482\frac{k}{\epsilon}. \tag{26}$$

The constant appearing in this equation was derived from a Langevin equation and was the agreed form by all those who participated in this 'test case' comparison [22].

The LES for a simple shear was carried out by first tracking particles through an isotropic flow field (again generated by LES) until they had reached equilibrium with the flow. Only then was the shear applied; this gives rise to some initial correlation between the particle and fluid velocities which is highlighted in the final row of table 1. Further, it should be noted that a constant and uniform mean shear rate was applied to both the fluid and the particle phase ($\alpha = 50$ s^{-1}); this was to avoid the added complication of the crossing trajectory effect. Thus the mean particle velocity and the mean fluid velocity, $\bar{\boldsymbol{u}}$ can be expressed as $\bar{v}_i = \alpha\delta_{i1}x_2 = \bar{u}_i$, and (24) reduces to

$$\frac{\mathrm{d}}{\mathrm{d}t}\overline{v_i'v_j'} = -2\beta\overline{v_i'v_j'} - \alpha\delta_{i1}(\overline{v_j'v_2'} + \lambda_{2j}) - \alpha\delta_{j1}(\overline{v_i'v_2'} + \lambda_{2i}) + \mu_{ij} + \mu_{ji}. \tag{27}$$

These are three coupled ordinary differential equations and are solved numerically with the form for $\boldsymbol{\lambda}$ and $\boldsymbol{\mu}$ given in appendix C. However, since both $\boldsymbol{\lambda}$ and $\boldsymbol{\mu}$ depend on $\overline{\boldsymbol{u}'\boldsymbol{u}'}$ and τ, which are developing and thus changing with time, the differential equations, (27), were solved in a piecewise manner. This is best explained as follows:

(1) At the time t_1, at which the fluid statistics are given, read in $\overline{\boldsymbol{u}'\boldsymbol{u}'}$, k and ϵ.
(2) Evaluate $\langle \boldsymbol{ff}\rangle$ and τ using (25) and (26) respectively. Use these expressions in the equations for $\boldsymbol{\lambda}$ and $\boldsymbol{\mu}$.
(3) Integrate the differential equations (27) from t_1 to the next time at which data is given, t_2, keeping $\langle \boldsymbol{ff}\rangle$ and τ fixed to the values calculated at t_1.

This procedure is now repeated until the final data point is reached.

† Due to the homogeneity of the problem considered, all spatial gradients other than the mean shearing are zero.

Table 1. Particle initial conditions for simple shear flow.

Particle diameter (μm)	5	30	60
Mean particle relaxation time (s)	1.95×10^{-4}	6.5×10^{-3}	2.4×10^{-2}
Particle Reynolds stresses, $\overline{v_i' v_j'}$ (m^2 s^{-2})	$0.078\delta_{ij}$	$0.064\delta_{ij}$	$0.040\delta_{ij}$
Fluid–particle velocity correlation, $\overline{u_i' v_j'}$ (m^2 s^{-2})	$0.078\delta_{ij}$	$0.064\delta_{ij}$	$0.040\delta_{ij}$

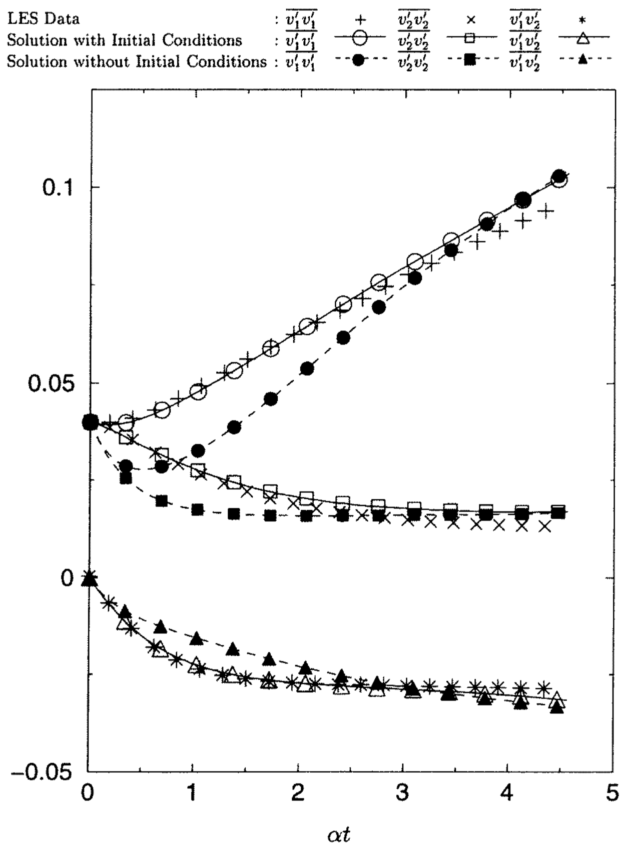

Figure 1. Particle Reynolds stresses for 60 μm particle.

The particle Reynolds stresses obtained from the pdf kinetic equation (both with and without initial correlation effects) are compared with those from the LES results in figures 1–3. For all cases, taking into account the initial effects gives very good agreement between the LES data and that obtained from the pdf kinetic equation. However, if these initial effects are neglected, only the 5 μm and 30 μm particles give good agreement; for the 60 μm particles, the $\overline{v_1'v_1'}$ component of the Reynolds stresses is underpredicted initially, though the agreement gets better for longer times. This is due to how quickly the particles respond to the fluid motion. Thus, initial correlation effects appear to be important for transient calculations or for particles with large response times but not so crucial for long time behaviour or for particles with small response times.

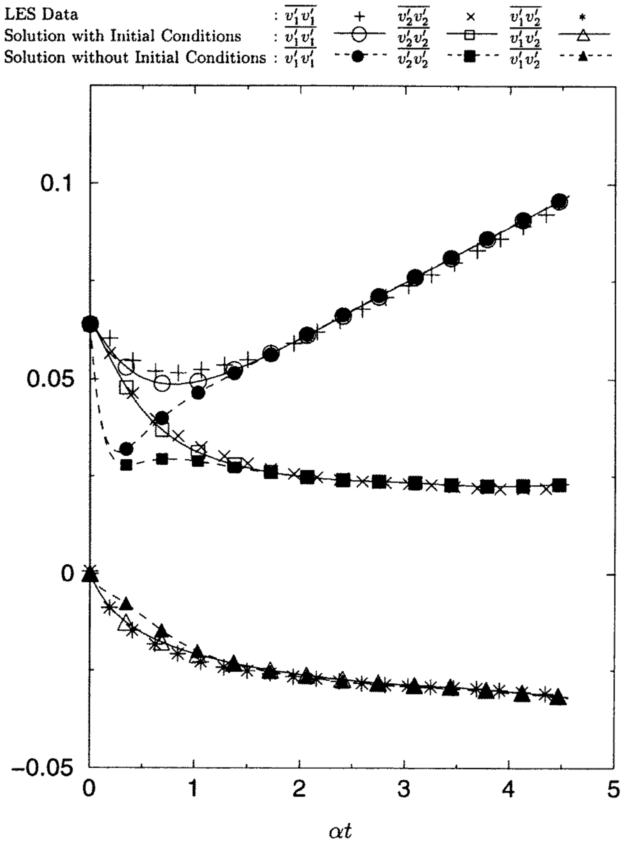

Figure 2. Particle Reynolds stresses for 30 μm particle.

6. Conclusions

In the context of particle laden flows, a new method has been developed to derive the pdf kinetic equation for the transport of particles in a turbulent flow. This is based on a functional formalism; specifically, both the particle position and velocity are considered to be functionals of the fluctuating component of the aerodynamic driving force. From the particle equations of motion, Liouville's equation for the particle phase space density can be derived from first principles. On averaging over all realizable states of the fluctuating aerodynamic driving force, this leads to a closure problem for the phase space diffusion current. However, results from functional calculus can be immediately applied, and for the case of a Gaussian random aerodynamic driving force, the closure model presented here is identical to that obtained from the LHDI approximation used elsewhere. It was also shown that the classical Fokker–Planck equation was a special case of the pdf kinetic equation under the additional assumption that the random aerodynamic driving force was a white noise process. The functional formalism was also extended to consider initial correlation effects. As a practical example of the theory presented here, solutions to the pdf kinetic equation were compared with results obtained from particle tracking in a developing simple shear generated by LES. It was seen that for a variety of particle sizes the agreement was excellent, and that initial correlation effects were important for particles with large response times.

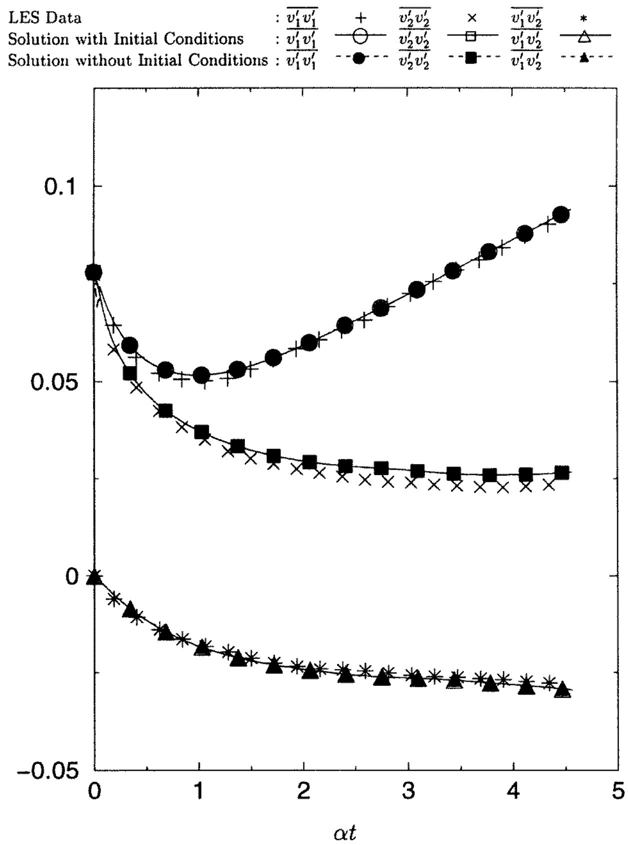

Figure 3. Particle Reynolds stresses for 5 μm particle.

Unlike the LHDI approximation, it would be straightforward to include other physical mechanisms acting on the particle, such as an exchange of mass and temperature between the two phases. This would only involve modifying the fine-grained phase space density function to include these additional particle variables. Further, it should be possible to take into account the other terms which appear in the more general equations of motion for a particle (such as the Basset history term, added mass term etc). This work can also be extended to include particle–particle interactions which would then make the study of aggregation and fragmentation possible.

Appendix A. Functionals and their derivatives

Throughout this paper, functionals and functional derivatives are used quite substantially. Here, brief definitions of these are given. For a more detailed and rigorous account see, for example [25, 26].

Definition A.1. *We say that a quantity* Φ *is a* functional *of the function* $\theta(t)$ *in the interval* (a, b), *when it depends on all the values taken by* $\theta(t)$ *when* t *varies in the interval* (a, b) *and we will write* $\Phi[\theta(t)]$ *for the functional* Φ.

Definition A.2. *The functional* $\Phi[\theta(t)]$ *is said to be* differentiable *with respect to* $\theta(t)$*, if when a small increment* $\delta\theta(t)$ *is added to* $\theta(t)$*, the principal part of the increment* $\delta\Phi[\theta(t)]$ *of this functional is linearly dependent on* $\delta\theta(t)$*, i.e.*

$$\begin{aligned}\delta\Phi[\theta(t)] &= \Phi[\theta(t)+\delta\theta(t)] - \Phi[\theta(t)]\\ &= \int_a^b \Phi'[\theta(t)]\delta\theta(t)\,\mathrm{d}t + \mathrm{o}\left(\int_a^b |\delta\theta(t)|\,\mathrm{d}t\right)\end{aligned} \tag{A1}$$

and $\Phi'[\theta(t)]$ *at the point* $t = t_1$ *is called the* functional derivative *of* $\Phi[\theta(t)]$ *with respect to* $\theta(t)$ *at the point* $t = t_1$*. Also,* $\delta\Phi[\theta(t)]$*, is called the* differential *or* first variation *of the functional* $\Phi[\theta(t)]$.

Taking into account that $\Phi'[\theta(t)]$ is the coefficient of $\delta\theta(t)\,\mathrm{d}t$ in the linear part of the differential $\delta\Phi[\theta(t)]$, it is convenient to adopt the notation

$$\frac{\delta\Phi[\theta(t)]}{\delta\theta(t)\,\mathrm{d}t} = \Phi'[\theta(t)] \tag{A2}$$

for the functional derivative. This notation stresses that the functional derivative is a double limit

$$\frac{\delta\Phi[\theta(t)]}{\delta\theta(t)\,\mathrm{d}t} = \lim_{\substack{|\delta\theta(t)|\to 0\\ \Delta t\to 0}} \frac{\Phi[\theta(t)+\delta\theta(t)] - \Phi[\theta(t)]}{\int_{\Delta t}\delta\theta(t)\,\mathrm{d}t}$$

where $\delta\theta(t)$ now indicates a function that is non-zero only in a small interval of length Δt surrounding the point t.

The simplest example of a differentiable functional is

$$\Phi[\theta(t)] = \int_a^b A(s)\theta(s)\,\mathrm{d}s$$

from which the differential is

$$\delta\Phi[\theta(t)] = \int_a^b A(s)\delta\theta(s)\,\mathrm{d}s + \mathrm{o}\left(\int_a^b |\delta\theta(t)|\,\mathrm{d}t\right)$$

and so from (A1) and (A2), the functional derivative of this functional is

$$\frac{\delta\Phi[\theta(t)]}{\delta\theta(s)\,\mathrm{d}s} = A(s).$$

Appendix B. Evaluation of the phase space diffusion current

In this appendix, a closed expression for the phase space diffusion current $\langle f_i(\boldsymbol{x},t)W(\boldsymbol{x},\boldsymbol{v},t)\rangle$ is obtained. In order to illustrate the derivation using the method outlined in section 3, the cases of homogeneous and inhomogeneous flows are considered separately.

B.1 Homogeneous flow

The first case to be considered will be homogeneous flow with no mean aerodynamic driving force, nor any external forces. It will also be assumed for the moment that $\boldsymbol{\beta}$ is a real symmetric tensor with constant elements and so can be diagonalized with elements β^i, say. In this case the particle equation of motion reduces to

$$\frac{\mathrm{d}x_i^p}{\mathrm{d}t} = v_i^p \tag{B1}$$

$$\frac{\mathrm{d}v_i^p}{\mathrm{d}t} = -\beta^i v_i^p + f_i(\boldsymbol{x}^p, t). \tag{B2}$$

Solving these for $\boldsymbol{x}^p$ gives

$$x_i^p(t) = c_i^0 - \frac{c_i^1}{\beta^i}\mathrm{e}^{-\beta^i t} + \int_0^t \mathrm{e}^{-\beta^i s}\int_0^s \mathrm{e}^{\beta^i r} f_i(\boldsymbol{x}^p, r)\,\mathrm{d}r\,\mathrm{d}s$$

where c^0 and c^1 are constants which are determined from the initial conditions which we assume here to be given at $t = 0$. Using this equation, the functional derivative can be evaluated, and is

$$\begin{aligned}\frac{\delta x_i^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}x'\,\mathrm{d}t'} &= \int_0^t \mathrm{e}^{-\beta^i s}\int_0^s \mathrm{e}^{\beta^i r}\frac{\delta f_i(\boldsymbol{x}^p, r)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}x'\,\mathrm{d}t'}\,\mathrm{d}r\,\mathrm{d}s \\ &= \begin{cases} 0 & \text{if} \quad t < t' \\ \dfrac{\delta_{ij}}{\beta^i}\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')(1 - \mathrm{e}^{-\beta^i(t-t')}) & \text{if} \quad t \geqslant t'. \end{cases}\end{aligned} \tag{B3}$$

Similarly, either by solving (B2) for $\boldsymbol{v}^p$, or using (B3) and

$$\frac{\delta v_k^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} = \frac{\mathrm{d}}{\mathrm{d}t}\frac{\delta x_k^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} \tag{B4}$$

it is seen that

$$\frac{\delta v_i^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} = \delta_{ij}\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')\mathrm{e}^{-\beta^i(t-t')}. \tag{B5}$$

These results coincide with those given in [27, 28] when $\beta^i = \beta$. However, in the latter paper, the equation of motion being considered includes an external force which is a function of position, and as will be seen in the next section, the forms given in that paper for (B3) and (B5) should have been modified to take this into account.

Having found the functional derivatives of both $\boldsymbol{x}^p$ and $\boldsymbol{v}^p$, we can now evaluate the phase space diffusion current, $\langle f_i(\boldsymbol{x}, t)W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle$. Recall that from (9) and (10), this involves the functional derivative of $W(\boldsymbol{x}, \boldsymbol{v}, t)$ which was expressed in terms of functional derivatives of $\boldsymbol{x}^p$ and $\boldsymbol{v}^p$ through (11). Thus, using (B3) and (B5),

$$\begin{aligned}\langle f_i(\boldsymbol{x}, t)W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = &-\int \mathrm{d}\boldsymbol{x}'\int_0^t \mathrm{d}t'\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}', t')\rangle \\ &\times\left[\frac{\partial}{\partial x_k}\left\langle\frac{1}{\beta^k}\delta_{jk}(1 - \mathrm{e}^{-\beta^k(t-t')})\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')W\right\rangle\right. \\ &\left.+\frac{\partial}{\partial v_k}\langle\delta_{jk}\mathrm{e}^{-\beta^k(t-t')}\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')W\rangle\right] \\ = &-\frac{\partial}{\partial x_j}\int \mathrm{d}\boldsymbol{x}'\int_a^t \mathrm{d}t'\frac{1}{\beta^j}(1 - \mathrm{e}^{-\beta^j(t-t')})\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}', t')\rangle\langle\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')W\rangle \\ &-\frac{\partial}{\partial v_j}\int \mathrm{d}\boldsymbol{x}'\int_0^t \mathrm{d}t'\mathrm{e}^{-\beta^j(t-t')}\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}', t')\rangle\langle\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')W\rangle \\ &+\int \mathrm{d}\boldsymbol{x}'\int_0^t \mathrm{d}t'\frac{1}{\beta^j}(1 - \mathrm{e}^{-\beta^j(t-t')})\left\langle\frac{\partial f_i(\boldsymbol{x}, t)}{\partial x_j}f_j(\boldsymbol{x}', t')\right\rangle\langle\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')W\rangle.\end{aligned} \tag{B6}$$

In (B6) a new unknown has been introduced: $\langle\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')W\rangle$. Since $\langle W\rangle$ is the probability of the particle position and velocity being $\boldsymbol{x}$ and $\boldsymbol{v}$ at time t, this unknown can be considered as the joint probability of the particle passing through $\boldsymbol{x}'$ at t' and $(\boldsymbol{x}, \boldsymbol{v})$ at time t. This joint probability can also be written in terms of conditional probabilities

$$\langle\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')W\rangle = \langle\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')|\boldsymbol{x}^p(t) = \boldsymbol{x}\boldsymbol{v}^p(t) = \boldsymbol{v}\rangle\langle W\rangle \tag{B7}$$

where the first term on the right-hand side can be considered as the probability that the particle is at $\boldsymbol{x}'$ at t' given that (conditional upon) it is at $(\boldsymbol{x}, \boldsymbol{v})$ at time t. Alternatively, it may be considered as the probability of transition from the state $(\boldsymbol{x}, \boldsymbol{v})$ at time t to the state $\boldsymbol{x}'$ at t'. Substituting (B7) into (B6), it can be seen that the phase space diffusion current has been expressed in terms of $\langle W\rangle$, the Eulerian statistics of $\boldsymbol{f}$ (which are assumed known) and the transition probability. Thus, the closure problem has been transferred into finding an expression for this latter term.

This problem can be avoided if the transition probability is independent of $\boldsymbol{f}$ for then the spatial integral in (B6) can be performed. Under certain circumstances [29] the probability distribution of the particle paths and $\boldsymbol{f}(\boldsymbol{x}, t)$ can be assumed to be independent (Corrsin's hypothesis). This assumption has been shown to be quite accurate in numerical simulations of particle motion in turbulent flows [30]. As an alternative approach, if $\beta\tau \ll 1$, where τ is the timescale of the fluctuating fluid motion, then $\langle \delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')|\boldsymbol{x}^p(t) = \boldsymbol{x}; \boldsymbol{v}^p(t) = \boldsymbol{v}\rangle$ can be replaced by $\langle \delta(\boldsymbol{x}^p(\boldsymbol{x}, \boldsymbol{v}, t|t') - \boldsymbol{x}')\rangle$ where $\boldsymbol{x}^p(\boldsymbol{x}, \boldsymbol{v}, t|t')$ represents the solution of (B1) and (B2) in the absence of $\boldsymbol{f}$ and with the 'initial' conditions $\boldsymbol{x}^p(\boldsymbol{x}, \boldsymbol{v}, t|t) = \boldsymbol{x}$ and $\boldsymbol{v}^p(\boldsymbol{x}, \boldsymbol{v}, t|t) = \boldsymbol{v}$. This approximation is exactly that found in [18]. It is also that used in the LHDI approach, where a transformation is carried out in terms of phase space variables which solve the particle equations of motion which are devoid of $\boldsymbol{f}$ (see [7] for details). Using either of the above approaches, so that the spatial integrals in (B6) can be evaluated, gives

$$\begin{aligned}\langle f_i(\boldsymbol{x}, t)W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = &-\frac{\partial}{\partial x_j}\int_0^t \mathrm{d}t' \frac{1}{\beta^j}(1 - \mathrm{e}^{-\beta^j(t-t')})\langle f_i(\boldsymbol{x}, t) f_j(\boldsymbol{x}^p(\boldsymbol{x}, \boldsymbol{v}, t|t'), t')\rangle\langle W\rangle \\ &-\frac{\partial}{\partial v_j}\int_0^t \mathrm{d}t' \mathrm{e}^{-\beta^j(t-t')}\langle f_i(\boldsymbol{x}, t) f_j(\boldsymbol{x}^p(\boldsymbol{x}, \boldsymbol{v}, t|t'), t')\rangle\langle W\rangle \\ &+\int_0^t \mathrm{d}t' \frac{1}{\beta^j}(1 - \mathrm{e}^{-\beta^j(t-t')})\left\langle \frac{\partial f_i(\boldsymbol{x}, t)}{\partial x_j} f_j(\boldsymbol{x}^p(\boldsymbol{x}, \boldsymbol{v}, t|t'), t')\right\rangle\langle W\rangle.\end{aligned}$$

Writing $\boldsymbol{f}(\boldsymbol{x}, \boldsymbol{v}, t|t')$ for $\boldsymbol{f}(\boldsymbol{x}^p(\boldsymbol{x}, \boldsymbol{v}, t|t'), t')$, the phase space diffusion current, $\langle \boldsymbol{f}W\rangle$, can be written as

$$\langle f_i(\boldsymbol{x}, t)W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = -\left[\frac{\partial}{\partial x_j}\lambda_{ji} + \frac{\partial}{\partial v_j}\mu_{ji} + \gamma_i\right] P(\boldsymbol{x}, \boldsymbol{v}, t)$$

where

$$\lambda_{ji} = \int_0^t \mathrm{d}t' \frac{1}{\beta^j}(1 - \mathrm{e}^{-\beta^j(t-t')})\langle f_j(\boldsymbol{x}, \boldsymbol{v}, t|t') f_i(\boldsymbol{x}, t)\rangle \quad \text{(B8)}$$

$$\mu_{ji} = \int_0^t \mathrm{d}t' \mathrm{e}^{-\beta^j(t-t')}\langle f_j(\boldsymbol{x}, \boldsymbol{v}, t|t') f_i(\boldsymbol{x}, t)\rangle \quad \text{(B9)}$$

$$\gamma_i = -\int_0^t \mathrm{d}t' \frac{1}{\beta^j}(1 - \mathrm{e}^{-\beta^j(t-t')})\left\langle f_j(\boldsymbol{x}, \boldsymbol{v}, t|t')\frac{\partial f_i(\boldsymbol{x}, t)}{\partial x_j}\right\rangle. \quad \text{(B10)}$$

On comparing these expressions with the equivalent expressions derived from the LHDI approximation given in [7], it is seen that the two forms for $\langle \boldsymbol{f}W\rangle$ are identical. Thus the final form of the pdf kinetic equation for homogeneous flow is

$$\frac{\partial P}{\partial t} + v_i\frac{\partial P}{\partial x_i} - \frac{\partial}{\partial v_i}(\beta^i v_i P) = \frac{\partial}{\partial v_i}\left[\frac{\partial}{\partial v_j}(\mu_{ji}P) + \frac{\partial}{\partial x_j}(\lambda_{ji}P) + \gamma_i P\right] \quad \text{(B11)}$$

with the tensors $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$ and $\boldsymbol{\gamma}$ given by (B8)–(B10).

It should be stressed that if we had been instead considering homogeneous flows with a Gaussian random process $\boldsymbol{f}(t)$ and $\boldsymbol{f}(\boldsymbol{x}, t)$ the above difficulty would not have arisen. In this case there would have been no delta function in (B3) and (B5), and no spatial integral in (B6).

The resulting pdf kinetic equation would thus have been the same as (B11) but without the γ term and with

$$\lambda_{ji} = \int_0^t \mathrm{d}t' \frac{1}{\beta^j}(1 - \mathrm{e}^{-\beta^j(t-t')})\langle f_j(t') f_i(t)\rangle$$
$$\mu_{ji} = \int_0^t \mathrm{d}t' \mathrm{e}^{-\beta^j(t-t')}\langle f_j(t') f_i(t)\rangle.$$

This agrees with the results given in [6].

Finally, to show that the above formulation contains the classical Fokker–Planck equation of Brownian motion, we assume $\boldsymbol{f}$ to be a white noise process,

$$\langle f_i(\boldsymbol{x}, t) f_j(\boldsymbol{x}', t')\rangle = F_{ij}(\boldsymbol{x}, \boldsymbol{x}')\delta(t - t')$$

where $F(\boldsymbol{x}, \boldsymbol{x}')$ is some known function. The time integrals in (B6) can be performed first, and this leads to

$$\langle f_i(\boldsymbol{x}, t) W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = -\frac{1}{2}\frac{\partial}{\partial v_j}\int \mathrm{d}\boldsymbol{x}' F_{ij}(\boldsymbol{x}, \boldsymbol{x}')\langle \delta(\boldsymbol{x}^p(t) - \boldsymbol{x}')|\boldsymbol{x}^p(t) = \boldsymbol{x}; \boldsymbol{v}^p(t) = \boldsymbol{v}\rangle\langle W\rangle$$

where the factor of $\frac{1}{2}$ arises since the singularity in the (time) delta function is one of the limits in the time integration. From the conditions imposed on the transition probability, this becomes

$$\langle f_i(\boldsymbol{x}, t) W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = -\frac{1}{2}\frac{\partial}{\partial v_j} F_{ij}(\boldsymbol{x}, \boldsymbol{x})\langle W\rangle.$$

Thus, as was stated in the introduction, the classical Fokker–Planck equation of Brownian motion is a special case of the more general form considered here.

B.2 Inhomogeneous flow

The method will now be extended to consider non-uniform flows, that is where the equation of motion is of the form

$$\frac{\mathrm{d}x_i^p}{\mathrm{d}t} = v_i^p \tag{B12}$$
$$\frac{\mathrm{d}v_i^p}{\mathrm{d}t} = -\beta_{ij}(\boldsymbol{x}^p, t)v_j^p + F_i(\boldsymbol{x}^p, t) + f_i(\boldsymbol{x}^p, t). \tag{B13}$$

Using these equations, an expression for $\langle \boldsymbol{f}(\boldsymbol{x}, t) W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle$ can now be derived. Recall that this requires the evaluation of the functional derivatives of both $\boldsymbol{x}^p$ and $\boldsymbol{v}^p$ with respect to $\boldsymbol{f}$. Two ways in which these can be derived will now be discussed. From (B4) we need only find an expression for the functional derivative of $\boldsymbol{x}^p$ with respect to $\boldsymbol{f}$, since the functional derivative of $\boldsymbol{v}^p$ with respect to $\boldsymbol{f}$, is the derivative with respect to t of the expression for the functional derivative of $\boldsymbol{x}^p$. To this end, we rewrite (B12) and (B13) in the form

$$\frac{\mathrm{d}^2 x_i^p}{\mathrm{d}t^2} = -\beta_{ij}(\boldsymbol{x}^p, t)\frac{\mathrm{d}x_j^p}{\mathrm{d}t} + F_i(\boldsymbol{x}^p, t) + f_i(\boldsymbol{x}^p, t). \tag{B14}$$

The infinitesimal response function. The first method presented here is an extension of the idea of the 'infinitesimal response function' as discussed in Leslie [31]. The evolution of $\boldsymbol{x}^p(t)$ is described by (B12) and (B13); that is, a particular realization of the (turbulent) particle motion under the influence of $\boldsymbol{f}(\boldsymbol{x}^p, t)$, a particular realization of the fluctuating driving force. Now suppose there is a rather similar system in which the particle position is $\boldsymbol{x}^p(t) + \Delta\boldsymbol{x}^p(t)$ and the driving force is $\boldsymbol{f}(\boldsymbol{x}^p, t) + \Delta\boldsymbol{f}(\boldsymbol{x}^p, t)$. The equation of motion for the second system is, from (B14),

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}(x_i^p + \Delta x_i^p) = -\beta_{ij}(\boldsymbol{x}^p + \Delta\boldsymbol{x}^p, t)\frac{\mathrm{d}}{\mathrm{d}t}(x_j^p + \Delta x_j^p) + F_i(\boldsymbol{x}^p + \Delta\boldsymbol{x}^p, t) + f_i(\boldsymbol{x}^p, t) + \Delta f_i(\boldsymbol{x}^p, t). \tag{B15}$$

Subtracting (B14) from (B15) and expanding the functions of $\boldsymbol{x}^p + \Delta\boldsymbol{x}^p$ as a Taylors series about $\boldsymbol{x}^p$, gives

$$\frac{\mathrm{d}^2\Delta x_i^p}{\mathrm{d}t^2} = -\beta_{ij}\frac{\mathrm{d}\Delta x_j^p}{\mathrm{d}t} - \frac{\partial\beta_{ij}}{\partial x_k^p}\Delta x_k^p\frac{\mathrm{d}x_j^p}{\mathrm{d}t} + \frac{\partial F_i}{\partial x_j^p}\Delta x_j^p + \Delta f_i(\boldsymbol{x}^p, t) + \mathrm{O}((\Delta\boldsymbol{x}^p)^2). \tag{B16}$$

We now suppose that the 'disturbing force', $\Delta\boldsymbol{f}$, which is responsible for the difference between the two systems, is small enough so that the terms $\mathrm{O}((\Delta\boldsymbol{x}^p)^2)$ can be ignored. Also, in this case, identifying $\Delta\boldsymbol{x}^p$ and $\Delta\boldsymbol{f}$ as the variations in $\boldsymbol{x}^p$ and $\boldsymbol{f}$ respectively, that is as $\delta\boldsymbol{x}^p$ and $\delta\boldsymbol{f}$, (B16) becomes

$$\frac{\mathrm{d}^2\delta x_i^p}{\mathrm{d}t^2} = -\beta_{ij}\frac{\mathrm{d}\delta x_j^p}{\mathrm{d}t} - \frac{\partial\beta_{ij}}{\partial x_k^p}\delta x_k^p\frac{\mathrm{d}x_j^p}{\mathrm{d}t} + \frac{\partial F_i}{\partial x_j^p}\delta x_j^p + \delta f_i(\boldsymbol{x}^p, t). \tag{B17}$$

This approximation is only exact if $\delta\boldsymbol{f}$ is infinitesimally small. Thus, the solution to (B17) is the response to an infinitesimal disturbance. As can be seen, (B17) is a linear ordinary differential equation for $\delta\boldsymbol{x}^p$, and as such has the general solution

$$\delta x_i^p(t) = \int_0^t \delta f_j(\boldsymbol{x}^p(t'), t')G_{ji}(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t)\,\mathrm{d}t' \tag{B18}$$

where $G_{ji}(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t)$ satisfies

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}G_{ji} + \beta_{in}\frac{\mathrm{d}}{\mathrm{d}t}G_{jn} + G_{jk}\frac{\partial\beta_{in}}{\partial x_k^p}\frac{\mathrm{d}x_n^p}{\mathrm{d}t} - G_{jk}\frac{\partial F_i}{\partial x_k^p} = \delta_{ji}\delta(t - t').$$

If it is assumed that the solution $\boldsymbol{x}^p$ is determined by the initial conditions at $t = 0$, then the functions G_{ji} vanish unless $t \geqslant t'$; that is they can be considered as retarded Green's functions. The G_{ji} are similar to the generalized response functions appearing in [12]; this will be further highlighted in the following section.

From (B18), we can now find the functional derivative of $\boldsymbol{x}^p$ with respect to $\boldsymbol{f}$. First we write (B18) in the form

$$\delta x_i^p(t) = \int_0^t \mathrm{d}t' \int \mathrm{d}\boldsymbol{x}'\delta f_j(\boldsymbol{x}', t')G_{ji}(\boldsymbol{x}', t'; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}').$$

Then

$$\frac{\delta x_i^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} = G_{ji}(\boldsymbol{x}', t'; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}') \tag{B19}$$

and using (B4), we also have

$$\frac{\delta v_i^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} = \frac{\mathrm{d}}{\mathrm{d}t}G_{ji}(\boldsymbol{x}', t'; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}'). \tag{B20}$$

Substituting (B19) and (B20) into (11), and the resultant expression into (9), the phase space diffusion current can thus be written as

$$\langle f_i(\boldsymbol{x}, t)W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = -\int \mathrm{d}\boldsymbol{x}' \int_0^t \mathrm{d}t'\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}', t')\rangle$$

$$
\begin{aligned}
&\times\left[\frac{\partial}{\partial x_k}\langle G_{jk}(\boldsymbol{x}',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')W\rangle\right.\\
&\left.+\frac{\partial}{\partial v_k}\left\langle\frac{\mathrm{d}}{\mathrm{d}t}G_{jk}(x',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')W\right\rangle\right]\\
=&-\frac{\partial}{\partial x_k}\int\mathrm{d}\boldsymbol{x}'\int_0^t\mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_j(\boldsymbol{x}',t')\rangle\langle G_{jk}(\boldsymbol{x}',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')W\rangle\\
&-\frac{\partial}{\partial v_k}\int\mathrm{d}\boldsymbol{x}'\int_0^t\mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_j(\boldsymbol{x}',t')\rangle\langle\dot{G}_{jk}(\boldsymbol{x}',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')W\rangle\\
&+\int\mathrm{d}\boldsymbol{x}'\int_0^t\mathrm{d}t'\left\langle\frac{\partial f_i(\boldsymbol{x},t)}{\partial x_k}f_j(\boldsymbol{x}',t')\right\rangle\langle G_{jk}(\boldsymbol{x}',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')W\rangle
\end{aligned}
\tag{B21}
$$

where $\dot{G}_{ji}=\mathrm{d}G_{ji}/\mathrm{d}t$.

As in the homogeneous case, by considering the above as joint probabilities, the terms in the angled brackets can be rewritten as follows:

$$
\begin{aligned}
\langle G_{jk}(\boldsymbol{x}',t';\boldsymbol{x}^p(t),t)\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')W\rangle &= \langle G_{jk}(\boldsymbol{x}',t';\boldsymbol{x},t)\rangle\langle\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')W\rangle\\
&= G_{jk}(\boldsymbol{x}',t';\boldsymbol{x},t)\langle\delta(\boldsymbol{x}^p(t')-\boldsymbol{x}')|\boldsymbol{x}^p(t)=x;\boldsymbol{v}^p(t)=\boldsymbol{v}\rangle\langle W\rangle
\end{aligned}
$$

and similarly for the terms involving $\dot{G}_{ji}$. The first line is obtained by expressing joint probabilities in terms of conditional ones; the second line follows because of the conditions on $\boldsymbol{G}$; and the third line is obtained by using the transition probability and by noting that $\boldsymbol{G}$ is not a fluctuating quantity since (1) by the method of construction of the particle equations of motion as given in [7], both β and $\boldsymbol{F}$ are dependent only on the mean values of the particle and fluid velocity and thus not explicitly on $\boldsymbol{f}$ and (2) $\boldsymbol{G}$, from the second line in the above equation, does not depend on $\boldsymbol{x}^p$ but only on $\boldsymbol{x}'$ and $\boldsymbol{x}$.

Finally, by making the same approximations for the transition probability as was done in the homogeneous case, and again writing $\boldsymbol{f}(\boldsymbol{x},\boldsymbol{v},t|t')$ for $\boldsymbol{f}(\boldsymbol{x}^p(\boldsymbol{x},\boldsymbol{v},t|t'),t')$, where $\boldsymbol{x}^p(\boldsymbol{x},\boldsymbol{v},t|t')$ is the solution to the equations of motion in the absence of $\boldsymbol{f}$, we have

$$
\begin{aligned}
\langle f_i(\boldsymbol{x},t)W(\boldsymbol{x},\boldsymbol{v},t)\rangle =& -\frac{\partial}{\partial x_k}\int_0^t\mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_j(\boldsymbol{x},\boldsymbol{v},t|t')\rangle G_{jk}(\boldsymbol{x}',t';\boldsymbol{x},t)\langle W\rangle\\
&-\frac{\partial}{\partial v_k}\int_0^t\mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_j(\boldsymbol{x},\boldsymbol{v},t|t')\rangle\dot{G}_{jk}(\boldsymbol{x}',t';\boldsymbol{x},t)\langle W\rangle\\
&+\int_0^t\mathrm{d}t'\left\langle\frac{\partial f_i(\boldsymbol{x},t)}{\partial x_k}f_j(\boldsymbol{x},\boldsymbol{v},t|t')\right\rangle G_{jk}(\boldsymbol{x}',t';\boldsymbol{x},t)\langle W\rangle.
\end{aligned}
\tag{B22}
$$

Writing $G_{jk}(t'|t)$ for $G_{jk}(\boldsymbol{x}',t';\boldsymbol{x},t)$, (B22) can be rewritten as

$$
\langle f_i(\boldsymbol{x},t)W(\boldsymbol{x},\boldsymbol{v},t)\rangle = -\left[\frac{\partial}{\partial x_j}\lambda_{ji}+\frac{\partial}{\partial v_j}\mu_{ji}+\gamma_i\right]P(\boldsymbol{x},\boldsymbol{v},t) \tag{B23}
$$

where

$$
\lambda_{ji}=\int_0^t\mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_k(\boldsymbol{x},\boldsymbol{v},t|t')\rangle G_{kj}(t'|t) \tag{B24}
$$

$$
\mu_{ji}=\int_0^t\mathrm{d}t'\langle f_i(\boldsymbol{x},t)f_k(\boldsymbol{x},\boldsymbol{v},t|t')\rangle\frac{\mathrm{d}}{\mathrm{d}t}G_{kj}(t'|t) \tag{B25}
$$

$$
\gamma_i=-\int_0^t\mathrm{d}t'\left\langle\frac{\partial f_i(\boldsymbol{x},t)}{\partial x_j}f_k(\boldsymbol{x},\boldsymbol{v},t|t')\right\rangle G_{kj}(t'|t) \tag{B26}
$$

and where $G_{ji}(t'|t)$ satisfies

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}G_{ji} + \beta_{in}\frac{\mathrm{d}}{\mathrm{d}t}G_{jn} + G_{jk}\frac{\partial\beta_{in}}{\partial x_k}\frac{\mathrm{d}x_n}{\mathrm{d}t} - G_{jk}\frac{\partial F_i}{\partial x_k} = \delta_{ji}\delta(t-t'). \tag{B27}$$

Comparing these expressions with the equivalent ones derived from the LHDI approximation for inhomogeneous flow given in [7], it is seen that the two forms for $\langle \boldsymbol{f} W\rangle$ are again identical. Thus the final form of the pdf kinetic equation for inhomogeneous flow is

$$\frac{\partial P}{\partial t} + v_i\frac{\partial P}{\partial x_i} - \frac{\partial}{\partial v_i}(\beta_{ij}v_j P) + \frac{\partial}{\partial v_i}(F_i P) = \frac{\partial}{\partial v_i}\left[\frac{\partial}{\partial v_j}(\mu_{ji}P) + \frac{\partial}{\partial x_j}(\lambda_{ji}P) + \gamma_i P\right]$$

but with the tensors, $\boldsymbol{\lambda}$, $\boldsymbol{\mu}$ and $\boldsymbol{\gamma}$ given by (B24)–(B26) above.

Green's function representation. Another approach in deriving the functional derivatives of $\boldsymbol{x}^p$ and $\boldsymbol{v}^p$ with respect to $\boldsymbol{f}$, is to define, following [12],

$$G_{ji}(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t) = \frac{\delta x_i^p(t)}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'}. \tag{B28}$$

Taking the functional derivative of (B14) with respect to $\boldsymbol{f}$, yields

$$\frac{\delta}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'}\frac{\mathrm{d}^2 x_i^p}{\mathrm{d}t^2} = -\frac{\delta}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'}\left(\beta_{ik}(\boldsymbol{x}^p(t), t)\frac{\mathrm{d}x_k^p}{\mathrm{d}t}\right) + \frac{\delta F_i(\boldsymbol{x}^p(t), t)}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t} + \frac{\delta f_i(\boldsymbol{x}^p(t), t)}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'}. \tag{B29}$$

Noting that the functional derivative with respect to $\boldsymbol{f}(\boldsymbol{x}^p(t'), t')$ commutes with the time derivatives appearing in (B29), this becomes

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}\frac{\delta x_i^p}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'} = -\beta_{ik}\frac{\mathrm{d}}{\mathrm{d}t}\frac{\delta x_k^p}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'} - \frac{\mathrm{d}x_k^p}{\mathrm{d}t}\frac{\delta\beta_{ik}}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'} + \frac{\delta F_i(\boldsymbol{x}^p(t), t)}{\delta f_j(\boldsymbol{x}^p(t'), t')\,\mathrm{d}t'} + \delta_{ij}\delta(t-t'). \tag{B30}$$

On using the chain rule to take the functional derivative of $\boldsymbol{\beta}$ and $\boldsymbol{F}$, and also using the definition given in (B28), (B30) becomes

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}G_{ji} = -\beta_{ik}\frac{\mathrm{d}}{\mathrm{d}t}G_{jk} - \frac{\mathrm{d}x_k^p}{\mathrm{d}t}\frac{\partial\beta_{ik}}{\partial x_m^p}G_{jm} + \frac{\partial F_i}{\partial x_k^p}G_{jk} + \delta_{ij}\delta(t-t').$$

Note, however, that we wish to evaluate $\delta x_i^p(t)/\delta f_j(\boldsymbol{x}'(t'), t')\,\mathrm{d}t'\,\mathrm{d}\boldsymbol{x}'$. From (B28) it can be seen that

$$\delta x_i^p(t) = \int_0^t \mathrm{d}t' \int \mathrm{d}\boldsymbol{x}'\delta f_j(\boldsymbol{x}', t')G_{ji}(\boldsymbol{x}', t'; \boldsymbol{x}^p(t), t))\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}')$$

from which follows

$$\frac{\delta x_i^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}t'\,\mathrm{d}\boldsymbol{x}'} = G_{ji}(\boldsymbol{x}', t'; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}').$$

This equation is exactly the same as was derived before using the infinitesimal response function method (cf (B19)). Thus that argument is now repeated with exactly the same final equation derived for the phase space diffusion current as given by (B23)–(B27).

Appendix C. The dispersion tensors

In order to derive solutions to the pdf kinetic equation, explicit forms of the dispersion tensors $\boldsymbol{\mu}$ and $\boldsymbol{\lambda}$ are required. In general, these will be functions of position, $\boldsymbol{x}$ and velocity $\boldsymbol{v}$ as well as time t. Nonetheless, under certain assumptions, explicit expressions for $\boldsymbol{\mu}$ and $\boldsymbol{\lambda}$ may be obtained as functions of time only.

In section 5 we are interested in the case where the underlying flow field is a simple shear, and this is what will be concentrated on here. However, it is not difficult to extend the details given below to other (linear) flow fields. Also, we include the initial correlation effects as outlined in section 4; see the discussion before (27). That is we set

$$x_i^p(0) = 0 \qquad \dot{x}_i^p = \frac{a}{\beta} f_i(\boldsymbol{x}^p(0), 0) \tag{C1}$$

with a a known constant and, as in section 5, we have assumed that $\beta_{ij} = \beta\delta_{ij}$, where β^{-1} is the particle response time. From the analysis given in section 4, it is seen from (20)

$$B_{ji} = 0 \qquad A_{ji} = \frac{a}{\beta}\delta_{ij}\delta(t').$$

Thus (23) gives

$$G_{ji}^2 = \frac{a}{\beta}\delta(t')G_{ji}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t)$$

where G_{ji}^1 satisfies (21). Thus,

$$G_{ji}(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t) = G_{ji}^1(\boldsymbol{x}^p(t'), t'; \boldsymbol{x}^p(t), t) + \frac{a}{\beta}\delta(t')G_{ji}^1(\boldsymbol{x}^p(0), 0; \boldsymbol{x}^p(t), t).$$

Substituting this into (B19) and (B20) gives

$$\begin{aligned}\frac{\delta x_i^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} &= G_{ji}^1(\boldsymbol{x}', t'; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}') \\ &\quad + \frac{a}{\beta}\delta(t')G_{ji}^1(\boldsymbol{x}', 0; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(0) - \boldsymbol{x}') \\ \frac{\delta v_i^p(t)}{\delta f_j(\boldsymbol{x}', t')\,\mathrm{d}\boldsymbol{x}'\,\mathrm{d}t'} &= \frac{\mathrm{d}}{\mathrm{d}t}G_{ji}^1(\boldsymbol{x}', t'; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(t') - \boldsymbol{x}') \\ &\quad + \frac{a}{\beta}\delta(t')\frac{\mathrm{d}}{\mathrm{d}t}G_{ji}^1(\boldsymbol{x}', 0; \boldsymbol{x}^p(t), t)\delta(\boldsymbol{x}^p(0) - \boldsymbol{x}').\end{aligned}$$

Using the above and (B21), along with the same arguments that lead to (B22), we find that the phase space diffusion current is

$$\begin{aligned}\langle f_i(\boldsymbol{x}, t)W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle &= -\frac{\partial}{\partial x_k}\int_0^t \mathrm{d}t'\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}, \boldsymbol{v}, t|t')\rangle G_{jk}^1(\boldsymbol{x}', t'; \boldsymbol{x}, t)\langle W\rangle \\ &\quad - \frac{a}{\beta}\frac{\partial}{\partial x_k}\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}, t)f_j(\boldsymbol{x}, \boldsymbol{v}, t|0)\rangle G_{jk}^1(\boldsymbol{x}', 0; \boldsymbol{x}, t)\langle W\rangle \\ &\quad - \frac{\partial}{\partial v_k}\int_0^t \mathrm{d}t'\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}, \boldsymbol{v}, t|t')\rangle \dot{G}_{jk}^1(\boldsymbol{x}', t'; \boldsymbol{x}, t)\langle W\rangle \\ &\quad - \frac{a}{\beta}\frac{\partial}{\partial v_k}\langle f_i(\boldsymbol{x}, t)f_j(\boldsymbol{x}, \boldsymbol{v}, t|0)\rangle \dot{G}_{jk}^1(\boldsymbol{x}', 0; \boldsymbol{x}, t)\langle W\rangle \\ &\quad + \int_0^t \mathrm{d}t'\left\langle \frac{\partial f_i(\boldsymbol{x}, t)}{\partial x_k} f_j(\boldsymbol{x}, \boldsymbol{v}, t|t')\right\rangle G_{jk}^1(\boldsymbol{x}', t'; \boldsymbol{x}, t)\langle W\rangle \\ &\quad + \frac{a}{\beta}\left\langle \frac{\partial f_i(\boldsymbol{x}, t)}{\partial x_k} f_j(\boldsymbol{x}, \boldsymbol{v}, t|0)\right\rangle G_{jk}^1(\boldsymbol{x}', 0; \boldsymbol{x}, t)\langle W\rangle.\end{aligned} \tag{C2}$$

As before, writing $G^1_{jk}(t'|t)$ for $G^1_{jk}(\boldsymbol{x}', t'; \boldsymbol{x}, t)$, (C2) can be rewritten as

$$\langle f_i(\boldsymbol{x}, t) W(\boldsymbol{x}, \boldsymbol{v}, t)\rangle = -\left[\frac{\partial}{\partial x_j}\lambda_{ji} + \frac{\partial}{\partial v_j}\mu_{ji} + \gamma_i\right] P(\boldsymbol{x}, \boldsymbol{v}, t)$$

where now

$$\lambda_{ji} = \int_0^t \mathrm{d}t' \langle f_i(\boldsymbol{x}, t) f_k(\boldsymbol{x}, \boldsymbol{v}, t|t')\rangle G^1_{kj}(t'|t) + \frac{a}{\beta}\langle f_i(\boldsymbol{x}, t) f_k(\boldsymbol{x}, \boldsymbol{v}, t|0)\rangle G^1_{kj}(0|t)\langle W\rangle \tag{C3}$$

$$\mu_{ji} = \int_0^t \mathrm{d}t' \langle f_i(\boldsymbol{x}, t) f_k(\boldsymbol{x}, \boldsymbol{v}, t|t')\rangle \frac{\mathrm{d}}{\mathrm{d}t} G^1_{kj}(t'|t) + \frac{a}{\beta}\langle f_i(\boldsymbol{x}, t) f_k(\boldsymbol{x}, \boldsymbol{v}, t|0)\rangle \frac{\mathrm{d}}{\mathrm{d}t} G^1_{kj}(0|t)\langle W\rangle \tag{C4}$$

$$\gamma_i = -\int_0^t \mathrm{d}t' \left\langle \frac{\partial f_i(\boldsymbol{x}, t)}{\partial x_j} f_k(\boldsymbol{x}, \boldsymbol{v}, t|t')\right\rangle G^1_{kj}(t'|t) - \frac{a}{\beta}\left\langle \frac{\partial f_i(\boldsymbol{x}, t)}{\partial x_k} f_k(\boldsymbol{x}, \boldsymbol{v}, t|0)\right\rangle G^1_{kj}(0|t)\langle W\rangle \tag{C5}$$

and where $G^1_{ji}(t'|t)$ satisfies

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2} G^1_{ji} + \beta \frac{\mathrm{d}}{\mathrm{d}t} G^1_{ji} - G^1_{jk}\frac{\partial F_i}{\partial x_k} = \delta_{ji}\delta(t - t'). \tag{C6}$$

To proceed, the correlation $\langle \boldsymbol{f}(\boldsymbol{x}, t)\boldsymbol{f}(\boldsymbol{x}, \boldsymbol{v}, t|t')\rangle$ must be evaluated somehow. Since the flow field in section 5 is homogeneous we neglect the variations with respect to $\boldsymbol{x}$ and $\boldsymbol{v}$. We further assume that the resultant correlation $\langle f_i(t) f_k(t|s)\rangle$ is stationary and has an exponential decay, i.e.

$$\langle f_i(t) f_k(t|s)\rangle = \langle f_i f_k\rangle \mathrm{e}^{-(s-t)/\tau}$$

where τ is a constant and represents the fluid integral timescale. Since the flow field is linear, we see from (C6) that we can replace $G^1_{ij}(t'|t)$ by $G^1_{ij}(t - t')$ in (C3)–(C5). Also, due to homogeneity, $\gamma = 0$.

Under these assumptions μ and λ are given by

$$\lambda_{ji}(t) = \langle f_i f_k\rangle \int_0^t G_{kj}(s)\mathrm{e}^{-s/\tau}\,\mathrm{d}s + \frac{a}{\beta}\langle f_i f_k\rangle \mathrm{e}^{-t/\tau} G_{kj}(t) \tag{C7}$$

$$\mu_{ji}(t) = \langle f_i f_k\rangle \int_0^t \frac{\mathrm{d}G_{kj}(s)}{\mathrm{d}s}\mathrm{e}^{-s/\tau}\,\mathrm{d}s + \frac{a}{\beta}\langle f_i f_k\rangle \mathrm{e}^{-t/\tau}\frac{\mathrm{d}G_{kj}(t)}{\mathrm{d}t} \tag{C8}$$

with $\boldsymbol{G}(t)$ obtained by solving

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2} G_{ji} + \beta \frac{\mathrm{d}}{\mathrm{d}t} G_{ji} - G_{jk}\frac{\partial F_i}{\partial x_k} = \delta_{ji}\delta(t). \tag{C9}$$

In the simple shear flow, the mean aerodynamic driving force acting on the particle is given by

$$\langle \boldsymbol{F}\rangle = \beta\bar{\boldsymbol{u}} = \alpha\beta(x_2, 0) \tag{C10}$$

where it has been assumed that the only force acting on the particle is Stokes drag; this also implies $\boldsymbol{f} = \beta\boldsymbol{u}'$ where $\boldsymbol{u}'$ is the fluctuating fluid velocity. In (C10), α is the shear gradient and $\boldsymbol{x} = (x_1, x_2)$ represents position. In the absence of any external force, (C9) gives

$$\frac{\mathrm{d}^2\boldsymbol{G}}{\mathrm{d}t^2} = -\beta\frac{\mathrm{d}\boldsymbol{G}}{\mathrm{d}t} + \alpha\beta\boldsymbol{H} + \delta(t)I$$

where

$$\boldsymbol{H} = \begin{pmatrix} G_{12} & 0 \\ G_{22} & 0 \end{pmatrix}.$$

This has the solution

$$G(t) = \begin{pmatrix} \frac{1}{\beta}(1 - e^{-\beta t}) & 0 \\ \frac{\alpha}{\beta^2}[2(e^{-\beta t} - 1) + \beta t(1 + e^{-\beta t})] & \frac{1}{\beta}(1 - e^{-\beta t}) \end{pmatrix}.$$

These are now substituted in (C7) and (C8) to obtain $\lambda(t)$ and $\mu(t)$:

$$\begin{aligned}
\lambda_{ij}(t) = \langle f_i f_j\rangle & \frac{\tau}{\beta(\beta\tau+1)}[(\beta\tau+1)(1-e^{-t/\tau}) + e^{-(\beta\tau+1)t/\tau} - 1)] \\
& +\delta_{i1}\langle f_2 f_j\rangle \frac{2\alpha\tau}{\beta^2(\beta\tau+1)}[(1-e^{-(\beta\tau+1)t/\tau}) + (\beta\tau+1)(e^{-t/\tau} - 1)] \\
& +\delta_{i1}\langle f_2 f_j\rangle \frac{\alpha\tau^2}{\beta(\beta\tau+1)^2}[(1-e^{-(\beta\tau+1)t/\tau}) - (\beta\tau+1)^2(e^{-t/\tau} - 1)] \\
& -\delta_{i1}\langle f_2 f_j\rangle \frac{\alpha\tau t}{\beta(\beta\tau+1)}[(\beta\tau+1)e^{-t/\tau} + e^{-(\beta\tau+1)t/\tau}] \\
& +\langle f_i f_j\rangle \frac{a}{\beta^2} e^{-t/\tau}(1-e^{-\beta t}) + \delta_{i1}\langle f_2 f_j\rangle \frac{a\alpha}{\beta^3} e^{-t/\tau}[2(e^{-\beta t} - 1) + \beta t(1+e^{-\beta t}] \\
\mu_{ij}(t) = \langle f_i f_j\rangle & \frac{\tau}{\beta\tau+1}(1-e^{-(\beta\tau+1)t/\tau}) \\
& +\delta_{i1}\langle f_2 f_j\rangle \frac{\alpha\tau}{(\beta\tau+1)^2} e^{-(\beta\tau+1)t/\tau}[t(\beta\tau+1) + \tau(1-e^{(\beta\tau+1)t/\tau})] \\
& +\delta_{i1}\langle f_2 f_j\rangle \frac{\alpha\tau}{\beta(\beta\tau+1)}[(\beta\tau+1)(1-e^{-t/\tau}) + (e^{-(\beta\tau+1)t/\tau} - 1)] \\
& +\langle f_i f_j\rangle \frac{a}{\beta} e^{-(\beta\tau+1)t/\tau} + \delta_{i1}\langle f_2 f_j\rangle \frac{a\alpha}{\beta^2} e^{-t/\tau}[1 - e^{-\beta t}(1+\beta t)].
\end{aligned}$$

From the initial conditions, (C1), with $\boldsymbol{f} = \beta\boldsymbol{u}'$, we have $v_i = au'_i$. Multiplying both sides by u'_j and averaging gives $\overline{u'_j v'_i} = a\overline{u'_j u'_i}$. This equation enables us to determine a since both $\overline{u'_j u'_i}$ and $\overline{u'_j v'_i}$ are known at $t = 0$ from the LES data with the latter given in table 1.

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