J. Fluid Mech. (1986), *vol.* 165, *pp.* 373–400
Printed in Great Britain

Effects of turbulence structure, molecular diffusion and source size on scalar fluctuations in homogeneous turbulence

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(Received 8 June 1983 and in revised form 5 November 1985)

A Lagrangian stochastic model of two-point displacements which includes explicitly the effects of molecular diffusion and viscosity is developed from the marked-particle model of Durbin (1980) and used to study the influence of these molecular processes on scalar fluctuations in stationary homogeneous turbulence. It is shown that for the homogeneous scalar field resulting from a uniform-gradient source distribution or for a cloud produced by a source large compared with the Kolmogorov microscale, the variance of scalar fluctuations $\overline{\theta'^2}$ is independent of the molecular diffusivity for large Reynolds number Re provided that the Prandtl number Pr is finite. In these circumstances $\overline{\theta'^2}$ can be calculated from marked-particle-pair statistics.

For source sizes that are not large compared with the Kolmogorov microscale, $\overline{\theta'^2}$ depends explicitly on the effect of molecular diffusion under the action of the straining motion on the Kolmogorov microscale. The model is consistent with Saffman's (1960) calculation for the mean concentration near such a small source and with Townsend's (1951) measurements on small heat spots. For the fluctuation field in stationary homogeneous turbulence these small-scale processes remain important far from the source.

It is shown that dissipation of $\overline{\theta'^2}$ is intimately connected to the process of relative dispersion and that the non-dissipative model for $\overline{\theta'^2}$ discussed by Corrsin (1952) and Chatwin & Sullivan (1979) corresponds to the point-sample, infinite-Pr limit of the two-point theory.

The two-point model is also used to examine the effect of instrumental averaging on $\overline{\theta'^2}$. For finite Pr, the reduction in $\overline{\theta'^2}$ due to a fixed sampling volume is eventually negligible because the lengthscale of $\overline{\theta'^2}$ grows with time. For infinite Pr, the linear-strain field of the turbulence on the Kolmogorov microscale generates an increasingly fine-scale structure in the scalar field. Then a finite sampling volume reduces $\overline{\theta'^2}$ but not as much as the effect of reducing Pr to a finite value. A finite sampling volume and infinite Pr is not necessarily equivalent to a finite value of Pr and a point sample.

Timescales for important stages in the development of the scalar field in a cloud or downwind of a continuous source (for example, the onset of dissipation or the stage at which fluctuations are dominated by internal structure or 'streakiness' within the cloud rather than bulk motion or 'meandering') have been estimated. For small sources and large Re these timescales are significantly less than the integral timescale t_L. Many real flows evolve on the timescale t_L so that the present results for stationary

homogeneous turbulence should apply to such flows for small sources and large Re, a situation typical of the atmosphere.

1. Introduction

This paper deals with the dispersion and mixing of a passive scalar quantity (e.g. heat or matter) in stationary homogeneous turbulence. Our reasons for limiting attention to such an idealized flow field are two-fold. First there are a number of exact kinematic results available with which to compare our model results. Secondly, such a flow is of practical interest since in a range of real problems (e.g. dispersion from an elevated source in a boundary layer; dispersion from a line source in decaying grid turbulence or grid turbulence with a mean shear) the initial stages of the evolution of the scalar field proceed as in stationary homogeneous turbulence.

It follows from Saffman's (1960) analysis of the combined action of molecular and turbulent diffusion that the mean concentration $\overline{\theta}$ resulting from the release of a small quantity of contaminant into a turbulent flow is determined by the turbulent velocity field alone for times much beyond $Pe^{-1}t_{\mathrm{L}}$, where the Peclet number $Pe = \sigma_w L/\kappa$, is the ratio of the turbulent and molecular diffusivities and σ_w, L and $t_{\mathrm{L}} = \sigma_w/L$ are turbulence velocity, length and Lagrangian timescales respectively. However, Saffman's (1960) analysis is valid only up to times of $O(Re^{-\frac{1}{2}}t_{\mathrm{L}})$, where the Reynolds number $Re = \sigma_w L/\nu$, and ν is the kinematic viscosity. At larger times after release, there is no precise estimate for the order of magnitude of the effect of molecular diffusion, but it is found experimentally that when $Pe \gg 1$ the mean field can be calculated accurately by considering the motion of marked fluid particles which follow the motion of the fluid continuum and conserve the scalar quantity. (Simulation of molecular diffusion by a small-scale random walk confirms this hypothesis (Drummond, Duane & Horgan, 1984; Stapountzis *et al.* 1986)).

However, the variance of scalar fluctuations, denoted by $\overline{\theta'^2} = \overline{\theta^2} - \overline{\theta}^2$ is fundamentally influenced by molecular diffusion even when $Pe \gg 1$. The magnitude of $\overline{\theta'^2}$ is controlled by the rate ϵ_θ at which molecular diffusion dissipates scalar fluctuations by reducing local scalar gradients. ϵ_θ is given by

$$\epsilon_\theta = 2\kappa\overline{(\nabla\theta')^2}, \tag{1.1}$$

where the variance of the temperature gradient $\overline{(\nabla\theta')^2}$ is controlled by molecular diffusion and by the smallest scales of the velocity field. These in turn are controlled in part by ν so at first sight $\overline{\theta'^2}$ and ϵ_θ appear to depend on the magnitudes of κ and ν. However, in many cases the values of $\overline{\theta'^2}$ and ϵ_θ only depend on the fact that κ and ν are not actually zero. For example, when no external lengthscale is imposed on the scalar field (i.e. scalar fluctuations are produced by homogeneous turbulence working on a uniform-gradient mean scalar field) the fluctuation field is also homogeneous and for large Pe and Reynolds number the scales on which fluctuations are produced are sufficiently far removed from the scales on which dissipation occurs for similarity scaling arguments to be used (Batchelor 1959; Batchelor, Howells & Townsend 1959). Then ϵ_θ and $\overline{\theta'^2}$ are independent of κ and ν and

$$\epsilon_\theta = \frac{\overline{\theta'^2}\,\sigma_w}{L_\theta}, \tag{1.2}$$

where L_θ is a lengthscale determined by the large-scale properties of the turbulence.

However, in many cases of practical interest an external lengthscale is imposed on the scalar field by a finite source lengthscale σ_0 (for example, the radius of a chimney emitting effluent). Then the scale ratio σ_0/L influences the evolution of the scalar field.

If σ_0 is $O(L)$ or greater the scaling arguments referred to above, and hence (1.2), should still apply. However σ_0 may be comparable with the microscales on which molecular processes act. (For velocity fluctuations the Kolmogorov microscale is $\eta = (\nu^3/\epsilon)^{\frac{1}{4}} \sim Re^{-\frac{3}{4}} L$, where ϵ is the rate of dissipation of turbulent kinetic energy per unit mass, and for scalar fluctuations there are different microscales depending on the value of the Prandtl number $Pr = \nu/\kappa$. For $Pr < 1$ this is the diffusion cut-off length, $\lambda_c = (\kappa^3/\epsilon)^{\frac{1}{4}} \sim Pe^{-\frac{3}{4}} L$ and when $Pr > 1$ the diffusive/straining microscale is $\lambda_c = (\kappa^2\nu/\epsilon)^{\frac{1}{4}} \sim Re^{-\frac{1}{4}} Pe^{-\frac{1}{2}} L$ (Townsend 1976, p. 346)). Then, initially at least, both $\overline{\theta'^2}$ and ϵ_θ depend explicitly on κ and ν. The main aim of this paper is to examine the extent to which σ_0/L, κ and ν explicitly influence the evolution of the θ-field.

Exact results are available for the case $\kappa = 0$ (or more particularly, $\epsilon_\theta = 0$). It can be shown by Eulerian methods (Chatwin & Sullivan 1979) or from marked-particle statistics (Corrsin 1952; Durbin 1980) that a cloud of material dispersing in incompressible turbulence conserves $\int \theta^2 \, \mathrm{d}V$ (where the integral is over all space) and that at large time

$$\frac{\overline{\theta'^2}}{\bar{\theta}^2} \to O\left(\frac{\sigma}{\sigma_0}\right)^3 \to \infty \quad \text{as } t \to \infty, \tag{1.3}$$

where $\sigma(t)$ is a measure of the size of the cloud. The fact that this limit does not accord with observation strongly suggests that molecular diffusion cannot be neglected.

The only exact solutions for the evolution of a scalar field under the combined action of molecular diffusion and turbulence are for very short times ($t \ll t_k = (\nu/\epsilon)^{\frac{1}{2}}$) and for very small initial volumes ($\sigma_0 \ll \eta$) when the turbulence can be approximated by a random linear-strain field (Townsend 1954; Saffman 1960; Chatwin & Sullivan 1979). A recent extension using path integral techniques has been developed by Drummond (1982). All other calculations involve some assumptions about the joint statistics of the velocities of fluid elements and the contaminant molecules. We seek here to use such an approximate calculation and first briefly review the range of possible approaches.

Chatwin & Sullivan (1979) suggested that the linear-strain results may be valid for $t \gg t_k$ and $\sigma \gg \eta$ with the result that ultimately (for a diffusing cloud)

$$\frac{\overline{\theta'^2}}{\bar{\theta}^2} \to O\left(\frac{\sigma}{\lambda_c}\right)^n \to \infty \quad \text{as } t \to \infty, \tag{1.4}$$

where n lies between 1 and 2. This is unrealistic because the linear-strain model takes no account of the effect of the energetic eddies, which on scales larger than η fold back onto themselves the strands and sheets of contaminant drawn out by the strain field. This is an essential feature of turbulent mixing which in practice increases the rate of dissipation sufficiently to prevent the divergent growth of fluctuations evident in (1.4).

Quite different results for the change of $\overline{\theta'^2}/\bar{\theta}^2$ were obtained from calculations of the motion of pairs of marked fluid particles. Even then differences were found depending on the assumptions about these relative motions. Durbin (1980) assumed (following Richardson 1926) that pairs of particles separate at a rate that is a random function but dependent on their *instantaneous* distance apart. For a planar cloud this model implies that, as $t \to \infty$,

$$\int \overline{\theta^2} \, \mathrm{d}V \to 0 \tag{1.5a}$$

and

$$\frac{\overline{\theta'^2}}{\bar{\theta}^2} \sim \left(\frac{\sigma_0}{L}\right)^{-\frac{1}{3}} - 1 \quad \text{if } \sigma_0 \ll L. \tag{1.5b}$$

The model accounts for molecular dissipation in the mixing of pairs of fluid elements with different concentrations; this is implicit in the result (1.5*a*). The probability distribution of particle spacing is highly non-Gaussian and leads to contaminant clouds having a very streaky structure.

On the other hand if it is assumed, following Batchelor (1952), that the particles move apart at a rate dependent on the *average* distance apart of all particles in the ensemble at that time, the distribution of pair separations is Gaussian and Sawford (1983*a*) has shown that this implies, as $t \to \infty$,

$$\frac{\overline{\theta'^2}}{\bar{\theta}^2} \sim \tfrac{1}{2}\rho^2 \sim t^{-1}, \tag{1.6}$$

where ρ is the correlation between the positions of the two particles. Thomson's (1985) recent extension of Durbin's model includes a mean (as well as fluctuating) separation velocity and also predicts that $\overline{\theta'^2}/\bar{\theta}^2$ vanishes like (1.6). These models are readily applied to an arbitrary scalar source distribution.

Various approaches ranging from the heuristic treatments of Richardson (1926) and Batchelor (1952) to the modern two-point turbulence closure theories (Lundgren 1981; Herring *et al.* 1982; Larchevêque & Lesieur 1981) lead to a diffusion equation for the pair separation. The two-point closure models also give an equation for the pair diffusivity. This approach has been restricted to decaying homogeneous scalar fields and has not yet been applied to the practical problems involving inhomogeneous scalar fields which are of interest here. Whenever scalar fluctuations are produced by mean-field gradients it is necessary also to consider centre-of-mass motions (in fact the full two-particle displacement probability density function (p.d.f.)).

Various second-order Eulerian closures have been proposed in which the dissipation term is modelled explicitly in terms of the large-scale properties of the turbulence. Sykes, Lewellen & Parker (1984), used (1.1) and assumed appropriate similarity forms for L_θ while Newman, Launder & Lumley (1981) closed an equation for $\mathrm{d}\epsilon_\theta/\mathrm{d}t$. The model of Sykes *et al.* (1984) has been applied to the dispersion of a cloud and at large times predicts a balance between production and dissipation of $\overline{\theta'^2}$ with

$$\frac{\overline{\theta'^2}}{\bar{\theta}^2} \sim t^{-\frac{1}{2}} \quad \text{as } t \to \infty, \tag{1.7}$$

i.e. at large time the relative intensity of fluctuation decreases as in the prediction (1.6). This is quite different to the predictions of steady or growing intensity in (1.5) or (1.3) respectively. A fundamental problem with second-order Eulerian closure schemes in general is that each release must be treated separately and independently (Sykes *et al.* 1984; Lumley 1983), i.e. correlations between the θ' fields from separate releases cannot be properly accounted.

The simplest approach to calculating scalar fluctuations is that these are produced by the rapid mixing between fluid elements having the same concentration as the local mean concentration a distance L apart, so that $\overline{\theta'^2}$ is proportional to the local *mean* concentration gradient, i.e. $\overline{\theta'^2} \sim L^2(\nabla\bar{\theta})^2$. In that case, for a planar cloud where $\bar{\theta} = \theta_0 \exp(-z^2/2\sigma^2(t))$,

$$\frac{\overline{\theta'^2}}{\bar{\theta}^2} \sim \frac{z^2}{\sigma^4(t)} \sim z^2 t^{-2} \quad \text{as } t \to \infty. \tag{1.8}$$

This implies that fluctuations vanish at the centre of the cloud where $\nabla\bar{\theta} = 0$, a result not borne out by experiments (Warhaft 1984; Stapountzis *et al.* 1986).

It is apparent from this brief review that existing theories for the evolution of scalar

fields in turbulence are based on many different assumptions and differ widely in their predictions for the asymptotic level of fluctuations. The experimental evidence does not conclusively distinguish between these theories because homogeneous turbulence decays and the scale of turbulence increases. Under these conditions the Lagrangian models and the Eulerian second-order closure model predict that $\overline{\theta'^2}/\overline{\theta}^2$ asymptotes to a constant in agreement with the grid-turbulence experiments of Warhaft (1984) and Stapountzis *et al.* (1986).

We have noted that Eulerian models implicitly or explicitly make assumptions about the scalar field as well as the velocity field whereas the Lagrangian models' assumptions concern the statistics of the displacement of marked particles and so are independent of the scalar field. The latter are thus potentially of greater generality provided that their uncertainties can be defined.

Of the Lagrangian models, the non-Gaussian model of Durbin (1980) is the most successful so far in describing a number of wind-tunnel experiments for the evolution of $\overline{\theta'^2}$ as a function of the lengthscale of the source, which affects $\overline{\theta'^2}$ for times much larger than the integral length scale L/σ_w (Durbin 1982), and for the behaviour of $\overline{\theta'^2}$ behind a single line source (Stapountzis *et al.* (1986)). In addition, molecular processes can be explicitly included through a very natural extension to Durbin's model so that their effect on ϵ_θ is *predicted*. Durbin's (1980) model thus represents a useful starting point for the present study.

Durbin (1980) justified the neglect of explicit molecular effects in his model by arguing that for Pr of $O(1)$, large Re and σ_0 of $O(L)$

(*a*) small-scale structure of the scalar field is eliminated by molecular diffusion (and its interaction with the turbulence) so that large scales contain all the variance;

(*b*) these large scales are only weakly influenced (to $O(Re^{-1})$) by molecular processes;

(*c*) turbulent mixing between pairs of marked particles with different concentrations (caused by their separation at previous times) determines the rate of dissipation of $\overline{\theta'^2}$, which is predicted without any detailed consideration of how microscale mixing occurs.

These kinds of assumptions are also used (more or less explicitly) in the other analyses of scalar fluctuations already mentioned. By modelling the molecular processes at the smallest scale we are able to test explicitly these assumptions for a range of source size σ_0 and a range of Prandtl number Pr. The aim is to examine and justify the extent to which moments of the scalar field θ can be calculated solely from the displacement of particles, an approach that has led to many interesting and practical results (Durbin 1980, 1982; Sawford 1983*b*). We focus our attention on answering the following questions, our answers to which are summarized in the concluding section.

1. The exact results of Townsend (1954), Saffman (1960) and Chatwin & Sullivan (1979) show molecular effects to be explicitly important for small sources and small time. Are there circumstances when molecular effects persist for all time? Under what circumstances is dissipation correctly accounted for by marked-particle statistics?

2. How does the intensity of scalar fluctuations in a cloud, $s = [\overline{\theta'^2}/\overline{\theta}^2]^{\frac{1}{2}}$, vary when t is large? Does Durbin's model prediction that s is constant still hold when molecular processes are important? As well as being of practical importance, this is also a fundamental question since if s is of $O(1)$, then local mixing-length arguments (based on a mixing length dependent on the turbulence scales) cannot describe the eventual development of clouds and plumes. It means that there is always almost uncontaminated space between contaminated spaces.

3. Durbin's model for marked-particle motions predicts σ_0/L to be an important parameter which influences the evolution of fluctuations even at large time. How is this prediction influenced by the explicit inclusion of molecular processes?

4. What is the importance of the fluid Prandtl number and how do the exact results available for $\kappa = 0$ fit into the overall picture?

5. At what stages in the dispersion of a cloud does dissipation become important? On what timescale does $\overline{\theta'^2}$ develop? We would like to relate these stages to the natural timescales of the turbulence, the integral timescale t_L and the Kolmogorov timescale t_k, to timescales that involve the imposed lengthscale ratio, σ_0/L, and to timescales over which the assumption of stationary homogeneous turbulence breaks down in real turbulent flows.

6. What is the effect of instrumental averaging? Durbin (1980) argued that it plays a similar role to molecular diffusion in eliminating fluctuations on small scales, although the justification for using the statistics of marked particles to calculate $\overline{\theta'^2}$ is less clear on these grounds since instrumental averaging achieves instantaneously what molecular diffusion does continuously.

Answers to these questions represent the main focus of the present paper. However, in extending Durbin's model we can also analyse in detail laboratory-scale turbulent-diffusion experiments where Re is insufficiently large to separate the energy-containing and dissipative scales of the turbulence and σ_0 is of $O(\eta)$. This is considered by Stapountzis *et al.* (1986).

2. Model development

2.1. *Statistical theory*

We consider dispersion from instantaneous plane sources in three-dimensional stationary homogeneous turbulence. Since the concept of a marked fluid particle is no longer useful once we explicitly include molecular diffusion we formulate our theory in terms of the statistics of the displacement of marked molecules that have a random thermal or Brownian motion relative to the fluid continuum. Expressions for moments of the scalar field are derived in the Appendix. They are formally identical to those derived for marked-particle statistics (see e.g. Monin & Yaglom 1971, p. 589; Batchelor 1952). In particular, the ensemble mean concentration is (with the restrictions on the source distribution noted above)

$$\overline{\theta}(z,t) = \int_{-\infty}^{\infty} P_1(z',0;z,t)\,S(z')\,\mathrm{d}z' \tag{2.1}$$

and the two-point covariance at a single time is

$$\overline{\theta(z_1,t)\,\theta(z_2,t)} = \int_{-\infty}^{\infty}\int P_2(z_1',z_2',0;z_1,z_2,t)\,S(z_1')\,S(z_2')\,\mathrm{d}z_1'\,\mathrm{d}z_2'. \tag{2.2}$$

$S(z)$ is the source distribution and P_1 and P_2 are one- and two-point displacement p.d.f's. These equations can be interpreted in terms of dispersion of marked molecules either forwards in time from the source location $(z',0)$ to the sampling location (z,t), or backwards in time from the sampling location to the source. For incompressible flow, Lundgren (1981) and Egbert & Baker (1984) show these interpretations to be equivalent. Durbin's (1980) model and the extension we develop here treat only one component of the velocity field. As in other diffusion analyses of one component of velocity, the incompressibility constraint is not considered. In that case Sawford (1984) has noted that the reverse-dispersion interpretation is appropriate since it deals

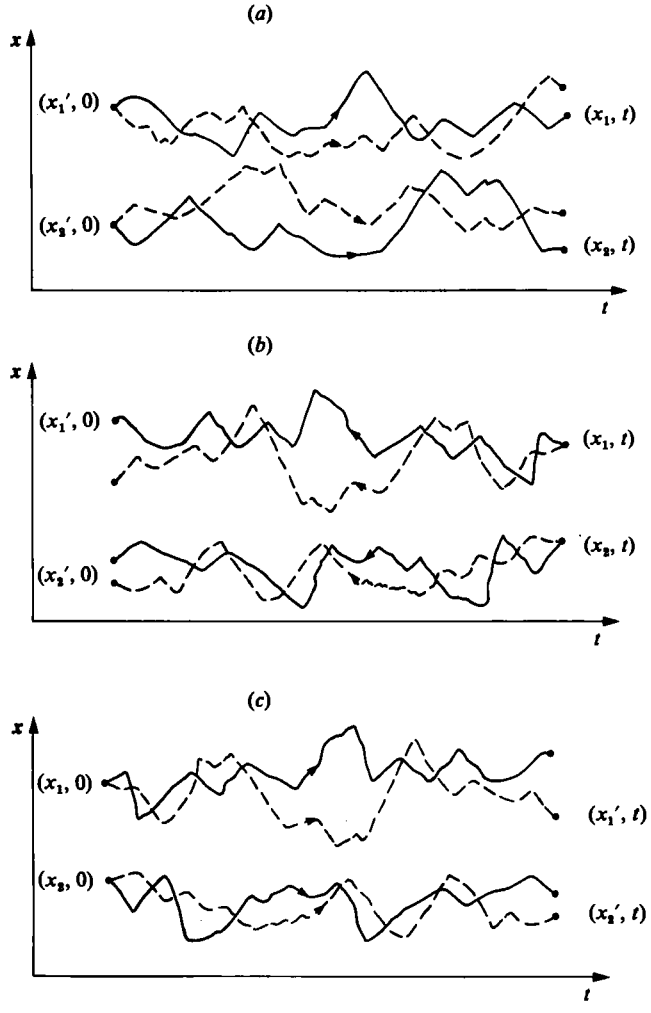

FIGURE 1. Schematic showing two realizations of the trajectories of a pair of particles (*a*) forwards in time from given initial points x_1', x_2'; (*b*) backwards in time from given final points x_1, x_2; (*c*) trajectories from the given points x_1, x_2 after the time transformation (2.3).

with the mass-specific concentration and so eliminates spurious concentration fluctuations which would otherwise (i.e. in the forward interpretation) arise from fluctuations in the total fluid density. This point is brought out more fully in the Appendix. Note that our notation in (2.1) and (2.2) emphasizes the reversed-dispersion interpretation. Thus $P_2(z_1', z_2', 0; z_1, z_2, t)$ is the probability density that molecules at z_1 and z_2 at time t *came* from z_1' and z_2' at time $t = 0$.

In order to avoid the inconvenience of negative travel times as molecules disperse backwards in time, we take advantage of stationarity to write

$$P_1(z_1', 0; z_1, t) = P_1(z_1', t; z_1, 0)$$

and

$$P_2(z_1', z_2', 0; z_1, z_2, t) = P_2(z_1', z_2', t; z_1, z_2, 0). \tag{2.3}$$

and thus we have the convenience of actually carrying out the dispersion calculation forwards in time. Note though that dispersion still commences at the field points z_1

and z_2 and finishes at the source points z_1' and z_2'. The concept of reversed dispersion and the time transformation (2.3) are illustrated schematically in figure 1.

Here we are most interested in the mean-square concentration field $\overline{\theta^2}(z,t)$ at the point z and thus in the relative dispersion of molecules in the limit in which their initial separation vanishes. Formally

$$\overline{\theta'^2}(z,t) = \int_{-\infty}^{\infty}\int P_2(z_1', z_2', t; z, z, 0)\, S(z_1')\, S(z_2')\, \mathrm{d}z_1'\, \mathrm{d}z_2'. \tag{2.4}$$

For a finite sampling volume the initial conditions on the dispersion process are different since the pair of molecules can then be anywhere within the sample volume at the measurement time. The effect of sample volume on $\overline{\theta'^2}$ is discussed in §4.

For later reference we also write here the corresponding result for Corrsin's (1952) one-particle analysis

$$\overline{\theta_{\mathrm{u}}^2}(z,t) = \int P_1(z', t; z, 0)\, S^2(z')\, \mathrm{d}z', \tag{2.5}$$

where we have followed Durbin's (1980) notation in the use of the subscript u. The relationship between this theory (which ignores dissipation of $\overline{\theta'^2}$ and leads to the result (1.3)) and the two-point theory is discussed in §4.

2.2. *Molecular trajectories*

Equations (2.1) and (2.2) are exact formal expressions relating concentration statistics to the displacement statistics of molecules. An exact specification of P_1 and P_2 is, of course, not generally possible. Instead we propose to develop a model for molecular trajectories from which the statistics P_1 and P_2, and hence $\overline{\theta}$ and $\overline{\theta'^2}$, can be generated.

Following Saffman (1960), at any instant the motion of a single marked molecule can be partitioned into a turbulent component (the motion of the fluid particle containing the molecule at that time) and a random thermal or Brownian component. Since the timescale for molecular collisions is much smaller than the smallest timescale of the turbulence (the Kolmogorov timescale t_{k}) the Brownian motion is random and independent of the turbulent motion, so that (for a single molecule)

$$\mathrm{d}z = w_{\mathrm{p}}\, \mathrm{d}t + \sqrt{(2\kappa)}\, \mathrm{d}W_{\mathrm{d}} \tag{2.6}$$

where $\mathrm{d}W_{\mathrm{d}}$ is a Gaussian white-noise process with zero mean value and variance $\mathrm{d}t$. Here w_{p} is the velocity of the fluid particle containing the marked molecule at the given instant (i.e. the continuum velocity of the fluid) and we need to simulate the statistics of those fluid particles along the trajectory of the marked molecule.

Since molecular diffusion proceeds independently for a pair of molecules, by analogy with (2.4) we can write

$$\mathrm{d}\Delta = \mathrm{d}\Delta_{\mathrm{p}} + \sqrt{(2\kappa)}\, \mathrm{d}W_{\mathrm{d}}' \tag{2.7}$$

and

$$\mathrm{d}\Sigma = \mathrm{d}\Sigma_{\mathrm{p}} + \sqrt{(2\kappa)}\, \mathrm{d}W_{\mathrm{d}}'', \tag{2.8}$$

where $\mathrm{d}W_{\mathrm{d}}'$ and $\mathrm{d}W_{\mathrm{d}}''$ are independent Gaussian white-noise processes. For convenience, (2.7) and (2.8) have been written in 'separation', $\Delta = (z_1 - z_2)/\sqrt{2}$, and 'centre-of-mass', $\Sigma = (z_1 + z_2)/\sqrt{2}$, coordinates.

Equations (2.5) and (2.6) show how molecular diffusion enters our problem. In the following sections we review Durbin's (1980) model for $\mathrm{d}\Delta_p/\mathrm{d}t$ and $\mathrm{d}\Sigma_{\mathrm{p}}/\mathrm{d}t$ for turbulence with an infinite inertial subrange and then describe our extension to include the effect of viscous dissipation on the turbulence.

2.3. *Durbin's (1980) model for particle-pair trajectories*

Durbin (1980) ignored the molecular diffusion terms in (2.7) and (2.8) and modelled the fluid-particle motions in turbulence with an inertial subrange extending to infinitely large wavenumbers, i.e. he also ignored the effect of viscous dissipation on the turbulence. Dropping the subscript p, his model is

$$\frac{\mathrm{d}\Delta}{\mathrm{d}t} = R^{\frac{1}{2}}(\Delta)\, U' \tag{2.9}$$

and

$$\frac{\mathrm{d}\Sigma}{\mathrm{d}t} = [(2-R(\Delta)]^{\frac{1}{2}}\, U'', \tag{2.10}$$

where $2\sigma_w R(\Delta)$ is an Eulerian structure function (Townsend 1976, p. 11), U' and U'' are independent Uhlenbeck Ornstein (UO) random variables, and we now identify σ_w as the standard deviation of the w-component of the vertical velocity. The use of an Eulerian structure function in (2.9) and (2.10) implies an independence hypothesis similar to that of Corrsin (1959) and is correct in both the small- and large-time limits. For convenience we refer to the normalized quantity $R(\Delta)$ as the structure function.

The UO process by which U' and U'' are generated

$$\mathrm{d}U = -U/t_{\mathrm{L}}\,\mathrm{d}t + \sigma_w\left(\frac{2}{t_{\mathrm{L}}}\right)^{\frac{1}{2}}\mathrm{d}W_t' \tag{2.11}$$

is a stationary Gaussian Markov process with mean zero, variance σ_w^2 and correlation function

$$\frac{\overline{U(\tau)\,U(\tau+t)}}{\sigma_w^2} = \exp\left(-\frac{t}{t_{\mathrm{L}}}\right), \tag{2.12}$$

and thus introduces 'memory' to the turbulent motion of each particle through the Lagrangian integral timescale t_{L}. The auto-correlation (2.12) has the appropriate Lagrangian inertial-range form, $1-t/t_{\mathrm{L}}$ for $t \ll t_{\mathrm{L}}$, (Monin & Yaglom 1975, p. 359) and vanishes for $t \gg t_{\mathrm{L}}$. Thus each particle 'forgets' its original velocity after a time $t \gg t_{\mathrm{L}}$. The dispersion of the UO position variable, $\zeta = \int_0^t U'(t)\,\mathrm{d}t'$ is

$$\sigma_\zeta^2 = 2\sigma_w^2 t_{\mathrm{L}}^2\left[\exp\left(-\frac{t}{t_{\mathrm{L}}}\right)+\frac{t}{t_{\mathrm{L}}}-1\right] \tag{2.13}$$

and the particle positions, z_1 and z_2, are normally distributed with variance

$$\sigma_{z_1}^2 = \sigma_{z_2}^2 = \sigma_\zeta^2. \tag{2.14}$$

For the Eulerian structure function, Durbin (1980) used

$$R(\Delta) = [\Delta^2/(\Delta^2+L^2)]^{\frac{1}{3}}, \tag{2.15}$$

where we now identify L as the Eulerian integral lengthscale. The form (2.5) is a simple interpolation between the inertial-subrange limit

$$R(\Delta) = \left(\frac{\Delta}{L}\right)^{\frac{2}{3}} = \frac{C\epsilon\Delta^{\frac{2}{3}}}{2\sigma_w^2} \quad (\Delta \ll L) \tag{2.16}$$

(Monin & Yaglom 1975, p. 353) and the large-scale limit of vanishing correlation,

$$R(\Delta) \to 1 \quad \left(\frac{\Delta}{L} \to \infty\right) \tag{2.17}$$

In (2.6) the semi-empirical result (Townsend 1976, p. 61)

$$\epsilon = 0.8\sigma_w^3/L \tag{2.18}$$

has been used with $C \approx 2\frac{1}{3}$.

It is instructive to examine the influence of $R(\Delta)$ on the relative motion of the pair of particles. At small separation $R(\Delta)$ is small and so $\mathrm{d}\Delta/\mathrm{d}t$ tends to be small, reflecting the fact that on average small eddies, with only a small fraction of the turbulent energy, are responsible for separating the particles. Of course, in any realization the positions of large eddies can be such as to influence $\mathrm{d}\Delta/\mathrm{d}t$ and cause the particles to separate more rapidly, and this possiblity is accommodated by the fact that U' is a random velocity which may be large for a particular realization. At large separations where $R(\Delta) \to 1$, $\mathrm{d}\Delta/\mathrm{d}t$ is independent of Δ, the pair of particles move independently and turbulence on all scales influences the rate of separation of the particles. Thus the structure function determines the proportion of turbulence energy that is effective in separating particles, the residue being responsible for centre-of-mass motion as is clear from (2.10). One consequence of this filtering process is the accelerating nature of relative dispersion in the inertial range where the form (2.16) ensures the growth law $\overline{\Delta^2} \sim t^3$ (Batchelor 1950) which applies for $t \ll t_{\mathrm{L}}$. At larger time, $t \gg t_{\mathrm{L}}$, $\overline{\Delta^2} \gg L^2$ so that $R(\Delta) \to 1$ for most pairs in the ensemble and $\overline{\Delta^2} \to \sigma_z^2$, the single-particle dispersion. Equivalently the correlation between the positions of the two particles vanishes,

$$\frac{\overline{z_1 z_2}}{\sigma_z^2} = 1 - \frac{\overline{\Delta^2}}{\sigma_z^2} \to 0 \quad (t \gg t_{\mathrm{L}}). \tag{2.19}$$

In the context of scalar fluctuations an important property of (2.9) is that it is a *nonlinear* stochastic differential equation – the rate of separation depends on the instantaneous separation through the factor $R(\Delta)$. As a consequence Δ is not a Gaussian variable and, although at large time the motion of the two particles is uncorrelated, their displacements are not independent. Sawford (1983*a*) showed that this property is essential for the maintenance of internal structure within a dispersing cloud. Although models in which $\mathrm{d}\Delta/\mathrm{d}t$ depends on $\overline{\Delta^2}$ (rather than Δ itself) can be formulated to give the inertial-range t^3 law for $\overline{\Delta^2}$ (Sawford 1982; Lamb 1981) such models are Gaussian in the sense that P_2 and the distribution of separations are Gaussian and, at large time, smooth-out structure in the scalar field leaving only those fluctuations due to bulk motion of the cloud. (Field observations by Jones (1982) demonstrate the highly intermittent and unsmoothed-out structure of plumes even when they are thin enough to meander around.)

2.4. *Viscous-dissipation-range structure*

Within the viscous-dissipation range, the Eulerian structure function has the form (Monin & Yaglom 1975, p. 353)

$$R(\Delta) \sim \frac{\epsilon}{\nu}\frac{\Delta^2}{\sigma_w^2} \quad (\Delta \ll \eta). \tag{2.20}$$

Here we extend Durbin's model to include the effect of viscous dissipation on the flow field by a modification to (2.15) to include the limit (2.20). In particular we replace (2.15) by

$$R(\Delta) = \left(\frac{\Delta^2}{\eta^2/\alpha^3 + \Delta^2}\right)^{\frac{2}{3}} \left(\frac{\Delta^2}{L^2 + \Delta^2}\right)^{\frac{1}{3}}. \tag{2.21}$$

for $\Delta \ll \eta$ this reduces to (2.20) with a constant of proportionality $1.16\alpha^2$, while for $\Delta \gg \eta$ (2.15) is recovered. We deal with the numerical value of α in the next section.

2.5. *Comparison with exact results*

For small time such that $t \ll t_k \sim Re^{-\frac{1}{2}} t_L$, $U'(t)$ can be replaced by $U'(0)$ in (2.7), where $U'(0)$ is Gaussian with $\overline{U'(0)} = 0$ and $\overline{U'^2(0)} = \sigma_w^2$. If we also restrict attention to small initial separations, $\Delta(0) \ll \eta$, then (2.7), (2.9) and (2.21) simplify to

$$\mathrm{d}\Delta = \alpha' \Delta U'(0)\,\mathrm{d}t + (2\kappa)^{\frac{1}{2}}\,\mathrm{d}W'_{\mathrm{d}}, \tag{2.22}$$

where

$$\alpha' = \frac{\alpha}{L}\left(\frac{L}{\eta}\right)^{\frac{4}{3}}. \tag{2.23}$$

Integrating (2.22) and averaging over both W'_{d} and $U'(0)$ we have

$$\overline{\Delta^2} - \Delta_0^2 = 2\Delta_0^2 \alpha'^2 \sigma_w^2 t^2 + 2\kappa t + \tfrac{4}{3}\kappa \alpha'^2 \sigma_w^2 t^3 + O(t^4), \tag{2.24}$$

which agrees with Saffman's (1960) exact result for the dispersion of a spot of contaminant about its centre-of-mass,

$$\overline{\Delta^2} - \Delta_0^2 = \tfrac{1}{3}\Delta_0^2 \left(\frac{t}{t_k}\right)^2 + 2\kappa t + \tfrac{2}{9}\kappa \frac{t^3}{t_k^2} + O(t^4), \tag{2.25}$$

provided we choose

$$\alpha'^2 \sigma_w^2 = \frac{1}{6t_k^2}, \tag{2.26}$$

or from (2.18) and (2.23)

$$\alpha^2 = 0.128. \tag{2.27}$$

Thus our model not only reproduces the leading-order terms for the growth of a puff due to the strain field acting over the finite initial separation and to molecular diffusion (the first two terms on the right-hand side of (2.25)) but also correctly represents the leading term due to the interaction of molecular diffusion with the turbulence (the third term).

The numerical value (2.27) for α means that the dissipation correction in (2.21) is effective over separations considerably greater than η. For example, $R(\Delta)$ is reduced by 10 % for $\Delta/\eta = 11$. This is consistent with the measurements by Townsend (1951) which show that the linear-strain flow is applicable to about 15η.

2.6. *Numerical details*

Pairs of molecular trajectories can be found by solving finite-difference versions of (2.7)–(2.11) with (2.21). In the finite-difference approximation $\mathrm{d}W'_t$ is replaced by $(\Delta t)^{\frac{1}{2}}\chi'_n$, where $t = n\Delta t$ and $[\chi'_n]$ is a set of independent standard Gaussian random variables. As indicated in figure 1, we solve for the source positions z'_1, z'_2 given the field position. Having found z'_1 and z'_2 we assign probability densities $S(z'_1), S(z'_2)$ that the molecules are contaminant species. In this way $S_n(z'_1)$ and $S_n(z'_2)$ for the nth molecule pair can be found as a function of the time of measurement (travel time) and position.

Then

$$\overline{\theta^2} = \frac{1}{N}\sum_1^N S_n(z'_1)\,S_n(z'_2), \tag{2.28}$$

$$\bar{\theta} = \frac{1}{2N}\sum_1^N S_n(z'_1) + S_n(z'_2), \tag{2.29}$$

are computed at fixed t, where N is the total number of pair trajectories.

There are three constraints on the size of the time step based on the requirements $\mathrm{d}U' \ll U'$, $\mathrm{d}\Delta \ll \Delta$ and $\mathrm{d}\Sigma \ll \Sigma$. From (2.9)–(2.11) these constraints are

$$\mathrm{d}t/t_{\mathrm{L}} \ll 1, \tag{2.30}$$

$$\mathrm{d}t/t_{\mathrm{L}} \ll (\Delta/L)/R_{\Delta}^{\frac{1}{2}}(\Delta), \tag{2.31}$$

$$\mathrm{d}t/t_{\mathrm{L}} \ll \sigma_{\zeta}/L. \tag{2.32}$$

Of these, (2.30) and (2.31) are the controlling conditions for $t > t_{\mathrm{L}}$ and $t < t_{\mathrm{L}}$ respectively. From (2.13) the condition (2.32) reduces to $\mathrm{d}t \ll (tt_{\mathrm{L}})^{\frac{1}{2}}$ for $t \gg t_{\mathrm{L}}$ and hence is automatically satisfied. For $t \ll t_{\mathrm{L}}$ (2.32) reduces to $\mathrm{d}t \ll t$ and so requires the obvious condition that the initial step size be small compared with the smallest time of interest.

3. Distribution of separations

The distribution of separations of molecules $P(\Delta, t; \Delta_0, 0)$ is of interest because it reflects directly the important effects of viscosity and molecular diffusion and also because it plays a central role in the theory of scalar fluctuations. Since the UO variables U' and ζ' are Gaussian random variables it follows from (2.9) that (Durbin 1980)

$$P(\Delta,t; \Delta_0, 0) = \frac{R_{\Delta}^{-\frac{1}{2}}(\Delta)}{(2\pi)^{\frac{1}{2}}\sigma_{\zeta}} \exp\left(\frac{-[G(\Delta)-G(\Delta_0)]^2}{2\sigma_{\zeta}^2}\right), \tag{3.1}$$

where

$$G(\Delta)-G(\Delta_0) = \int_{\Delta_0}^{\Delta} R_{\Delta}^{-\frac{1}{2}}(\Delta')\,\mathrm{d}\Delta'.$$

Because (3.1) contains both fixed (Δ_0, η and L) and variable (σ_{ζ}) lengthscales $P(\Delta, t; \Delta_0, 0)$ is not generally self-similar. However, with increasing time the exponential factor in (3.1) becomes negligibly different from unity on an ever-larger scale since σ_{ζ} increases with time. In particular,

$$(2\pi)^{\frac{1}{2}}\sigma_{\zeta} P(\Delta, t; \Delta_0, 0) \to R_{\Delta}^{-\frac{1}{2}}(\Delta) \tag{3.2}$$

for $\sigma_{\zeta} \gg |G(\Delta)-G(\Delta_0)|$, i.e. with increasing time $(2\pi)^{\frac{1}{2}}\sigma_{\zeta} P(\Delta, t; \Delta_0, 0)$ approaches a time-independent asymptote on an ever-increasing scale.

In order to provide a reference against which to compare our present model we briefly summarize some important features of $P(\Delta, t; \Delta_0, 0)$ for Durbin's marked-particle model, i.e. (3.1) with the structure function (2.15). We noted in §2 that, for that model, Δ is not normally distributed. Indeed on small scales, $\Delta \ll L$, (3.2) reduces to

$$(2\pi)^{\frac{1}{2}}\sigma_{\zeta} P(\Delta, t; \Delta_0, 0) \to \left(\frac{\Delta}{L}\right)^{-\frac{1}{3}}. \tag{3.3}$$

Compared with a normal distribution this weak singularity at the origin represents enhanced small-scale structure in the flow field and corresponds physically to the cascade of energy to small scales through the inertial subrange. Sawford (1983*a*) has shown that this non-Gaussian feature of $P(\Delta, t; \Delta_0, 0)$ is responsible for structure in the distribution of material *within* a dispersing cloud.

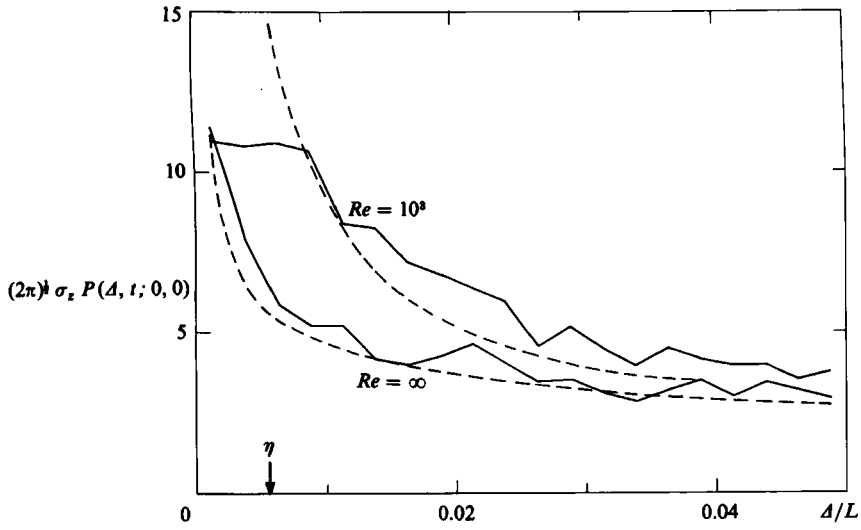

FIGURE 2. Small-scale asymptotic form of the separation p.d.f. for Durbin's (1980) model ($Re = \infty$) and the present model ($Re = 10^3$). ——, numerical computations averaged over $t/t_L = 2$, 5 and 10 using $N = 3 \times 10^4$ realizations. – – –, $R^{-\frac{1}{2}}(\Delta)$ from (2.15) and (2.21).

For the extended model developed in this paper, the structure function (2.21) results in the strong singularity†

$$(2\pi)^{\frac{1}{2}}\sigma_\zeta P(\Delta, t; \Delta_0, 0) \to (\alpha Re^{\frac{1}{2}}\Delta)^{-1} \tag{3.4}$$

within the dissipation subrange, $\Delta \ll \eta$. Ignoring for the moment the effects of molecular diffusion, we see from (3.4) that the effect of viscous dissipation is to enhance $P(\Delta, t; \Delta_0, 0)$ relative to the inertial-range form. The physical explanation is that viscosity greatly reduces the turbulent kinetic energy available to separate material points on these scales. Implications for the scalar field are discussed in detail in §4, but we anticipate that the increase in $P(\Delta, t; \Delta_0, 0)$ for $\Delta \ll \eta$ corresponds to an increase in the small-scale structure of the scalar field as reflected, for example, by the Batchelor (1959) viscous–convective subrange of the scalar variance spectrum.

We have not obtained an analytic expression for the effect of molecular diffusion on the distribution of separations and so draw on results from numerical integrations. From our understanding of the role of molecular diffusion in eliminating small-scale structure of the scalar field we expect molecular diffusion to smooth $P(\Delta, t; \Delta_0, 0)$ on small scales. This is confirmed by figure 2. There we have plotted $(2\pi)^{\frac{1}{2}}\sigma_z P(\Delta, t; 0, 0)$ over separations $\Delta \ll L$ for our new model at $Re = 10^3$ and $Pe = 10^3$. For comparison we have also plotted results from integration of Durbin's inertial-range model. In both cases these results represent the average over times $t/t_L = 2, 5$ and 10 for which $(2\pi)^{\frac{1}{2}}\sigma_z P(\Delta, t; 0, 0)$ has essentially converged to its large-time asymptote on these scales. Statistics were accumulated over $N = 3 \times 10^4$ pair trajectories. Figure 2 also shows as broken lines $R^{-\frac{1}{2}}(\Delta)$ corresponding to (2.15) and (2.21) and the Kolmogorov lengthscale for $Re = 10^3$. For the inertial-range model the numerical results asymptote to $R^{-\frac{1}{2}}(\Delta)$ in agreement with (3.2). For $Re = 10^3$ the effect of viscous dissipation is

† In fact the singular form (3.4) extends to arbitrarily small Δ with increasing time, but at any finite time the exponential factor in (3.1) with the structure function (2.21) causes $P(\Delta, t; \Delta_0, 0)$ to vanish at the origin.

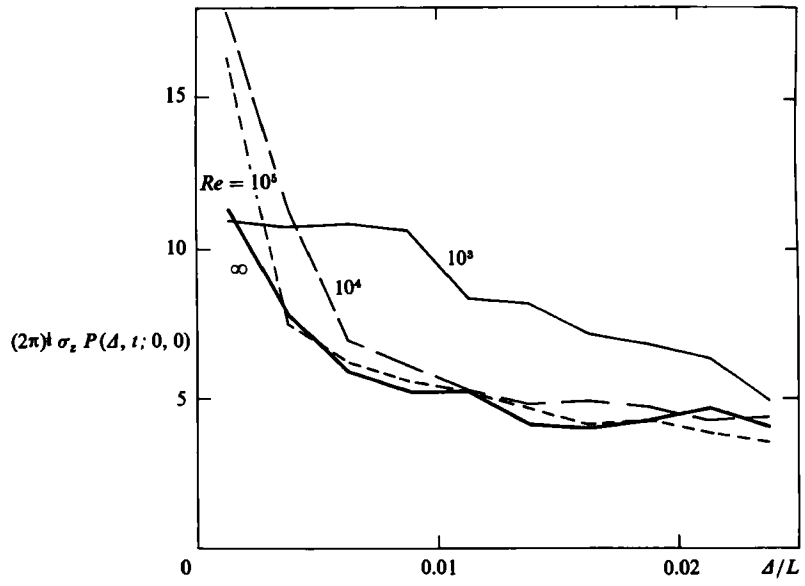

FIGURE 3. Effect of Re on the small-scale asymptotic form of the separation p.d.f. Numerical computations as in figure 2.

reflected through the $R^{-\frac{1}{2}}(\Delta)$ asymptote and the strong Δ^{-1} singularity of (3.4) as an increase in $(2\pi)^{\frac{1}{2}}\sigma_z P(\Delta, t; 0, 0)$ for separations $\leqslant 10\eta$. For this $Pr = 1$ case, molecular diffusion smooths the Δ^{-1} singularity for separations of $O(\eta)$. Thus molecular diffusion and viscosity have opposing effects; viscosity enhances $P(\Delta, t; 0, 0)$ on small scales and diffusion smooths and reduces it.

These effects are confined to scales $\leqslant 10\eta$, the range of influence of viscous dissipation being determined by the constant α in (2.21). Thus as is illustrated in figure 3 they occur on diminishing scales (relative to L) as η/L decreases with increasing Re. For $Pr = 1$ we conclude that the inertial-range marked-particle model accurately represents $P(\Delta, t; 0, 0)$ at any arbitrarily small scale for sufficiently high Re.

More generally the lengthscale for the interaction of molecular diffusion and the turbulent flow field (the conduction cutoff length) is a function of Pr, while the direct effect of viscous dissipation occurs on the scale η regardless of Pr. Thus, as is shown in figure 4, the balance between the competing effects of viscous dissipation and molecular diffusion is a function of Pr. Since $\lambda_c \ll \eta$ for large Pr the smoothing effect of molecular diffusion, although still effective in eliminating the Δ^{-1} singularity for $\Delta \leqslant \lambda_c$, does not balance the enhancement due to the linear-strain field for $\lambda_c \leqslant \Delta \leqslant 10\eta$. On the other hand $\lambda_c \gg \eta$ for small Pr. Then viscosity and the linear-strain field are not important and molecular diffusion interacts directly with the inertial-range eddies, smoothing the $\Delta^{-\frac{1}{3}}$ singularity of (3.3). We see from figure 4 that these effects diminish with increasing scale and, although they are more dramatic at small scales for Pr not of $O(1)$, it is clear that at fixed finite Pr the inertial-range model is again accurate to arbitrarily small scales at sufficiently high Re.

In summary, the most important findings of this section are:

(i) small scales of $P(\Delta, t; 0, 0)$ are strongly affected by molecular diffusion and viscosity and these effects are permanent;

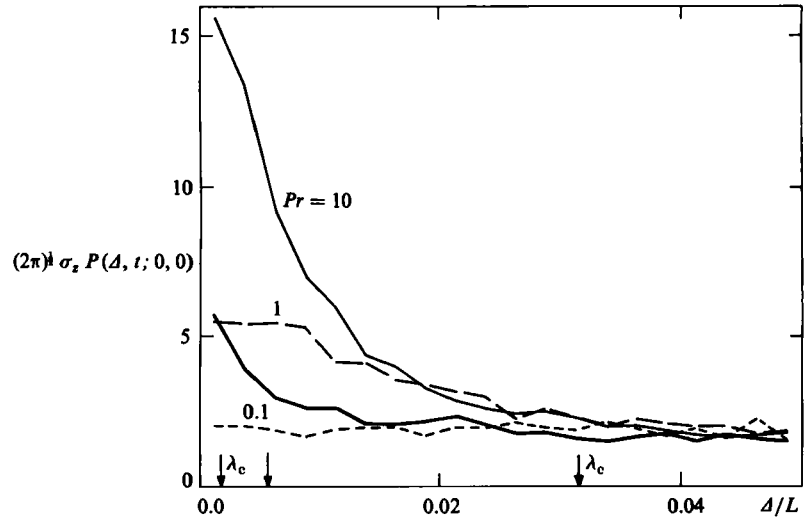

FIGURE 4. Effect of Pr on the small-scale asymptotic form of the separation p.d.f. Numerical computations as in figure 2. (——) Durbin's (1980) model ($Re = \infty$).

(ii) $P(\Delta, t; 0, 0)$ is strongly non-Gaussian for $\Delta \ll L$;

(iii) for fixed Re, molecular effects become more pronounced as Pr departs increasingly from unity;

(iv) Durbin's inertial-range model accurately represents $P(\Delta, t; 0, 0)$ on arbitrarily small scales at sufficiently high Re (in the general framework of the approach used here).

4. Concentration fluctuations

4.1. *Uniform-gradient source*

Although in this paper our interest is directed mainly to sources with a finite lengthscale we digress briefly to consider the case of fluctuations in a homogeneous scalar field resulting from a uniform-gradient source. This is a relatively simple problem which can be seen as a convenient test case for our model. With a source distribution of the form

$$S(z) = mz, \tag{4.1}$$

it follows immediately from (2.1) that

$$\bar{\theta}(z, t) = mz,$$

i.e. the mean gradient is constant for all time. Taking $z = 0$ so that $\bar{\theta} = 0$ we obtain from (2.4)

$$\frac{\overline{\theta'^2}(0, t)}{m^2} = \int P_2(z_1, z_2, t; 0, 0, 0)\, z_1\, z_2\, \mathrm{d}z_1\, \mathrm{d}z_2$$

$$= \overline{z_1(t)\, z_2(t)} \tag{4.2a}$$

$$= \tfrac{1}{2}(\overline{\Sigma^2} - \overline{\Delta^2}) \tag{4.2b}$$

$$= (\sigma_z^2 - \overline{\Delta^2}). \tag{4.2c}$$

For convenience we have dropped the primes in (4.2) (but recall that we are considering dispersion from the measurement point to the source).

Equations (4.2) are exact and do not depend on details of the trajectory model. It is illuminating to compare them with the Eulerian conservation equation which in the present case reduces to

$$\frac{\partial\overline{\theta'^2}}{\partial t} = -2\overline{\theta' w'}\frac{\partial\bar{\theta}}{\partial z} - 2\kappa\overline{(\nabla\theta')^2}. \tag{4.3}$$

The first term on the right represents production of fluctuations from the mean field while the second represents dissipation by molecular diffusion. Since the diffusivity is $\frac{1}{2}\partial\sigma_z^2/\partial t$ the production term in (4.3) is just $m^2\,\partial\sigma_z^2/\partial t$ and so matches the first term on the right-hand side of (4.2c). Comparison of (4.2c) and (4.3) then shows

$$\epsilon_\theta = 2\kappa\overline{(\nabla\theta')^2} = m^2\frac{\partial\overline{\Delta^2}}{\partial t}. \tag{4.4}$$

This equation is also exact and is a clear demonstration of the connection between the relative dispersion of material points and the dissipation of scalar variance. It also shows how a marked-particle relative-dispersion model can predict non-zero dissipation.

Corrsin's (1952) one-particle analysis ignores dissipation ($\overline{\Delta^2} = 0$) so that

$$\frac{\overline{\theta'^2_u}(0,t)}{m^2} = \sigma_z^2, \tag{4.5}$$

which is correct at small times, $t \ll t_{\mathrm{L}}$, when $\sigma_z^2 \gg \overline{\Delta^2}$. At large times $\sigma_z \sim t^{\frac{1}{2}}$ and (4.5) predicts

$$\frac{\overline{\theta'^2_u}(0,t)}{m^2} = O(t) \quad (t \gg t_{\mathrm{L}}). \tag{4.6}$$

However in stationary homogeneous turbulence $\overline{\Delta^2} \to \sigma_z^2$ at large times so that dissipation and production balance and the evolution of $\overline{\theta'^2}$ depends on the next-order terms. According to Durbin's inertial-range model these are of $O(t^{\frac{1}{2}})$, so that

$$\frac{\overline{\theta'^2}}{m^2} = O(t^{\frac{1}{2}}) \quad (t \gg t_{\mathrm{L}}). \tag{4.7}$$

The influence of molecular processes on these predictions has been examined through computations with our extended model. Figure 5 shows $\overline{\theta'^2}/m^2$ calculated over $N = 10^4$ pair trajectories out to $t/t_{\mathrm{L}} = 50$. The heavy solid and broken lines correspond to calculations using the inertial-range model and to Corrsin's one-particle model respectively, while the faint lines represent various calculations with the present model.

Despite the statistical uncertainty and the limited integration time of these calculations it is clear from figure 5 that the qualitative prediction (4.7) of Durbin's model is not dramatically altered by explicit inclusion of molecular diffusion and viscosity. Specifically, for $Pr = 1$ there is only about 10% change in $\overline{\theta'^2}/m^2$ for $Re = 10^3$ and a calculation at $Re = 10^5$ (not shown) was not significantly different from Durbin's model. As Pr departs increasingly from unity molecular processes have a greater influence on $\overline{\theta'^2}$. In the small-time limit all calculations converge to Corrsin's one-particle result (4.5). As would be expected, dissipation increases with decreasing Pr.

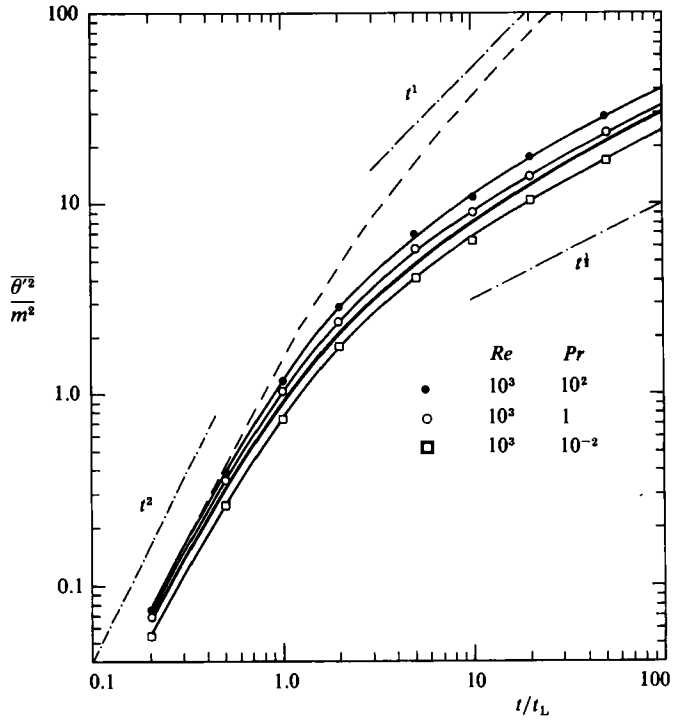

FIGURE 5. Fluctuation variance for the uniform-gradient source distribution (4.1). ——, Durbin's (1980) model ($Re = \infty$); – – –, Corrsin's (1952) one-particle model.

It has been suggested by Garrett (1983) that (4.7) is implausible and instead that $\overline{\theta'^2} \to$ const. for large t. This result is based on the semi-empirical relationship (1.2) with $\epsilon_\theta \to$ const. and $L_\theta = L$. On the other hand, (4.7) implies that the lengthscale appropriate to the scalar field at large time is the single-particle spread, i.e.

$$L_\theta \sim \sigma_z \sim t^{\frac{1}{2}}. \tag{4.8}$$

Both Garrett's (1983) result and (4.7) predict less scalar variance than the one-particle result (4.6). Thus both imply mixing of scalar elements as they migrate from the source to the measurement point and differ only in the *extent* to which mixing occurs. However there is little point in pursuing this question since the limit is unrealizable. Figure 5, for example, shows that (4.7) is approached only after many t_L and so cannot be observed in experimental homogeneous turbulence which decays significantly over an integral timescale (see §4.4).

This ambiguity in the asymptotic form of L_θ and $\overline{\theta'^2}$ disappears for the case of decaying grid turbulence. Then $L_\theta \sim L \sim \sigma_z \sim t^{1-m}$ and $\sigma_w \sim t^{-m}$, which with (4.3) or (4.4) implies $\epsilon_\theta \sim t^{1-2m}$ and so from (1.2), $\overline{\theta'^2} \sim t^{2(1-m)}$. Because there is only one lengthscale in this case both our model and Garrett's (1983) argument produce the same result. With the experimental value $m \sim \frac{1}{2}$ typical of grid turbulence, $\overline{\theta'^2} \sim t$, as has been observed experimentally (Sirivat & Warhaft 1983).

4.2. *Finite cloud source*

In this section we consider the dispersion of an instantaneously released plane cloud of finite thickness. Appropriately interpreted (e.g. through Taylor's translation

hypothesis), our results should be relevant to more practical problems such as the dispersion of a continuous plume emitted from an industrial stack. For analytical convenience we consider a Gaussian source distribution

$$(2\pi)^{\frac{1}{2}}\sigma_0 S(z) = \exp\left(-\frac{z^2}{2\sigma_0^2}\right). \tag{4.9}$$

For Durbin's inertial-range model, Sawford (1983*a*) showed that Δ and Σ are to a good approximation independent (because the factor $[2-R(\Delta)]^{\frac{1}{2}}$ in (2.10) is only a weak function of Δ) and Σ is very nearly Gaussian with variance $(1+\rho)\sigma_z^2$. This approximation becomes exact in both the small- and large-time limits. It applies equally well to the present model since the factor $[2-R(\Delta)]^{\frac{1}{2}}$ remains a weak function of Δ and molecular diffusion is modelled as a Gaussian random process. Substituting from (4.9) into (2.4) and using this independence approximation we have

$$\overline{\theta^2}(z,t) = \int_{-\infty}^{\infty}\int P(\Delta',t,0,0)\,P(\Sigma',t;\Sigma,0)\,S(\Delta')\,S(\Sigma')\,\mathrm{d}\Delta'\,\mathrm{d}\Sigma', \tag{4.10}$$

where $P(\Sigma',t;\Sigma,0)$ is the 'centre-of-mass' p.d.f. Using the near-Gaussianity of $P(\Sigma',t;\Sigma,0)$, (4.10) can be integrated to give

$$\overline{\theta^2}(z,t) = [2\pi(\sigma_0^2+(1+\rho)\sigma_z^2)]^{-\frac{1}{2}}\exp\left[\frac{-z^2}{\sigma_0^2+(1+\rho)\sigma_z^2}\right]\int_{-\infty}^{\infty} P(\Delta',t;0,0)\,S(\Delta')\,\mathrm{d}\Delta'. \tag{4.11}$$

Thus to this approximation $\overline{\theta^2}(z,t)$ is a Gaussian function of z with variance $\frac{1}{2}[\sigma_0^2+(1+\rho)\sigma_z^2]$.

The quantity in which we are most interested is the intensity of fluctuations, $s = [\overline{\theta'^2}/\overline{\theta}^2]^{\frac{1}{2}}$. Restricting attention to the centreline, $z=0$, and using $\overline{\theta}(0,t) = [2\pi(\sigma_0^2+\sigma_z^2)]^{-\frac{1}{2}}$ which follows from (2.1) for a Gaussian one-point displacement p.d.f., (4.11) reduces to

$$\frac{\overline{\theta^2}(0,t)}{\overline{\theta}^2(0,t)} = s^2(0,t)+1 = \frac{(2\pi)^{\frac{1}{2}}(\sigma_0^2+\sigma_z^2)}{[\sigma_0^2+(1+\rho)\sigma_z^2]^{\frac{1}{2}}}\int_{-\infty}^{\infty} P(\Delta',t;0,0)\,S(\Delta')\,\mathrm{d}\Delta'. \tag{4.12}$$

This relation is most revealing about the role played by source size in determining the intensity of scalar fluctuations. For small time such that $\sigma_\Delta \ll \sigma_0$ and $\sigma_\Delta \ll \sigma_z$ (i.e. $\rho \approx 1$) (4.12) reduces to Corrsin's one-particle result

$$s_u^2(0,t) = \frac{\sigma_0^2+\sigma_z^2}{\sigma_0(\sigma_0^2+2\sigma_z^2)^{\frac{1}{2}}} - 1. \tag{4.13}$$

For large time such that $\sigma_\Delta \gg \sigma_0$ and $\sigma_\Delta \approx \sigma_z$ (i.e. $\rho \ll 1$) (4.12) reduces to

$$s^2(0,t) = (2\pi)^{\frac{1}{2}}\sigma_z\int_{-\infty}^{\infty} P(\Delta',t;0,0)\,S(\Delta')\,\mathrm{d}\Delta' - 1. \tag{4.14}$$

Then the source function acts as a filter on $P(\Delta,t;0,0)$ so that only those pairs with separations not much larger than σ_0 contribute significantly to the fluctuation field. Clearly the asymptotic behaviour of $s^2(0,t)$ is determined by that of $(2\pi)^{\frac{1}{2}}\sigma_z\,P(\Delta,t;0,0)$ on scales of $O(\sigma_0)$ and so will be strongly influenced by molecular processes when σ_0 is comparable with the lengthscales η or λ_c.

Figure 6(*a*) shows $s^2(0,t)$ as a function of time for $\sigma_0/L = 0.01$ and 0.2, $Re = 10^3$ and 10^5 and $Pr = 1$, while figure 6(*b*) shows corresponding calculations for $Pr = 0.1$, 1 and 10 at $Re = 10^3$. In both cases calculations for the one-particle model and the

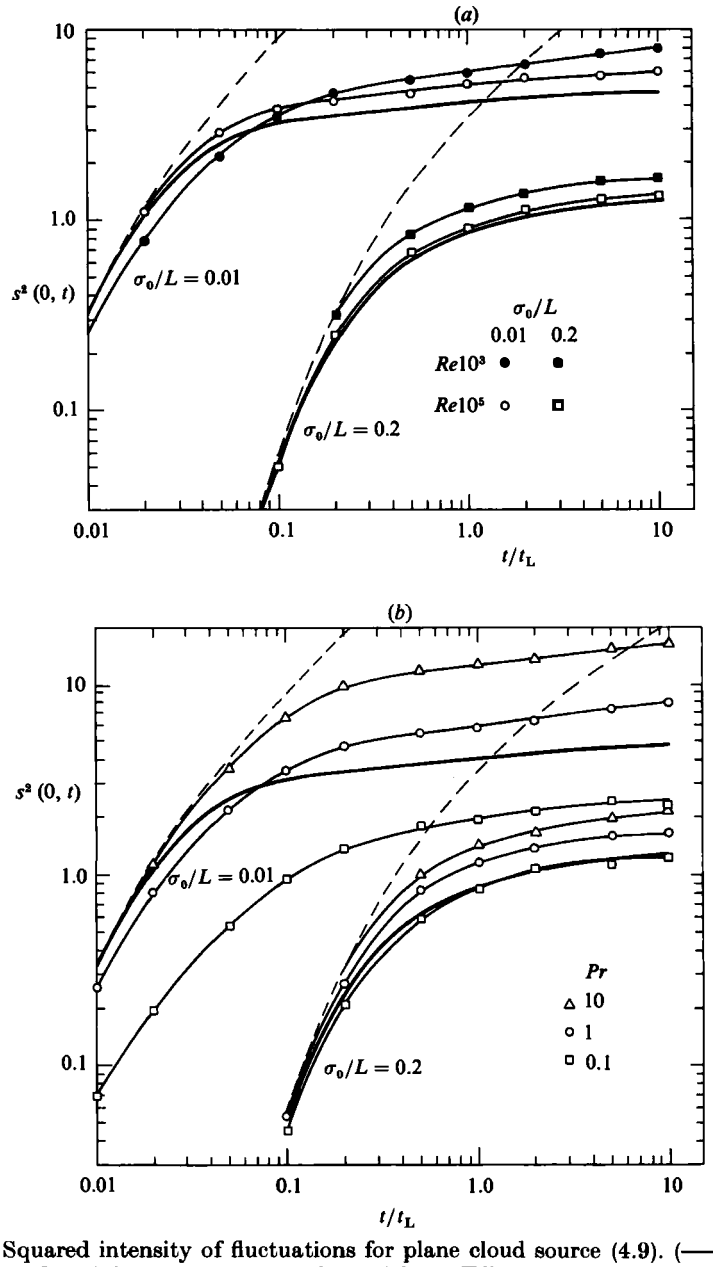

FIGURE 6. Squared intensity of fluctuations for plane cloud source (4.9). (——) Durbin's (1980) model; (– – –) Corrsin's (1952) one-particle model. (*a*) Effect of Re and source size at $Pr = 1$; (*b*) effect of Pr and source size at $Re = 10^3$.

inertial-range model are shown for comparison. Statistics were accumulated over $N = 3 \times 10^4$ pair trajectories.

All calculations approach the one-particle result at small times. At large times the two-point calculations reflect, through (4.14), the asymptotic features of $(2\pi)^{\frac{1}{2}}\sigma_z P(\Delta, t; 0, 0)$. The intensity of fluctuations asymptotes to a constant value as a result of the self-preserving, non-Gaussian asymptote of the separation p.d.f. and is a function of σ_0, Re and Pr. For fixed Pr the essential parameter determining the

extent of departure from the inertial-range model is the lengthscale ratio σ_0/η, the ratio of the scale on which separations contribute to fluctuations to the scale affected by molecular processes. Thus the inertial-range model is increasingly accurate with increasing Re and increasing σ_0. For example, the $Re = 10^5$, $\sigma_0/L = 0.2$ calculation in figure 6(*a*) indicates that the inertial-range model is accurate to $\sim 10\%$ for $\sigma_0/\eta \sim 10^3$. Departures from the inertial-range model are accentuated as Pr departs increasingly from unity but still diminish with increasing Re or σ_0. We see also from figure 6(*b*) that for a given range of σ_0 the source-size dependence of $s^2(0,t)$ diminishes with decreasing Pr, i.e. with the ratio σ_0/λ_c. In fact for $\sigma_0 \leqslant \lambda_c$ molecular diffusion smooths $P(\Delta, t; 0, 0,)$ (which is thus independent of Δ) over those scales which contribute to $s^2(0,t)$ and the source-size dependence is eliminated. For the present model then, in contrast to the inertial-range model (for which $s^2(0,t) \sim (\sigma_0/L)^{-\frac{1}{3}}$ for $\sigma_0 \ll L$), the 'point-source' limit is well-defined.

Finally in this section we consider the dissipation of scalar fluctuations. From the Eulerian conservation equation for $\overline{\theta^2}$ for the present case in which the scalar field is inhomogeneous in the z-direction we have

$$\left\langle \frac{\partial \overline{\theta^2}(z,t)}{\partial t} \right\rangle = -2\kappa \langle \overline{(\nabla\theta)^2} \rangle, \tag{4.15}$$

where we have used $\langle\ \rangle$ to denote the global integral $\int_{-\infty}^{\infty} \mathrm{d}z$. The term on the right differs from the global rate of dissipation of scalar fluctuations $\langle \epsilon_\theta \rangle$ by a term involving the mean gradient, $2\kappa\langle(\nabla\bar{\theta})^2\rangle$. Because this term is negligible at high Pe, for the cases we consider $\langle \epsilon_\theta \rangle$ can be calculated to an excellent approximation from the left-hand side of (4.15). In fact we calculate the nett global dissipation

$$\int_0^t \langle \epsilon_\theta \rangle \, \mathrm{d}t = \langle \overline{\theta^2}(0) \rangle - \langle \overline{\theta^2}(t) \rangle. \tag{4.16}$$

Figure 7 shows $\langle \overline{\theta^2}(t) \rangle$ as a function of time for $Re = 10^3$ and 10^5 and $\sigma_0/L = 0.01$ and 0.2. The one-particle and inertial-range results are shown as in figure 6(*a*). The most striking feature is that the inertial-range model 'dissipates' scalar fluctuations and that it accurately predicts the dissipation at high Re for σ_0/L of $O(1)$. Again we can refine this requirement to σ_0/η of $O(10^3)$. Figure 7 basically reflects the departure of $s^2(0,t)$ from the one-particle calculation as shown in figure 6(*a*), so it is obvious from figure 6(*b*) that dissipation increases with decreasing Pr.

In summary, the interesting qualitative features of Durbin's inertial-range model are preserved in our extended model in that $s^2(0,t)$ asymptotes to a constant large-time limit which is a function of source-size. The important qualitative difference is that the source-size dependence is eliminated by molecular diffusion for $\sigma_0 \leqslant \lambda_c$. Durbin's use of marked-particle statistics to calculate $\overline{\theta'^2}$ for large Re and σ_0/L of $O(1)$ for $Pr = 1$ is justified and indeed we have refined the condition on σ_0 and have included cases where Pr is not of $O(1)$. For small sources, $\sigma_0 \sim \eta$ or λ_c, explicit molecular effects cause significant and persistent departures from the inertial-range model.

4.3. *The effect of a finite sampling volume*

In the context of the two-point Lagrangian theory, concentration fluctuations over a finite sampling volume can be calculated from the statistics of pairs of molecules which disperse from within this volume back to the source (i.e. in general with non-zero initial separation). Therefore we can discuss the effect of a finite sample in terms of modifications to the point-sample results already obtained by comparing $P(\Delta, t; \Delta_0, 0)$ and $P(\Delta, t; 0, 0)$ where Δ_0 is a representative separation of pairs of molecules within

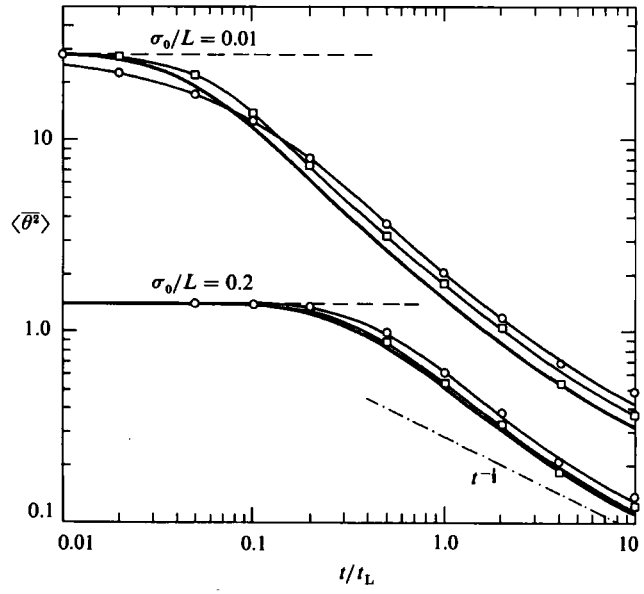

FIGURE 7. Effect of Re and source size on the global integral of the mean-square scalar field (essentially the complement of the nett global dissipation). ——, Durbin's (1980) model; – – –, Corrsin's (1952) one-particle model; ○, $Re = 10^3$; □, 10^5.

the sample volume. For the inertial-range model we showed in §3 that for sufficiently large time $(2\pi)^{\frac{1}{2}}\sigma_z P(\Delta, t; \Delta_0, 0)$ tends to the self-preserving form $R^{-\frac{1}{2}}(\Delta)$ on ever-increasing scales, independent of Δ_0. Thus although $\overline{\theta'^2}$ can depend on the sampling volume for small times, ultimately it does not. This conclusion can be interpreted as a consequence of the fact that the lengthscale of fluctuations increases with time according to (4.8) so that the reduction in $\overline{\theta'^2}$ due to averaging over a fixed volume eventually is negligible.

When molecular diffusion and viscosity are explicitly modelled we can no longer write an analytic form like (3.1) but we have confirmed numerically that again $P(\Delta, t; \Delta_0, 0)$ ultimately 'forgets' the initial separation Δ_0 and $\overline{\theta'^2}$ approaches the 'point-sample' limit. Thus although molecular diffusion affects $\overline{\theta'^2}$ at all times, instrumental averaging does not. Note that since λ_c is the smallest scale on which scalar fluctuations occur, in practice the point-sample limit is valid at all times for $\Delta_0 \ll \lambda_c$. Then the first term in (2.25) is negligible compared with the diffusive-growth terms.

That instrumental averaging does not play quite the same role as molecular diffusion can also be seen by examining a hypothetical fluid in which $\kappa = 0$ at some finite Re, i.e. the $Pr = \infty$ limit.

Consider first the 'point-sample' case (i.e. the limit $\Delta_0 \to 0$). Putting $\kappa = 0$ in (2.25) we see that for any finite time $\overline{\Delta^2}$ can be made arbitrarily small by choosing Δ_0 sufficiently small, i.e. in the point-sample limit the linear-strain flow field cannot separate a pair of particles in a finite time. Consequently the pair always move together and

$$P_2(z_1', z_2', t; z, z, 0) = P_1(z_1', t; z, 0)\,\delta(z_1' - z_2'), \tag{4.17}$$

so that (2.4) reduces to Corrsin's one-particle theory, i.e. it (and the equivalent Eulerian treatment of Chatwin & Sullivan (1979) in which dissipation is ignored) correspond to the 'point-sample', infinite Pr limit.

For a finite sample (i.e. finite Δ_0) (2.25) shows that material points separate even in the absence of molecular diffusion. The separation p.d.f. is now given by (3.1) with the generalized structure function (2.21). It follows from (2.9) that within the dissipation-range limit (2.20) Δ always remains the same sign as Δ_0, i.e. fluid-particle trajectories do not cross and $P(\Delta, t; \Delta_0, 0)$ is a one-sided p.d.f. (For finite Pr, molecular diffusion causes trajectories to cross.)

Although we cannot evaluate (3.1) analytically we are able to make order-of-magnitude estimates of scalar variance. Equation (3.1) can be integrated (using the transformation $\zeta = \int R^{-\frac{1}{2}}(\Delta)\,\mathrm{d}\Delta$) to show that equal numbers of pairs have separations greater than and less than Δ_0. In fact it is easily shown that for sufficiently large time half the pairs have separations smaller than any arbitrarily small value and half have separations greater than any arbitrarily large value. The former arises because there is no smoothing due to molecular diffusion and the linear velocity field produces increasingly fine structure through the Δ^{-1} singularity (3.4). At the same time the dispersive action of the energetic eddies generates increasingly large separations. Since the large scales determine $\overline{\Delta^2}$ (and fine scales contribute negligibly) we deduce that asymptotically $\overline{\Delta^2} \to \frac{1}{2}\sigma_z^2$. It follows immediately from (4.2c) that for a uniform-gradient source

$$\frac{\overline{\theta'^2}}{m^2} \sim \tfrac{1}{2}\sigma_z^2 \quad (t \to \infty). \tag{4.18}$$

That is, for $Pr = \infty$ where there is no molecular dissipation of scalar variance, half of the variance predicted by the one-particle model is eliminated by instrumental averaging. This is ultimately true for any finite sampling volume since at sufficiently large time half the fluctuation variance occurs on an arbitrarily small scale.

For the dispersing cloud discussed in §4.2 a similar asymptotic result obtains where instrumental averaging eliminates a fixed fraction of the variance predicted by the one-particle model, (4.13).

We conclude that although instrumental averaging reduces the scalar variance in the absence of molecular diffusion, its action is not equivalent to molecular diffusion. In particular Durbin's (1980) argument that the smoothing effect of instrumental averaging justifies the use of marked-particle statistics to calculate $\overline{\theta'^2}$ is not correct.

4.4. *Timescales for the development of a cloud or plume*

In general the development of scalar fluctuations in a dispersing cloud is controlled by the scales of the turbulence σ_w and L, the molecular properties of the fluid, κ and ν, the lengthscale imposed by the source σ_0 and the distance z of the measurement point from the centreline. For simplicity here we consider development along the centreline, $z = 0$. We consider a number of cases according to the magnitude of σ_0 relative to η, λ_c and L.

Figures 6 and 7 highlight two aspects of the evolution of $\overline{\theta'^2}$, the onset of dissipation as indicated by departure from the one-particle result and the approach to the large-time asymptote. Here we consider timescales for these processes and for the disappearance of a sampling-volume effect.

Consider first the onset of dissipation. From (4.16) and (4.11)

$$\frac{1}{\overline{\theta^2}(0)} \int_0^t \langle \epsilon_\theta \rangle \,\mathrm{d}t = 1 - (2\pi)^{\frac{1}{2}} \sigma_0 \int_{-\infty}^{\infty} P(\Delta', t; 0, 0)\, S(\Delta')\,\mathrm{d}\Delta'. \tag{4.19}$$

For $\overline{\Delta^2} \ll \sigma_0^2$ the Gaussian source function can be expanded in powers of Δ'^2 to give

$$\frac{1}{\overline{\theta^2}(0)} \int_0^t \langle \epsilon_\theta \rangle \, \mathrm{d}t = \frac{\overline{\Delta^2}}{2\sigma_0^2} + O(\overline{\Delta^4}). \tag{4.20}$$

Thus, as for the homogeneous case with a uniform-gradient source, dissipation is intimately connected with relative dispersion. Equation (4.20) shows that nett dissipation becomes significant after a time of $O(t_\mathrm{d})$ when $\frac{1}{2}\overline{\Delta^2}$ is a significant fraction of σ_0^2, i.e. in the same time it takes for a spot (or 'point' source) to grow to a fraction of the actual source size.

For $\sigma_0 \ll \lambda_\mathrm{c}$, $\overline{\Delta^2}$ is dominated by the diffusive-growth term $2\kappa t$ (see 2.25) and

$$t_\mathrm{d} = \frac{\sigma_0^2}{\lambda_\mathrm{c}^2} Re^{-\frac{1}{2}} t_\mathrm{L}. \tag{4.21}$$

For $\lambda_\mathrm{c} \ll \sigma_0 \ll \eta$ (which implies $Pr \gg 1$) the small-time result (2.25) can be extended using Saffman's (1960) linear-strain calculation which gives (for $\Delta_0 = 0$)

$$\overline{\Delta^2} = \frac{1}{\sqrt{3}} \lambda_\mathrm{c}^2 \left[4 \exp\left(\frac{1}{\sqrt{3}} \frac{t}{t_\mathrm{k}} \right) - \exp\left(\frac{2}{\sqrt{3}} \frac{t}{t_\mathrm{k}} \right) - 3 \right] \tag{4.22}$$

and so

$$t_\mathrm{d} = \ln 1(\sigma_0/\lambda_\mathrm{c}) \, Re^{-\frac{1}{2}} t_\mathrm{L}. \tag{4.23}$$

Within the inertial subrange $(\eta, \lambda_\mathrm{c}) \ll \sigma_0 \ll L$, we have $\overline{\Delta^2} \sim \epsilon t^3$, the well-known t^3 law (Batchelor 1950) from which

$$t_\mathrm{d} = \sigma_0^{\frac{2}{3}} \epsilon^{\frac{1}{3}} \sim (\sigma_0/L)^{\frac{2}{3}} t_\mathrm{L}. \tag{4.24}$$

Finally, for $\sigma_0 \gg L$, $\overline{\Delta^2} \sim \sigma_w^2 t t_\mathrm{L}$ and

$$t_\mathrm{d} \sim (\sigma_0^2/L^2) \, t_\mathrm{L}. \tag{4.25}$$

The effect of a finite sampling volume diminishes after a time t_m when relative dispersion is comparable with the sample length, i.e. $\overline{\Delta^2} - \Delta_0^2 \sim \Delta_0^2$. Application of this criterion parallels that for the onset of dissipation and timescales t_m corresponding to (4.21), (4.24) and (4.25) but with σ_0 replaced by Δ_0 are easily derived. However for $\lambda_\mathrm{c} \ll \Delta_0 \ll \eta$, dispersion is initially dominated by the linear-strain field acting over the separation Δ_0 (rather than by molecular diffusion) and proceeds on the timescale t_k, i.e. $t_\mathrm{m} = Re^{-\frac{1}{2}} t_\mathrm{L}$.

Consider now the approach of $\overline{\theta'^2}(0,t)$ (and $s^2(0,t)$) to their large-time asymptotes. Equations (4.11) or (4.12) show that two timescales are involved. The first, which we denote by t_a, is that on which $P(\Delta, t; 0, 0)$ approaches its asymptote for separations of $O(\sigma_0)$. According to (2.31) all separations within the dissipation subrange evolve on the timescale $t_\mathrm{k} \sim Re^{-\frac{1}{2}} t_\mathrm{L}$ so that for $\sigma_0 \ll \eta$ and $Pr \geqslant 1$

$$t_\mathrm{a} = Re^{-\frac{1}{2}} t_\mathrm{L}. \tag{4.26}$$

However for $Pr \ll 1$, dissipation-subrange structure is not important and molecular diffusion interacts directly with the inertial-subrange eddies on the timescale $(\kappa/\epsilon)^{\frac{1}{2}}$. Therefore for $\sigma_0 \ll \lambda_\mathrm{c}$ and $Pr \ll 1$

$$t_\mathrm{a} = Pe^{-\frac{1}{2}} t_\mathrm{L}. \tag{4.27}$$

For inertial-range sources $(\eta, \lambda_\mathrm{c}) \ll \sigma_0 \ll L$, $P(\Delta, t; 0, 0)$ approaches the large-time asymptote (3.3) for Δ of $O(\sigma_0)$ when $G(\sigma_0) \sim \sigma_0^{\frac{2}{3}} L^{\frac{1}{3}} \gg \sigma_z \sim \sigma_w t$, so that

$$t_\mathrm{a} = (\sigma_0/L)^{\frac{2}{3}} t_\mathrm{L}. \tag{4.28}$$

Finally for large sources, $\sigma_0 \gg L$, $G(\sigma_0) \sim \sigma_0 \gg \sigma_z \sim \sigma_w (t t_L)^{\frac{1}{2}}$ implies that

$$t_a = (\sigma_0^2/L^2)\, t_L. \tag{4.29}$$

The second timescale on which $\overline{\theta'^2}(0,t)$ evolves is that over which $\rho \to 0$, i.e. the integral timescale t_L. For small sources and large Re, $t_a \ll t_L$ and (4.12) shows that $s^2(0,t)$ is within a factor of $(1+\rho)^{\frac{1}{2}} \leqslant \sqrt{2}$ of the asymptote (4.13) for $t \sim t_a$, i.e. s^2 increases rapidly to within $\leqslant \sqrt{2}$ of its final value and then evolves more slowly. This two-stage evolution is clear in figures 6 and 7. In the initial stage when $t \ll t_a$, the rapid growth of fluctuations is dominated by bulk motion of the cloud (meandering) whereas in the final stage, $t \gg t_a$, fluctuations are produced by internal structure within the cloud and the meandering contribution is negligible (Sawford 1983*a*). The important point to be made here is that evolution of the intensity of fluctuations along the centreline for small sources is essentially complete (to within a factor of $\sqrt{2}$) after a time t_a much smaller than the integral timescale.

All the results presented so far in this paper have been for stationary homogeneous turbulence. These conditions are well-approximated in the initial stages of dispersion in real turbulent flows. However whether or not the large-time asymptotic predictions of our model are relevant in such flows depends on the relative magnitudes of the timescale t_a and that over which the approximation of stationary homogeneous turbulence breaks down.

The effects of non-stationarity or non-homogeneity cannot be ignored when the change in any turbulence scale (e.g. Δt_L) is comparable in magnitude with its value at the source ($t_{L,0}$). For example in grid turbulence t_L is the most rapidly changing scale and varies almost linearly with downstream distance ($\approx$ travel time if the streamwise intensity of turbulence is small). Non-stationarity (or streamwise non-homogeneity) therefore has a significant effect on dispersion after a time t of $O(t_{L,0})$. In a neutral boundary layer both L and t_L vary approximately linearly with height above the boundary. For a small source at height H this vertical non-homogeneity is no longer negligible when $\sigma_z = \sigma_w t \sim L(H)$, i.e. again when t is of $O(t_{L,0})$. Similarly for sheared homogeneous turbulence, L varies approximately linearly with downstream distance so again non-stationarity is important for t of $O(t_{L,0})$. Note though that since the mean shear itself also affects the scalar fluctuations on the timescale $(\partial \overline{U}/\partial z)^{-1}$ (Durbin 1980; Sawford 1983*b*) in this case we also require $(\partial \overline{U}/\partial z)\, t_{L,0} \geqslant 1$.

It follows that the asymptotic predictions of our model are relevant for these real flows for small sources and high Re for which $t_a \ll t_{L,0}$. In these circumstances, scalar fluctuations in the cloud undergo fairly complete development before the effects of mean shear, non-stationarity and non-homogeneity of the turbulence become important. This may often be the case in the atmosphere where Re is very large (of $O(10^8)$) and L may be of $O(1000$ m) (e.g. in the convective boundary layer). The situation is less clear in laboratory flows where Re is much lower.

5. Conclusions

We have extended Durbin's (1980) marked-particle pair-trajectory model in order to examine the effects of molecular diffusion on the scalar fluctuation variance in stationary homogeneous turbulence.

Our conclusions, which focus on the questions raised in the Introduction, are broadly independent of the details of the model.

1. Molecular processes influence the spreading of pairs of molecules from a source for all time where their separation is of the order of the velocity or diffusive microscale

(η, λ_c). However, viscosity and molecular diffusion have opposing effects – the linear-strain turbulence associated with viscous dissipation tends to enhance small-scale structure while molecular diffusion smooths and reduces it.

For both a uniform-gradient source and a dispersing cloud, dissipation of $\overline{\theta'^2}$ is intimately connected to relative dispersion and is correctly predicted by marked-particle statistics at a large enough large Re. Molecular processes affect the dissipation when Pr is much greater than or less than 1. But for finite Pr, marked-particle statistics are still correct at large enough large Re, as can be seen from figures 6 and 7.

2. The intensity of scalar fluctuations, $s = [\overline{\theta'^2}/\overline{\theta}^2]^{\frac{1}{2}}$, tends to a constant value when t is large. Thus molecular processes do not eliminate the persistent streaky structure predicted by Durbin's model for clouds of contaminant.

3. For small sources (σ_0 of $O(\eta, \lambda_c)$, $\overline{\theta'^2}$ is determined by the small-scale structure controlled by molecular processes and so also depends on κ and ν for all time (figure 6). As a corollary, $\overline{\theta'^2}$ is independent of κ and ν for $\sigma_0 \gg (\eta, \lambda_c)$ at high Re for fixed σ_0/L and is therefore controlled by the statistics of marked particles. Thus the source-scale dependence, (1.5b), of Durbin's model is retained for $\sigma_0 \gg (\eta, \lambda_c)$.

4. Our results emphasize the importance of Durbin's (1980) 'outer limit' when using marked-particle statistics to calculate $\overline{\theta'^2}$ at finite Pr. Essentially this means that in neglecting molecular diffusion (by using marked-particles) one must also neglect viscous dissipation (i.e. the turbulence must be modelled as having an inertial subrange extending to infinitely small scales). At infinite Pr where molecular diffusion is zero but viscous-dissipation-range structure of the turbulence is retained, we show that in the 'point-sample' limit the 2-point theory reduces to Corrsin's (1952) one-particle analysis which is equivalent to Chatwin & Sullivan's (1979) Eulerian analysis with $\epsilon_\theta = 0$. Instrumental averaging reduces $\overline{\theta'^2}$ in this case but unlike molecular diffusion does not counter the tendency of the linear-strain turbulence to generate increasingly fine-scale structure.

5. Timescales for the development of $\overline{\theta'^2}$ in a dispersing cloud depend on σ_0 and, for small sources ($\sigma_0 \ll L$) and high Re, are much smaller than t_L. We showed that many real flows evolve on the timescale t_L so that the present results for stationary homogeneous turbulence are relevant to such flows for small sources and large Re. The inertial-range timescales (4.25) or (4.29) agree with Fackrell & Robins' (1982) measurements from small elevated sources in a wind-tunnel boundary layer which show that $\overline{\theta'^2}$ develops downstream on a timescale proportional to $\sigma_0^{0.7}$.

6. The reduction in $\overline{\theta'^2}$ due to instrumental averaging over a finite sampling volume is negligible once the relative dispersion process 'forgets' the initial separation Δ_0 imposed by the sampling volume. In other words instrumental averaging does not have the same effect as molecular diffusion in reducing $\overline{\theta'^2}$. The physical reason for this is that there is an increase in the lengthscale of scalar fluctuations with time so that the proportion of variance contained on scales of $O(\Delta_0)$ is eventually negligible. Estimates of the timescale t_m on which sampling effects are eliminated were given in §4.4, the most noteworthy point being that $t_m \ll t_L$ for $\Delta_0 \ll L$.

Some of our other findings are more dependent on the details of Durbin's (1980) model (and our extension). While it is possible to criticize the details of these models (for example the treatment of only one component of the turbulence) we note that our extended model reproduces the known exact three-dimensional results at small time and the exact results for a uniform-gradient scalar field and that the reversed-diffusion formalism compensates for the lack of a compressibility constraint. In addition, as outlined in the Introduction, there is some support for the predictions of Durbin's model from wind-tunnel experiments. Qualitatively these predictions are

not strongly altered by molecular diffusion. Our results for the uniform-gradient source are consistent with the continued growth $\overline{\theta'^2} \sim t^{\frac{1}{2}}$ at large time as predicted by Durbin's (1980) model. For a dispersing cloud the intensity of fluctuations is still predicted to be constant at large time (as was assumed by Csanady 1973) even for small sources when molecular effects are important. The major qualitative effect of molecular processes is the elimination of the source-size dependence of s for $\sigma_0 \ll \lambda_c$. This regularizes the 'point-source' limit and is likely to be a more general result.

B.L.S. was supported while at Cambridge in part by a grant from the Natural Environmental Research Council. We are grateful for comments on the paper by Dr P. A. Durbin, Professor P. C. Chatwin and the referees.

Appendix. Derivation of (2.2)

Consider a volume of fluid $(\Delta x)^3$ about the point $\boldsymbol{x}$ which, at the measurement time t, contains N molecules, N_m of which are labelled.† Then the proportion (specific concentration) of labelled molecules in the volume at $(\boldsymbol{x}, t)$ is

$$\theta(\boldsymbol{x}, t) = \frac{N_m}{N} = \frac{1}{N} \sum_{i=1}^{N} \gamma_i, \tag{A 1}$$

where $\gamma_i = 1$ for a labelled molecule and $\gamma_i = 0$ for an unlabelled molecule. Note that for elemental volumes, $\Delta x \ll \lambda_c$, the smallest scale over which the concentration varies, $\theta(\boldsymbol{x}, t)$ can be regarded as the instantaneous concentration at the point $\boldsymbol{x}$.

We calculate the concentration covariance for two separated points $(\boldsymbol{x}_1, t)$ and $(\boldsymbol{x}_2, t)$ for an instantaneous source distribution, $S(\boldsymbol{x})$ released at time 0. (Extension to more general cases is straightforward.) The instantaneous concentration product is

$$\theta(\boldsymbol{x}_1, t)\, \theta(\boldsymbol{x}_2, t) = \frac{1}{N^2} \sum_{i,j=1}^{N} \gamma_i \gamma_j, \tag{A 2}$$

Consider an arbitrary pair of molecules 1, 2 (one from each volume) which may or may not be labelled. Clearly, from A 2, the required covariance $\overline{\theta(\boldsymbol{x}_1, t)\, \theta(\boldsymbol{x}_2, t)}$ is related to $\overline{\gamma_1 \gamma_2}$, the probability that *both* molecules are labelled. We evaluate this probability in two stages using the reversed-diffusion concept illustrated in figure 1(*a*).

It is given that 1, 2 are 'at' $\boldsymbol{x}_1$ and $\boldsymbol{x}_2$ at time t and we ask what is the probability that they are labelled if they came from elemental volumes about the points $\boldsymbol{x}_1'$ and $\boldsymbol{x}_2'$ at time 0. Since the labelling of one molecule has no influence on the labelling of the other, and since at time 0 labelled molecules are distributed according to the source function, the conditional labelling covariance is just

$$\overline{\gamma_1 \gamma_2}|_{\boldsymbol{x}_1', \boldsymbol{x}_2', 0} = S(\boldsymbol{x}_1)\, S(\boldsymbol{x}_2). \tag{A 3}$$

The second stage of our calculation is to express the unconditional labelling covariance as the convolution of the conditional covariance (A 3) and the joint p.d.f. that molecules 1, 2 came from $\boldsymbol{x}_1'$ and $\boldsymbol{x}_2'$ at time 0 (given that they are at $\boldsymbol{x}_1, \boldsymbol{x}_2$ at time t), $P_2(\boldsymbol{x}_1', \boldsymbol{x}_2', 0; \boldsymbol{x}_1, \boldsymbol{x}_2, t)$. Thence

$$\overline{\gamma_1 \gamma_2} = \int P_2(\boldsymbol{x}_1', \boldsymbol{x}_2', 0; \boldsymbol{x}_1, \boldsymbol{x}_2, t)\, S(\boldsymbol{x}_1')\, S(\boldsymbol{x}_2')\, \mathrm{d}\boldsymbol{x}_1'\, \mathrm{d}\boldsymbol{x}_2'. \tag{A 4}$$

† The term 'labelled' is used to denote contaminant species, while the term 'marked' is reserved to indicate the set of molecules in whose displacement statistics we are interested, i.e. in this case *all* molecules in $(\Delta x)^3$.

This is the required result since for elemental volumes all pairs are statistically equivalent so that the sum in (A 2) is trivial. (A 4) reduces to (2.2) for a plane source distribution.

For a finite sampling volume, the statistics of any given pair depend on their locations within the volumes and averaging over pairs according to (A 2) must be carried out explicitly.

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