

The vanishing effect of molecular diffusivity on turbulent dispersion: implications for turbulent mixing and the scalar flux

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In 1921 G. I. Taylor introduced (with little discussion) the notion that the dispersion of a conserved passive scalar in a turbulent flow is determined by the motion of fluid particles (independent of the molecular diffusivity). Here, a hypothesis of diffusivity independence is introduced, which provides a sufficient condition for the validity of Taylor's approach. The hypothesis, which is supported by DNS data, is that, at high Reynolds number, the mean of the scalar conditional on the velocity is independent of the molecular diffusivity. From this hypothesis it is shown that (at high Reynolds number) the conditional Laplacian of the scalar is zero. This new result has several significant implications for models of turbulent mixing, and for the scalar flux. Primarily, a model of turbulent scalar mixing that is independent of velocity is inconsistent with the hypothesis, and gives rise to a spurious source or (more likely) sink of the scalar flux.

1. Introduction

In the paper 'Diffusion by continuous movements' Taylor (1921) postulated that in a turbulent flow the mean field $\langle\phi(\mathbf{x}, t)\rangle$ of a conserved passive scalar can be determined from the statistics of the motion of fluid particles. This observation forms the basis for studies of turbulent dispersion (e.g. Batchelor & Townsend 1956; Hunt 1985).

In second-moment closures for turbulent flows, the mean field $\langle\phi(\mathbf{x}, t)\rangle$ is determined differently, namely by solving the transport equation for $\langle\phi\rangle$. This equation involves the scalar flux (i.e. the velocity–scalar covariance $\langle\mathbf{u}\phi\rangle$) which is obtained as the solution to a modelled transport equation (see, for example, Launder 1990).

Yet another approach is to solve a modelled transport equation for the joint probability density function (PDF) of velocity and the scalar, and then to obtain the mean scalar $\langle\phi(\mathbf{x}, t)\rangle$ as a first moment of the joint PDF (see e.g. Pope 1985). Part of the modelled PDF equation is a mixing model which accounts for the effects of molecular diffusion.

A second (but less widely appreciated) contribution of Taylor's paper is the idea of using a stochastic model to describe the motion of fluid particles. Over the years, this idea has been extended as stochastic models have been proposed for other fluid-particle properties such as velocity and the conserved passive scalar ϕ (see e.g. Pope 1994). From these stochastic models, PDF equations can be obtained without further modelling assumptions; and, from the PDF equations, modelled second-moment

equations can also be obtained (Pope 1985, 1994; Durbin & Speziale 1994). There is, therefore, a close connection between these stochastic models and the mean scalar field $\langle\phi\rangle$ and scalar flux $\langle\mathbf{u}\phi\rangle$ obtained from PDF models, second-moment closures and the turbulent dispersion approach.

The purpose of this paper is to make these connections precise, and to study their implications. A principal result is that the expectation of the Laplacian $\nabla^2\phi(\mathbf{x}, t)$ conditioned on the velocity (i.e. $\langle\nabla^2\phi(\mathbf{x}, t)|\mathbf{U}(\mathbf{x}, t) = \mathbf{V}\rangle$) tends to zero at high Reynolds number. This result imposes a constraint on mixing models – one that is violated by all models currently in use. It also implies that a modelled scalar flux equation can be obtained from a stochastic model for fluid-particle velocity, independent of the modelling for the scalar ϕ .

Previous studies of the effects of molecular diffusivity on turbulent dispersion include the papers of Saffman (1960) and Sawford & Hunt (1986). Some of the ideas used in the development below stem from the recent works of Dreeben & Pope (1997), Fox (1996), Durbin & Shabany (1995) and Klimenko (1996).

2. Formulation

We consider the turbulent flow of a constant-property Newtonian fluid in an infinite domain. The velocity field $\mathbf{U}(\mathbf{x}, t)$ satisfies the continuity equation

$$\nabla \cdot \mathbf{U} = 0, \quad (1)$$

and the momentum equation

$$\frac{D\mathbf{U}}{Dt} = \frac{\partial\mathbf{U}}{\partial t} + \mathbf{U} \cdot \nabla\mathbf{U} = \mathbf{F}(\mathbf{x}, t) = \nu\nabla^2\mathbf{U} - \nabla p. \quad (2)$$

Here, ν and p are the kinematic viscosity and pressure, and $\mathbf{F}(\mathbf{x}, t)$ is written for the net specific force on the fluid that causes the acceleration $D\mathbf{U}/Dt$.

From the deterministic initial condition

$$\phi(\mathbf{x}, 0) = \phi_0(\mathbf{x}), \quad (3)$$

the field $\phi(\mathbf{x}, t)$ of a conserved passive scalar evolves by

$$\frac{D\phi}{Dt} = \Gamma\nabla^2\phi, \quad (4)$$

where Γ is the molecular diffusivity. For convenience, the initial condition $\phi_0(\mathbf{x})$ is specified to be non-negative, zero at infinity, and normalized so that its integral over the infinite domain is unity. (Thus, $\phi_0(\mathbf{x})$ has the properties of a PDF.) It follows from (4) that $\phi(\mathbf{x}, t)$ also possesses these properties for finite positive times.

The Schmidt number is $Sc \equiv \nu/\Gamma$, the Reynolds number is $Re \equiv \hat{u}L/\nu$, and the Péclet number is $Pe \equiv \hat{u}L/\Gamma = ReSc$, where \hat{u} is a characteristic r.m.s. turbulent velocity, and L is a characteristic turbulent integral length scale.

The Reynolds decomposition of the velocity and scalar fields are

$$\mathbf{U}(\mathbf{x}, t) = \langle\mathbf{U}(\mathbf{x}, t)\rangle + \mathbf{u}(\mathbf{x}, t), \quad (5)$$

and

$$\phi(\mathbf{x}, t) = \langle\phi(\mathbf{x}, t)\rangle + \phi'(\mathbf{x}, t), \quad (6)$$

so that the mean scalar equation is

$$\frac{\partial\langle\phi\rangle}{\partial t} + \nabla \cdot \langle\mathbf{U}\phi\rangle = \frac{\partial\langle\phi\rangle}{\partial t} + \langle\mathbf{U}\rangle \cdot \nabla\langle\phi\rangle + \nabla \cdot \langle\mathbf{u}\phi'\rangle = \Gamma\nabla^2\langle\phi\rangle. \quad (7)$$

At high Reynolds and Péclet numbers, transport by molecular diffusivity is negligibly small compared to mean-flow convection, and to convection by the turbulent scalar flux $\langle \mathbf{u}\phi' \rangle$.

3. Diffusing particles

Following Saffman (1960) and Sawford & Hunt (1986), the starting point for our description of turbulent dispersion is the concept of a diffusing particle, which is a mathematical model for the motion of a molecule. The position $\mathbf{X}^+(t)$ of a diffusing particle evolves by the stochastic differential equation (SDE)

$$d\mathbf{X}^+(t) = \mathbf{U}^+(t) dt + (2\Gamma)^{1/2} d\mathbf{W}(t), \tag{8}$$

where the particle's velocity $\mathbf{U}^+(t)$ is the local (continuum) fluid velocity,

$$\mathbf{U}^+(t) \equiv \mathbf{U}(\mathbf{X}^+[t], t), \tag{9}$$

and $\mathbf{W}(t)$ is an isotropic Wiener process. The initial particle position is random, with the joint probability density function (PDF) of $\mathbf{X}^+(0)$ being $\phi_0(\mathbf{x})$.

It is important to appreciate that there are three distinct sources of randomness. First, for a given realization of the flow and a given initial condition $\mathbf{X}^+(0)$, there is the randomness of the molecular motion expressed in the Wiener process $\mathbf{W}(t)$. Second, there is randomness in the initial condition $\mathbf{X}^+(0)$. And third, there is the randomness of the turbulence that is manifest in a different velocity field $\mathbf{U}(\mathbf{x}, t)$ in each realization. An average over the first two sources of randomness (for a particular realization of the flow) is denoted by $\langle \rangle_{\mathcal{S}}$; whereas $\langle \rangle$ denotes an average over all three sources.

For a given realization of the flow, $\theta(\mathbf{x}, t)$ is defined to be the PDF of $\mathbf{X}^+(t)$. In terms of Dirac delta functions, this can be written

$$\theta(\mathbf{x}, t) = \langle \delta(\mathbf{X}^+[t] - \mathbf{x}) \rangle_{\mathcal{S}}. \tag{10}$$

As stated above, the initial condition $\mathbf{X}^+(0)$ is random, with distribution $\phi_0(\mathbf{x})$, that is,

$$\theta(\mathbf{x}, 0) = \phi_0(\mathbf{x}). \tag{11}$$

It follows from (8) and (10) that θ evolves by

$$\frac{\partial \theta}{\partial t} + \mathbf{U} \cdot \nabla \theta = \Gamma \nabla^2 \theta \tag{12}$$

(see e.g. Pope 1985; Gardiner 1985).

The above two equations that determine $\theta(\mathbf{x}, t)$ are identical to (3) and (4) that determine $\phi(\mathbf{x}, t)$. This establishes that the density $\theta(\mathbf{x}, t)$ of particles evolving according to the SDE, (8), provides an exact mathematical analogy for the conserved passive scalar $\phi(\mathbf{x}, t)$: on each realization of the flow, $\theta(\mathbf{x}, t)$ and $\phi(\mathbf{x}, t)$ are identical.

Diffusing particles are defined (by (8)) in terms of the diffusivity Γ . If Γ is set to zero, then the particles become fluid particles, whose motion is due solely to the motion of the fluid.

For a given value of Γ and for a given realization of the flow, the fields $\phi(\mathbf{x}, t)$ and $\theta(\mathbf{x}, t)$ are numerically identical. But we continue to distinguish between them because they represent fundamentally different quantities. The conserved passive scalar ϕ is a physical quantity, whose governing equation (4) stems from Fick's or Fourier's empirical law, and Γ is an empirically determined material property. In contrast, θ

is a purely mathematical construction, based on (8), in which Γ is a non-negative parameter that can be specified at will.

4. Equivalent statistics

Because the fields $\theta(\mathbf{x}, t)$ and $\phi(\mathbf{x}, t)$ are identical, so also are their statistics jointly with velocity. The relevant joint statistics of $\phi(\mathbf{x}, t)$ and $\mathbf{U}(\mathbf{x}, t)$ are first defined, and then they are related to those of $\mathbf{X}^+(t)$ and $\mathbf{U}^+(t)$.

With $\mathbf{V} = \{V_1, V_2, V_3\}$ and ψ being sample-space variables corresponding to \mathbf{U} and ϕ , the one-point one-time joint PDF of \mathbf{U} and ϕ is denoted by $f(\mathbf{V}, \psi; \mathbf{x}, t)$. In terms of delta functions it is

$$f(\mathbf{V}, \psi; \mathbf{x}, t) = \langle \delta(\mathbf{U}[\mathbf{x}, t] - \mathbf{V}) \delta(\phi[\mathbf{x}, t] - \psi) \rangle. \quad (13)$$

The PDF of velocity is

$$f_u(\mathbf{V}; \mathbf{x}, t) = \langle \delta(\mathbf{U}[\mathbf{x}, t] - \mathbf{V}) \rangle. \quad (14)$$

The focus of the development is on the quantity $g(\mathbf{V}; \mathbf{x}, t)$ which has the following equivalent definitions:

$$\begin{aligned} g(\mathbf{V}; \mathbf{x}, t) &\equiv \int_{-\infty}^{\infty} \psi f(\mathbf{V}, \psi; \mathbf{x}, t) d\psi \\ &= \langle \phi(\mathbf{x}, t) \delta(\mathbf{U}[\mathbf{x}, t] - \mathbf{V}) \rangle \\ &= \langle \phi(\mathbf{x}, t) | \mathbf{U}(\mathbf{x}, t) = \mathbf{V} \rangle f_u(\mathbf{V}; \mathbf{x}, t). \end{aligned} \quad (15)$$

Essentially, g contains information about the expectation of ϕ conditional on the velocity, i.e. $\langle \phi(\mathbf{x}, t) | \mathbf{U}(\mathbf{x}, t) = \mathbf{V} \rangle$ which is abbreviated to $\langle \phi | \mathbf{V} \rangle$.

Turning now to the properties of diffusing particles, for a given realization of the flow, the fine-grained, one-time, single-particle joint PDF of $\mathbf{X}^+(t)$ and $\mathbf{U}^+(t)$ is defined as

$$h'(\mathbf{V}, \mathbf{x}; t) \equiv \langle \delta(\mathbf{X}^+[t] - \mathbf{x}) \delta(\mathbf{U}^+[t] - \mathbf{V}) \rangle_{\mathcal{S}}. \quad (16)$$

Averaging over all realizations, we obtain the joint PDF of $\mathbf{X}^+(t)$ and $\mathbf{U}^+(t)$:

$$h(\mathbf{V}, \mathbf{x}; t) = \langle h'(\mathbf{V}, \mathbf{x}; t) \rangle = \langle \delta(\mathbf{X}^+[t] - \mathbf{x}) \delta(\mathbf{U}^+[t] - \mathbf{V}) \rangle. \quad (17)$$

The correspondence between the statistics of $\phi(\mathbf{x}, t)$ and $\mathbf{U}(\mathbf{x}, t)$, and those of $\mathbf{X}^+(t)$ and $\mathbf{U}^+(t)$ is that $g(\mathbf{V}; \mathbf{x}, t)$ is equal to $h(\mathbf{V}, \mathbf{x}; t)$. Starting from (17), this observation stems from the following steps:

$$\begin{aligned} h(\mathbf{V}, \mathbf{x}, t) &= \langle \langle \delta(\mathbf{X}^+[t] - \mathbf{x}) \delta(\mathbf{U}[\mathbf{X}^+(t), t] - \mathbf{V}) \rangle_{\mathcal{S}} \rangle \\ &= \langle \langle \delta(\mathbf{X}^+[t] - \mathbf{x}) \rangle_{\mathcal{S}} \delta(\mathbf{U}[\mathbf{x}, t] - \mathbf{V}) \rangle \\ &= \langle \theta(\mathbf{x}, t) \delta(\mathbf{U}[\mathbf{x}, t] - \mathbf{V}) \rangle = \langle \theta | \mathbf{V} \rangle f_u. \end{aligned} \quad (18)$$

(In the first line, (9) is substituted for $\mathbf{U}^+(t)$. In the second line, the sifting property of the delta function is used to replace $\mathbf{X}^+(t)$ by \mathbf{x} ; and then the fact that $\mathbf{U}(\mathbf{x}, t)$ is non-random for a given realization is used to remove $\delta(\mathbf{U} - \mathbf{V})$ from the inner expectation. The last line follows from the definition of θ , (10).) Thus, given the equality of $\phi(\mathbf{x}, t)$ and $\theta(\mathbf{x}, t)$, a comparison of (15) and (18) indeed shows that g and h are equal, and furthermore that $\langle \phi | \mathbf{V} \rangle$ equals $\langle \theta | \mathbf{V} \rangle$.

In the Appendix the exact evolution equations are derived for g (equation (A 3)), h (A 10), $\langle \mathbf{U} \phi \rangle$ (A 6) and $\langle \mathbf{U} \theta \rangle$ (A 12).

5. High-Reynolds number limit

5.1. Hypothesis of diffusivity independence

We introduce a hypothesis, which formalizes and extends a notion used by Taylor (1921) and which has generally been accepted ever since: ‘At high Reynolds number and high Péclet number, the conditional mean scalar field $\langle \phi(\mathbf{x}, t) | \mathbf{U}(\mathbf{x}, t) = \mathbf{V} \rangle$ is independent of the magnitude of the molecular diffusivity Γ (except close to singularities arising from initial or boundary conditions).’ Since the density of diffusing particles $\theta(\mathbf{x}, t)$ is identical to $\phi(\mathbf{x}, t)$, the hypothesis applies equally to $\langle \theta | \mathbf{V} \rangle$. And, in the particular case $\Gamma = 0$, θ is the density of fluid particles. Hence, an immediate corollary is that, subject to the same conditions: ‘The conditional mean $\langle \phi | \mathbf{V} \rangle$ of a diffusive scalar ($\Gamma > 0$) is identical to the conditional density of fluid particles, i.e. $\langle \theta | \mathbf{V} \rangle$ (for $\Gamma = 0$).’

These statements about the conditional mean apply also to the unconditional mean $\langle \phi \rangle$ and to the scalar flux $\langle \mathbf{U}\phi \rangle$ – since these quantities are determined by $\langle \phi | \mathbf{V} \rangle$ and f_u .

In essence, Taylor (1921) assumes $\langle \phi \rangle$ (for $\Gamma > 0$) to be identical to $\langle \theta \rangle$ (for $\Gamma = 0$), which implies the equality of $\langle \mathbf{u}\phi \rangle$ and $\langle \mathbf{u}\theta \rangle$. (Taylor does not explicitly state any requirements such as high Reynolds number.)

As mentioned, the above hypothesis is generally accepted (for $\langle \phi \rangle$ and $\langle \mathbf{U}\phi \rangle$ at least), and it is in accord with experimental data (e.g. data on mean concentration fields in free shear flows). These data are not reviewed here. A more stringent test of the hypothesis is given in §5.3.

It should be appreciated that the diffusivity-independence hypothesis, especially with $\Gamma = 0$, applies only to the conditional mean $\langle \phi | \mathbf{V} \rangle$. Other statistics, the scalar variance and PDF in particular, have qualitatively different behaviours for $\Gamma = 0$ and $\Gamma > 0$.

5.2. Implications for the conditional Laplacian

With respect to the development in previous sections, the implication of the hypothesis is that (subject to the specified conditions) $g(\mathbf{V}; \mathbf{x}, t)$ for a diffusive scalar ($\Gamma > 0$) is identical to $h(\mathbf{V}, \mathbf{x}; t)$ for fluid particles ($\Gamma = 0$), and consequently that these two quantities evolve in the same way. A comparison of the evolution equation for g (A 3) with that for h (A 10) with $\Gamma = 0$ shows that there is a term-by-term correspondence, except for the term $f_u \langle \Gamma \nabla^2 \phi | \mathbf{V} \rangle$ in (A 3). An implication of the hypothesis is, therefore, that this term vanishes at high Reynolds and Péclet number. More precisely,

$$\lim_{Re, Pe \rightarrow \infty} \left\{ \frac{L}{\hat{u}\hat{\phi}} \langle \Gamma \nabla^2 \phi | \mathbf{V} \rangle \right\} = 0, \tag{19}$$

where the conditional Laplacian is normalized by the characteristic r.m.s. velocity \hat{u} , scalar $\hat{\phi}$, and integral length scale L .

The normalized conditional Laplacian in (19) can be re-expressed as

$$\frac{L}{\hat{u}\hat{\phi}} \langle \Gamma \nabla^2 \phi | \mathbf{V} \rangle = \left\{ \frac{L/\hat{u}}{\hat{\phi}^2 / \langle \Gamma \nabla \phi \cdot \nabla \phi \rangle} \right\} \frac{\hat{\phi} \langle \nabla^2 \phi | \mathbf{V} \rangle}{\langle \nabla \phi \cdot \nabla \phi \rangle}. \tag{20}$$

The term in braces is the ratio of the mechanical time scale L/\hat{u} to the scalar dissipation time scale. This quantity is of order one in the limit considered, so that

(19) can be rewritten

$$\lim_{Re, Pe \rightarrow \infty} \left\{ \frac{\hat{\phi} \langle \nabla^2 \phi | V \rangle}{\langle \nabla \phi \cdot \nabla \phi \rangle} \right\} = 0. \quad (21)$$

(In the term-by-term correspondence between (A 3) and (A 10), the terms $\langle \phi \mathbf{F} | V \rangle$ and $\langle \theta \mathbf{F} | V \rangle$ correspond. However, the argument above leading to (19) is weakened by the fact that there is no proof of the equality of these two conditional expectations, i.e. $\langle \phi \mathbf{F} | V \rangle$ for $\Gamma > 0$ and $\langle \theta \mathbf{F} | V \rangle$ for $\Gamma = 0$.)

5.3. Evidence from direct numerical simulations

Overholt & Pope (1996) performed direct numerical simulations (DNS) of a conserved passive scalar ($Sc = 0.7$) with an imposed uniform mean scalar gradient in homogeneous isotropic turbulence (with $\langle U \rangle = 0$). Their measurements of $\langle \nabla^2 \phi | V, \psi \rangle$ are used to test (21).

Two observations from the DNS are that both $\langle \phi | V \rangle$ and $\langle \nabla^2 \phi | V, \psi \rangle$ are very well approximated as linear functions of V and ψ . (Here V is the sample-space variable corresponding to the component of velocity U in the direction of the mean scalar gradient.) Specifically

$$\langle \phi | V \rangle = \langle \phi \rangle + \frac{\hat{\phi}}{\hat{u}} \rho_{u\phi} V, \quad (22)$$

and

$$\frac{\hat{\phi} \langle \nabla^2 \phi | V, \psi \rangle}{\langle \nabla \phi \cdot \nabla \phi \rangle} = r \frac{V}{\hat{u}} - (1 + r \rho_{u\phi}) \frac{(\psi - \langle \phi \rangle)}{\hat{\phi}}, \quad (23)$$

where \hat{u} and $\hat{\phi}$ are the standard deviations of U and ϕ , $\rho_{u\phi}$ is the correlation coefficient

$$\rho_{u\phi} = \frac{\langle u \phi' \rangle}{\hat{u} \hat{\phi}}, \quad (24)$$

and the non-dimensional parameter r is determined from the DNS data.

These equations can be manipulated to yield

$$\frac{\hat{\phi} \langle \nabla^2 \phi | V, \psi \rangle}{\langle \nabla \phi \cdot \nabla \phi \rangle} = \frac{1}{1 - \rho_{u\phi}^2 (1 - \zeta)} \left\{ \frac{\zeta \langle \phi \rangle + [1 - \zeta] \langle \phi | V \rangle - \psi}{\hat{\phi}} \right\}, \quad (25)$$

where

$$\zeta = 1 - \frac{r}{\rho_{u\phi} (1 + r \rho_{u\phi})}. \quad (26)$$

And from (25) we obtain

$$\frac{\hat{\phi} \langle \nabla^2 \phi | V \rangle}{\langle \nabla \phi \cdot \nabla \phi \rangle} = \frac{\zeta}{1 - \rho_{u\phi}^2 (1 - \zeta)} \left\{ \frac{\langle \phi \rangle - \langle \phi | V \rangle}{\hat{\phi}} \right\}. \quad (27)$$

It is evident from (21) and (27) that the DNS data are consistent with the hypothesis providing that

$$\lim_{Re, Pe \rightarrow \infty} \zeta = 0. \quad (28)$$

Figure 1 shows the value of ζ obtained from the DNS as a function of the Taylor-scale Reynolds number R_λ . There is every indication that indeed ζ tends to zero, in accord with the hypothesis.

It would, of course, be valuable to assess the accuracy of the linear estimations ((22) and (23)) and of the value of ζ in other flows and at higher Reynolds numbers.

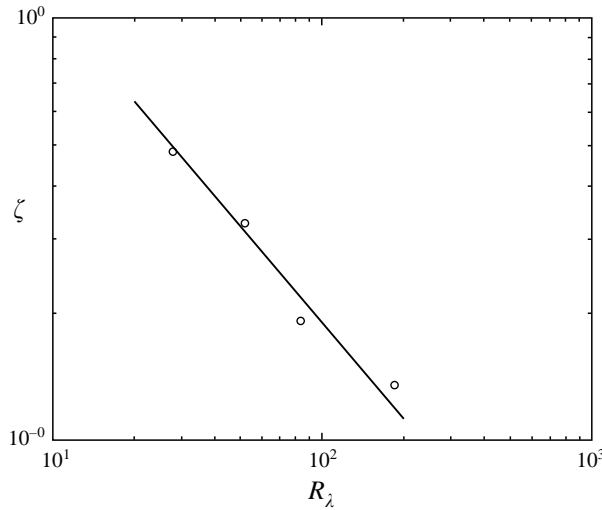


FIGURE 1. The quantity ζ appearing in (25)–(28) as a function of the Taylor-scale Reynolds number R_λ . From the direct numerical simulations of Overholt & Pope (1996).

5.4. *Scalar-flux, dissipation and local isotropy*

The equation for the scalar flux $\langle \mathbf{U}\phi \rangle$ (A 6) contains a dissipation term of the form

$$\epsilon_{u\phi} \equiv (\nu + \Gamma)\langle \nabla\phi \cdot \nabla\mathbf{u} \rangle. \tag{29}$$

A further deduction from the linear model (25) is

$$\frac{\hat{\phi} \langle \nabla\phi \cdot \nabla\mathbf{u} \rangle}{\hat{u} \langle \nabla\phi \cdot \nabla\phi \rangle} = \frac{-\zeta \rho_{u\phi}}{1 - \rho_{u\phi}^2 [1 - \zeta]}. \tag{30}$$

Thus, at high Reynolds and Péclet number, as ζ tends to zero, so also does the scalar-flux dissipation $\epsilon_{u\phi}$. This is confirmed directly by the DNS data of Overholt & Pope (1996).

At high Reynolds number, if the velocity and scalar fields are locally isotropic, then $\langle \nabla\phi \cdot \nabla\mathbf{U} \rangle$ is zero. For this reason, $\epsilon_{u\phi}$ is generally assumed to be zero at high Reynolds number. Notice that the assumption of local isotropy together with the assumption that $\langle \nabla^2\phi | \mathbf{V} \rangle$ is linear in \mathbf{V} implies that $\langle \nabla^2\phi | \mathbf{V} \rangle$ is zero. But by itself, local isotropy does not imply that $\langle \nabla^2\phi | \mathbf{V} \rangle$ is zero.

In recent years, the local isotropy of scalar fields at high Reynolds number has been called in question (e.g. Sreenivasan 1991). The clearest departure from local isotropy is the persistence of the scalar derivative skewness in isotropic turbulence with an imposed mean scalar gradient (e.g. Tong & Warhaft 1994; Overholt & Pope 1996). Consequently, it would be valuable to test (in high-Reynolds-number flows) the diffusivity-independence hypothesis, and its implication that the correlation coefficient between $\nabla\phi$ and $\nabla\mathbf{u}$ tends to zero.

6. **Implications for stochastic models**

The position, velocity and scalar value of a fluid particle are denoted by $\mathbf{X}^+(t)$, $\mathbf{U}^+(t)$ and $\phi^+(t) \equiv \phi(\mathbf{X}^+[t], t)$. We now introduce stochastic models for these quantities, denoted by $\mathbf{X}^*(t)$, $\mathbf{U}^*(t)$ and $\phi^*(t)$. If the models were perfect, then the joint statistics

of \mathbf{X}^* , \mathbf{U}^* and ϕ^* would be identical to those of \mathbf{X}^+ , \mathbf{U}^+ and ϕ^+ . The purpose of this section is to deduce the implications for these models of the hypothesis of diffusivity independence (which applies at high Reynolds and Péclet number).

By definition of a fluid particle, the position $\mathbf{X}^*(t)$ evolves by

$$d\mathbf{X}_i^*(t) = U_i^*(t) dt. \quad (31)$$

For the velocity $\mathbf{U}^*(t)$ and scalar $\phi^*(t)$, the models considered are reasonably general diffusion processes:

$$dU_i^*(t) = A_i(\mathbf{U}^*[t], \mathbf{X}^*[t], t) dt + B_{ij}(\mathbf{U}^*[t], \mathbf{X}^*[t], t) dW_j, \quad (32)$$

and

$$d\phi^*(t) = a(\mathbf{U}^*[t], \phi^*[t], \mathbf{X}^*[t], t) dt + b(\mathbf{U}^*[t], \phi^*[t], \mathbf{X}^*[t], t) dW', \quad (33)$$

where $\mathbf{W}(t)$ and $W'(t)$ are independent Wiener processes. For the scalar ϕ^* , the drift coefficient a and the diffusion coefficient b are completely general. But, because the scalar is passive, in the velocity equation it would be inappropriate to allow the drift coefficient A_i or the diffusion coefficient B_{ij} to depend on ϕ^* .

These general models are analysed below. But, to fix ideas, we mention that the simplest model for $\mathbf{U}^*(t)$ is the simplified Langevin model (SLM) (Haworth & Pope 1986; Pope 1994); and the simplest model for $\phi^*(t)$ is the IEM or LMSE model (Villermaux & Devillon 1972; Dopazo & O'Brien 1974). The IEM (interaction by exchange with the mean) model is

$$d\phi^*(t) = -\Omega(\mathbf{X}^*[t], t)(\phi^*(t) - \langle \phi^*(t) | \mathbf{X}^*(t) \rangle) dt, \quad (34)$$

where $\Omega(\mathbf{x}, t)$ is a specified mixing frequency, and $\langle \phi^*(t) | \mathbf{X}^*(t) \rangle$ is the model equivalent of the mean $\langle \phi(\mathbf{x}, t) \rangle$ evaluated at the particle location.

From the stochastic model equations, (31)–(33), it is straightforward to derive model evolution equations for a variety of statistics: this is done in the Appendix. For each statistic defined above (e.g. f , g , h) the corresponding model statistics are indicated by an asterisk (e.g. f^* , g^* , h^*).

According to the hypothesis of diffusivity independence, at high Reynolds and Péclet number, g and h are equal. The stochastic models are consistent with this limit if, and only if, the evolution equations for g^* and h^* are equivalent. It is shown in the Appendix that these equations are indeed equivalent if, and only if, the conditional mean drift of $\phi^*(t)$ is zero, i.e.

$$\langle a^* | \mathbf{V}, \mathbf{x} \rangle \equiv \langle a(\mathbf{U}^*[t], \phi^*[t], \mathbf{X}^*[t], t) | \mathbf{U}^*[t] = \mathbf{V}, \mathbf{X}^*[t] = \mathbf{x} \rangle = 0. \quad (35)$$

A consistent model therefore satisfies the condition $\langle a^* | \mathbf{V}, \mathbf{x} \rangle = 0$ in the high-Reynolds-number limit. (Note that there is no condition imposed on the diffusion coefficient b .)

Most mixing models (i.e. specifications of a and b) take no account of the particle velocity $\mathbf{U}^*(t)$, and therefore in general do not satisfy $\langle a^* | \mathbf{V}, \mathbf{x} \rangle = 0$. For example, for the IEM model (34), we have

$$\langle a^* | \mathbf{V}, \mathbf{x} \rangle = -\Omega(\langle \phi^* | \mathbf{V}, \mathbf{x} \rangle - \langle \phi^* | \mathbf{x} \rangle). \quad (36)$$

Only in special cases, e.g. when \mathbf{U}^* and ϕ^* are independent, is $\langle a^* | \mathbf{V}, \mathbf{x} \rangle$ zero.

Mixing models that involve a dependence on velocity have been proposed by Pope (1985) and Song (1987). Pope (1994) observed that the IEM model is inconsistent

with local isotropy, whereas the alternative

$$d\phi^* = -\Omega(\phi^* - \langle \phi^* | \mathbf{V}, \mathbf{x} \rangle) dt, \quad (37)$$

is consistent. This model can be called IECM – interaction by exchange with the conditional mean. It is immediately apparent that the IECM model (37) indeed satisfies $\langle a^* | \mathbf{V}, \mathbf{x} \rangle = 0$. It can be argued that a mixing model should ideally be local in physical space, velocity space and composition space (Norris & Pope 1990; Masri, Subramaniam & Pope 1996). Compared to IEM, the IECM model has the virtue of being local in velocity space.

Fox (1996) proposed a blend of the IEM and IECM models, namely

$$d\phi^* = -\Omega(\phi^* - \zeta \langle \phi^* | \mathbf{x} \rangle - [1 - \zeta] \langle \phi^* | \mathbf{V}, \mathbf{x} \rangle) dt. \quad (38)$$

It may be observed that this is equivalent to the linear model (25) that accurately represents the DNS data (since $\langle d\phi^*/dt | \phi^* = \psi, \mathbf{V}, \mathbf{x} \rangle$ obtained from (38) is the same as the right-hand side of (25)). This model is in accord with the independence-of-diffusivity hypothesis provided that ζ tends to zero in the high-Reynolds-number limit (cf. figure 1).

There are two important observations to make about the modelled equation (A 22) for the scalar flux $\langle U_j^* \phi^* \rangle$. First, it contains the term $\langle U_j^* a^* \rangle$ which vanishes if the condition $\langle a^* | \mathbf{V}, \mathbf{x} \rangle = 0$ is satisfied. If, on the other hand, a mixing model does not satisfy $\langle a^* | \mathbf{V}, \mathbf{x} \rangle = 0$ in the high-Reynolds-number limit, then it induces the spurious source $\langle a^* U^* | \mathbf{x} \rangle$ in the scalar flux equation. For example, in the IEM model, this source is $-\Omega \langle \phi^* U^* | \mathbf{x} \rangle$.

The second observation from (A 22), again if the condition $\langle a^* | \mathbf{V}, \mathbf{x} \rangle = 0$ is satisfied, is that of all the stochastic model coefficients – A_i, B_{ij}, a and b – the scalar flux is affected only by A . As pointed out by Durbin & Shabany (1995), this suggests that in developing improved stochastic models, A is best determined by reference to high-Reynolds-number scalar flux data, rather than from Reynolds-stress data. Once A is determined, then B can be determined by reference to Reynolds-stress data. In practice, however, much of the available experimental and DNS data are at moderate Reynolds numbers, at which $\langle a^* | \mathbf{V}, \mathbf{x} \rangle$ is not negligible.

Finally we observe that from a stochastic model for fluid-particle velocity alone (i.e. given A and B) there follow high-Reynolds-number model equations for the Reynolds stresses and scalar fluxes that are both realizable and consistent with the diffusivity-independence hypothesis.

7. Conclusion

Taylor (1921) introduced the fundamental idea that the dispersion of a conserved passive scalar in turbulent flow is determined by the motion of fluid particles (independent of the molecular diffusivity). Here, this idea is formalized through the hypothesis of diffusivity independence, namely: At high Reynolds number and high Péclet number, the conditional mean scalar field $\langle \phi(\mathbf{x}, t) | \mathbf{U}(\mathbf{x}, t) = \mathbf{V} \rangle$ is independent of the magnitude of the molecular diffusivity.

It then follows (subject to the same conditions) that the mean $\langle \phi \rangle$ and conditional mean $\langle \phi | \mathbf{V} \rangle$ of a diffusive scalar are identical to the density $\langle \theta \rangle$ and conditional density $\langle \theta | \mathbf{V} \rangle$ of fluid particles.

From the correspondence between $\langle \phi | \mathbf{V} \rangle$ and $\langle \theta | \mathbf{V} \rangle$, it is deduced that the conditional Laplacian $\langle \Gamma \nabla^2 \phi | \mathbf{V} \rangle$ tends to zero in the high-Reynolds-number limit. Evidence from DNS supports this deduction (figure 1).

A consequence for turbulent mixing models (33) is that the conditional mean drift of the scalar $\langle a^* | \mathbf{V}, \mathbf{x} \rangle$ should vanish at high Reynolds number. The IEM model (34) is incorrect in this respect, but a variant – IECM (interaction by exchange with the condition mean, (37)) – is correct.

In the high-Reynolds-number limit, the scalar flux $\langle U\phi \rangle$ is determined entirely by the fluid-particle motion. Stochastic models for fluid-particle properties are consistent with this observation if, and only if, the condition $\langle a^* | \mathbf{V}, \mathbf{x} \rangle = 0$ is satisfied. Model equations for the Reynolds stresses and scalar fluxes can be obtained from a stochastic model for velocity. The model equations thus obtained are consistent with the hypothesis of diffusivity independence and they guarantee realizability.

The results obtained here can be used to guide the development of improved stochastic models and turbulence models. It should be appreciated, however, that most experiments and direct numerical simulations are some way from the high-Reynolds-number limit at which the present results apply.

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Appendix

A.1. Evolution equation for $g(\mathbf{V}; \mathbf{x}, t)$

The evolution equation for the joint PDF of $\mathbf{U}(\mathbf{x}, t)$ and $\phi(\mathbf{x}, t)$, i.e. $f(\mathbf{V}, \psi; \mathbf{x}, t)$ defined by (13), is

$$\frac{\partial f}{\partial t} + V_i \frac{\partial f}{\partial x_i} + \frac{\partial}{\partial V_i} \left\{ f \left\langle \frac{DU_i}{Dt} | \mathbf{V}, \psi \right\rangle \right\} + \frac{\partial}{\partial \psi} \left\{ f \left\langle \frac{D\phi}{Dt} | \mathbf{V}, \psi \right\rangle \right\} = 0. \quad (\text{A } 1)$$

Here, for any quantity $Q(\mathbf{x}, t)$, the mean conditioned on $\mathbf{U}(\mathbf{x}, t)$ and $\phi(\mathbf{x}, t)$ is written $\langle Q(\mathbf{x}, t) | \mathbf{U}(\mathbf{x}, t) = \mathbf{V}, \phi(\mathbf{x}, t) = \psi \rangle$ and is abbreviated to $\langle Q | \mathbf{V}, \psi \rangle$. With DU/Dt given by (2), and $D\phi/Dt$ by (4), the PDF equation becomes

$$\frac{\partial f}{\partial t} + V_i \frac{\partial f}{\partial x_i} + \frac{\partial}{\partial V_i} \{ f \langle F_i | \mathbf{V}, \psi \rangle \} + \frac{\partial}{\partial \psi} \{ f \Gamma \langle \nabla^2 \phi | \mathbf{V}, \psi \rangle \} = 0. \quad (\text{A } 2)$$

The equation for $g(\mathbf{V}; \mathbf{x}, t)$ (defined by (15)) is obtained by multiplying (A 2) by ψ and integrating over all ψ . The result is

$$\frac{\partial g}{\partial t} + V_i \frac{\partial g}{\partial x_i} + \frac{\partial}{\partial V_i} \{ f_u \langle \phi F_i | \mathbf{V} \rangle \} - f_u \Gamma \langle \nabla^2 \phi | \mathbf{V} \rangle = 0. \quad (\text{A } 3)$$

With $f_u(\mathbf{V}; \mathbf{x}, t)$ being the PDF of $\mathbf{U}(\mathbf{x}, t)$, and with $f_{\phi|u}(\psi | \mathbf{V}; \mathbf{x}, t)$ being the PDF of $\phi(\mathbf{x}, t)$ conditioned on $\mathbf{U}(\mathbf{x}, t)$, the manipulations leading to the above equation are, for example,

$$\int_{-\infty}^{\infty} \psi f \langle \mathbf{F} | \mathbf{V}, \psi \rangle d\psi = \int_{-\infty}^{\infty} f_u f_{\phi|u} \langle \phi \mathbf{F} | \mathbf{V}, \psi \rangle d\psi = f_u \langle \phi \mathbf{F} | \mathbf{V} \rangle. \quad (\text{A } 4)$$

The evolution equation for the scalar flux

$$\langle U_j \phi \rangle = \langle U_j \rangle \langle \phi \rangle + \langle u_j \phi' \rangle, \quad (\text{A } 5)$$

can be obtained by multiplying (A 3) by V_j and integrating over all \mathbf{V} – or, more

simply, directly from the equations for \mathbf{U} and ϕ . The result is

$$\begin{aligned} \frac{\partial}{\partial t} \langle U_j \phi \rangle + \frac{\partial}{\partial x_i} \langle U_i U_j \phi \rangle &= - \left\langle \phi \frac{\partial p}{\partial x_j} \right\rangle + \Gamma \nabla^2 \langle U_j \phi \rangle \\ &+ (v - \Gamma) \frac{\partial}{\partial x_i} \left\langle \phi \frac{\partial U_j}{\partial x_i} \right\rangle - (v + \Gamma) \left\langle \frac{\partial \phi}{\partial x_i} \frac{\partial U_j}{\partial x_i} \right\rangle. \end{aligned} \quad (\text{A } 6)$$

A.2. Evolution equation for $h(\mathbf{V}, \mathbf{x}; t)$

Because $\mathbf{X}^+(t)$ evolves by a diffusion process (8), so also does $\mathbf{U}^+(t)$. From (9), the infinitesimal increment in $\mathbf{U}^+(t)$ is

$$\begin{aligned} dU_i^+ &= \left(\frac{\partial U_i}{\partial t} \right)^+ dt + \left(\frac{\partial U_i}{\partial x_j} \right)^+ dX_j^+ + \frac{1}{2} \left(\frac{\partial^2 U_i}{\partial x_j \partial x_k} \right)^+ dX_j^+ dX_k^+ \\ &= F_i^+ dt + (2\Gamma)^{1/2} \left(\frac{\partial U_i}{\partial x_j} \right)^+ dW_j + \Gamma (\nabla^2 U_i)^+ dt, \end{aligned} \quad (\text{A } 7)$$

where the superscript + indicates, for example,

$$F_i^+ = F_i(\mathbf{X}^+[t], t), \quad (\text{A } 8)$$

and (2) and (8) have been invoked. Even though the velocity field $\mathbf{U}(\mathbf{x}, t)$ is smooth and differentiable, $\mathbf{U}^+(t)$ inherits from $\mathbf{X}^+(t)$ randomness and a lack of differentiability.

The evolution equation for $h(\mathbf{V}, \mathbf{x}; t)$ is obtained from its definition and from the SDE's for $\mathbf{X}^+(t)$ and $\mathbf{U}^+(t)$. In this derivation, the following manipulation (similar to 18) is used:

$$\begin{aligned} \langle Q^+(t) \delta(\mathbf{X}^+[t] - \mathbf{x}) \delta(\mathbf{U}^+[t] - \mathbf{V}) \rangle &= \langle Q(\mathbf{x}, t) \delta(\mathbf{X}^+[t] - \mathbf{x}) \delta(\mathbf{U}(\mathbf{x}, t) - \mathbf{V}) \rangle \\ &= \langle Q(\mathbf{x}, t) \theta(\mathbf{x}, t) \delta(\mathbf{U}(\mathbf{x}, t) - \mathbf{V}) \rangle \\ &= \langle Q \theta | \mathbf{V} \rangle f_{\mathbf{u}}. \end{aligned} \quad (\text{A } 9)$$

The equation thus obtained is

$$\begin{aligned} \frac{\partial h}{\partial t} + V_i \frac{\partial h}{\partial x_i} + \frac{\partial}{\partial V_i} \{ f_{\mathbf{u}} \langle \theta F_i + \Gamma \theta \nabla^2 U_i | \mathbf{V} \rangle \} &= \Gamma \nabla^2 h \\ + 2\Gamma \frac{\partial^2}{\partial V_i \partial x_k} \left\{ f_{\mathbf{u}} \left\langle \theta \frac{\partial U_i}{\partial x_k} | \mathbf{V} \right\rangle \right\} &+ \Gamma \frac{\partial^2}{\partial V_i \partial V_j} \left\{ f_{\mathbf{u}} \left\langle \theta \frac{\partial U_i}{\partial x_k} \frac{\partial U_j}{\partial x_k} | \mathbf{V} \right\rangle \right\}. \end{aligned} \quad (\text{A } 10)$$

By multiplying the above equation by V_j and integrating over all \mathbf{V} we obtain the equation for the particle convective flux $\langle U_j \theta \rangle$:

$$\frac{\partial}{\partial t} \langle U_j \theta \rangle + \frac{\partial}{\partial x_i} \langle U_i U_j \theta \rangle - \langle \theta F_j \rangle - \Gamma \langle \theta \nabla^2 U_j \rangle = \Gamma \nabla^2 \langle U_j \theta \rangle - 2\Gamma \frac{\partial}{\partial x_k} \langle \theta \frac{\partial U_j}{\partial x_k} \rangle, \quad (\text{A } 11)$$

which can be re-expressed as

$$\begin{aligned} \frac{\partial}{\partial t} \langle U_j \theta \rangle + \frac{\partial}{\partial x_i} \langle U_i U_j \theta \rangle &= - \left\langle \theta \frac{\partial p}{\partial x_j} \right\rangle + \Gamma \nabla^2 \langle U_j \theta \rangle \\ &+ (v - \Gamma) \frac{\partial}{\partial x_i} \left\langle \theta \frac{\partial U_j}{\partial x_i} \right\rangle - (v + \Gamma) \left\langle \frac{\partial \theta}{\partial x_i} \frac{\partial U_j}{\partial x_i} \right\rangle. \end{aligned} \quad (\text{A } 12)$$

Indeed, (A 10) for h can be put into the identical form to (A 3) for g by using the following identity which stems from the definition of h (17):

$$\begin{aligned} \nabla^2 h &= f_u \langle \nabla^2 \theta \mid \mathbf{V} \rangle + \frac{\partial}{\partial V_j} \{ f_u \langle \theta \nabla^2 U_j \mid \mathbf{V} \rangle \} \\ &\quad - 2 \frac{\partial^2}{\partial x_i \partial V_j} \left\{ f_u \left\langle \theta \frac{\partial U_j}{\partial x_i} \mid \mathbf{V} \right\rangle \right\} - \frac{\partial^2}{\partial V_j \partial V_k} \left\{ f_u \left\langle \theta \frac{\partial U_j}{\partial x_i} \frac{\partial U_k}{\partial x_i} \mid \mathbf{V} \right\rangle \right\}. \end{aligned} \quad (\text{A } 13)$$

It may be observed that this equation for $\langle U_j \theta \rangle$ is identical to that for $\langle U_j \phi \rangle$, (A 6).

A.3. Evolution equations from stochastic models

The model quantity corresponding to the joint PDF $f(\mathbf{V}, \psi; \mathbf{x}, t)$ is the joint PDF of $\mathbf{U}^*(t)$ and $\phi^*(t)$ conditional upon $\mathbf{X}^*(t) = \mathbf{x}$, which is denoted by $f^*(\mathbf{V}, \psi | \mathbf{x}; t)$.

The evolution equation for f^* obtained from the given stochastic models (31)–(33) is

$$\begin{aligned} \frac{\partial f^*}{\partial t} + V_i \frac{\partial f^*}{\partial x_i} + \frac{\partial}{\partial V_i} \{ f^* A_i(\mathbf{V}, \mathbf{x}, t) \} + \frac{\partial}{\partial \psi} \{ f^* a(\mathbf{V}, \psi, \mathbf{x}, t) \} \\ = \frac{1}{2} \frac{\partial^2}{\partial V_i \partial V_j} \{ f^* B_{ik} B_{jk} \} + \frac{1}{2} \frac{\partial^2}{\partial \psi^2} \{ f^* b^2 \} \end{aligned} \quad (\text{A } 14)$$

(where the arguments of \mathbf{B} and b are the same as those of \mathbf{A} and a , respectively).

The joint PDF of velocity, $f_u^*(\mathbf{V} | \mathbf{x}; t)$, and the quantity $g^*(\mathbf{V} | \mathbf{x}; t)$ are defined in obvious ways:

$$f_u^*(\mathbf{V} | \mathbf{x}; t) = \int_{-\infty}^{\infty} f^*(\mathbf{V}, \psi | \mathbf{x}; t) d\psi, \quad (\text{A } 15)$$

and

$$\begin{aligned} g^*(\mathbf{V} | \mathbf{x}; t) &= \int_{-\infty}^{\infty} \psi f^*(\mathbf{V}, \psi | \mathbf{x}; t) d\psi \\ &= f_u^*(\mathbf{V} | \mathbf{x}; t) \langle \phi^*(t) | \mathbf{U}^*(t) = \mathbf{V}, \mathbf{X}^*(t) = \mathbf{x} \rangle. \end{aligned} \quad (\text{A } 16)$$

The evolution equation for g^* is obtained by multiplying (A 14) by ψ and integrating:

$$\frac{\partial g^*}{\partial t} + V_i \frac{\partial g^*}{\partial x_i} + \frac{\partial}{\partial V_i} \{ g^* A_i(\mathbf{V}, \mathbf{x}, t) \} - f_u^* \langle a^* | \mathbf{V}, \mathbf{x} \rangle = \frac{1}{2} \frac{\partial^2}{\partial V_i \partial V_j} \{ g^* B_{ik} B_{jk} \}. \quad (\text{A } 17)$$

In this derivation, the last term on the right-hand side arises as

$$\int_{-\infty}^{\infty} f^*(\mathbf{V}, \psi | \mathbf{x}; t) a(\mathbf{V}, \psi, \mathbf{x}, t) d\psi, \quad (\text{A } 18)$$

and from the definition of conditional expectations it can be re-expressed as

$$f_u^*(\mathbf{V} | \mathbf{x}; t) \langle a(\mathbf{V}, \phi^*(t), \mathbf{x}, t) | \mathbf{U}^*(t) = \mathbf{V}, \mathbf{X}^*(t) = \mathbf{x} \rangle. \quad (\text{A } 19)$$

For brevity, this is written $f_u^* \langle a^* | \mathbf{V}, \mathbf{x} \rangle$, and $\langle a^* | \mathbf{V}, \mathbf{x} \rangle$ is referred to as ‘the conditional mean drift of ϕ^* ’.

The evolution equation for $h^*(\mathbf{V}, \mathbf{x}, t)$ is derived from its definition

$$h^*(\mathbf{V}, \mathbf{x}, t) \equiv \langle \delta(\mathbf{X}^*(t) - \mathbf{x}) \delta(\mathbf{U}^*(t) - \mathbf{V}) \rangle, \quad (\text{A } 20)$$

(cf. (17)), and from the stochastic model equations, (31) and (32). The equation for h^* thus obtained is identical to that for g^* (A 17) except for the omission of the term in $\langle a^* | \mathbf{V}, \mathbf{x} \rangle$.

The model quantity corresponding to the scalar flux $\langle U\phi \rangle$ is

$$\langle U^*(t)\phi^*(t)|X^*(t) = \mathbf{x} \rangle = \int_{-\infty}^{\infty} \int V \psi f^*(V, \psi | \mathbf{x}; t) dV d\psi, \quad (\text{A } 21)$$

which is abbreviated to $\langle U^* \phi^* \rangle$. From (A 14), the model equation obtained for $\langle U_j^* \phi^* \rangle$ is

$$\frac{\partial}{\partial t} \langle U_j^* \phi^* \rangle + \frac{\partial}{\partial x_i} \langle U_i^* U_j^* \phi^* \rangle = \langle \phi^* A_j^* \rangle + \langle U_j^* a^* \rangle, \quad (\text{A } 22)$$

where a^* is written for $a(U^*[t], \phi^*[t], X^*[t], t)$, and similarly for A^* .

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