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A stochastic model of two-particle dispersion and concentration fluctuations in homogeneous turbulence

By P. A. DURBIN
Department of Applied Mathematics and Theoretical Physics,
University of Cambridge, Silver Street, Cambridge CB3 9EW

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A new definition of concentration fluctuations in turbulent flows is proposed. The definition implicitly incorporates smearing effects of molecular diffusion and instrumental averaging. A stochastic model of two-particle dispersion, consistent with this definition, is formulated. The stochastic model is an extension of Taylor's (1921) model and is consistent with Richardson's $\frac{4}{3}$ law. Its predictions of concentration fluctuations are contrasted with predictions based on a more usual one-particle model.

The present model is used to predict fluctuations in three case studies. For example (case (i) of § 6), downstream of a linear concentration gradient we find $\overline{c'^2} = \frac{1}{2}m^2(\overline{Z^2} - \overline{\Delta^2})$. Here m is the linear gradient, $\overline{Z^2}$ is related to centre-of-mass dispersion and $\overline{\Delta^2}$ is related to relative dispersion (see equation (3.1)). The term $\frac{1}{2}m^2\overline{Z^2}$ represents net production of fluctuations by random centre-of-mass dispersion, whereas $\frac{1}{2}m^2\overline{\Delta^2}$ represents net destruction of fluctuations by relative dispersion. Only the first term is included in the usual one-particle model (Corrsin 1952).

1. Introduction

It is well known that pairs of particles released in a turbulent fluid tend, on average, to drift apart in consequence of random convection. Many turbulence phenomena can be understood in terms of the dynamics of these particle pairs; in the present work we shall be interested in describing fluctuations of the concentration of a dispersing passive contaminant via two-particle statistics. Our approach will be to postulate a stochastic model, similar to Taylor's (1921) random-walk model for one-particle dispersion, which reproduces certain known statistical properties of two-particle dispersion.

A need for mathematical description of concentration fluctuations arises in numerous applications including: prediction of air pollution; determination of reaction rates in turbulent chemical reactors; analysis of turbulent combustion; and simulation of 'temperature noise' downstream of non-uniform heat sources in turbulent flows. The fluctuations we shall be concerned with are those produced when an initially smooth distribution of contaminant is dispersed randomly by a turbulent flow. This is essentially a stochastic phenomenon. In that vein, a stochastic model can be enlightening.

Consider an initially small blob of contaminant in a turbulent fluid. At a later time there will be some probability of observing this blob at locations distant to its initial position. If a distant location happens to lie within the blob a positive concentration

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is recorded; otherwise, there is zero concentration. In this way concentration fluctuations are produced. This production of fluctuations is connected to dispersion of the blob's centre of mass by large turbulent eddies – to a first approximation.

We say 'to a first approximation' because, if the blob is small enough, most of the turbulent kinetic energy will be contained in eddies larger than the blob. These eddies convect the blob without destroying its integrity. However, some of the turbulent energy will reside in eddies comparable in size to the blob. These will mix the blob with its environment, thereby decreasing its concentration. The concentration measured at a fixed location will depend on the degree to which the blob has been mixed with the environment, as well as the probability of the point lying within the blob. The mixing process is stochastic, for it depends on the probability that turbulent eddies have mixed together two blobs (our initial blob and a blob from the environment). Hence it depends on the dispersion of the blobs *relative to each other*.

It is quite evident that a complete stochastic theory of concentration fluctuations needs to concern itself with relative dispersion. Below we give a brief, critical review of relative dispersion models. Attention is restricted to aspects relevant to the present modelling effort.

Modelling relative dispersion

The simplest model of relative dispersion appearing in the literature is Richardson's $\frac{4}{3}$ eddy-diffusion model (Batchelor 1952). The $\frac{4}{3}$ law can be justified by inertial range scaling. Inertial range scaling is an important ingredient of a relative dispersion model.

On the other hand, Richardson's use of a diffusion equation is indefensible. An assumption that relative dispersion can be described as a Markov process underlies the use of a diffusion equation. In other words, it is assumed that one can describe relative dispersion adequately by consideration only of scales of particle separation large compared with those of eddies doing the dispersing. But, at small separations, eddies of size comparable to the separation of the particles are responsible for their relative dispersion (Csanady 1973, § 4.3): a diffusion equation cannot be justified when the particle separation is less than the turbulence integral scale.

A different objection to Richardson's $\frac{4}{3}$ eddy diffusivity was raised by Batchelor (1952). Denoting the random particle separation by Δ, Batchelor claimed that it was improper to have an eddy diffusivity proportional to the *random* variable $|\Delta|^{\frac{4}{3}}$; he suggested, therefore, that it be made proportional to the *mean* quantity $\overline{|\Delta|^{\frac{4}{3}}}$. In the light of our present understanding of the theory of stochastic processes, Batchelor's argument seems specious; indeed, his form of diffusivity would imply that individual particle pairs were dispersed by 'average eddies', rather than by instantaneous random eddies as in Richardson's model.

Another diffusion model was proposed by Thiebaux (1975). His model is similar to Richardson's, though more general. It could be considered a special case in which the present stochastic model becomes Markovian, as is shown in appendix B.

Chatwin & Sullivan (1979), following several previous authors, considered a random linear-strain model of relative dispersion. Strictly speaking, this model is applicable only when particle separations are small compared to the Kolmogoroff microscale.†

† To be fair, Chatwin & Sullivan used the random-strain model to illustrate ideas which they feel have wide application.

Chatwin & Sullivan found that in this regime molecular diffusion played an important role.

When concentration fluctuations are measured experimentally the measuring devices used† may average concentrations over several microscales, and this, together with the fact that molecular smearing also reduces fine structure, must have bearing on any practical model of fluctuations. How can we connect our ideas about random convection and relative dispersion of contaminant blobs with this need to model 'smearing' processes?

To answer this question the connexion between relative dispersion concentration fluctuations must be quantified. A mathematical connexion between random particle paths, as described by the relative dispersion model, and concentrations, is provided by the statistical theory of dispersion. By delving into this theory we shall find the answer to the question raised in the last paragraph.

2. Statistical theory of concentration fluctuations

The body of this paper deals with a one-dimensional model; hence, in this section all equations will be written in their one-dimensional form.

Consider parcels of some passive contaminant released into a turbulent flow. In the absence of molecular diffusion, these parcels conserve their concentration. Thus the mean concentration of contaminant observed at any point in the flow is simply the sum of the concentrations assigned to particles at their source, times the probability of these particles reaching the observation point:

$$\bar{C}(z,t) = \iint_{-\infty}^{\infty} P_1(z,t;z',t')\, S(z',t')\, dz'\, dt' \tag{2.1}$$

(Monin & Yaglom 1971, §10). Here $P_1(z,t;z',t')$ is the probability density function (p.d.f.) that particles leaving z' at time t' arrive at z at time t. The quantity $S(z',t')$ is the source concentration.

A similar line of reasoning leads to the expression

$$\overline{C(z_1,t)\, C(z_2,t)} = \iiiint_{-\infty}^{\infty} P_2(z_1,z_2,t;z_1',z_2',t_1',t_2')\, S(z_1',t_1')\, S(z_2',t_2')\, dz_1'\, dz_2'\, dt_1'\, dt_2' \tag{2.2}$$

for the two-point covariance. Here P_2 is the two-particle p.d.f. with an interpretation similar to P_1, though it depends on the *simultaneous* motion of two particles (Monin & Yaglom, §24). Of course,

$$P_1(z_1,t;z_1',t_1') = \iint_{-\infty}^{\infty} P_2(z_1,z_2,t;z_1',z_2',t_1',t_2')\, dz_2'\, dt_2' \tag{2.3}$$

and

$$\iint_{-\infty}^{\infty} P_1(z,t;z',t')\, dz'\, dt' = 1. \tag{2.4}$$

If $|z_1-z_2|$ is large compared to the Kolmogoroff scale, $\eta \equiv (\nu^3/\epsilon)^{\frac{1}{4}}$, then P_2 is determined primarily by the 'outer region' dynamics; i.e. by eddies of inertial range size or larger. (N.B. In this paper we refer to an 'outer region', with dimension on the order of the turbulence integral scale, L, and to an 'inner region' with dimension

† In some applications – such as to pollutants which affect plants or humans – 'instrumental averaging' is not a technological limitation; rather, it is an intrinsic constraint.

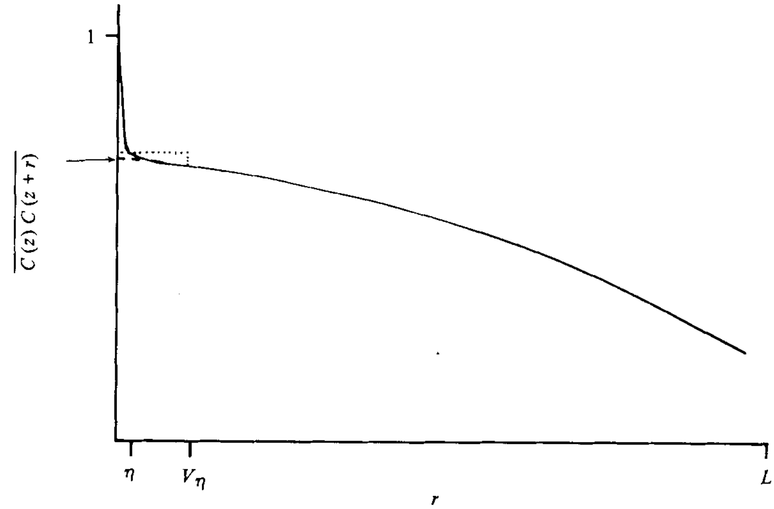

FIGURE 1. Suggesting the structure of the function $\overline{C(z)\;C(z+r)}$: with smoothing absent (solid line); and with the inner-region structure smoothed over the region V_η (dotted line). The 'outer limit' is marked by an arrow and the 'inner limit' is taken as 1.

$O(\eta)$.) Now, $\overline{C^2}(z,t)$ can be defined by letting $z_1, z_2 \to z$ in (2.2). Howeveı (in the absence of molecular diffusion, at least), this limit will be discontinuous in the sense that the limit as $|z_1 - z_2| \to 0$ in the outer region will not be the same as that in the inner region. (In the outer region $|z_1 - z_2| \to 0$ means $|z_1 - z_2|/L \to 0$. In the inner region it means $|z_1 - z_2|/\eta \to 0$.) This is illustrated by figure 1. Because of smearing by molecular action or finite measurement-probe size, the definition of $\overline{C^2}$ in terms of the outer limit should, in most realistic situations, be the more appropriate. We adopt this definition.

Our definition of $\overline{C^2}$ can alternatively be obtained by averaging $\overline{C(z_1)\,C(z_2)}$ (dropping the time-dependence for convenience) over a region, V_η, $O(\eta)$: this makes its connexion with instrumental smoothing clearer.

$$\begin{aligned}\overline{C^2} &\equiv V_\eta^{-2} \iint_{V_\eta} \overline{C(z_1)\,C(z_2)}\, dz_1\, dz_2 \\ &= V_\eta^{-2} \iint_{-\infty}^{\infty} \iint_{V_\eta} P_2(z_1, z_2; z_1' z_2')\, S(z_1')\, S(z_2')\, dz_1\, dz_2\, dz_1'\, dz_2' \\ &\approx \iint_{-\infty}^{\infty} \lim_{|z_1 - z_2|/L \to 0} P_2(z_1, z_2; z_1', z_2')\, S(z_1')\, S(z_2')\, dz_1'\, dz_2'. \end{aligned} \tag{2.5}$$

The last step follows from the fact that the outer limit is assumed to be a good approximation to the averaged inner region (see figure 1). With this definition understood, (2.5) can be written

$$\overline{C^2}(z) = \iint_{-\infty}^{\infty} P_2(z; z_1', z_2')\, S(z_1')\, S(z_2')\, dz_1'\, dz_2'. \tag{2.6}$$

In a slightly different context, Lamb (1976) proposed using (2.6) for prediction of air-pollution episodes.

Of course, averaging over some V_η is not equivalent to diffusive smoothing, although there is a loose connexion. Our definition really follows from an assertion that molecular diffusion makes the covariance (figure 1) smooth at the origin, on the scale L.

Figure 1 might be called our fundamental hypothesis: it therefore merits further discussion. The solid line illustrates the structure of $\overline{C(z)\,C(z+r)}$ in the absence of molecular diffusion. The height and sharpness of the peak near $r = 0$ will increase with time as small-scale turbulent straining produces increasingly finer structure in the contaminant field – figure (79) in Monin & Yaglom (§ 10.2) is a graphic illustration of this. Any contaminant with finite molecular diffusivity will not permit this peak to appear. Rather, the fine structure will be smeared by diffusion. If the contaminant does not diffuse by molecular action, the peak will appear and grow as just described. However, any measurement probe with dimension V_η will produce a smearing effect similar to diffusion; although it will do instantaneously what diffusion does gradually.

There are two assumptions implicit in figure 1 and the theory so far presented: (i) a V_η exists such that $L \gg V_\eta$ and (ii) smearing makes $\overline{C(z)\,C(z+r)}$ smooth at $r = 0$. Neither (i), nor equations (2.1) and (2.2), can be true unless (iii) the turbulent Péclet number, $u'L/\kappa$, is $\gg 1$. Assumption (ii) can only be true if (iv) the Prandtl number is $O(1)$ (or if a measurement device with dimension larger than η is used). The condition (iii) ensures that the large-scale structure of $\overline{C(z)\,C(z+r)}$ is not significantly affected by molecular diffusion, while (iv) ensures that V_η, or an effective V_η in the diffusive case, will be large compared to the width of the peak. In these circumstances (i) and (ii) will, to a good approximation, be justified. Also, $\overline{C^2}$ should then be reasonably independent of the dimensions of measuring devices.

If the processes that smear the peak in figure 1 were ignored, then $\overline{C^2}$ would be given by the inner limit of (2.2). We will refer to $\lim_{r/\eta \to 0} \overline{C(z)\,C(z+r)}$ as the *usual* definition of $\overline{C^2}$, to distinguish it from (2.5), and because it has been used to define $\overline{C^2}$ by several authors (Corrsin 1952; Csanady 1973; Thiebaux 1975; Chatwin & Sullivan 1979). Since

$$\lim_{|z_1 - z_2|/\eta \to 0} P_2(z_1, z_2; z_1', z_2') = P_1(z_1; z_1')\,\delta(z_1' - z_2'), \tag{2.7}$$

the *usual* definition of $\overline{C^2}$ is

$$\overline{C_u^2} = \int_{-\infty}^{\infty} P_1(z; z')\,S^2(z')\,dz'. \tag{2.8}$$

Equation (2.7) says that this usual definition does not include turbulent mixing processes associated with relative dispersion: our new definition (2.6) does. It is easily proved that relative dispersion reduces fluctuations. Particles one and two are indistinguishable so the inequality

$$\iint_{-\infty}^{\infty} P_2(z; z_1', z_2')\,(S(z_2') - S(z_2'))^2\,dz_1'\,dz_2' \geqslant 0 \tag{2.9}$$

together with (2.3) proves that

$$\overline{C^2} \leqslant \overline{C_u^2}, \tag{2.10}$$

equality holding when S is identically constant. Taking $\overline{c'^2} = \overline{C^2} - \bar{C}^2$ as a measure of fluctuation amplitude, (2.10) shows that the present definition leads to lower concentration fluctuations than the usual definition.

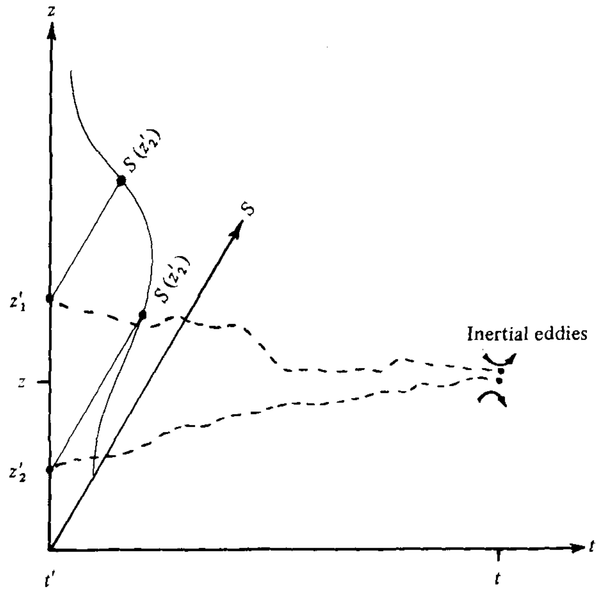

FIGURE 2. Illustrates dispersion by inertial eddies and our procedure for calculating the 'inner limit' of the two-point covariance. The dashed lines follow two particles, from their origins at z_1' and z_2', as they are swept together, at z, by turbulent eddies. This is what we call 'mixing of blobs'. In calculations the particles' paths are traced backwards from z to z_1' and z_2'. In this reversed diffusion, particles are dispersed by turbulence. Hence in this figure relative dispersion and mixing are interchangeable: the distinction is only by the direction in which the dotted lines are traced. Initially, dispersion is effected by inertial eddies. Because of dispersion $S(z_1') \neq S(z_2')$ mixing occurs and fluctuations are reduced, see (2.10).

If $S(z')$ has a uniform value of N over the region V, and letting

$$f(z) = \int_V P_1(z, z')\, dz', \tag{2.11}$$

(2.8) and (2.1) lead to the familiar equation (Csanady 1973, cha. 7)

$$\overline{c_u'^2} \equiv \overline{C_u^2} - \bar{C}^2 = N^2(f - f^2). \tag{2.12}$$

J. Foss and S. Corrsin (1979, private communication from Foss) have compared (2.12) with appropriate experimental data and found their experimental values of $\overline{c'^2}$ to be considerably lower than (2.12) suggests. Experimentally, then, (2.10) is also observed.

Consider two particles which have, at time t, some small but finite separation. Travelling backwards in time, their positions may at time t', $t' \ll t$, become very nearly independent. (See figure 2 and § 6. Figure 2 shows why one wants to trace trajectories backwards in time: their final location is given while their initial positions are random.) The probability density function of particles 1 and 2 is $P_2(z, t; z_1', z_2', t_1', t_2')$. If mixing were to make z_1 and z_2 *completely* independent, then

$$\left.\begin{aligned} &P_2(z, t; z_1', z_2', t_1', t_2') \to P_1(z, t; z_1', t_2')\, P_1(z, t; z_2', t_2'),\\ \text{so, by (2.6),}\quad &\overline{C^2} \to \bar{C}^2\\ \text{and}\quad &\overline{c'^2} \to 0. \end{aligned}\right\} \tag{2.13}$$

In this case turbulent stirring would eradicate fluctuations (in the sense of (2.5)).

Consider, again, our pair of particles at time t: for t' close to t – or if turbulent mixing is very inefficient – their separation will remain small. $P_2(z_1', z_2', t')$ will be close to $P_1(z_1', t')\,\delta(z_2'-z_1')$, so (2.8) and its consequence (2.12) will apply. Thus, (2.8) and (2.13) provide extreme limits for $\overline{c'^2}$. In general $\overline{c'^2}$ will be intermediate to these values.

The question posed near the end of § 1 has now been answered: by defining the mean-square concentration fluctuation as the covariance between fluctuations at two points with separation larger than η (or, more correctly, $o(L)$) it is made to depend on relative dispersion, and smearing processes have been represented. If the dotted line in figure 1 is correct then it does not matter that our definition of $\overline{C^2}$ involves two points which, on the scale η, are distinct. However, for modelling purposes it is quite important that these points be distinct, for then particles located at them can be dispersed by inertial-range eddies. This is clarified by the following: When particles are separated by a distance Δ very much less than η a linear strain model gives $d\Delta/dt \propto \Delta$ for their relative velocity. If $\Delta \rightarrow 0$ as $t \rightarrow 0$ then $\Delta \equiv 0$; hence the significance of not letting $\Delta/\eta \rightarrow 0$. In the inertial range $d\Delta/dt \propto |\Delta|^{\frac{1}{3}}$. Thus $|\Delta| \propto t^{\frac{3}{2}}$ if $\Delta \rightarrow 0$ as $t \rightarrow 0$; hence inertial range eddies can disperse particles even when $\Delta/L \rightarrow 0$. In figure 2 this dispersion is tied up with concentration fluctuations.

To apply the theory described in this section a method for determining P_2 and calculating its integral (2.6) is required. In § 3 we formulate a stochastic model of relative dispersion for this purpose. After analysing this model in §§ 4 and 5, its application to concentration fluctuations is illustrated in § 6.

3. Formulation of the stochastic model

Consider two particles, at positions z_1 and z_2, being dispersed in a turbulent fluid. The velocities of these particles consist of a common part plus a relative drift. It is convenient, therefore, to formulate our stochastic dynamic model in terms of the variables

$$\left.\begin{aligned} Z &= (z_1+z_2)/\surd 2, \\ \Delta &= (z_1-z_2)/\surd 2. \end{aligned}\right\} \tag{3.1}$$

These can be loosely thought of as 'centre-of-mass' and 'relative' co-ordinates. The $\surd 2$, rather than 2, appears in the denominator of (3.1) for mathematical reasons which become clear later (see equations (3.5) and (6.16)).

Extension of Taylor's model

Taylor (1921) proposed a model for the random velocity of a single particle in homogeneous turbulence which (in the limit of his $\Delta t \rightarrow 0$) is equivalent to the Langevin equation (Wax 1954):

$$dU = \frac{-U\,dt}{T_L} + \sigma_w \left(\frac{2}{T_L}\right)^{\frac{1}{2}} dW_t; \quad U(t=0) = \sigma_w \mathcal{N}. \tag{3.2}$$

Here T_L is the Lagrangian integral time scale, σ_w the r.m.s. fluctuating velocity (see Taylor 1921), dW_t a Gaussian white noise process and $\mathcal{N}$ is a mean-zero, variance-one (i.e. 'standard') Gaussian random variable. The letter U is used to denote velocity in (3.2) because the solution to (3.2) is commonly called an Uhlenbeck–Ornstein (UO) process. The UO process is a stationary Gaussian–Markov process with mean-zero and variance σ_w; hence, it has a correlation function

$$\frac{\overline{U_{t+\tau}U_t}}{\sigma_w^2} = \exp[-|\tau|/T_L]. \tag{3.3}$$

The particle position obtained by integrating $U(t)$ with respect to t may be thought of as a correlated random walk, although a more apt terminology is random flight (Chandrasekhar, in Wax 1954).

To extend Taylor's model to two interdependent, simultaneously dispersing, particles we postulate the model equations

$$\left.\begin{aligned} dZ/dt &= [\alpha(\Delta)+\beta(\Delta)]\,U^{(1)}(t),\\ d\Delta/dt &= [\alpha(\Delta)-\beta(\Delta)]\,U^{(2)}(t).\end{aligned}\right\} \tag{3.4}$$

Here $U^{(1)}$ and $U^{(2)}$ are independent UO processes (determined by (3.2) with two independent white-noise processes). The coefficients α and β have been made functions of Δ alone since in this paper we will be considering homogeneous turbulence. Their Δ-dependence will be determined below. This model is non-Markovian.† (In other words, the finite correlation time-scale of the turbulent velocity field has been taken into account.)

An equivalent form of (3.4) is

$$\left.\begin{aligned} dz_1/dt &= \alpha U'+\beta U'',\\ dz_2/dt &= \alpha U''+\beta U',\end{aligned}\right\} \tag{3.5}$$

where

$$U' = (U^{(1)}+U^{(2)})/\surd 2,$$
$$U'' = (U^{(1)}-U^{(2)})/\surd 2.$$

Note that, by the independence and Gaussianity of $U^{(1)}$ and $U^{(2)}$, U' (respectively U'') is an UO process, equivalent to $U^{(1)}$ or $U^{(2)}$, and independent of U'' (respectively U').

A physical interpretation of (3.5) is that $\alpha U'$ represents a contribution to dz_1/dt from eddies located near particle 1 and $\beta U''$ represents a contribution from eddies near particle 2. Therefore, we must have $\alpha \geqslant \beta$. Only eddies near particle 2 of size comparable to, or larger than, Δ contribute to dz_1/dt. Therefore, when $\Delta\to\infty$, $\beta\to 0$. Further considerations, which appear in appendix A, completely specify α and β. The specifications given there are

$$\left.\begin{aligned} \alpha^2 &= \tfrac{1}{2}[1+(2R(\Delta)-R^2(\Delta))^{\frac{1}{2}}],\\ \beta^2 &= 1-\alpha^2,\end{aligned}\right\} \tag{3.6}$$

where

$$R(\Delta) = \left(\frac{\Delta^2}{L^2+\Delta^2}\right)^{\frac{1}{3}}. \tag{3.7}$$

The function $R(\Delta)$ is related to the structure function (Townsend 1976, p. 11), or covariance function of the turbulent *relative* velocity field. As $\Delta\to 0$, $R(\Delta)\to|\Delta|^{\frac{2}{3}}$, in

† In an appropriate phase space it is Markovian – see appendix B. This 'concealed' Markovianity simplifies the model, without losing the, physically necessary, non-Markovianity in Z, Δ space.

consequence of inertial range scaling, and, as $\Delta \to \infty$, $R(\Delta) \to 1$, because at large separation particles move independently. The constraint $\alpha^2+\beta^2 = 1$ ensures that the variance of the turbulent velocity is correct.

We next seek analytic solutions to the model (3.4). Ultimately we will want to use an extension of this model to inhomogeneous turbulent flows, and this will require fully numerical analysis; but in the present paper attention is restricted to the homogeneous case, where partial analytical solutions can be found. Numerical results will also be given in § 6.

It is quite an easy matter to integrate the second of (3.4) and thereby to obtain a particle separation p.d.f. This density function, in itself, has little meaning for us; because it deals with the distribution of particles. However, its moments will tell us something about the concentration fluctuations in which we are interested.

4. Particle separation p.d.f.

The UO *position* process obtained by integrating the velocity process, $U(t)$, with respect to t is a Gaussian random variable, ξ, with mean zero and variance

$$\sigma_x^2 = 2\sigma_w^2 T_L^2(e^{-t/T_L}-1+t/T_L) \tag{4.1}$$

(Wax 1954). Let

$$G(\Delta) = \int_0^\Delta (\alpha(\Delta')-\beta(\Delta'))^{-1}\, d\Delta'.$$

If (3.4) is integrated it is found that

$$G(\Delta)-G(\Delta_0) \equiv \int_{\Delta_0}^{\Delta} \frac{d\Delta'}{\alpha(\Delta')-\beta(\Delta')} = \xi. \tag{4.2}$$

Hence $G(\Delta)$ is a Gaussian random variable. The appropriate p.d.f. for Δ is therefore

$$P(\Delta_0, 0; \Delta, t) = \frac{\exp[-\{G(\Delta)-G(\Delta_0)\}^2/2\sigma_x^2]}{(2\pi)^{\frac{1}{2}}\sigma_x(\alpha(\Delta)-\beta(\Delta))}. \tag{4.3}$$

The factor of $(\alpha-\beta)^{-1}$ is the Jacobian, $|d\xi/d\Delta|$, of the transformation: $\xi \leftrightarrow \Delta$.

As $\Delta \to \infty$, $G \to \Delta$ and $\alpha-\beta \to 1$ so that (4.3) becomes approximately Gaussian at large Δ. Thus, we recover the 'Batchelor–Obukhov' hypothesis of Gaussianity (Monin & Yaglom, § 24.4); but the variance of this Gaussian distribution is σ_x^2 not $\overline{\Delta^2}$! This is because Gaussianity only applies when $\Delta \gg L$, where energy-containing eddies are active. When inertial range eddies come into play ($\Delta = o(L)$) (4.3) departs from a Gaussian form.

As $\Delta \to 0, \alpha-\beta \to R^{\frac{1}{2}} \to |\Delta/L|^{\frac{1}{3}}$; $P(\Delta)$ is therefore (integrably) infinite at $\Delta = 0$. The 'perfectly correlated' model (2.7) is more strongly singular at $\Delta = 0$; while the 'uncorrelated' model (2.13) is not singular. Hence (4.3) corresponds to an intermediate degree of 'correlation'.

We have mentioned the 'Batchelor–Obukhov' hypothesis above. Obukhov's approach to relative dispersion (see pp. 571–573 of Monin & Yaglom 1971), like our own, was based on a stochastic model. A comparison of his and our approaches is made in appendix B. There it is suggested that his representation of dispersion is not correct at small separation. Hence, his Gaussian form for $P(\Delta)$ should not be correct as $\Delta/L \to 0$. Furthermore, Obukhov's model forces $\overline{\Delta^2}$ to vary as t^3 (see § 5) by letting the turbulence be non-stationary. Batchelor's deduction of Gaussianity has already been criticized in the introduction. Our new p.d.f. could also be criticized; however, we believe it to contain more of the relevant physics than do other models.

A solution for $P(\Delta)$ was presented above partly because it reveals the nature of our model but, more importantly, partly because it enters implicitly into the following discussions of mean-square dispersion and concentration fluctuations.

5. Mean-square dispersion

A large amount of the literature on (one- and two-particle) dispersion deals with prediction of quantities like $\overline{Z^2}$ and $\overline{\Delta^2}$; the usual reasoning being that $\frac{1}{2}(\overline{Z^2}+\overline{\Delta^2})$ provides a measure of the dispersion of an *ensemble* of clouds of contaminant, while $\overline{\Delta^2}$ is indicative of the dispersion of *individual* clouds. In the present application these quantities are related to the production of concentration fluctuations downstream of a source. Although we shall not do so here, well-known theoretical asymptotic values of $\overline{\Delta^2}$ and $\overline{Z^2}$ can be derived from our model. This fact suggests our model is a plausible interpolation between these limits.

The asymptotes which can be derived for our model are:

$$\lim_{\sigma_x\to 0}\begin{cases}\overline{\Delta^2}/L^2 = \frac{16}{27}(2/\pi)^{\frac{1}{2}}(\sigma_x/L)^3,\\ \overline{Z^2}/L^2 = 2\,(\sigma_x/L)^2;\end{cases} \tag{5.1}$$

$$\lim_{\sigma_x\to \infty}\begin{cases}\overline{\Delta^2} = \sigma_x^2 - O(L\sigma_x),\\ \overline{Z^2} = \sigma_x^2 + O(L\sigma_x);\end{cases} \tag{5.2}$$

here σ_x is given by (4.1). These asymptotic behaviours are those one expects (Monin & Yaglom 1971, § 24).

In Taylor's (1921) one-particle model the mean moment of inertia of a cloud of dispersing contaminant was always equal to σ_x^2. That model represented the cloud by a single dispersing particle. In our two-particle model the moment of inertia is

$$\frac{\overline{z_1^2}+\overline{z_2^2}}{2} = \frac{\overline{Z^2}+\overline{\Delta^2}}{2}.$$

As $\sigma_x \to 0$, or $\sigma_x \to \infty$

$$\frac{\overline{Z^2}+\overline{\Delta^2}}{2} \to \sigma_x^2$$

to lowest order; but, because the cloud has finite spatial scale, higher-order corrections to this value (as given, e.g., by (5.1) or (5.2)) occur when σ_x is finite. Although these higher-order terms are not important for predicting mean concentrations (or other functionals of P_1), they are required for prediction of concentration fluctuations: the next section shows why this is true.

6. Concentration fluctuations

In this section calculations of $\overline{C}$, $\overline{c'^2}$ and $\overline{c_u'^2}$, via equations (2.1), (2.6) and (2.8), are presented. The sources are instantaneous, of the form $S(z,t) = S(z)\,\delta(t)$. Three prescriptions of $S(z)$ will be considered: (i) uniform gradient, $S(z) = mz$; (ii) step profile,

$S(z) = \mathrm{sgn}\,(z)$; (iii) line source. We will present both numerical and analytical results. The following briefly describes how numerical calculations were performed.

Numerical method

Pairs of particle trajectories can be found by solving finite-difference versions of (3.2) and (3.5). In the finite-difference approximation dW_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. Thus, in finite-difference form

$$\left.\begin{aligned} U^{(i)}_{n+1} &= \left(1-\frac{\Delta T}{T_L}\right)U^{(i)}_n + \sigma_w\sqrt{\left(\frac{2\Delta T}{T_L}-\frac{\Delta T^2}{T_L^2}\right)}\,\chi^{(i)}_n, \\ z^{(1)}_{n+1} &= z^{(1)}_n + \frac{\Delta T}{2}\,[\alpha_n(U^{(1)}_n + U^{(1)}_{n+1}) + \beta_n(U^{(2)}_n + U^{(2)}_{n+1})], \end{aligned}\right\} \qquad (6.1)$$

with a similar equation for $z^{(2)}_n$. (The $(\Delta T/T_L)^2$ under the square-root sign in the first equation is a small term. It is required if the turbulence is to be strictly stationary (Durbin 1980).) Owing to the randomness, we experienced no difficulties with stability. We found that our results were independent of the magnitude of ΔT provided it was much smaller than T_L. It was also found that centring α and β (i.e. replacing them by $\alpha_{n+\frac{1}{2}}$ and $\beta_{n+\frac{1}{2}}$ in (6.1)) had no discernible effect on results.

In practice one solves for the *initial* positions of the particles, z'_1, z'_2 at time 0, given their *final* positions, $z_1 = z_2 = z$ at time t (see figure 2). This concept of reversed diffusion (valid in stationary turbulence) was introduced by Corrsin (1952). Having found z'_1 and z'_2 one then assigns particles 1 and 2 the concentrations $C^{(1)}(z,t) = S(z'_1)$, $C^{(2)}(z,t) = S(z'_2)$. In this way $C^{(1)}_n(z,t)$ and $C^{(2)}_n(z,t)$, for the nth particle pair, can be found as a function of measurement time t, and position z.

$$\left.\begin{aligned} \overline{C^2} &= \frac{1}{N}\sum_1^N C^{(1)}_n C^{(2)}_n, \\ \overline{C^2_u} &= \frac{1}{2N}\sum_1^N C^{(1)2}_n + C^{(2)2}_n, \\ \bar{C} &= \frac{1}{2N}\sum_1^N C^{(1)}_n + C^{(2)}_n, \\ \overline{c'^2} &= \overline{C^2} - \frac{1}{N^2}\sum_1^N C^{(1)}_n \sum_1^N C^{(2)}_n, \\ \overline{c'^2_u} &= \overline{C^2_u} - \bar{C}^2 \end{aligned}\right\} \qquad (6.2)$$

are computed at fixed (z,t). Formulae (6.2) correspond to the statistical theory presented in § 2.

When statistics are computed from a sample of N particles they have an uncertainty of order $N^{-\frac{1}{2}}$. Numerical results were obtained for large values of N (500 to 1000). Results for $\bar{C}$ and $\overline{c'^2_u}$ tended, therefore, to have fairly small relative uncertainties. $\overline{c'^2}$, on the other hand, often had significant uncertainty. Estimates of its standard deviation

$$\sigma = \left[\frac{1}{N}\sum_1^N (C^{(1)}_n C^{(2)}_n - \overline{C^2})^2\right]^{\frac{1}{2}} N^{-\frac{1}{2}}, \qquad (6.3)$$

$N \gg 1$, have been included in figures 3, 6 and 7 with our numerical results. On the basis of the central limit theorem, $\pm$ one standard deviation from $\overline{c'^2}$ delimits a 68 % confidence interval.

Case (i): *Initially uniform gradient*

If the source function in (2.1), (2.6) and (2.8) has uniform gradient, $S = mz$, then fluctuating concentrations will be expressed in terms of mean-square dispersion. Corrsin (1952) gave a one-particle analysis showing that the mean gradient is constant for all time

$$d\bar{C}/dz = m, \tag{6.4}$$

and that $\overline{c_u'^2}$ grows according to

$$\overline{c_u'^2} = m^2\sigma_x^2. \tag{6.5}$$

This follows e.g. from (2.8) if $P_1(z = 0, z') = ((2\pi)^{\frac{1}{2}}\sigma_x)^{-1}\exp[-z'^2/2\sigma_x^2]$ and $S = mz'$. Next we derive $\overline{c'^2}$ as given by the present analysis.

Substituting $S = mz$ into (2.6) and setting $z = 0$, so that $\bar{C} = 0$,

$$\overline{c'^2} = m^2\iint_{-\infty}^{\infty} P_2(z = 0, t; z_1', z_2')\, z_1' z_2'\, dz_1'\, dz_2'.$$

The transformation (3.1) allows this to be rewritten in terms of Δ and Z:

$$\overline{c'^2} = \frac{m^2}{2}\iint_{-\infty}^{\infty} P_2(z = 0, t; \Delta', Z')\,(Z'+\Delta')\,(Z'-\Delta')\, dZ'\, d\Delta' = \frac{m^2}{2}(\overline{Z^2}-\overline{\Delta^2}). \tag{6.6}$$

Again, this formula connects fluctuations to mean-square dispersion; but now mixing between blobs of contaminant – the $\overline{\Delta^2}$ term – is included, as well as centre-of-mass dispersion.

We find according to (4.1) and (6.5)

$$\overline{c_u'^2} \to 2m^2\sigma_w^2 T_L t = O(t);$$

while, according to (5.2) and (6.6),

$$c_u'^2 \to m^2 O(L\sigma_x) = O(t^{\frac{1}{2}})$$

as $t \to \infty$. Thus, (6.6) predicts much lower levels of $\overline{c'^2}$ than does (6.5). As $t \to 0$

$$\overline{c'^2} \to \overline{c_u'^2} \to m^2(\sigma_w t)^2.$$

Until now, we have described the phenomenology of our model in terms of 'blobs of contaminant'. Equation (6.6) can be alternatively described in Eulerian conceptions of 'production' and 'dissipation' of fluctuations.

Eulerian interpretation. Our statistical theory and stochastic model follow what is commonly called the *Lagrangian* approach to dispersion. The alternative, *Eulerian* approach starts from the conservation equation

$$\frac{\partial c}{\partial t} + +\mathbf{u}\,.\,\nabla c = k\nabla^2 c$$

for contaminant concentration. It then follows that

$$\frac{\partial \overline{c'^2}}{\partial t} = \begin{cases} -\,2\overline{c'w'}m - 2\kappa\overline{\nabla c'\,.\,\nabla c'} \\ \quad P \quad\quad - \quad\quad \epsilon_c \end{cases} \tag{6.7}$$

(Csanady 1973, §7.5; the term $\nabla . \overline{\mathbf{u}'c'^2}$ is zero owing to homogeneity) describes the evolution of $\overline{c'^2}$. The term P represents production and ϵ_c represents dissipation of fluctuations. It is interesting to interpret Corrsin's formula (6.5) and formula (6.6) in terms of the symbols P and ϵ_c.

As Corrsin himself noted, his formula (6.5) contains only the production term $P = m^2 d\sigma_x^2/dt$: ϵ_c is zero. Similarly, the first term in (6.6) is the production

$$P = \tfrac{1}{2}m^2 d\overline{Z^2}/dt.$$

(Note that, if $\overline{\Delta^2} = 0$, $\overline{Z^2} = 2\sigma_x$, so this is the two-particle analogue of Corrsin's formula for P.) However, ϵ_c is not zero in (6.6): it is equal to $\frac{1}{2}m^2 d\overline{\Delta^2}/dt$.

Because of its explicit form, $2\kappa\overline{|\nabla c'|^2}$, the *rate* of dissipation, ϵ_c, is usually ascribed to destruction of small-scale gradients, which were produced by larger-scale motions. The present, Lagrangian, analysis does not represent these small-scale processes. However, they are rate-limited by the larger-scale processes which our analysis does describe. For this reason the *rate* of dissipation does not enter our Lagrangian analysis; but the analogue of *net* dissipation does. It is what we have described as mixing between blobs of contaminant. Can the Lagrangian and Eulerian views be reconciled?

When two particles such as those depicted in figure 2 come together they juxtapose contaminant parcels with a concentration difference $2^{\frac{1}{2}} m\Delta$. In the present view, these particles approach to within a distance of $O(\eta)$ of each other before molecular processes smooth them together. Thus gradients of $O(m\Delta/\eta)$ are produced and the fluctuating dissipation rate is $\epsilon_c' = O(\kappa m^2\Delta^2/\eta^2)$. But, gradients are smoothed on the short time-scale η^2/κ (short because $\eta^2/\kappa \ll T_L$).

The mean net dissipation is therefore

$$\overline{\int_0^t \epsilon_c' \, dt} \sim \overline{\int_0^{\eta^2/\kappa} \kappa m^2 \Delta^2/\eta^2 \, dt} \sim \left(\frac{\kappa m^2 \overline{\Delta^2}}{\eta^2}\right) \eta^2/\kappa = m^2\overline{\Delta^2}, \tag{6.8}$$

which agrees with (6.6). The integration in (6.8) is over the time-scale η^2/κ, on which t is constant with respect to T_L.

If we let $\epsilon_c = \overline{\epsilon_c'} = \kappa m^2 \overline{\Delta^2}/\eta^2$ and compute net dissipation from (6.7) we get

$$\int_0^t \epsilon_c \, dt \sim \int_0^t \kappa m^2 \overline{\Delta^2}/\eta^2 \, dt \neq m^2\overline{\Delta^2}.$$

In fact, this last integral is $O((m^2\overline{\Delta^2}T_L)/(\eta^2/\kappa))$; *or*, a factor of $T_L\kappa/\eta^2$ larger than (6.8). The disparity between the Eulerian and Lagrangian estimates lies in the fact that dissipation appears continuously on the time-scale η^2/κ but is intermittent on the scale T_L (on which we are considering the evolution of $\overline{c'^2}$). One must take account of intemittency and revise the estimate of ϵ_c from $\kappa m^2\overline{\Delta^2}/\eta^2$ down to $m^2 d\overline{\Delta^2}/dt$. Only then can one recover (6.6) from (6.7). Below (6.6) its asymptotic behaviours were described. Its intermediate behaviour was found by numerical simulation.

Numerical results. In figure 3 numerical evaluations of $\overline{c_u'^2}/m^2L^2$ and $\overline{c'^2}/m^2L^2$ as functions of t/T_L are presented. Here, and in all our calculations, we have used $L = \sigma_w T_L$. The upper solid curve in figure 3 is $\overline{c_u'^2}$. Although it was computed via the two-particle model (3.5), this curve is practically identical with the value, $(\sigma_x/L)^2$, that Corrsin (1952) predicted for $\overline{c_u'^2}/m^2L^2$ using a one-particle model. Indeed, we have not

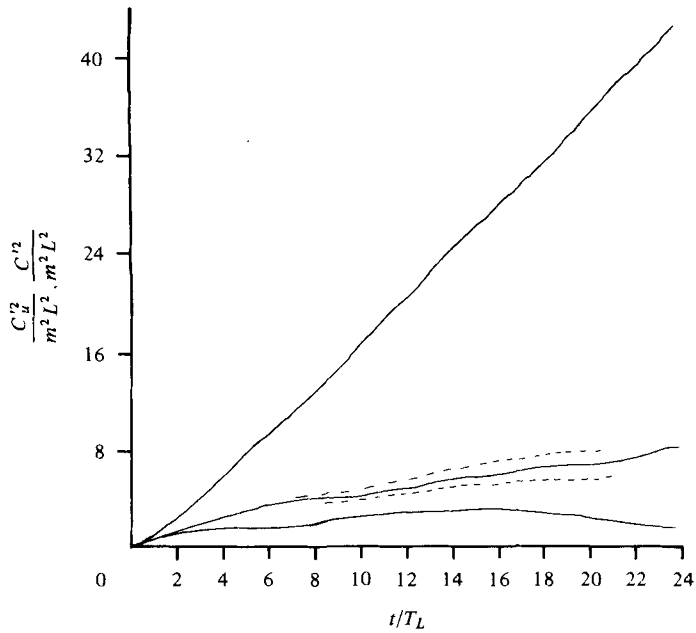

FIGURE 3. Comparison of $\overline{c'^2}$ computed from various models: the upper solid curve is $\overline{c_u'^2}$, computed via one-particle statistics; the middle solid curve is $\overline{c'^2}$, the present two-particle definition; the lowest solid curve is $\overline{c'^2}$ computed with a non-dimensional mean shear of 2·5. The dashed lines, about the middle curve, provide a 68 % confidence interval for that curve. All curves are averages over 1000 particle pairs and initially the concentration gradient is uniform.

plotted $(\sigma_x/L)^2$, as given by (4.1), in the figure because it would obscure the numerically simulated curve.

In the last paragraph of § 5 it was remarked that the higher-order correlations introduced by the simultaneous dispersion of two particles are irrelevant when computing quantities dependent on the single particle p.d.f. In our numerical modelling we have generally found that values of $\overline{C}$ and $\overline{c_u'^2}$ (which depend only on P_1) computed via the two-particle model (3.5) are indistinguishable from values computed via the simpler one-particle model

$$dz_1/dt = U(t). \tag{6.9}$$

Equation (6.9) is, of course, Taylor's model for one-particle dispersion. That our two-particle model reproduces the one-particle model, in appropriate situations, is an adequacy of its formulation.

The middle solid curve in figure 3 is $\overline{c'^2}$: the dotted lines around it are 68 % confidence intervals (cf. 6.3). As $t/T_L \to 0, \overline{c'^2} \to \overline{c_u'^2}$, as expected; while as $t/T_L \to \infty, \overline{c'^2} \ll \overline{c_u'^2}$. Definition (2.6) does, indeed, result in lower fluctuations than (2.8).

The upper solid curve in figure 3 is, according to (6.2),

$$\frac{\overline{z_1^2}+\overline{z_2^2}}{2L^2} = \frac{\overline{\Delta^2}+\overline{Z^2}}{2L^2};$$

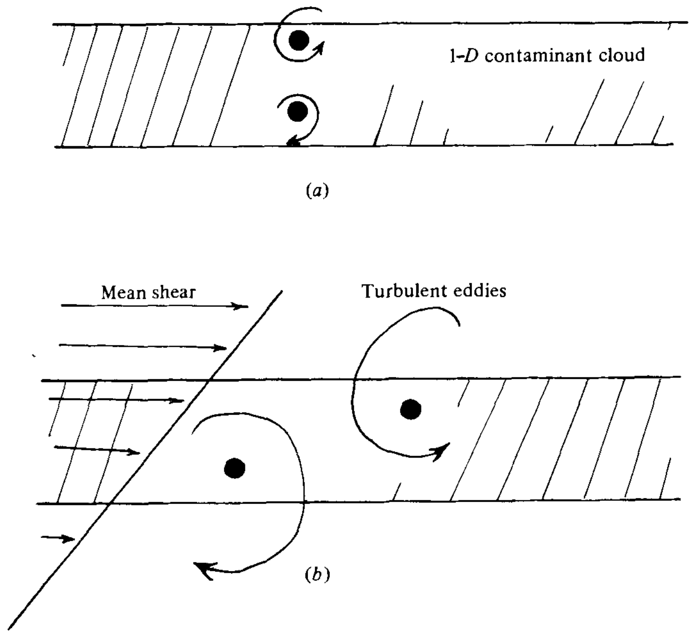

FIGURE 4. The effect of mean shearing on dispersion of one-dimensional clouds: In figure (*a*) there is no shear and small eddies disperse particle pairs. In (*b*) mean shear separates particles enabling larger, more energetic, eddies to come into play. These larger eddies mix the cloud more efficiently, decorrelate particle positions and reduce concentration fluctuations.

while the middle curve is

$$\frac{\overline{z_1 z_2}}{L^2} = \frac{\overline{Z^2} - \overline{\Delta^2}}{2L^2}.$$

Hence, values of $\overline{Z^2}$ or $\overline{\Delta^2}$ can be obtained by adding or subtracting these curves.

We have just seen that, in an unbounded fluid with uniform initial gradient, production of fluctuations can overwhelm dissipation so that $\overline{c'^2}$ grows indefinitely. In a bounded flow, or in the presence of (uniform) mean shearing this is not necessarily true. The case of mean shearing is treated below; applications of our model to inhomogeneous, bounded flows will be described in a future paper.

The effect of mean shearing. A further consequence of making $\overline{c'^2}$ dependent on the correlation between particle pairs is that mean shearing will reduce $\overline{c'^2}$. In Corrsin's one-particle analysis mean shear has no effect. The present paper is restricted to homogeneous turbulence so we consider a uniform mean shear.

Mean shear increases the separation between particle pairs (see figure 4), thereby helping to decorrelate their random velocities. This, in turn, leads to decorrelation between the positions – or, equivalently, to increased turbulent mixing and to reduction of fluctuations (see (2.13)).

The common approximation of ignoring streamwise turbulent velocity will be made and the mean x velocity $U = \gamma z$ used. Then the relative x displacement of a particle pair follows from

$$\frac{d}{dt}(x_1 - x_2) = \gamma(z_1 - z_2) = \sqrt{2}\,\gamma\Delta(t) \tag{6.10}$$

and the solution to (3.4) for $\Delta(t)$.

Very briefly, R of equation (3.7) must be replaced by a component of an isotropic correlation function for incompressible turbulence (Monin & Yaglom, § 21.4). Thus,

$$R = \left(\frac{\epsilon^2}{L^2+\Sigma^2}\right)^{\frac{1}{3}}\left[1+\frac{L^2\Delta_x^2}{(L^2+\Sigma^2)\,\Sigma^2}\right], \tag{6.11}$$

where

$$\Delta_x \equiv \frac{x_1-x_2}{\surd 2} \quad \text{and} \quad \Sigma^2 = \Delta^2+\Delta_x^2.$$

The lowest solid curve in figure 3 is a numerical calculation with non-dimensional shear $\hat{\gamma} = \gamma L/\sigma_w$, equal to 2·5; $\overline{c'^2}$ is reduced from its value when $\hat{\gamma} = 0$.

As $t \rightarrow 0$, $\overline{\Delta_x^2} \rightarrow O(t^5) \ll \overline{\Delta^2}$, so that dispersion takes place initially as it would in the absence of shear. As $t \rightarrow \infty$ pairs of particles tend to move independently and their individual motions can be approximated by a Markov process (Monin & Yaglom, § 10.3). Thus, letting $\Delta \approx \sigma_w\,(2T_L)^{\frac{1}{2}}\,W_t$, substituting into (6.10), integrating, squaring and averaging gives, to lowest order,

$$\lim_{t/T_L\rightarrow\infty} \overline{\Delta_x^2} = \tfrac{4}{3}\gamma^2 T_L\,\sigma_w^2\,t^3. \tag{6.12}$$

It follows that, as $t \rightarrow \infty$, $\overline{\Delta_x^2}/\overline{\Delta^2} \rightarrow O(t^2)$; so the effect of mean shearing dominates relative dispersion. Curiously $\overline{\Delta^2}+\overline{\Delta_x^2} \rightarrow O(t^3)$ in both limits $t \rightarrow 0$ and $t \rightarrow \infty$! A similar result is described in Csanady (1973, § 5.13): it may explain the prevalence of t^3 behaviour in observational data on relative dispersion.

We have not found the exact asymptotic behaviour of $\overline{c'^2}$ as $t \rightarrow \infty$ in this case; but (6.12) and our numerical calculations suggest that $\overline{c'^2}$ tends asymptotically to zero. Physically, this is because, on average, the mean shear causes larger separation between particle pairs so that larger eddies disperse them. These larger eddies can stir the fluid more efficiently, thereby reducing concentration fluctuations (see figure 4).

Case (ii): *Step profile*

In this case the source function is $S(z) = \operatorname{sgn}(z)$. The quantities $\bar{C}$ and $\overline{c_u'^2}$ depend on P_1. We use the dictum (see above) that our two-particle model will closely reproduce the results of the one-particle model (6.9) for these quantities. Equation (6.9) is simply the classical UO model of Brownian motion (Wax 1954). Since dW_t is Gaussian white noise it follows that z_1, being a linear summation of Gaussian noise, is also Gaussian. Hence

$$P_1(z;z') = \frac{\exp\left[-(z-z')/2\sigma_x^2\right]}{(2\pi)^{\frac{1}{2}}\,\sigma_x}. \tag{6.13}$$

Substituting (6.13) into (2.1) and equating $S(z')$ to $\operatorname{sgn}(z')$ gives

$$\bar{C}(z,t) = \operatorname{erf}(|z|/\surd(2)\,\sigma_x)\operatorname{sgn}(z). \tag{6.14}$$

Because $\overline{C_u'^2} \equiv 1$, (6.14) implies

$$\overline{c_u'^2} = 1-\operatorname{erf}^2(z/\surd(2)\,\sigma_x). \tag{6.15}$$

A numerical calculation of $\bar{C}$ at the cross-stream position $z/L = 2$ is shown in figure 5 along with the analytical formula (6.15). The numerical curve is quite erratic because S is discontinuous; but the numerical results agree closely with (6.14): our dictum is obeyed.

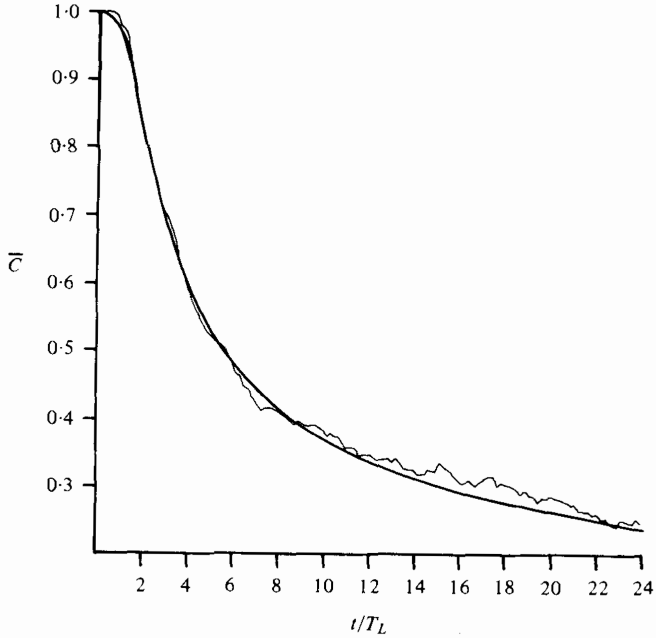

FIGURE 5. Mean concentration at $z/L = 2$ as a function of time when the initial concentration is a step function. The smooth curve is the analytic solution (6.14) and the irregular curve was produced by averaging numerical simulations of 800 particle pairs.

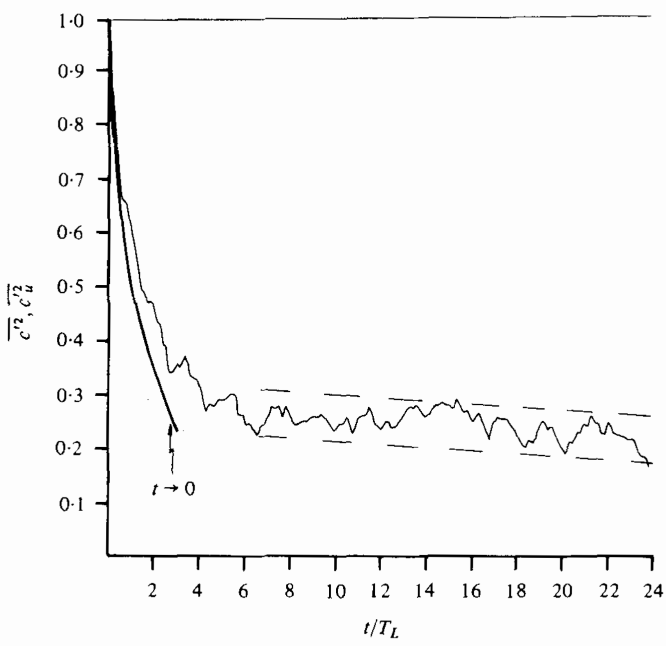

FIGURE 6. $\overline{c_u'^2}$ (straight line) and $\overline{c'^2}$ (irregular line) at $z/L = 0$ as functions of time when the initial concentration profile is a step function. The small t asymptote to $\overline{c'^2}$ (6.18) and the 68 % confidence interval (dashed lines) are also shown. 500 particle pairs were used to compute numerical statistics.

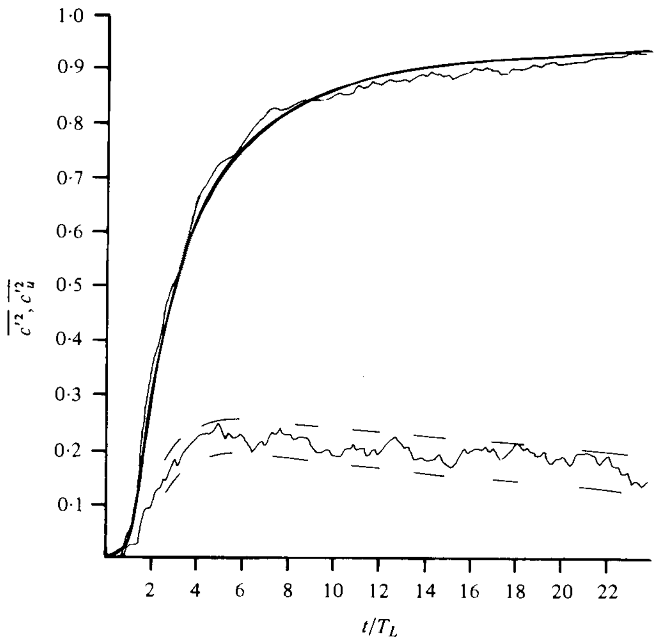

FIGURE 7. As in figure 6, but now $z/L = 2$ and 800 particle pairs are used. We have also plotted (6.15) for comparison with $\overline{c_u'^2}$ (upper curves).

At $z/L = 0, \bar{C} \equiv 0, \overline{c_u'^2} \equiv 1$. The numerical model, of course, also gives $\overline{c_u'^2} = 1$; as shown by the upper line in figure 6. The upper curves in figure 7 are (6.15) compared with numerical evaluations of $\overline{c_u'^2}$ (equation (6.2)) at $z/L = 2$.

The one-particle model is quite tractable. Unfortunately, two-particle statistics are more difficult to treat. We revert to the practice of analysing asymptotic behaviours and filling in details by numerical simulation.

Substituting a step profile for S into (2.6) and noting that $P_2(z_1', z_2') = P_2(z_2', z_1')$ since the particles are indistinguishable, gives

$$\overline{C^2} = 2\left\{\int_0^\infty \int_{z_1}^\infty + \int_{-\infty}^0 \int_{z_1}^0 - \int_{-\infty}^0 \int_0^\infty\right\} P_2(z, t; z_1', z_2')\, dz_2'\, dz_1'. \tag{6.16}$$

This can be re-expressed in terms of Z and Δ by noting that the Z, Δ co-ordinate system is obtained by rotating the z_1, z_2 system through $\frac{1}{4}\pi$ radians (see (3.1)):

$$\begin{aligned}\overline{C^2} &= 2\int_0^\infty \left\{\int_\Delta^\infty + \int_{-\infty}^{-\Delta} - \int_{-\Delta}^{\Delta}\right\} P_2(z, t; Z', \Delta')\, dZ'\, d\Delta' \\ &= 1 - 4\int_0^\infty \int_{-\Delta}^{\Delta} P_2(z, t; Z', \Delta')\, dZ'\, d\Delta'. \end{aligned} \tag{6.17}$$

The asymptotic behaviour of this last integral as $t/T_L \to 0$, when $z/L = 0$, can be shown to be

$$\overline{c'^2} = 1 - 0{\cdot}5282(\sigma_x/L)^{\frac{1}{2}}. \tag{6.18}$$

This is compared, in figure 6, with a numerical simulation of $\overline{c'^2}$ at $z = 0$. It works reasonably well out to $t/T_L \sim 3$.

If $\overline{c_u'^2}$ is considered the squared fluctuation intensity which would appear in the absence of dissipation and $\overline{c'^2}$ that which appears when dissipation is present then, at $z/L = 0$,

$$\overline{\int_0^t \epsilon_c' \, dt} = \overline{c_u'^2} - \overline{c'^2} \approx 0{\cdot}5282(\sigma_x/L)^{\frac{1}{2}}, \tag{6.19}$$

when $t/T_L \to 0$. In figure 6 this dissipation causes dramatic reduction of $\overline{c'^2}$.

As $t/T_L \to \infty$ asymptotic analysis suggests that $\overline{c'^2}$ tends to zero as $(\sigma_x/L)^{-1}$, or as $(t/T_L)^{-\frac{1}{2}}$. Thus, dissipation is capable, in this case, of eventually overwhelming production.

In figure 7 $\overline{c'^2}$ and $\overline{c_u'^2}$ at $z/L = 2$ are compared. As $t/T_L \to 0$ both are exponentially small. When t/T_L crosses ~ 1 both start to grow; $\overline{c_u'^2}$ tends toward its asymptotic value of 1; $\overline{c'^2}$ first increases and then, as asymptotic analysis suggests, begins a slow descent toward zero. This slow descent is partially obscured by the noisiness of the computed statistics.

The discontinuous profile for S generally produces quite noisy curves for $\overline{c'^2}$. In figures 6 and 7 the dashed lines are estimated 68 % confidence intervals: the time traces of $\overline{c'^2}$ fill this interval.

Case (iii): *Line source*

The theory described in § 2 is designed for *distributed* initial sources. However, an analysis for *line* sources can be made by first assuming they have some finite size, $\delta \gg \eta$, and then letting $\delta \to 0$. The analysis of this case is similar to that of case (ii).

Let

$$S(z) = \begin{cases} N & \text{for} \quad |z| \leqslant \delta, \\ 0 & \text{for} \quad |z| > \delta \end{cases} \tag{6.20}$$

and assume $\delta \ll \sigma_x$. The mean concentration is found, from the one-particle model (6.9), to be

$$\bar{C} = \frac{N}{2}\{\operatorname{erf}(\delta - z/\sqrt{(2)}\,\sigma_x) + \operatorname{erf}(\delta + z/\sqrt{(2)}\,\sigma_x)\}$$

$$\approx \left(\frac{2}{\pi}\right)^{\frac{1}{2}} \frac{N\delta}{\sigma_x} e^{-z^2/2\sigma_x^2}. \tag{6.21}$$

Equation (2.12) applies in the present case: substituting $Nf = \bar{C}$ into it gives

$$\overline{c_u'^2} = N\bar{C} - \bar{C}^2$$

or, from (6.21),

$$s^2(z,t) \equiv \frac{\overline{c_u'^2}}{\bar{C}^2} = \left(\frac{\pi}{2}\right)^{\frac{1}{2}} \frac{\sigma_x}{\delta} e^{z^2/2\sigma_x^2} - 1. \tag{6.22}$$

$s(z,t)$ is the intensity of fluctuations relative to the mean. Thus, according to the usual definition, at $z = 0$

$$s(0,t) = \left(\left(\frac{\pi}{2}\right)^{\frac{1}{2}} \frac{\sigma_x}{\delta} - 1\right)^{\frac{1}{2}}. \tag{6.23}$$

Because $\sigma_x \gg \delta$ the relative fluctuation intensity grows as $t^{\frac{1}{2}}$ when $t/T_L \to 0$ and as $t^{\frac{1}{4}}$ when $t/T_L \to \infty$.

Now we proceed to analyse the two-particle model. After rearrangement, definition (2.6), with $S(z)$ given by (6.20), can be written

$$\overline{C^2} = 2N^2 \int_0^{\sqrt{(2)}\delta} \int_{-\sqrt{(2)}\delta+\Delta'}^{\sqrt{(2)}\delta-\Delta'} P_2(\Delta', Z')\, dZ'\, d\Delta'. \tag{6.24}$$

An analysis of (6.24), which we omit for brevity, leads to

$$\lim_{G(\sqrt{(2)}\delta)/\sigma_x \to 0}\ \lim_{\delta/L \to 0} \overline{C^2} = \frac{0{\cdot}9N^2\sqrt{(2)}\,L^2}{\pi\sigma_x^2}\left(\frac{\sqrt{(2)}\,\delta}{L}\right)^{\frac{5}{3}} e^{-z/2\sigma_x^2}. \tag{6.25}$$†

Combining (6.21) and (6.25) gives

$$s^2(z,t) = \frac{\overline{c'^2}}{\bar{C}^2} = 0{\cdot}9\sqrt{2}\left(\frac{L}{\sqrt{(2)}\,\delta}\right)^{\frac{1}{3}} e^{z^2/2\sigma_x^2} - 1; \tag{6.26}$$

or, on $z = 0$,

$$s^2(0,t) = -0{\cdot}9\sqrt{2}(L/\sqrt{(2)}\,\delta)^{\frac{1}{3}} - 1. \tag{6.27}$$

Thus, with our new definition for $\overline{C^2}$ the relative fluctuation intensity does not continue to grow as $t \to \infty$, but attains the 'equilibrium' value (6.27). In fact, (6.26) indicates that $s(z,t)$ is a function only of the similarity variable $z/\sqrt{(2)}\,\sigma_x$ and not of σ_x itself; it does, however, depend on $(L/\delta)^{\frac{1}{3}}$. Because of this δ-dependence we conclude that: 'discussions of concentration fluctuations are useless unless they take account of the finite initial size of the cloud. (In particular there is no point in considering an initial point source.)' (Chatwin & Sullivan 1979, p. 341). However, the present argument leading to this conclusion differs from Chatwin & Sullivan's. Their argument would lead to (6.23) in one-dimensional dispersion; *or* to s depending on σ_x/δ.

7. Discussion

What are the significances of the limiting conditions imposed in (6.25)? When δ/L is not small departures from a self-preserving form may occur; however, we expect these to be minimal. (Of course, if δ/L is not small the $\frac{1}{3}$ law (6.27) is not valid.) Rather, the finite magnitude of δ/L will be important at an early stage when fluctuations are being produced. (6.26) applies only when fluctuations are decaying; but, for any finite-sized source, there is a time during which fluctuations are produced. This time increases as δ/L increases.

On the contrary, if $G(\sqrt{(2)}\,\delta)/\sigma_x$ is not small the self-preserving form will not yet have been attained. This is true even though the production stage may be completed. In other words, we may be on an 'elbow' connecting the production stage to the self-preserving stage.

Note that $G(\sqrt{(2)}\,\delta)/\sigma_w$ is the time-scale required for mixing between the contaminant cloud and the environment (see equations (3.4) and (4.2)). The ratio σ_x/σ_w is a time-scale for bulk turbulent stirring. Thus $G(\sqrt{(2)}\,\delta)/\sigma_x$ is an indicator of the probability that contaminant has *not* mixed with surrounding fluid, but has been

† Strictly, this result assumes the further asymptotic limit $\Delta^2 \ll L^2$ or $(t/T_L)^3 \ll 1$, but it is approximately true at larger t.

bodily dispersed. As $G(\surd(2)\,\delta)/\sigma_x$ decreases the probability of mixing increases. When we speak of dissipation of fluctuations we are referring to this mixing with environmental fluid: the condition $G(\surd(2)\,\delta)/\sigma_x \ll 1$ ensures that enough mixing has occurred for this dissipative process to have reached an asymptotic stage.

The limiting condition for (6.21) to be valid has a more straightforward interpretation. In the mean, contaminant is dispersed by bodily convection. The condition $\delta/\sigma_x \ll 1$ ensures that a blob has, on average, been convected several blob radii and hence that the dispersion mechanism has reached an asymptotic stage.

Murthy & Csanady (1971) made measurements of $s(z,t)$ for dye plumes dispersing in Lake Huron. They concluded (see their figure 12) that s was a self-preserving function of $z/\surd(2)\,\sigma_x$ and that $s(z=0)$ was independent of σ_x. (In their experiments σ_x/δ varied by a factor of 2.) Qualitatively, these conclusions favour our new theory, (6.26), over the usual theory (6.22).

Recent experiments by Gad-el-Hak & Morton (1979) on concentration fluctuations in an axisymmetric smoke plume dispersing in isotropic turbulence also produced a self-similar form for $s(z,t)$. Their profiles of $s(z/\surd(2)\,\sigma_x)$ were qualitatively similar to those of Murthy & Csanady. In particular, both sets of data showed the exponential increase of s with increasing $|z/\surd(2)\,\sigma_x|$ that (2.26) predicts.

Csanady (1967) has previously proposed a self-preserving form for $s(z,t)$. His proposal was based on a simple closure model for $\overline{c'^2}$. Csanaday did not include any explicit dependence of s on δ/L. One could argue, however, that his undetermined parameter, α, is a function of $(\delta/L)^{\frac{1}{3}}$, such that $\alpha \to 0$ as $(\delta/L)^{\frac{1}{3}} \to 0$. His figure 1 then agrees with our conclusion that $s(z=0)$ increases as $(\delta/L)^{\frac{1}{3}}$.

Appendix A. Specification of α and β in (3.5)

The coefficients α and β must be specified so that our model reproduces known asymptotic limits. A heuristic argument leading to forms of α and β is given here; in §5 it was shown that the model does, indeed, reproduce the correct asymptotes.

When $\Delta \to 0$ the two particles in equations (3.5) must have the same velocity so $\alpha(0) = \beta(0)$. If we require that $\overline{(dZ/dt)^2} = 2\sigma_w^2$ when $\Delta = 0$ we find $\alpha(0) = \beta(0) = 1/\surd(2)$. When $\Delta \to \infty$, the particle velocities become uncoupled and behave as those of independently moving particles, so $\alpha(\infty) = 1$ and $\beta(\infty) = 0$. In both these limits

$$\alpha^2 + \beta^2 = 1; \tag{A 1}$$

and (A 1) seems a reasonable model constraint, no matter what value Δ has. Further justification for this constraint is given in equation (B 2) of appendix B.

Now, were α and β constant, or were our model Markovian, then

$$\frac{\overline{dz_1\,dz_2}}{(\overline{dz_1^2}\,\overline{dz_2^2})^{\frac{1}{2}}} = \frac{2\alpha\beta}{\alpha^2+\beta^2} = 2\alpha\beta \tag{A 2}$$

would follow from (3.5) and the fact that U' and U'' are independent and statistically identical. In the present context α and β vary slowly on the time scale of the random velocity fluctuations, so (A 2) is a good approximation. Equation (A 2) relates $2\alpha\beta$ to a velocity correlation function. When $\Delta = 0$

$$\frac{\overline{dz_1\,dz_2}}{(\overline{dz_1^2}\,\overline{dz_2^2})^{\frac{1}{2}}} = 1;$$

therefore a convenient form for (A 2) is

$$2\alpha\beta = 1 - R(\Delta), \tag{A 3}$$

where

$$R(\Delta) = 1 - \frac{\overline{dz_1\,dz_2}}{(\overline{dz_1^2}\,\overline{dz_2^2})^{\frac{1}{2}}}$$

is called the 'structure function' (Townsend 1976, p. 11); $1 \geqslant R(\Delta) \geqslant 0$. The function $R(\Delta)$ represents the fraction of the mean squared particle velocity associated with relative drift: obviously $R(0) = 0$ and $R(\infty) = 1$. By (A 1) and (A 3)

$$\left.\begin{aligned} \alpha^2 &= \tfrac{1}{2}(1 + (2R - R^2)^{\frac{1}{2}}), \\ \beta^2 &= \tfrac{1}{2}(1 - (2R - R^2)^{\frac{1}{2}}), \end{aligned}\right\} \tag{A 4}$$

thus $\alpha \geqslant \beta$.

A feature of the model (3.5) is that large-scale turbulence components evolve on the finite time scale, T_L; while small-scale components, as embodied in $\alpha(\Delta)$ and $\beta(\Delta)$, adjust to values appropriate to the instantaneous value of Δ. If Δ is in the inertial range,

$$R(\Delta) = C\epsilon^{\frac{2}{3}}|\Delta|^{\frac{2}{3}}/(2\sigma_w^2)\ (= \text{structure function}/2\sigma_w^2, \text{ Townsend, p. 96}). \tag{A 5}$$

Townsend gives $C \approx 2^{\frac{4}{3}}$, when the present definition of Δ is used. With the semi-empirical formula

$$\epsilon = A\sigma_w^3/L, \quad A \approx 0{\cdot}8 \tag{A 6}$$

(Townsend, p. 61), where L is the turbulence integral scale, (A 5) becomes

$$R(\Delta) = \frac{CA^{\frac{2}{3}}}{2}\left|\frac{\Delta}{L}\right|^{\frac{2}{3}} \approx \left|\frac{\Delta}{L}\right|^{\frac{2}{3}}. \tag{A 7}$$

Equation (A 7) is an approximation when $\Delta/L \to 0$ and hence provides the proper 'outer limit' of R at small Δ. In general, we need an interpolation formula between this limit for small Δ and the limit $R(\infty) = 1$; we will use

$$R(\Delta) = \left(\frac{\Delta^2}{L^2 + \Delta^2}\right)^{\frac{1}{3}}. \tag{A 8}$$

Equations (A 8) and (A 4) prescribe α and β. In appendix B it is shown that these prescriptions are, in a sense, in accord with Richardson's and Thiebaux's diffusion models.

Appendix B. Comparison with other models

Models of relative dispersion proposed by Richardson, Thiebaux and Obukhov have been mentioned in the text. In this appendix these models are discussed in the context of the present model.

Connexion with Richardson's and Thiebaux's diffusion models

The stochastic model (3.4), or (3.5), is non-Markovian because the UO process has a finite memory time. If T_L is allowed to tend to zero, $U(t)$ reduces to the white-noise process, $\sigma_w(2T_L)^{\frac{1}{2}}\,dW_t$, and the model becomes Markovian. Of course, the limit $T_L \to 0$ is trivial in the present context for, to be consistent, one should also let $L \to 0$ in (A 8) so that, in this limit, the particles move independently ($\alpha \equiv 1, \beta \equiv 0$). For the sake of argument, L will be allowed to remain finite.

In the present context, a *Markov* process is equivalent to a *diffusion* process. The 'diffusivity' for the Markov process corresponding to (3.4) with $U(t)$ replaced by $\sigma_w(2T_L)^{\frac{1}{2}}\,dW_t$ is

$$\left(\frac{K_{ij}}{\sigma_w^2 T_L}\right) = \begin{pmatrix} (\alpha+\beta)^2 & 0 \\ 0 & (\alpha-\beta)^2 \end{pmatrix}. \tag{B 1}$$

As $|\Delta|/L \to 0$, $K_{22} \to O(|\Delta|^{\frac{4}{3}})$; hence a connexion between the present model and Richardson's $\frac{4}{3}$ law.

Corresponding to (3.5) one finds

$$\left(\frac{K_{ij}}{\sigma_w^2 T_L}\right) = \begin{pmatrix} \alpha^2+\beta^2 & 2\alpha\beta \\ 2\alpha\beta & \alpha^2+\beta^2 \end{pmatrix}. \tag{B 2}$$

This is the form of K proposed by Thiebaux, provided $\alpha^2+\beta^2 = 1$ (equation (A 1)), for the diagonal elements must be the (homogeneous) one-particle diffusivity. The term K_{12} parametrizes the correlation between velocity fluctuations at z_1 and z_2, as was remarked by Thiebaux (see (A 2)). The diffusion limit of our model is a 'trivial' limit so it will not be pursued further.

Comparison with Obukhov's model

We will use the present notations and numerical constants in this comparison. Readers unfamiliar with technical details of the theory of stochastic differential equations (Arnold 1974) may prefer to ignore this appendix.

Obukhov's model is one of 'Markovian diffusion in phase space' (Monin & Yaglom, § 24.4). This phase space is the two-dimensional (in the present context) space (Δ, V), where $V = d\Delta/dt$. The present model is also Markovian in the space (Δ, U), where we have dropped a superscript 2 from U. Obukhov presented his model in the form of the Fokker–Planck equation

$$\frac{\partial}{\partial t}P(\Delta, V) + V\frac{\partial}{\partial \Delta}P(\Delta, V) = \frac{\sigma_w^2}{T_L}\frac{\partial^2}{(\partial V)^2}P(\Delta, V), \tag{B 3}$$

but the corresponding stochastic differential equations,

$$\left.\begin{aligned} dV &= \sigma_w\left(\frac{2}{T_L}\right)^{\frac{1}{2}} dW_t, \\ d\Delta/dt &= V(t), \end{aligned}\right\} \tag{B 4}$$

are easily inferred. Compare (B 4) with (3.2) and (3.4). Two shortcomings of (B 4) are readily seen: (i) the rate of decorrelation of the velocity fluctuations, $-V/T_L$, has been set to zero in the first of (B 4); (ii) the rate of relative dispersion has been made independent of particle separation in the second of (B 4). Indeed, by (B 4), V is simply the *Wiener process* $\sigma_w(2/T_L)^{\frac{1}{2}}\,W_t$ so $\overline{V^2} = 2\sigma_w^2 t/T_L$. This is far from the stationary random velocity, $\overline{U^2} = \sigma_w^2$, of the Uhlenbeck–Ornstein process used in the present model.

It does not seem, therefore, that the proper physics are contained in (B 4): and, in this sense, it is fortuitous that, when the Wiener process V is integrated to find Δ, the t^3 law, $\Delta^2 = \overline{\sigma_w^2 t^3/3T_L}$, is obtained. One could not expect (B 4) to model higher-order statistics, or $P(\Delta)$, correctly. In §6 (see equation (6.10)) it is proposed that equations like (B 4) are a more appropriate model of the asymptotic form of relative (streamwise) dispersion produced by homogeneous turbulence in a mean shear flow.

To complete our comparison with Obukhov the phase-space Fokker–Planck equation for (3.2) and (3.4) is given below:

$$\frac{\partial}{\partial t}P(\Delta, U)+U\frac{\partial}{\partial \Delta}[(\alpha(\Delta)-\beta(\Delta))P(\Delta, U)]-\frac{1}{T_L}\frac{\partial}{\partial U}[UP(\Delta, U)] = \frac{\sigma_w^2}{T_L}\frac{\partial^2}{(\partial U)^2}P(\Delta, U). \tag{B 5}$$

This can be compared with (B 3).

The marginal distribution,

$$P(U) = \frac{1}{(2\pi)^{\frac{1}{2}}\sigma_w}\exp[-u^2/2\sigma_w^2]$$

(Wax 1954) for the UO velocity process follows from (B 5), as does the marginal distribution (4.3).

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