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On diffusion theory in turbulence

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Abstract. The Fokker-Planck equation for the probability density of fluid particle position in inhomogeneous unsteady turbulent flow is derived. The equation is obtained starting from the general kinematic relationship between velocity and displacement of a fluid particle and applying exact asymptotic analysis. For (almost) incompressible flow the equation reduces to the convection diffusion equation and the equation pertaining to the scalar gradient hypothesis. In this way the connection is established with eddy diffusivity models, widely used in numerical codes of computational fluid dynamics. It is further shown that within the accuracy of the approximation scheme of the diffusion limit, diffusion constants can equally be based on coarse-grained Lagrangian statistics as defined by Kolmogorov or on Eulerian statistics in a frame that moves with the mean Eulerian velocity as proposed by Burgers. The results presented for diffusion theory are the leading terms of asymptotic expansions. Truncated terms are higher-order spatial derivatives of the probability density or of the scalar mean value with coefficients based on cumulants higher than second order of fluid velocities and their derivatives. The magnitude of these terms has been assessed by employing scaling rules of turbulent flows in pipes and channels, turbulent boundary layers, turbulent jets, wakes and mixing layers, grid turbulence, convective layers and canopy turbulence. It reveals that a true diffusion limit does not exist. Although truncated terms can be of limited magnitude, a limit process by which these terms become vanishingly small and by which the diffusion approximation would become exact does not occur for any of the cases of turbulent flow considered. Applying the concepts of diffusion theory resorts to employing approximate methods of analysis.

Key words: turbulence, diffusion, Fokker-Planck equation

1. Introduction

Fluid flow in nature and engineering applications is nearly always turbulent. Velocities, pressures and other fluid-mechanical variables fluctuate randomly in time and space. These fluctuations strongly influence the ability of the fluid to transport matter, momentum and heat. Fundamental understanding of this process is a key target of research in turbulence.

Transport by turbulent flow is generally dominated by the large-scale components of the turbulence. Theories of large-scale turbulence are due to Taylor [1, 2], Prandtl [3] Von Karman [4] and others. They originate from the assumed resemblance between turbulent fluctuations and molecular chaos. Expressions for transport of matter, momentum and energy are formulated analogous to the phenomenological descriptions of diffusion of matter, viscous transport of momentum and conduction of heat by random motion of molecules. Constants of proportionality can be measured and can be used to predict parameters of turbulent transport in similar flow configurations.

Though widely used, the phenomenological theories of large-scale turbulence are semi-empirical. The phenomenological descriptions, *viz.* the gradient hypotheses of mass, momentum and energy, are postulated but their validity has yet to be proven. Furthermore, in these theories the statistical parameters of turbulence are treated at a modest level. More exact and detailed theories exist for specific areas. Taylor [5] developed the concept of homogeneous isotropic turbulence, a mathematical idealization by which it is possible to derive expressions for statistical quantities which explain important aspects of turbulent flow. In his investigation of diffusion by continuous movements in homogeneous isotropic fields [6], diffusion of foreign matter was shown to be related to the time correlation of the random velocity of tagged or marked fluid particles. This essentially Lagrangian-based description enabled the observations of the spatial spread downstream of a point source to be explained. Comprehensive treatises on homogeneous isotropic turbulence and diffusion in homogeneous turbulence have been given by Batchelor [7, 8]. The application of homogeneous isotropic turbulence including certain generalizations to real turbulent flow was facilitated by Kolmogorov [9, 10]. His similarity hypotheses provided the basis for the description of the small-scale components of turbulent flow at very high Reynolds number according to the concept of locally homogeneous isotropic turbulence, see [11, Chapter 8].

Several other efforts have been made to contribute to the theoretical development of statistical turbulence: see, *e.g.*, the textbooks of Monin and Yaglom [12, 11], Hinze [13] and Tennekes and Lumley [14] or the review article of Hunt [15]. Monin [16] and Lundgren [17] were the first to formulate and study the dynamical equations for the time evolution of the probability density distribution of Eulerian fluid velocity fluctuations. A promising approach involves the application of Markov theory. Under the assumption that evolution times are large compared to the correlation time of the underlying stochastic process, the variables of turbulent flow are approximated by those of a Markov process. This enables the probability density of the stochastic variable to be described by a Fokker-Planck equation. Alternatively, the behaviour of the variable in time can be described by an equivalent Langevin or fluctuation equation in which random forcing occurs through white-noise. Application of the Markov approximation to the displacement of a marked fluid particle in homogeneous uniform turbulent flow yields the diffusion equation.The Markov approach has further been applied to the stochastic process of velocities of marked particles in turbulent flow: *e.g.* [18, 19, 20, 21].

Despite the numerous efforts and high level of mathematical methods employed, fundamental questions on the theory of statistical turbulence have remained unanswered. Though widely used in numerical codes of computational fluid dynamics, the correctness of the phenomenological theories of large scale turbulence has been subjected to doubts: see [14, Section 2.2] and [22, Section 4.4]. Equally, the validity of diffusion theory or the Markov approximation for fluid particle displacement, which is thought to underlie the phenomenological theories, has been questioned [18]. It is still unclear how and under which limit conditions the general stochastic process of turbulent flow might reduce to that of diffusion theory or the Markov approximation of particle displacement and what the precise relationship of these theories is with the phenomenological theories.

Uncertainty on the position of diffusion theory can be attributed to the common practice of assuming Fokker-Planck and Langevin equations rather than deriving them from general principles. It has among others therefore remained unnoticed that for the nonlinear fluctuation equations of turbulent flow, the limit of long evolution times is not sufficient to arrive at the Markov process of fluid particle displacement. As the present analysis will show, conditions are also imposed on the magnitudes of the nonlinear terms apparent in case of real inhomogeneous turbulent flow. The form of these terms influences the validity and form of the Fokker-Planck equation and the Langevin equation associated with it.

Another source of uncertainty is the representation of the coefficients in the postulated Fokker-Planck and Langevin equations. These are usually given in terms of the Lagrangian statistics of coarse-grained fluid particle motion. The determination of these coefficients in inhomogeneous turbulent flow is practically impossible. It gives rise to a practice in which coefficients are treated as parameters to be chosen. Their values are determined by fitting predictions to measurements. As the coefficients are space-dependent and sometimes time-dependent as well, falsification of the thus established theory is an almost impossible task. Forecasts in situations other than those tested are surrounded by uncertainty as a result. Efforts to specify the coefficients in terms of better measurable Eulerian-based statistics have only been successful for the Langevin equation for particle velocity in isotropic grid turbulence; *e.g.* [19, 20, 23].

The present analysis is aimed at avoiding the above shortcomings. Instead of postulating the Fokker-Planck equation is, derived starting from the general kinematic relationship between particle velocity and displacement and applying exact asymptotic analysis. In this way conclusive answers are given on the various questions of diffusion theory including its validity in turbulent fluid flow.

The derivation of the Fokker-Planck equation for particle position is given in Section 2, while elementary forms of the equation and the equations pertaining to the gradient hypothesis are derived in Section 3. The connection between Lagrangian and Eulerian statistics appropriate for the diffusion limit is treated in Section 4. The conditions under which the diffusion limit holds and the extent to which these are satisfied for actual cases of turbulent flow are discussed in Section 5. Conclusions are summarized in Section 6.

2. Derivation of the Fokker-Planck equation

The objective is to derive descriptions for the probability-density distribution of the position of a discrete fluid particle in inhomogeneous unsteady turbulent flow. Here a discrete fluid particle is a fluid particle that is marked at some point without disturbing the flow field, or a substance that is added at some point and moves with the fluid as if it is part of it. Fluid velocities are described as

$$u_\nu(\mathbf{x}, t) = u_\nu^0(\mathbf{x}, t) + \varepsilon u_\nu^{'}(\mathbf{x}, t), \text{ for } \nu = 1, 2, 3, \tag{1}$$

where $u_\nu^0(\mathbf{x}, t)$ is the ensemble mean velocity,

$$u_\nu^0(\mathbf{x}, t) = \langle u_\nu(\mathbf{x}, t)\rangle\ , \tag{2}$$

$\varepsilon u_\nu^{'}(\mathbf{x}, t)$ is the fluctuating component of fluid velocity, t is time and $\mathbf{x}$ is space coordinate. The stochastic process $u_\nu(\mathbf{x}, t)$ has finite correlation time. In accordance with the assumed possibility of unsteadiness of the turbulent flow, ensemble averages involving $u_\nu(\mathbf{x}, t)$ are allowed to vary in a deterministic manner with time. Ensemble average here means mean value at a specific position and a specific moment in time having repeated the process many times. When ergodicity is adopted, for stationary processes ensemble averages can be taken equal to time averages which are easier to assess in practice.

In the subsequent analysis a theoretical approach is presented which is based on the application of asymptotic expansions involving powers of ε. Equation (1) may suggest that this implies restriction to fluctuating velocities, which are small compared to mean velocity, but this is not the case. Rather than mean velocity, fluctuating velocities are required to be small compared to L/τ_c, where τ_c is correlation time of fluid particle velocity and L external length scale [11] or integral scale [14]: *e.g.*, thickness of a turbulent boundary layer, radius of a pipe or width of a channel in which turbulent flow takes place, mesh spacing of a grid producing turbulence, or cross-sectional dimension of a turbulent jet, wake or mixing layer. This can be indicated by

$$\varepsilon \ll 1, \tag{3}$$

where

$$\varepsilon = \frac{\tau_c \tilde{u}}{L}, \tag{4}$$

$\tilde{u}$ representing the order of magnitude of the fluctuating part of fluid velocity, *e.g.*, the standard deviation of fluid velocity. Condition (3) implies that the random displacement of a fluid particle measured over a time period which corresponds to the correlation time of fluid particle velocity is required to be small compared to the external length scale. While displacements due to random velocities are required to be small during periods of correlation, this restriction is not imposed on displacement by the mean fluid motion; it can be large or small. The parameter ε corresponds to the Kubo number employed in stochastic analysis [24] or the inhomogeneity index referred to in the modeling of atmospheric turbulence [20]. For the diffusion limit to hold, it appears necessary that $\varepsilon \ll 1$. Under this condition results pertinent to the diffusion limit are obtained as the leading terms of perturbation expansions involving powers of ε. These are presented in the present and subsequent sections. The necessity of $\varepsilon \ll 1$ and the extent to which this condition is satisfied for actual cases of turbulent fluid flow are discussed in Section 5.

The position $x_\nu(t)$ of any passively marked fluid particle initially at $x_\nu(0)$ at $t = 0$ is described by the equation

$$\dot{x}_\nu(t) = u_\nu^0(\mathbf{x}(t), t) + \varepsilon u_\nu^{'}(\mathbf{x}(t), t), \tag{5}$$

where the dot denotes differentiation with respect to time and where fluid velocities are evaluated at the position of the particle. In (5), $\mathbf{x}(t)$ denotes the Lagrangian position of the marked particle which varies with time, this is in contrast to $\mathbf{x}$ which denotes fixed spatial position in an Eulerian specification of variables. The case of zero perturbed flow, *i.e.*, $\varepsilon = 0$ in (5), is used to implement a transformation of variables. To this end $\mathbf{y}(t; \mathbf{z})$ denotes the solution of the equation of the mean flow

$$\dot{y}_\nu(t) = u_\nu^0(\mathbf{y}(t), t), \tag{6}$$

subject to the initial condition

$$y_\nu(0) = z_\nu. \tag{7}$$

Next, a new variable $\mathbf{x}^*(t)$ is defined by $\mathbf{x}(t) = \mathbf{y}(t; \mathbf{x}^*(t))$: for each moment in time t its value is such that, when starting from position $x_\nu^*(t)$ and following a path according to the mean flow field, one arrives at time t at the same position as the particle which starts from $x_\nu(0)$ and follows the path described by (5). Noting that

$$\dot{x}_\nu(t) = u_\nu^0(\mathbf{x}(t), t) + \dot{x}_\mu^*(t) \frac{\partial y_\nu}{\partial z_\mu}(t; \mathbf{x}^*(t)) \tag{8}$$

with the Einstein summation convention being employed, one obtains from (5) for $x_\mu^*(t)$ the equations of motion

$$\dot{x}^*_\mu(t) = \varepsilon u^{*'}_\mu(\mathbf{x}^*(t), t), \tag{9}$$

subject to the initial condition that $x^*_\mu(0) = x_\mu(0)$. The velocity $u^{*'}_\mu = u^{*'}_\mu(\mathbf{x}^*(t), t)$ is defined by

$$u^{*'}_\mu = \left(\frac{\partial y}{\partial z}(t; \mathbf{x}^*(t))\right)^{-1}_{\mu k} u^{'}_k(\mathbf{y}(t; \mathbf{x}^*(t)), t). \tag{10}$$

The net result of the above transformation of variables is that a fluctuation equation for the transformed variable $\mathbf{x}^*$ has been obtained in which the terms on the right-hand side are $O(\varepsilon)$: *cf.* (9). For such a fluctuation equation a Fokker-Planck or diffusion equation has been derived by Stratonovich [25]. It evolves as the leading terms of an asymptotic expansion involving powers of ε considering times that are large compared to the correlation time of the stochastic process underlying the right-hand side of (9): [25, Sections 4.8 and 4.9]. Truncating terms of $O(\varepsilon^3)$ and larger, one can write the equation as (*cf.* [25, Equation 4.194]

$$\begin{aligned}\frac{\partial p^*}{\partial t} \; = \; & -\varepsilon \frac{\partial}{\partial x^*_\nu}(\left\langle u^{*'}_\nu \right\rangle p^*) \\ & -\varepsilon^2 \int_0^\infty \mathrm{d}\tau \frac{\partial}{\partial x^*_\nu}(\langle\langle \frac{\partial u^{*'}_\nu}{\partial x^*_\mu}(\mathbf{x}^*, t) u^{*'}_\mu(\mathbf{x}^*, t-\tau)\rangle\rangle p^*) \\ & +\varepsilon^2 \int_0^\infty \mathrm{d}\tau \frac{\partial^2}{\partial x^*_\nu \partial x^*_\mu}(\langle\langle u^{*'}_\nu(\mathbf{x}^*, t) u^{*'}_\nu(\mathbf{x}^*, t-\tau)\rangle\rangle p^*),\end{aligned} \tag{11}$$

where $\langle\langle\cdot\rangle\rangle$ denotes the cumulant. Equation (11) can be simplified to

$$\frac{\partial p^*}{\partial t} = \varepsilon^2 \frac{\partial}{\partial x^*_\nu}\left(\left(\int_0^\infty \mathrm{d}\tau \left\langle u^{*'}_\nu(\mathbf{x}^*, t) \frac{\partial}{\partial x^*_\mu}(u^{*'}_\mu(\mathbf{x}^*, t-\tau)\right\rangle p^*)\right)\right), \tag{12}$$

where $p^* = p^*(\mathbf{x}^*, t)$ is transient probability density distribution; *i.e.*, the probability that $\mathbf{x}^*(t)$ has a value between $\mathbf{x}^*$ and $\mathbf{x}^* + \mathrm{d}\mathbf{x}^*$ is $p^*(\mathbf{x}^*, t)\mathrm{d}\mathbf{x}^*$. In the step from (11) to (12) use has been made of the property that $u^{*'}_\mu$ has zero mean. It is noted that statistical averages on the right-hand side of (12) are taken with $\mathbf{x}^*$ fixed, *i.e.*, $\mathbf{x}^*(t) \equiv \mathbf{x}^*$.

Equation (12) for $p^*(\mathbf{x}^*, t)$ can be transformed into a Fokker-Planck equation for $p(\mathbf{x}, t)$ using relationships between x^*_ν and x_ν

$$x_\nu(\mathbf{x}^*, t) = x_\nu, \qquad x_\nu(\mathbf{x}^*, t-\tau) = x^{-\tau}_\nu. \tag{13}$$

Here, $\mathbf{x}^{-\tau}$ is the position of a fluid particle at time $t-\tau$ while being at $\mathbf{x}$ at time t when moving according to mean Eulerian fluid velocity. In accordance with (6) and (7) it is determined by the equations

$$\dot{x}^t_\nu = u^0_\nu(\mathbf{x}^t, t), \qquad x^0_\nu = x_\nu. \tag{14}$$

Probability densities are related to each other by

$$p^*(\mathbf{x}^*, t) = \left|\frac{\mathrm{d}\mathbf{x}}{\mathrm{d}\mathbf{x}^*}\right| p(\mathbf{x}, t), \tag{15}$$

where $|\mathrm{d}\mathbf{x}/\mathrm{d}\mathbf{x}^*|$ is the Jacobian of the co-ordinate transformation $\mathbf{x}^* \rightarrow \mathbf{x}$. Furthermore

$$\frac{\partial p^*}{\partial t} = \left|\frac{d\mathbf{x}}{d\mathbf{x}^*}\right| \left\{\frac{\partial p}{\partial t} + \frac{\partial}{\partial x_\nu}\left\{u_\nu^0(\mathbf{x}, t)p\right\}\right\}, \tag{16}$$

and

$$\left|\frac{d\mathbf{x}}{d\mathbf{x}^*}\right| = \left|\frac{d\mathbf{x}}{d\mathbf{x}^{-\tau}}\right| \left|\frac{d\mathbf{x}^{-\tau}}{d\mathbf{x}^*}\right|, \tag{17}$$

$$\frac{\partial}{\partial x_j^*}\left\{\frac{\partial x_j^*}{\partial x_k} u_k^{'}(\mathbf{x}, t)\left|\frac{d\mathbf{x}}{d\mathbf{x}^*}\right|\right\} = \left|\frac{d\mathbf{x}}{d\mathbf{x}^*}\right| \frac{\partial u_l^{'}(\mathbf{x}, t)}{\partial x_l}, \tag{18}$$

so that

$$\begin{aligned} &\left|\frac{d\mathbf{x}^*}{d\mathbf{x}}\right| \frac{\partial}{\partial x_\nu^*}\left\{u_\nu^{*'}(\mathbf{x}^*, t)\frac{\partial}{\partial x_\mu^*}\left\{u_\mu^{*'}(\mathbf{x}^*, t-\tau)\left|\frac{d\mathbf{x}}{d\mathbf{x}^*}\right|\right\}\right\} \\ &= \left|\frac{d\mathbf{x}^*}{d\mathbf{x}}\right| \frac{\partial}{\partial x_\nu^*}\left\{\frac{\partial x_\nu^*}{\partial x_i} u_i^{'}(\mathbf{x}, t)\frac{\partial}{\partial x_j^*}\left\{\frac{\partial x_j^*}{\partial x_\mu^{-\tau}} u_\mu^{'}(\mathbf{x}^{-\tau}, t-\tau)\left|\frac{d\mathbf{x}}{d\mathbf{x}^*}\right|\right\}\right\} \\ &= \frac{\partial}{\partial x_m}\left\{\left|\frac{d\mathbf{x}^{-\tau}}{d\mathbf{x}}\right| u_m^{'}(\mathbf{x}, t)\frac{\partial}{\partial x_n^{-\tau}}\left\{u_n^{'}(\mathbf{x}^{-\tau}, t-\tau)\left|\frac{d\mathbf{x}}{d\mathbf{x}^{-\tau}}\right|\right\}\right\} \end{aligned} \tag{19}$$

Upon substituting the above relations in (12) one obtains for $p = p(\mathbf{x}, t)$ the equation

$$\begin{aligned} \frac{\partial p}{\partial t} = &-\frac{\partial}{\partial x_\nu}(u_\nu^0(\mathbf{x}, t)p) \\ &+\varepsilon^2\frac{\partial}{\partial x_\nu}\left(\int_0^\infty d\tau\left\langle u_\nu^{'}(\mathbf{x}, t)\left|\frac{d\mathbf{x}^{-\tau}}{d\mathbf{x}}\right| \frac{\partial}{\partial x_\mu^{-\tau}}\left(u_\mu^{'}(\mathbf{x}^{-\tau}, t-\tau)\right\rangle\left|\frac{d\mathbf{x}}{d\mathbf{x}^{-\tau}}\right| p\right)\right) \end{aligned} \tag{20}$$

Equation (20) is the Fokker-Planck equation for the probability density of particle position in inhomogeneous unsteady turbulent flow of the type described by (1)-(5). The equation has been arrived at starting from the Lagrangian specification of the position of a marked fluid particle: *cf.* (5). The derivation involves the description of the random displacements relative to coordinates moving with the mean flow field: *cf.* (6) and (7). In this way Stratonovich's formulation [25] of the diffusion or Fokker-Planck equation becomes applicable. This formulation is limited to stochastic processes in which values of both mean and fluctuating components remain small over time periods of correlation: *cf.* (9). Upon retransforming to fixed coordinates using (13)–(19), Stratonovich's formulation has been extended to the more general case of stochastic processes with large mean values. The extension is of direct relevance to applications of turbulent fluid flow where undisturbed mean velocities are generally large. The Fokker-Planck equation (20) will be reduced to a more elementary form in the next section.

An alternative derivation of the Fokker-Planck equation has been given by Van Kampen [24, 29]. His derivation can be described in a sketchy way as follows. Instead of starting from the Lagrangian specification of particle position, his derivation starts from conservation of admixture. For any particular realization of the random turbulent flow field the concentration field of admixture $\psi = \psi(\mathbf{x}, t)$ in regions not containing the admixture sources is given by the equation

$$\frac{\partial \psi}{\partial t} = -\frac{\partial}{\partial x_\nu}\left\{u_\nu(\mathbf{x}, t)\psi\right\}, \tag{21}$$

subject to suitably chosen boundary conditions [12, Section 10.1]. The mean concentration field is related to the probability density distribution $p(\mathbf{x}, t)$ of passive tracer or marked fluid particles as

$$\langle \psi \rangle = p(\mathbf{x}, t), \tag{22}$$

where as before angled brackets denote ensemble average. Equation (21) is a linear differential equation for ψ with randomly fluctuating coefficients $u_\nu(\mathbf{x}, t)$. It can also be written as

$$\dot{\psi} = A\psi, \tag{23}$$

where the dot denotes differentiation with respect to time and $A = A(\mathbf{x}, t)$ is a stochastic operator defined as

$$A(x, t)\psi = -\frac{\partial}{\partial x_\nu} \{u_\nu(\mathbf{x}, t)\psi\}. \tag{24}$$

When (23) is treated as an ordinary fluctuation equation and any dependency on $\mathbf{x}$, $\psi = \psi(t)$, $A = A(t)$, is disregarded, it is possible to derive an explicit expression for the ensemble average of ψ [24, 26–28]. The expression involves a perturbation expansion with the magnitude of the fluctuations as small parameter. The coefficients in the expansion are the cumulants of $A(t)$. When (24) is adopted the result can be transformed into a spatially dependent diffusion-type equation for $p(\mathbf{x}, t)$: see [29, Equation 19.6] and [24, Equation XVI (5.11)]. The equation is equal to (20), except that in Van Kampen's derivation attention has been confined to stationary stochastic processes $u'_\nu(\mathbf{x}, t)$. In this connection it is noted that turbulence behind a grid is an example of a non-stationary process when viewed from an observer who moves with the mean flow.

3. Elementary forms of the Fokker-Planck equation and derivation of the gradient hypothesis

The Fokker-Planck equation derived so far, *viz.* (20), is rather exotic in appearance. To reduce the equation to a more familiar form write (20) in the alternative form

$$\begin{aligned} \frac{\partial p}{\partial t} = &-\frac{\partial}{\partial x_\nu}\left(\left(u^0_\nu(\mathbf{x}, t) + \varepsilon^2 \int_0^\infty \mathrm{d}\tau \left\langle \frac{\partial u'_\nu(\mathbf{x}, t)}{\partial x^{-\tau}_\mu} u'_\mu(\mathbf{x}^{-\tau}, t-\tau)\right\rangle\right) p\right) \\ &+\varepsilon^2 \frac{\partial}{\partial x_\nu}\left(\int_0^\infty \mathrm{d}\tau \left|\frac{\mathrm{d}\mathbf{x}^{-\tau}}{d\mathbf{x}}\right| \frac{\partial}{\partial x^{-\tau}_\mu}\left(\left\langle u'_\nu(\mathbf{x}, t) u'_\mu(\mathbf{x}^{-\tau}, t-\tau)\right\rangle \left|\frac{\mathrm{d}\mathbf{x}}{\mathrm{d}\mathbf{x}^{-\tau}}\right| p\right)\right). \end{aligned} \tag{25}$$

Employing (18), one can show that

$$\begin{aligned} &\left|\frac{\mathrm{d}\mathbf{x}^{-\tau}}{\mathrm{d}\mathbf{x}}\right| \frac{\partial}{\partial x^{-\tau}_\mu}\left(\left\langle u'_\nu(\mathbf{x}, t) u'_\mu(\mathbf{x}^{-\tau}, t-\tau)\right\rangle \left|\frac{\mathrm{d}\mathbf{x}}{\mathrm{d}\mathbf{x}^{-\tau}}\right| p\right) = \\ &= \frac{\partial}{\partial x_\mu}\left(\frac{\partial x_\mu}{\partial x^{-\tau}_i}\left\langle u'_\nu(\mathbf{x}, t) u'_i(\mathbf{x}^{-\tau}, t-\tau)\right\rangle p\right). \end{aligned} \tag{26}$$

Furthermore,

$$\left\langle \frac{\partial u_\nu'(\mathbf{x},t)}{\partial x_\mu^{-\tau}} u_\mu'(\mathbf{x}^{-\tau}, t-\tau) \right\rangle = \frac{\partial}{\partial x_\mu}\left(\frac{\partial x_\mu}{\partial x_i^{-\tau}} \left\langle u_\nu'(\mathbf{x},t) u_i'(\mathbf{x}^{-\tau}, t-\tau)\right\rangle \right) - u_\nu'(\mathbf{x},t) \frac{\partial u_\mu'(\mathbf{x}^{-\tau}, t-\tau)}{\partial x_\mu^{-\tau}} \tag{27}$$

When (26) and (27) are implemented into the right-hand side of (25), the Fokker-Planck equation takes the form

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial x_\nu}\left(\left(u_\nu^0 + \varepsilon^2 c_\nu\right) p \right) + \varepsilon^2 \frac{\partial}{\partial x_\nu}\left(K_{\nu\mu} \frac{\partial p}{\partial x_\mu} \right). \tag{28}$$

where $c_\nu = c_\nu(\mathbf{x}, t)$ is drift due to compressibility defined as

$$c_\nu = -\int_0^\infty \mathrm{d}\tau \left\langle u_\nu'(\mathbf{x},t) \frac{\partial u_\mu'(\mathbf{x}^{-\tau}, t-\tau)}{\partial x_\mu^{-\tau}} \right\rangle, \tag{29}$$

while $K_{\nu\mu} = K_{\nu\mu}(\mathbf{x}, t)$ is a general diffusivity tensor defined as

$$K_{\nu\mu} = \int_0^\infty \mathrm{d}\tau \frac{\partial x_\mu}{\partial x_i^{-\tau}} \kappa_{\nu i}, \tag{30}$$

with $\kappa_{\nu i} = \kappa_{\nu i}(\mathbf{x}, t, \tau)$ being space-time Eulerian correlation:

$$\kappa_{\nu i} = \left\langle u_\nu'(\mathbf{x},t) u_i'(\mathbf{x}^{-\tau}, t-\tau) \right\rangle; \tag{31}$$

i.e., the time correlation of Eulerian fluid velocities measured in a frame that moves with the mean Eulerian fluid velocity (*cf.* (28)). The drift term in (28) represents the effect of compressibility. For incompressible flow

$$\frac{\partial u_i'(\mathbf{x},t)}{\partial x_i} = 0, \tag{32}$$

so that $c_\nu = 0$. The result is that Fokker-Planck equation (28) reduces to a conventional convection-diffusion equation (see also [29, Equation 19.9])

$$\frac{\partial p}{\partial t} = -u_\nu^0 \frac{\partial p}{\partial x_\nu} + \varepsilon^2 \frac{\partial}{\partial x_\nu}(K_{\nu\mu} \frac{\partial p}{\partial x_\mu}) \tag{33}$$

The equation holds for inhomogeneous non-uniform turbulent flows for which $\varepsilon \ll 1$ and for which the incompressibility assumption is justified, *i.e.*, for flows involving velocities which are less than the velocity of sound such that the square of the Mach number is much smaller than unity.

The above result shows that for (almost) incompressible flow and provided that $\varepsilon \ll 1$ the phenomenological description of dispersion due to turbulent fluctuations can be described analogous to that of molecular diffusion. The diffusion constant $K_{\nu\mu}$ is a tensor whose elements are determined by space-time correlations of Eulerian-based fluid velocities. In the expression for $K_{\nu\mu}$, account is taken of inhomogeneity of the fluctuating flow field through the term $K_{\nu\mu}$ in (30), and non-uniformity of the mean flow field through the term $(\partial/\partial x_i^{-\tau})x_\mu$ in (30). In regions of (almost) uniform mean flow, *e.g.*, in the centre of a pipe, or for diffusivities $K_{\nu\mu}$ in which the value of μ coincides with a direction perpendicular to the mean flow one can take $(\partial/\partial x_i^{-\tau})x_\mu = \delta_{i\mu}$ where $\delta_{i\mu}$ is the Kronecker delta, so that (30) reduces to

$$K_{\nu\mu} = \int_0^\infty \mathrm{d}\tau \kappa_{\nu\mu}. \tag{34}$$

That is, the diffusion tensor is directly determined by the time correlations of Eulerian velocities in corresponding directions assessed in a frame that moves with the mean velocity. The term $(\partial/\partial x_i^{-\tau})x_\mu$ in (30) is unequal to $\delta_{i\mu}$ in case of non-uniform mean flow and values of μ which coincide with the direction of the mean flow. In that case the value of $K_{\nu\mu}$ for any particular ν and μ depends on correlations between fluctuating velocities in directions other than those corresponding to the values μ taken in evaluating $K_{\nu\mu}$. The factor $(\partial/\partial x_i^{-\tau})x_\mu$ quantifies both dispersion by Eulerian velocity fluctuations in corresponding directions and dispersion by changes in mean velocity when following a marked particle through non-uniform mean flow. Interestingly, (34), that is the expression for the diffusivity which holds when $(\partial/\partial x_i^{-\tau})x_\mu = \delta_{i\mu}$, is in line with previously made and largely empirically based proposals which started with Burgers's idea [30] to approximate Lagrangian-based correlations by Eulerian correlations in a frame that moves with the mean velocity. Using such correlations to determine particle dispersion in almost uniform mean flow was further brought forward in]31–35], including some preliminary experimental results for fully developed turbulent flow in the interior of a pipe in support of it. Experimental investigations on space-time correlations have further been reported in [36–38].

Substituting the expression for u_ν^0 given by (1) in (21) and taking the mean, one obtains for conservation of admixture in mean sense the equation

$$\frac{\partial \langle \psi \rangle}{\partial t} = -\frac{\partial \left(u_\nu^0 \langle \psi \rangle\right)}{\partial x_\nu} - \varepsilon \frac{\partial \left\langle u_\nu' \psi \right\rangle}{\partial x_\nu}. \tag{35}$$

Noting that $\langle \psi \rangle = p$, the above equation combined with (28) yields for $\left\langle u_\nu' \psi \right\rangle$ the relation

$$\left\langle u_\nu' \psi \right\rangle = \varepsilon c_\nu - \varepsilon K_{\nu\mu} \frac{\partial \langle \psi \rangle}{\partial x_\mu}, \tag{36}$$

which for (almost) incompressible flow reduces to

$$\left\langle u_\nu' \psi \right\rangle = -\varepsilon K_{\nu\mu} \frac{\partial \langle \psi \rangle}{\partial x_\mu}. \tag{37}$$

With this result the scalar gradient hypothesis has been obtained. It is of tensor form with the coefficients defined by (30). The result is valid under the same conditions as those identified for the convection-diffusion equation, *i.e.*, $\varepsilon \ll 1$ and incompressible flow. As for low-Mach-number-flow temperature fluctuations and admixture concentration are described by the same equation [12, Section 1.5], the above result also confirms under the same conditions the gradient hypothesis for temperature with $K_{\nu\mu}$ defined by (30) being turbulent thermal diffusivity.

4. Lagrangian-Eulerian statistical relationships

A common procedure is to postulate the Fokker-Planck equation under the assumption that evolution times are large compared to the correlation time of the underlying stochastic process. The coefficients of the equation are given in terms of coarse-grained Lagrangian-based statistical summaries of the process. The objective of the present analysis is to relate this version of the Fokker-Planck equation with the Fokker-Planck equation with Eulerian-based coefficients derived in the previous sections. For this purpose asymptotic expansions based on powers of ε are developed which describe Lagrangian-based statistical properties in terms of fixed-point velocity statistics in a frame that moves with the mean Eulerian velocity. In this way it will be shown that for the conditions under which the diffusion limit holds, *i.e.*, $\varepsilon \ll 1$, Lagrangian and Eulerian-based versions of diffusion theory are fully compatible with each other. Later (in Section 5) it will be shown that for turbulent flow $\varepsilon = \mathcal{O}(1)$: *i.e.*, the conditions under which the diffusion limit holds ($\varepsilon \ll 1$) are not satisfied. The importance of the presented Lagrangian-Eulerian statistical relationships is that, though inaccurate when $\varepsilon = \mathcal{O}(1)$, they still hold to order of magnitude. Hence, they provide the possibility to estimate orders of magnitude of Lagrangian-based statistical parameters by their better assessable and measurable Eulerian counterparts. Another important conclusion to be derived from the present results is that the conditions of validity of diffusion theory are the same for both the Eulerian and the Lagrangian representations. The presented relationships enable the Eulerian formulation to be transformed into the Lagrangian formulation. The latter is subject to the same truncation error as the former, which is assessed in Section 5.

Given the fluctuation equation (9), the evolution of the displacement variable x_μ^* starting from the initial condition $x_\mu^*(0)$ at $t = 0$ can be described by the expansion [25, Equation 4.187]:

$$
\begin{aligned}
x_\mu^*(t) &= x_\mu^*(0) + \varepsilon \int_0^t u_\mu^{*'}(\mathbf{x}_0^*, t_1)\mathrm{d}t_1 \\
&\quad + \varepsilon^2 \int_0^t \mathrm{d}t_1 \frac{\partial u_\mu^{*'}}{\partial x_m^*}(\mathbf{x}_0^*, t_1) \int_0^{t_1} u_m^{*'}(\mathbf{x}_0^*, t_2)\mathrm{d}t_2 + O(\varepsilon^3)
\end{aligned}
\tag{38}
$$

where $\mathbf{x}_0^* = \mathbf{x}^*(0)$ .The above expansion leads to expressions for $x_\mu^*(t)$ which are based on fixed-point statistics. Upon transformion to the inertial frame, it leads to expressions based on Eulerian velocity statistics in a frame that moves with the mean Eulerian velocity. Differentiation of (38) with respect to time yields

$$
\dot{x}_\mu^*(t) = \varepsilon u_\mu^{*'}(\mathbf{x}_0^*, t) + \varepsilon^2 \frac{\partial u_\mu^{*'}(\mathbf{x}_0^*, t)}{\partial x_{m0}^*} \int_0^t u_m^{*'}(\mathbf{x}_0^*, t_1)\mathrm{d}t_1 + O(\varepsilon^3) \tag{39}
$$

which can be used to derive an expansion for fluctuating particle velocity in the fixed frame as follows

$$
\begin{aligned}
&u_\nu^{'}(\mathbf{x}(t), t) = \varepsilon^{-1}\dot{x}_\mu^*(t)\frac{\partial y_\nu}{\partial z_\mu}(t; \mathbf{x}^*(t)) = \frac{\partial y_\nu}{\partial z_\mu}(t; \mathbf{x}^*(t)) u_\mu^{*'}(\mathbf{x}_0^*, t) \\
&+\varepsilon \frac{\partial y_\nu}{\partial z_\mu}(t; \mathbf{x}^*(t)) \frac{\partial u_\mu^{*'}(\mathbf{x}_0^*, t)}{\partial x_{m0}^*} \int_0^t u_m^{*'}(\mathbf{x}_0^*, t_1)\mathrm{d}t_1 + O(\varepsilon^2) = \frac{\partial y_\nu}{\partial z_\mu}(t; \mathbf{x}_0^*) u_\mu^{*'}(\mathbf{x}_0^*, t)
\end{aligned}
\tag{40}
$$

$$+\varepsilon \frac{\partial}{\partial x^*_{m0}} \left(\frac{\partial y_\nu}{\partial z_\mu}(t; \mathbf{x}^*_0) u^{*'}_\mu(\mathbf{x}^*_0, t) \right) \int_0^t u^{*'}_m(\mathbf{x}^*_0, t_1) \mathrm{d}t_1 + O(\varepsilon^2) = u'_\nu(\mathbf{y}(t; \mathbf{x}^*_0), t)$$

$$+\varepsilon \frac{\partial u'_\nu(\mathbf{y}(t; \mathbf{x}^*_0), t)}{\partial x^*_{m0}} \int_0^t \left(\frac{\partial y}{\partial z}(t_1, \mathbf{x}^*_0) \right)^{-1}_{mk} u'_k(\mathbf{y}(t_1; \mathbf{x}^*_0), t_1) \mathrm{d}t_1 + O(\varepsilon^2)$$

$$= u'_\nu(\mathbf{x}^t_0, t) + \varepsilon \int_0^t \frac{\partial u'_\nu(\mathbf{x}^t_0, t)}{\partial x^\sigma_{k0}} u'_k(\mathbf{x}^\sigma_0, \sigma) \mathrm{d}\sigma + O(\varepsilon^2)$$

where application has been made of (6)–(10) and (13)–(14) and where $\mathbf{x}^t_0$ is position at time t while being at $\mathbf{x}_0$ at $t = 0$ moving with the mean Eulerian velocity. The above result can be generalized to any turbulent flow quantity $G_\mu(\mathbf{x}(t), t) = G_\mu(\mathbf{x}(t), t; \delta(t))$ which depends on $\mathbf{x}(t)$ and t with $\mathbf{x}(t)$ being given by (5) and on the stochastic process $\delta(t)$ underlying the fluctuating velocity field as

$$G_\mu(\mathbf{x}(t), t) = G_\mu(\mathbf{x}^t_0, t) + \varepsilon \int_0^t \frac{\partial G_\mu(\mathbf{x}^t_0, t)}{\partial x^\sigma_{k0}} u'_k(\mathbf{x}^\sigma_0, \sigma) \mathrm{d}\sigma + O(\varepsilon^2) \tag{41}$$

Taking the mean, one has

$$\left\langle G_\mu(\mathbf{x}(t), t) \right\rangle = \left\langle G_\mu(\mathbf{x}^t_0, t) \right\rangle + \varepsilon \int_0^t \left\langle \frac{\partial G_\mu(\mathbf{x}^t_0, t)}{\partial x^\sigma_{k0}} u'_k(\mathbf{x}^\sigma_0, \sigma) \right\rangle \mathrm{d}\sigma + O(\varepsilon^2) \tag{42}$$

Invoking $\mathbf{x}^t_0 = \mathbf{x}_1$ and $\sigma = t - \tau$ so that $\mathbf{x}^\sigma_0 = \mathbf{x}^{-\tau}_1$, one obtains

$$\left\langle G_\mu(\mathbf{x}(t), t) \right\rangle = \left\langle G_\mu(\mathbf{x}, t) \right\rangle + \varepsilon \int_0^t \left\langle \frac{\partial G_\mu(\mathbf{x}, t)}{\partial x^{-\tau}_k} u'_k(\mathbf{x}^{-\tau}, t - \tau) \right\rangle \mathrm{d}\tau + O(\varepsilon^2) \tag{43}$$

where $\mathbf{x}_1$ naturally apprearing in the terms on the right-hand side of this equation has been replaced by $\mathbf{x}$ on the grounds that this causes an error of $O(\varepsilon^2)$ only. As follows from (41), $\mathbf{x}$ and $\mathbf{x}_1$ differ by $O(\varepsilon)$ for periods of time during which there is correlation between particle velocities. Replacing $\mathbf{x}_1$ by $\mathbf{x}$ in the second term on the right-hand side therefore leads to an error of $O(\varepsilon^2)$ only. Furthermore, as u'_k is zero-mean mean values of functions $G_\mu(\mathbf{x}^t_0, t)$ differ $O(\varepsilon^2)$ from $G(\mathbf{x}, t)$: This follows from applying (41) to $\left\langle G_\mu(\mathbf{x}, t) \right\rangle$ instead of to $G_\mu(\mathbf{x}(t), t)$, that is assessing the evolution of the Eulerian mean $\left\langle G_\mu(\mathbf{x}, t) \right\rangle$, while following the path of a marked particle. When mean of left and right-hand side are taken, *i.e.*, the ensemble average of all Eulerian mean values then leads to the conclusion that $\left\langle G_\mu(\mathbf{x}^t_0, t) \right\rangle$ and $\left\langle G_\mu(\mathbf{x}, t) \right\rangle$ are equal to $O(\varepsilon^2)$. While (42) is suited to assess the evolution of statistical properties of $G_\mu(\mathbf{x}(t), t)$ with time for particles which start at position $\mathbf{x}_0$ at $t = 0$ moving in various directions, (43) is convenient to determine Lagrangian-based statistical properties of $G_\mu(\mathbf{x}(t), t)$ valid at position $\mathbf{x}$ at time t.

For stochastic processes including turbulent flow it is usually assumed that the correlation between stochastic variables rapidly decreases for $t > \tau_c$. Describing stochastic variables in the diffusion region, that is for times much larger than the correlation time, it enables to replace the upper bound t in the integral by ∞, so that

$$\left\langle G_\mu(\mathbf{x}(t),t)\right\rangle = \left\langle G_\mu(\mathbf{x},t)\right\rangle + \varepsilon \int_0^\infty \left\langle \frac{\partial G_\mu(\mathbf{x},t)}{\partial x_k^{-\tau}} u_k^{'}(\mathbf{x}^{-\tau}, t-\tau)\right\rangle \mathrm{d}\tau + O(\varepsilon^2) \tag{44}$$

This result is a generalisation of Stratonovich's analogous expansion for a stochastic variable described by a one-dimensional fluctuation equation with small random right-hand side: Equation (4.167) of [25]. It is the extension to stochastic variables described by systems of fluctuation equations with large mean values: *cf.* (5). While the first term on the right-hand side of (44) represents the mean of $G_\mu(\mathbf{x}(t),t)$ disregarding correlation between $\mathbf{x}(t)$ and the underlying stochastic process, the second term represents a correction due to correlation. The result is due to applying to $G_\mu(\mathbf{x}(t+\Delta t), t+\Delta t)$ the limit process $\Delta t \gg \tau_c$ prior to $\Delta t \to 0$: That is, applying coarse graining appropriate for the diffusion limit omitting short-time behavior corresponding to the time-scale τ_c and then letting $\Delta t \to 0$. It leads to the appearance of the correction term on the right-hand side of (44). This term would be absent when averaging properties of particles being at position $\mathbf{x}$ at time t without coarse graining. The term describes the situation shortly after marking when transients corresponding to the time-scale τ_c have vanished while the particle is still in the region where the same statistical properties apply. Relation (44) is exploited in subsequent parts of this section to express Lagrangian-based statistical representations appropriate for the diffusion limit in terms of Eulerian velocity statistics in a frame that moves with the mean Eulerian velocity.

The diffusion approximation applies to times after marking such that $t \gg \tau_c$. For times shortly after marking such that $t \sim \tau_c$, replacing t by ∞ in the integrals on the right-hand sides of the above equations leads to relative errors of unit order of magnitude in statistical parameters of particle dispersion. With increasing time after marking random particle displacement extends over larger and larger distances whereas the absolute error due to the diffusion approximation approaches a constant value. The situation is qualitatively not different from that of turbulent spread in homogeneous isotropic turbulence [6]. Applying the diffusion approximation the relative error in the prediction of variables like the variance of particle displacement is $O(\tau_c/t)$: That is, $O(\varepsilon)$ for $t \sim \varepsilon^{-1}\tau_c$, and $O(\varepsilon^2)$ for $t \sim \varepsilon^{-2}\tau_c$, which corresponds to the time scale where particle dispersion has extended to the external length scale. Although shortly after marking the error may be of appreciable magnitude compared to the actual spread at that time, it is small compared to the ultimate spread occurring at large times. If one is primarily interested in large scale diffusion occurring at long times the diffusion approximation can be applied without regard of short-time behaviour. If on the other hand one is interested in accurate predictions shortly after marking, the diffusion approximation may be too inaccurate. In that case one can resort to descriptions for particle dispersion starting from the fluctuation equations for particle velocity [18–20].

Averaging left and right-hand side of (5), taking $\left\langle u_\nu^0(\mathbf{x}(t),t)\right\rangle = u_\nu^0(\mathbf{x},t)$ and applying expansion (44) to evaluate $\left\langle u_\nu^{'}(\mathbf{x}(t),t)\right\rangle$, one obtains for the mean value of the velocity of fluid particles being at position $\mathbf{x}$ at time t the expression:

$$\langle \dot{x}_\nu(t)\rangle = u_\nu^0(\mathbf{x},t) + \varepsilon^2 \int_0^\infty \mathrm{d}\tau \left\langle \frac{\partial u_\nu^{'}(\mathbf{x},t)}{\partial x_\mu^{-\tau}} u_\mu^{'}(\mathbf{x}^{-\tau}, t-\tau)\right\rangle + O(\varepsilon^3) \tag{45}$$

The second term on the right-hand side of this equation represents drift. It is $O(\varepsilon^2)$ and describes the additional mean motion a particle is subjected to shortly after marking. As this term is of the same order of magnitude as the diffusion terms, it is to be included when relating the Fokker-Planck equation with Eulerian-based coefficients to the Fokker-Planck equation with coarse-grained Lagrangian-based coefficients. For incompressible flow (45) reduces to, to $O(\varepsilon^3)$,

$$\langle \dot{x}_\nu(t)\rangle = u^0_\nu(\mathbf{x}, t) + \varepsilon^2 \frac{\partial K_{\nu\mu}(\mathbf{x}, t)}{\partial x_\mu}, \tag{46}$$

where $K_{\nu\mu}(\mathbf{x}, t)$ is Eulerian-based diffusivity defined by (30). For the theoretical abstraction of homogeneous turbulence, the second term vanishes and mean values of Eulerian and Lagrangian velocities become equal. It is in line with the general result that for homogeneous turbulence in incompressible flow Eulerian and Lagrangian-based velocity statistics are equal [39, 12].

Applying coarse graining, a Lagrangian-based diffusivity $K^L_{\nu\mu} = K^L_{\nu\mu}(\mathbf{x}, t)$ can be defined along the lines of Kolmogorov as: (see Equation (10.53) of [12]),

$$K^L_{\nu\mu} = \tfrac{1}{2} \left\langle\!\left\langle \dot{\overline{x_\nu(t) x_\mu(t)}} \right\rangle\!\right\rangle . \tag{47}$$

Making use of

$$\begin{aligned} \left\langle\!\left\langle \dot{x}_\nu(t) x_\mu(t) \right\rangle\!\right\rangle = {} & \left\langle\!\left\langle u'_\nu(\mathbf{x}(t), t) x_\mu(t) \right\rangle\!\right\rangle = \\ & \left\langle u'_\nu(x(t), t) x_\mu(t) \right\rangle - \left\langle u'_\nu(x(t), t) \right\rangle x_\mu \end{aligned} \tag{48}$$

and applying expansion (44) to evaluate the terms on the right-hand side of this equation one finds

$$K^L_{\nu\mu} = \tfrac{1}{2}\varepsilon^2 (K_{\nu\mu} + K_{\mu\nu}) + O(\varepsilon^3). \tag{49}$$

It shows that apart from the multiplicative factor ε^2, with a relative error of $O(\varepsilon)$ the coarse-grained Lagrangian-based diffusivity and the symmetrical part of the Eulerian-based diffusivity are equal to each other.

Employing (26), (45), (49), (30) and (31), the Fokker-Planck equation (25) can be expressed with a relative error of $O(\varepsilon)$ as

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial x_\nu} (\langle \dot{x}_\nu \rangle\, p) + \frac{\partial^2}{\partial x_\nu \partial x_\mu} (K^L_{\nu\mu} p). \tag{50}$$

This is the conventional Kolmogorov formulation of the Fokker-Planck equation in which the coefficients are based on coarse-grained Lagrangian-based particle statistics. It shows that within the order of approximation for which the diffusion limit holds, the conventional formulation and the version with Eulerian-based coefficients are equal. Provided $\varepsilon \ll 1$ representations appropriate for the diffusion limit which are based on coarse-grained Lagrangian statistics are equivalent to those based on Eulerian statistics in a moving frame.

Another remarkable point is that the Lagrangian-based formulation of the Fokker-Planck equation as given by (50) does not automatically lead to the convection-diffusion equation and the scalar gradient hypothesis which forms the basis of the phenomenological theories of conservation of mass and heat. Only if one presumes the relation

$$\langle \dot{x}_\nu \rangle = \langle u_\nu \rangle + \frac{\partial K^L_{\nu\mu}}{\partial x_\mu}, \tag{51}$$

(50) can be converted to a convection-diffusion equation and thereby to the scalar gradient hypothesis. The Eulerian-based versions of the coefficients reveal the existence of such a relation, *i.e.* (46), and it is found to be valid only in case of incompressible flow.

5. Validity of diffusion theory in turbulent fluid flow

The Fokker-Planck equation and the convection diffusion equation and gradient hypothesis derived from it are the result of truncation of an asymptotic expansion involving powers of ε. The error due to truncation can be assessed by considering the terms of $O(\varepsilon^3)$ so far omitted in (20). For this purpose use is made of Van Kampen's and Terwiel's cumulant expansion [26–28] associated with ordinary fluctuation equation (23). Adopting their expression for the third cumulant of the ordinary process of (23) and transforming this to the present spatial problem using (24), one obtains for the terms of $O(\varepsilon^3)$ to be added to the right-hand side of Fokker-Planck Equation (20) the expression

$$-\varepsilon^3 \frac{\partial}{\partial x_\nu} \int_0^\infty \mathrm{d}\tau \int_0^\infty \mathrm{d}\sigma \left\langle u'_\nu(\mathbf{x}, t) \left| \frac{\mathrm{d}\mathbf{x}^{-\tau}}{d\mathbf{x}} \right| \frac{\partial}{\partial x_\mu^{-\tau}} u'_\mu(\mathbf{x}^{-\tau}, t-\tau) \right. \\ \left. \left| \frac{\mathrm{d}\mathbf{x}^{-\tau-\sigma}}{\mathrm{d}\mathbf{x}^{-\tau}} \right| \frac{\partial}{\partial x_k^{-\tau-\sigma}} u'_k(\mathbf{x}^{-\tau-\sigma}, t-\tau-\sigma) \right\rangle \left| \frac{\mathrm{d}\mathbf{x}}{\mathrm{d}\mathbf{x}^{-\tau-\sigma}} \right| p, \tag{52}$$

where it is noted that the operators $\partial/\partial x_\nu$, $\partial/\partial x_\mu^{-\tau}$ and $\partial/\partial x_k^{-\tau-\sigma}$ act on all terms behind them.

The terms in (52) consist of spatial derivatives higher than second order and involve triple and higher order correlations of fluid velocities and their derivatives. The error due to neglecting these terms in the Fokker-Planck equation can be assessed by comparing their magnitude with that of the second term on the right hand side of (20). It is noted that in the diffusion limit the magnitude of spatial derivatives of diffusional terms can be represented by L^{-1} where L corresponds to the external length scale of the flow. This characterisation is also applied to the derivatives of fluid velocities occurring in the triple correlation of (52). Furthermore it is noted that as a result of the normalisation of u'_ν by ε, the magnitude of u'_ν can be represented by U. One then finds that the magnitude of the third term in the expansion compares to the magnitude of the second term as $\varepsilon U \tau_c L^{-1}$ where τ_c is correlation time of the random process. The requirement $\varepsilon U \tau_c L^{-1} \ll 1$ is the same as requiring that the typical displacement due to fluctuations measured over a time period of correlation is small compared to the external length scale of the flow: *cf.* (3) and (4).

In case of the theoretical abstraction of stationary homogeneous turbulence in uniform mean flow statistical averages of the velocity field are constant in space. Lagrangian fluid particle velocities on the right hand side of (5) can be represented by quantities which vary randomly with time only. Solutions of the resulting linear stochastic differential equation can be approximated by those pertinent to the diffusion limit under the only and sufficient condition that $t \gg \tau_c$, viz. for times after marking much larger than the correlation time: Section 4.7 of [25]. For real turbulent flow, however, stochastic differential equation (5) is essentially nonlinear and the diffusion approximation only holds when in addition to $t \gg \tau_c$, $\varepsilon \ll 1$. The condition $t \gg \tau_c$ is well-known in diffusion approximations to turbulent flow: *e.g.*, Section 10.3 of [12] and [40, 41]. Conditions relating to ε, however, have not been noticed. For processes where Eulerian velocities and all their derivatives are Gaussian, cumulants higher than second order of velocities and their derivatives are equal to zero. It results in coefficients of derivatives higher than second order in the cumulant expansion leading to the Fokker-Planck equation which are all zero. Approximating the random process of particle dispersion by a continuous Markov process is then allowed if $t \gg \tau_c$ irrespective the magnitude of ε. But the assumption of Gaussianity of velocities and their derivatives does not hold for real turbulent flow. While velocities may be Gaussian, viscous-scale turbulence causes distributions of derivatives of velocities to be non-Gaussian including those for isotropic turbulence in uniform flow behind a grid: Section 8.1 of [8]. It implies that for the diffusion limit to hold in turbulent fluid flow under all circumstances ε must be of limited magnitude such that contributions of cumulants higher than second order can be disregarded. This conclusion equally applies to diffusion theory formulated in terms of coarse-grained Lagrangian-based statistics. Using the relationships of the previous section, Eulerian-based coefficients can be transformed into their Lagrangian-based counterparts. While doing so, the truncation error involved with the diffusion limit remains unaltered and still requires $\varepsilon \ll 1$. In this connection it is noted that also Planck's and Kolmogorov's original derivations of the Fokker-Planck equation for nonlinear stochastic processes implied the assumption of small or infinitely small jumps: VIII 1 of [24].

The diffusion approximation can be extended by one order in ε in case of approximately Gaussian velocity statistics and incompressible flow. Including the terms of $O(\varepsilon^3)$ given by (52) in the Fokker-Planck equation (20), using (32) and repeating some of the mathematical procedures of the previous sections, one finds as 'convection-diffusion equation' formulated to $O(\varepsilon^3)$ the equation

$$\begin{aligned}\frac{\partial p}{\partial t} = &-u_\nu^0 \frac{\partial p}{\partial x_\nu} + \varepsilon^2 \frac{\partial}{\partial x_\nu}\left((K_{\nu\mu} - \varepsilon K^1_{\nu\mu})\frac{\partial p}{\partial x_\mu}\right)\\ &-\varepsilon^3 \frac{\partial}{\partial x_\nu}(K_{\nu\mu\kappa}\frac{\partial^2 p}{\partial x_\mu \partial x_\kappa})\end{aligned} \tag{53}$$

where

$$K_{\nu\mu\kappa} = \int_0^\infty \int_0^\infty \mathrm{d}\sigma\, \mathrm{d}\tau \frac{\partial x_\mu}{\partial x_j^{-\tau}} \frac{\partial x_\kappa}{\partial x_i^{-\tau-\sigma}} \kappa_{\nu j i}, \tag{54}$$

and

$$\kappa_{\nu j i} = \left\langle u'_\nu(\mathbf{x}, t) u'_j(\mathbf{x}^{-\tau}, t-\tau) u'_i(\mathbf{x}^{-\tau-\sigma}, t-\tau-\sigma)\right\rangle. \tag{55}$$

while

$$\begin{aligned}K^1_{\nu\mu} = &\int_0^\infty \int_0^\infty \mathrm{d}\sigma\, \mathrm{d}\tau \left\langle u'_\nu(\mathbf{x}, t) \frac{\partial}{\partial x_j^{-\sigma}}\left\{\frac{\partial x_\mu}{\partial x_i^{-\tau-\sigma}} u'_i(\mathbf{x}^{-\tau-\sigma}, t-\tau-\sigma)\right\}\right.\\ &\left. u'_j(\mathbf{x}^{-\sigma}, t-\sigma)\right\rangle\end{aligned} \tag{56}$$

For Gaussian Eulerian velocity statistics triple correlation (55) is equal to zero. The third order derivative vanishes and (53) becomes an ordinary diffusion equation. The equation holds to $O(\varepsilon^3)$ provided the higher order diffusivity $K^1_{\nu\mu}$ defined by (56) is included. Similarly, the accuracy of the gradient hypothesis can be improved by one order in ε by replacing $K_{\nu\mu}$ by $K_{\nu\mu} - \varepsilon K^1_{\nu\mu}$ in (37). The higher order term is determined by triple time-correlations of velocities and derivatives of velocities in a frame that moves with the mean Eulerian velocity. Extension of the accuracy of the diffusion approximation by yet another order in ε, however, is not feasible. For Gaussian velocity statistics the diffusivity based on the fourth order cumulant will again be zero. But the coefficients of third order derivatives will contain fourth order cumulants in which derivatives of the velocity field occur similar to those in (42). In view of viscous scale turbulence such cumulants can not be expected to be zero or small. The equation can no longer be reduced to one with derivatives of second order as highest order. It is in accordance with the observation made previously that for the diffusion approximation to hold in turbulent fluid flow in all circumstances ε must be of limited magnitude such that the contributions of all cumulants involving velocities and their derivatives higher than second order can be disregarded.

A point not considered so far is the effect of small-scale or viscous-scale turbulence. Characteristic for turbulent flow is that the power spectra of stochastic variables reduce only slowly in magnitude with increasing frequency and wave-number up to the point where viscosity becomes important. It makes terms involving spatial derivatives of velocities apparent in (52) correspondingly large. At the same time fluctuations associated with small-scale turbulence correlate over very short times only. To assess the net effect use is made of Kolmogorov's scalings of length, time and velocity of small-scale turbulence (Equation (1.5.11) of [14]) thereby representing the mean energy dissipation rate by $\tilde{u}^2 U/L$. Orders of magnitude of small-scale and large-scale fluctuating velocity, time and length then compare to each other as $(\varepsilon^2\mathrm{Re})^{-1/4}$, $(\varepsilon^2\mathrm{Re})^{-1/2}$ and $(\varepsilon^2\mathrm{Re})^{-3/4}$, respectively, where Re is Reynolds number defined as $\mathrm{Re} = UL/\nu$ with ν being kinematic viscosity. Introducing these scalings in (52) and considering incompressible flow, only the order of magnitude of (52) based on small-scale turbulence compares to the order of magnitude of the diffusion term of (20) as $(\varepsilon^2\mathrm{Re})^{-1}$. Instead of using Kolmogorov scaling it may be considered more appropriate to assess the effect of small-scale turbulence on the basis of Tennekes' Eulerian-based scalings for small-scale turbulence taking account of large-scale advection of dissipative eddies [42]. The net effect is that the scaling of time reduces by a factor $(\varepsilon^2\mathrm{Re})^{-1/4}$. It all indicates that small-scale turbulence does not cause the presented expansions to break down as long as $\varepsilon^2\mathrm{Re}$ is sufficiently large. As in practical cases of turbulent flow Re is generally very large, the condition is generally satisfied. The validity of the diffusion theory is primarily determined by the magnitude of ε which represents the magnitude of the truncated terms according to large-scale turbulence.

Unfortunately, a limit process $\varepsilon \to 0$ by which the diffusion approximation would become exact does not seem to exist in turbulent fluid flow. For turbulence in wall-bounded shear flows such as turbulent boundary layers along walls and along the earth's surface and turbulent flows in pipes and channels the correlation time τ_c can be scaled according to $((\partial/\partial x_k)u_\nu^0)^{-1}$ which is the time by which fluid elements are deformed by the mean flow field. The same scaling is obtained under the assumption that the order of magnitudes of turbulent viscosities and diffusivities are the same and that the gradient hypothesis of momentum holds to order of magnitude: As the covariance of fluctuating velocities scales as $\tilde{u}^2$ and the diffusivity as $\tilde{u}^2\tau_c$, τ_c scales as $((\partial/\partial x_k)u_\nu^0)^{-1}$. Implementing Prandtl's velocity defect law appropriate for the core region of wall-bounded turbulent flow (Section XIXe of [43]) one finds $\tau_c \sim \kappa L u_*^{-1}$ where κ is von Karman's constant ($\kappa \sim 0.4$) and u_* is friction velocity. Noting that $u_* \sim \tilde{u}$ it follows from (4) that $\varepsilon \sim \kappa$. Although ε may be less than unity, it does not become vanishingly small in some limit process of parameters governing fluid flow, *e.g.*, $\mathrm{Re} \to \infty$. The same conclusion applies to the inertial sublayer. In this region gradients of the mean flow become steeper implying that correlation times become smaller. This effect is compensated by a corresponding decrease in size L of energetic eddies so that still $\varepsilon \sim \kappa$. In general it can be concluded that for wall-bounded turbulence a limit process by which $\varepsilon \to 0$ does not exist. It also implies that a limit process by which the truncated terms in the expansion leading to the gradient hypotheses vanish does not exist.

For turbulent jets, turbulent mixing layers and thermal plumes one may scale τ_c according to LU^{-1} where L is cross-sectional dimension and U magnitude of mean velocity producing the turbulence. Following (4) in this case $\varepsilon \sim \tilde{u}/U$ which is the ratio of fluctuating velocities to mean velocity. According to the results of measurements this ratio has values of typically one-fifth to one-tenth: chapter 4 of [14], chapter 6 of [13]. But also in these applications it does not seem possible to identify a limit process of flow parameters by which $\varepsilon \to 0$. Examples of flow where random displacements during periods of correlation are not small in comparison to the external length scale altogether are atmospheric turbulence within a crop or forest canopy [20], and turbulent flow in the convective boundary layer [44]. Also for far wake turbulence and turbulence behind a grid the limit $\varepsilon \to 0$ does not occur. In these applications mean velocity gradients decrease with distance from the obstacle producing the turbulence. At distances where mean velocities are almost uniform correlation times τ_c scale according to $L\tilde{u}^{-1}$ where $\tilde{u}$ corresponds to the magnitude of the fluctuating velocities. The result is that ε as defined by (4) is of unit order of magnitude and that the asymptotic procedure leading to the diffusion limit breaks down. Interestingly, the condition $t \gg \tau_c$ does not seem to hold here either. Seen from an observer which moves with the uniform mean flow, the turbulence decays with $\tau_c t^{-1}$. The limit process by which the diffusion limit is established is also the limit process by which the turbulence disappears.

6. Conclusions

The diffusion approximation leading to the Fokker-Planck equation for fluid particle position and the convection-diffusion equation and the scalar gradient hypothesis in case of incompressible flow is valid when $\varepsilon \ll 1$; that is, when displacements due to random fluid velocity measured over time periods where there exists correlation between velocities are small compared to the external length scale of the flow. For practical cases of turbulent flow, however $\varepsilon = \mathcal{O}(1)$: The limit process $\varepsilon \to 0$ by which the diffusion approximation becomes exact does not exist. Applying the concepts of diffusion theory resorts to employing approximate methods of analysis. This is what seems to happen in practice. Diffusion coefficients are treated as fit parameters which are assessed from case to case. They are determined by fitting experimental and theoretical results on the basis of resemblance of a specific output variable. As follows from considerations of higher order spatial derivatives being described by second order derivatives only, the predictive capacity of such a model will be limited when ε is not small. Good resemblance can only be expected to occur for the output variable for which the empirical model has been calibrated; and only under the conditions for which the calibration has taken place: same boundary conditions, same initial conditions and same flow configuration. Scaling rules, however, are not violated in such an approach. As ε is order unity, truncation errors are not small but become large neither. The effect of small-scale turbulence on the values of diffusion coefficients is small as the Reynolds number is generally large. The representation of coarse-grained Lagrangian statistics by Eulerian statistics in a moving frame is inaccurate for $\varepsilon = \mathcal{O}(1)$ and $t/\tau_c = \mathcal{O}(1)$, but correct to order of magnitude. It is within these constraints that diffusion theory can be applied as a semi-empirical concept or engineering approximation in the analysis of problems of turbulent flow. In the absence of a true limit process the theory cannot achieve the position it has obtained in the description of the macroscopic behaviour of molecular chaos.

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