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The relative diffusion of a cloud of passive contaminant in incompressible turbulent flow

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A problem of major practical interest is the variation with $\mathbf{x}$ and t of the statistical properties of $\Gamma(\mathbf{x}, t)$, the distribution of concentration of a contaminant in a cloud containing a finite quantity Q of contaminant, released in a specified way at $t = 0$ over a volume of order L_0^3. Of particular relevance is the case of relative diffusion (when $\mathbf{x}$ is measured throughout each realization relative to the centre of mass of the cloud), when important properties are $L(t)$, the linear dimension of the cloud, $C(\mathbf{x}, t)$, the ensemble mean concentration, $\overline{c^2}(\mathbf{x}, t)$, the variance of the concentration, and $p(\mathbf{y}, t)$, the distance-neighbour function. Much fundamental work has led to a knowledge of the way L varies with t, but not of the way the other properties vary. Hitherto therefore, prediction of such variation has normally used unjustifiable empirical concepts such as eddy diffusivities, but this is ultimately unsatisfactory, practically as well as theoretically. Hence the exact equations have been used to obtain a quite new description of the structure of a dispersing cloud, which it is hoped will serve as a basis for future practical work.

When $\kappa = 0$ (where κ is the molecular diffusivity) the magnitude of $p(\mathbf{y}, t)$ is of order Q/L^3 for most $\mathbf{y}$, but of order Q/L_0^3 when $|\mathbf{y}|$ is very small. By a variety of arguments it is shown that these facts can be explained (for many, if not all, flows) only if the distributions of C and $\overline{c^2}$, as well as that of p, have a core–bulk structure. In the bulk of the cloud C and $\overline{c^2}$ have magnitudes of order Q/L^3 and $Q^2/L_0^3 L^3$ respectively, but there is a core region of thickness decreasing to zero surrounding the centre of mass within which they have much greater magnitudes. In one case, examined in some detail, the magnitudes in the core are of order Q/L_0^3 and Q^2/L_0^6.

It is then shown that the core and bulk exist even in the real case when $\kappa \neq 0$. In the real case the core thickness no longer tends to zero but to a constant of order λ_c, the conduction cut-off length. As a consequence almost entirely of molecular diffusion acting in the core region, the magnitudes of C and $\overline{c^2}$ in both the core and the bulk decay to zero in a way which depends on the details of the fine-scale structure of the velocity field. Several examples of the decay are discussed.

1. Introduction

The spread of contaminant in turbulent flows is a major practical problem. It would therefore be extremely useful to be able to predict how the statistical properties

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of Γ, the distribution of concentration of contaminant in one realization, will vary in space and with time for a given release situation in a specific flow. Conceptually, the simplest such problem is the dispersion of a cloud of passive contaminant (arising from instantaneous release) in incompressible flow, and knowledge of this could in principle be used to analyse other problems with more complicated release conditions. However theoretical analyses of this problem have usually either used unfounded empirical concepts like eddy diffusivities or have considered somewhat artificial idealizations of the real problem in which, for example, the ensemble mean of Γ is taken to be homogeneous in space. In this paper a partial theoretical description of real problems is attempted, using the basic equations without empiricism. The description will be exclusively in terms of relative diffusion†; thus the origin of co-ordinates will be at the centre of mass of the contaminant distribution throughout each realization of the dispersion process. Relative diffusion has the practical advantage over absolute diffusion of less smearing during the averaging process so that the ensemble means determined from measurements are more closely related to the real values of Γ. A further advantage at high enough Reynolds number in real flows is that all of the eddies contributing significantly to relative diffusion lie within the inertial subrange. These eddies have a structure of universal form in contrast to the energy-containing eddies which largely dominate absolute diffusion (through meandering) but have a structure determined by the details of the flow geometry.

Suppose that at $t = 0$ a quantity Q of passive contaminant is released in an incompressible fluid, and that its initial spatial distribution is the same for each realization of the dispersion. Since the total quantity of contaminant is finite the statistical properties of Γ are inhomogeneous unlike the situations considered, e.g., by Batchelor (1959) and Kraichnan (1974). As will be seen, the magnitudes of the practically important properties of Γ considered in the present paper can be estimated only by allowing for this inhomogeneity. The equation governing $\Gamma(\mathbf{x}, t)$ (where, as explained above, $\mathbf{x}$ is measured relative to the moving centre of mass of the cloud) is

$$\partial\Gamma/\partial t + \nabla . (\boldsymbol{\Upsilon}\Gamma) = \kappa\nabla^2\Gamma, \tag{1.1}$$

where κ is the molecular diffusivity, and $\boldsymbol{\Upsilon} = \boldsymbol{\Upsilon}(\mathbf{x}, t)$ is the fluid velocity relative to the velocity of the centre of mass. By incompressibility,

$$\nabla . \boldsymbol{\Upsilon} = 0. \tag{1.2}$$

Both Γ and $\boldsymbol{\Upsilon}$ are random functions of space and time; in the usual way each can be written as the sum of its ensemble mean (denoted throughout this paper by an overbar) and fluctuation, viz.

$$\left.\begin{aligned} \Gamma &= C + c, \quad \text{where} \quad C = \overline{\Gamma}, \quad \bar{c} = 0; \\ \boldsymbol{\Upsilon} &= \mathbf{U} + \mathbf{u}, \quad \text{where} \quad \mathbf{U} = \overline{\boldsymbol{\Upsilon}}, \quad \bar{\mathbf{u}} = 0. \end{aligned}\right\} \tag{1.3}$$

The simplest, and practically most important, statistical properties of Γ are C and $\overline{c^2}$; thus the main theme of this paper is the way in which they vary in space and with time. One other property of Γ will be important in the discussion; this is $p(\mathbf{y}, t)$ where

$$p(\mathbf{y}, t) = Q^{-2} \int \overline{\Gamma(\mathbf{x})\,\Gamma(\mathbf{x}+\mathbf{y})}\, dV(\mathbf{x}), \tag{1.4}$$

† Although, with suitable amendments to definitions, some of the analysis in the paper applies also to absolute diffusion.

and is a modification of Richardson's distance-neighbour function (Richardson 1926). By mass conservation it follows immediately from (1.4) that

$$\int p(\mathbf{y}, t)\, dV(\mathbf{y}) = 1, \tag{1.5}$$

where, as throughout this paper, the integral is over all space occupied by the fluid.

The outstanding feature of a cloud is that (on the average) it spreads. A single measure of this spread is $L(t)$, where

$$L^2(t) = Q^{-1} \int |\mathbf{x}|^2 C(\mathbf{x}, t)\, dV(\mathbf{x}), \tag{1.6}$$

and is proportional to the trace of the relative cloud dispersion tensor (Batchelor 1952*a*). As the result of much work, summarized in § 24 of Monin & Yaglom (1975), it is known that, over a wide range of length scales encountered in nature, $L^2(t)$ satisfies approximately the law first partly enunciated by Richardson (1926), viz.

$$dL^2/dt = b\epsilon^{\frac{1}{3}} L^{\frac{4}{3}} \Rightarrow L^2 \approx (\tfrac{1}{3}b)^3 \epsilon t^3, \tag{1.7}$$

where b is a universal dimensionless constant and ϵ is the rate of dissipation of mechanical energy per unit mass. As a consequence of the spreading and of mass conservation, the magnitude of C decreases. If the spreading takes place at a uniform rate over the whole cloud, it follows further that

$$C = O(Q/L^3). \tag{1.8}$$

Observations by Townsend (1951) of a hot spot in the approximately isotropic and homogeneous turbulence generated by a grid were consistent with (1.8), and showed that the distribution of C was approximately self-similar over the spot. Also observations of diffusing plumes (summarized on p. 578 of Monin & Yaglom 1975) tend to support the two-dimensional analogue of (1.8).

The distribution of C is not sufficient to give many of the statistical properties of Γ that are important in practice. With toxic contaminants one is interested, for example, in the probability that, at a given place and time, a particular concentration is exceeded. In order to determine such probabilities one needs, at the very least, the distribution with $\mathbf{x}$ and t of $\overline{c^2}$, which, by (1.3), is the variance of Γ. There are no reported observations of $\overline{c^2}$ in clouds but experimental evidence from steady plumes (summarized on pp. 236–242 of Csanady 1973) suggests that the distribution of $\overline{c^2}$ is self-similar, and that $\overline{c^2}/C^2$ has a value at the centre which varies significantly from experiment to experiment (but is typically somewhat less than 0·5) and then increases outwards (reaching values of order 10 at distances from the centre of order L).

As with $\overline{c^2}$, there appear to be no observations of $p(\mathbf{y}, t)$ for clouds [where p is defined in (1.4)], but experiments on dye plumes in Lake Huron by Sullivan (1971) show that the appropriate analogue of p is, except near the centre, approximately self-similar (and indeed Gaussian). Assuming that these observations generalize to clouds, it follows from (1.5) that, except near $\mathbf{y} = 0$,

$$p(\mathbf{y}, t) = O(1/L^3). \tag{1.9}$$

It is important to realize that in all the observations discussed above the degree of spatial resolution of the distributions of Γ was limited.

Although it is known that structure on a scale of the same order of smallness as $(\nu\kappa^2/\epsilon)^{\frac{1}{4}}$, where ν is the kinematic viscosity, is profoundly influenced by κ (Batchelor 1959; Gibson & Schwarz 1963), it is not known how κ influences the overall characteristics of a finite cloud of contaminant. Important such characteristics are the magnitudes and distributions of the statistical properties defined above. The aim of this paper is to investigate such questions without invoking unjustified concepts like eddy diffusivities. In §§ 2 and 3 the investigation is for a cloud of marked fluid particles for the reason that it is important first to try to understand the simpler (though admittedly unrealistic) problem arising when there are no effects of κ. This is the traditional approach to such problems (see for example, p. 345 of Batchelor 1952*a*). The technique uses certain conserved quantities associated with Γ, and has some points in common with one described by Lumley (1964). The effects of κ on these results are investigated in § 4, and it is shown that these are profound.

2. The consequences of some conservation relations for a cloud of marked fluid particles

For a cloud of marked fluid particles there are no effects of molecular diffusivity. Hence the quantity of contaminant within each fluid particle remains constant during each realization so that, if f is any function,

$$\frac{D}{Dt}f(\Gamma) = 0. \tag{2.1}$$

Of course this also follows immediately from (1.1) with $\kappa = 0$. On integrating (2.1) over the flow field, there follows

$$\frac{d}{dt}\int f(\Gamma)\,dV = 0. \tag{2.2}$$

With $f(\Gamma) = \Gamma$, (2.2) gives the obvious result from mass conservation, viz.

$$\int \Gamma\,dV = Q, \tag{2.3}$$

and on taking the ensemble mean, using (1.3), there results that, for all time in each realization,

$$\int C\,dV = Q, \quad \int c\,dV = 0. \tag{2.4}$$

Slightly less obvious, and equally important, results are obtained by taking $f(\Gamma) = \Gamma^2$ in (2.2), so that

$$\int \Gamma^2 dV = Q^2/L_0^3, \tag{2.5}$$

where L_0^3 is determined entirely by the prescribed initial distribution, and is equal to the volume of marked fluid if this initial distribution is spatially uniform. Taking the ensemble mean gives

$$\int C^2(\mathbf{x},t)\,dV + \int \overline{c^2}(\mathbf{x},t)\,dV = Q^2/L_0^3. \tag{2.6}$$

Since the initial distribution is prescribed, $c(\mathbf{x},0) = 0$, so that further consequences of (2.6) are

$$\int C^2(\mathbf{x},0)\,dV = Q^2/L_0^3, \quad \int \overline{c^2}(\mathbf{x},0)\,dV = 0. \tag{2.7}$$

However, if, at time t, C is of order Q/L^3 over most of the cloud, as given by (1.8), it also follows from (2.6) that, as $t \to \infty$,

$$\int C^2(\mathbf{x},t)\,dV \to 0, \quad \int \overline{c^2}(\mathbf{x},t)\,dV \to Q^2/L_0^3. \tag{2.8}$$

Thus, while the total amount of 'Γ^2-stuff' in a cloud of marked fluid particles is conserved and is initially all C^2-stuff, it is ultimately all converted into $\overline{c^2}$-stuff.

Assuming that $\overline{c^2}$ has a magnitude which is of the same order over the whole cloud, it is clear from the second of (2.8) that ultimately

$$\overline{c^2} = O(Q^2/L_0^3 L^3). \tag{2.9}$$

Now experiments suggest that the estimate (1.8) for the magnitude of C is observed very soon after release, following an initial period of adjustment, and there seems no reason to doubt at this stage that the same will be true for this estimate of the magnitude of $\overline{c^2}$. Of course both estimates can be correct only approximately, though with an accuracy increasing with time. There must be other terms [presumably of order less than (1.8) or (2.9) by a power of L_0/L] to account for the transition from the initial to the ultimate state.

Another simple argument confirms the estimate (2.9). Consider one realization of the dispersion. By incompressibility the volume of marked fluid remains constant at its initial value which is of order L_0^3. But the orientation and shape of this volume are not fixed since they depend on the particular velocity field during this realization. What can be said is that the constant volume of marked fluid is somewhere in a region of space surrounding the centre of mass of volume of order L^3, with the statistical properties of the position of the marked fluid within this region being determined entirely by the statistical properties of the velocity field. Thus Γ has a value of order Q/L_0^3 within a volume of order L_0^3, but is elsewhere zero. Now the probability that any particular point within the region of volume of order L^3 lies in marked fluid is, for most points, of order L_0^3/L^3 so that the ensemble mean of any function $f(\Gamma)$ is of order $f(Q/L_0^3) \times L_0^3/L^3$. In particular C, the ensemble mean of Γ, is of order Q/L^3, consistent with (1.8), while $\overline{\Gamma^2}$ is of order $Q^2/L_0^3 L^3$. Since $\overline{\Gamma^2} = C^2 + \overline{c^2}$, and since $L \gg L_0$, the estimate (2.9) is recovered.

There are two immediate important consequences of the estimates of the magnitudes of C and $\overline{c^2}$. First, it follows from (2.9) that theoretical discussions of concentration fluctuations are valueless unless they take account of the finite initial size of the cloud†; this was not done by Csanady (1967). Second, $\overline{c^2}/C^2$ is proportional to $(L/L_0)^3$ and will vary significantly from one set of experiments to another unless L/L_0 is the same. Although this argument ignores the possible effects of κ, it gives one cause for the observed variation in $\overline{c^2}/C^2$ reported in § 1.

The above probability argument can also be applied to the function $p(\mathbf{y},t)$. For, in each realization,

$$\int \Gamma(\mathbf{x})\,\Gamma(\mathbf{x}+\mathbf{y})\,dV(\mathbf{x})$$

is of order $(Q^2/L_0^6)\,L_0^3 = Q^2/L_0^3$, so that its ensemble mean is, at least for most points, of order $(Q^2/L_0^3)\,(L_0^3/L^3) = Q^2/L^3$. Thus, using the definition of p in (1.4), the result (1.9) is obtained.

† In particular there is no point in considering an initial point source.

The experimental evidence reported in § 1 suggests that the distributions of C, $\overline{c^2}$ and p are approximately self-similar, at least to the degree of spatial resolution attained. In a case when the governing scales of turbulence are isotropic, which frequently occurs with relative diffusion in natural flows because of the wide range of length scales lying in the inertial subrange, it follows that, if (1.8), (2.9) and (1.9) hold everywhere,

$$C = \frac{Q}{L^3} F\left(\frac{|\mathbf{x}|}{L}\right); \quad \overline{c^2} = \frac{Q^2}{L_0^3 L^3} J\left(\frac{|\mathbf{x}|}{L}\right); \quad p = \frac{1}{L^3} H\left(\frac{|\mathbf{y}|}{L}\right). \tag{2.10}$$

3. The existence of a core for a cloud of marked fluid particles

The new estimate (2.9) for the magnitude of $\overline{c^2}$, and the equations (2.10) for the distributions of C, $\overline{c^2}$ and p (when these are self-similar and the turbulence is isotropic) were derived with certain reservations. The most notable of these are that inferences were drawn from observations in which the finest scale structure could not be resolved, and the possibility, mentioned in the probability arguments in § 2, that there are some points whose probability of lying in marked fluid is not of order L_0^3/L^3. In this section, where κ is still assumed to have no effect, the importance of these reservations is investigated.

When $\kappa = 0$, the function $p(\mathbf{y}, t)$ has a simple interpretation for the case when the initial distribution of contaminant is spatially uniform† (Richardson 1926; Batchelor 1952*a*). Then $L_0^6 p(\mathbf{y}, t)$ is the ensemble average volume of marked fluid within which the point $\mathbf{x}$ can lie so that the point $\mathbf{x} + \mathbf{y}$ also lies in marked fluid. It is then immediate, as Batchelor (1952*a*) pointed out, that, for all t,

$$p(0, t) = L_0^{-3} \quad \text{when} \quad \kappa = 0. \tag{3.1}$$

Notice that (3.1) also follows when (2.6) is used in (1.4), since $\overline{\Gamma^2}(\mathbf{x}) = C^2(\mathbf{x}) + \overline{c^2}(\mathbf{x})$. This gives a simple practical interpretation to the value of $p(0, t)$, as requested on p. 90 of Csanady (1973). This is that it is constant because the quantity of contaminant within each fluid particle remains equal to its initial value.

One striking and surprising consequence of (3.1) is that there must be a core region surrounding $\mathbf{y} = 0$ in which the magnitude of p changes from its constant value at $\mathbf{y} = 0$ given by (3.1) to decreasing bulk values of order L^{-3} given by (1.9). The normalization condition (1.5) presumably ensures that the thickness of this core region decreases with time. It also follows from (3.1) that the distribution of p can never be self-similar.

The result (3.1) is not consistent with the estimate (1.9). (It was however noted in § 1 that the observations by Sullivan (1971) from which (1.9) was inferred did not support it near $\mathbf{y} = 0$.) Therefore the probability argument used in § 2 to derive (1.9) must fail near $\mathbf{y} = 0$. Since, in relative diffusion, the origin is a special point in each realization, i.e. the centre of mass of the distribution of contaminant‡, the obvious reason for breakdown is that the probability of a point sufficiently near the origin lying in marked fluid is not of order L_0^3/L^3 as assumed, but, to be consistent with (3.1), of order 1. It will be shown in this section that this proposition is correct in many

† Although (3.1), and the subsequent argument, remain valid whatever the initial distribution.

‡ The centre of mass can be defined only if there is a finite quantity of contaminant, i.e. only if the statistical properties of Γ are inhomogeneous.

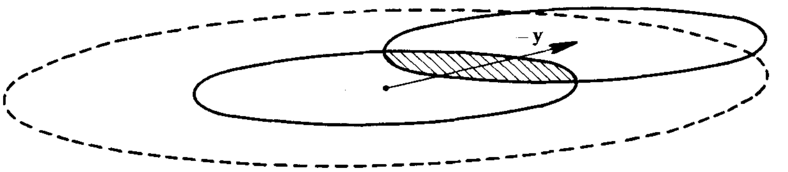

FIGURE 1. Sketch illustrating the value of $q(\mathbf{y}, t)$ defined in (3.6). The shaded region is that common to the two congruent ellipsoids, and is equal to $L_0^6\, q(\mathbf{y}, t)$. When $-\mathbf{y}$ lies outside the large ellipsoid, q is zero.

flows and has important consequences, particularly when effects of κ are considered (as they will be in § 4).

An exact solution

Although (3.1) holds in all flows, so that the distribution of p always has a core and can never be self-similar, it is valuable to examine its detailed structure for a class of exact solutions, especially for the insight this gives on the conjectured breakdown of the probability argument (used for the magnitudes of C and $\overline{c^2}$, as well as p) for points near the origin. This class of exact solutions is such that axes can be chosen for each realization so that, throughout that realization,

$$\boldsymbol{\Upsilon} = (\alpha_1 x_1, \alpha_2 x_2, \alpha_3 x_3), \tag{3.2}$$

where α_1, α_2, α_3 are the same for each realization of the ensemble, and will be taken to be constant in time. The sole random feature is thus that the directions of the axes with respect to which (3.2) holds differ from one realization to another; hence ensemble means are obtained by averaging over all possible directions, assumed here to be distributed uniformly in space so that $\boldsymbol{\Upsilon}$ has statistical isotropy. This class of exact solutions has been widely used with results that are often applicable more widely than anticipated (e.g. Townsend 1951; Batchelor 1959; Saffman 1963).

By incompressibility

$$\alpha_1 + \alpha_2 + \alpha_3 = 0. \tag{3.3}$$

Without loss of generality take

$$\alpha_1 \leqslant \alpha_2 \leqslant \alpha_3, \quad \alpha_1 < 0, \quad \alpha_3 > 0. \tag{3.4}$$

Then an initially spherical volume of marked fluid of radius a becomes after time t an ellipsoid of the same volume with semi-axes $a_i = a \exp(\alpha_i t)$ $(i = 1, 2, 3)$, where, by (3.4), for moderate and large t,

$$a_1 \ll a \ll a_3. \tag{3.5}$$

For simplicity consider the case when the initial marking is uniform (although the general conclusions below remain valid for all initial conditions except those in which for example the marked fluid forms a torus). Then it is easy to see that, for all time,

$$L_0^6 Q^{-2} \int \Gamma(\mathbf{x})\, \Gamma(\mathbf{x}+\mathbf{y})\, dV(\mathbf{x}) = L_0^6\, q(\mathbf{y}, t), \tag{3.6}$$

say, is the volume common to two ellipsoids; one is that in which the marked fluid lies, and the other has axes equal and parallel to those of the first, but its centre at the point with position vector $-\mathbf{y}$ relative to the centre of the first (see figure 1). As is obvious from figure 1, $q(\mathbf{y}, t)$ is zero unless

$$0 \leqslant d \leqslant 1 \quad \text{where} \quad \frac{y_1^2}{a_1^2} + \frac{y_2^2}{a_2^2} + \frac{y_3^2}{a_3^2} = 4d^2, \tag{3.7}$$

and an easy calculation, given in the appendix, shows that then

$$q(\mathbf{y},t) = \tfrac{1}{2}L_0^{-3}(2-3d+d^3), \tag{3.8}$$

where, by definition, $L_0^3 = \frac{4}{3}\pi a^3$. Now the ensemble mean of $q(\mathbf{y},t)$, obtained by averaging over all the possible and equiprobable orientations of the ellipsoids in space while keeping $\mathbf{y}$ unchanged, is $p(\mathbf{y},t)$, using (1.4) and (3.6). Obviously $p(\mathbf{y},t)$ is independent of the direction of $\mathbf{y}$, and, from (3.7), is non-zero provided $|\mathbf{y}| \leqslant 2a_3$; it is also clear that $L(t)$, the spread of the cloud, is of order $a_3 = a\exp(\alpha_3 t)$. It follows from (3.7) that the value of $q(\mathbf{y},t)$ is non-zero for all the ellipsoids considered in the averaging provided $|\mathbf{y}| \leqslant 2a_1$. Thus, using (3.8), $p(\mathbf{y},t)$ is of order L_0^{-3} for such $\mathbf{y}$ [with $p(0,t)$ equal to L_0^{-3}, in agreement with (3.1)]. However, if $|\mathbf{y}|$ is of order L (i.e. of order a_3) it also follows from (3.7) that $q(\mathbf{y},t)$ is non-zero only for a fraction of order L_0^3/L^3 of the ellipsoids so that, again using (3.8), $p(\mathbf{y},t)$ is of order L^{-3} for such $\mathbf{y}$ in agreement with (1.9). Detailed mathematics confirming these order of magnitude results is given in the appendix.

Consider now the way in which $\overline{\Gamma}(\mathbf{x},t)$ and $\overline{\Gamma^2}(\mathbf{x},t)$ depend on $\mathbf{x}$ when the marked fluid is initially distributed, not necessarily uniformly, over a sphere. By means of arguments analogous to those just used for p it follows that $\overline{\Gamma}$ is of order Q/L_0^3 and $\overline{\Gamma^2}$ is of order Q^2/L_0^6 when $|\mathbf{x}| \leqslant a_1$†, and that $\overline{\Gamma}$ is of order Q/L^3 and $\overline{\Gamma^2}$ is of order $Q^2/L_0^3L^3$ when $|\mathbf{x}|$ is of order L. Since $\overline{\Gamma} = C$ and $\overline{\Gamma^2} = C^2+\overline{c^2}$, it follows that [always assuming (3.5)]

$$C = O(Q/L_0^3),\quad \overline{c^2} = O(Q^2/L_0^6)\quad \text{for}\quad |\mathbf{x}| \leqslant a_1, \tag{3.9}$$

and that

$$C = O(Q/L^3),\quad \overline{c^2} = O(Q^2/L_0^3L^3)\quad \text{for}\quad |\mathbf{x}| = O(L). \tag{3.10}$$

Some detailed mathematics supporting these results is also given in the appendix.

Thus, for this particular velocity field, the distributions of C and $\overline{c^2}$ (as well as that of p) have a core of thickness tending to zero as $t\to\infty$ (since $a_1 = a\exp(\alpha_1 t)$ and $\alpha_1 < 0$) within which both C and $\overline{c^2}$ have magnitudes of order equal to those given by the initial distribution. Outside this core however, the magnitudes decrease to zero as t increases, with values in the bulk of the cloud given by (3.10), which is, of course, consistent with the earlier estimates (1.8) and (2.9).

More general velocity fields

It is now apparent how the probability argument used in § 2 to justify the estimates (1.8) and (2.9) for the magnitudes of C and $\overline{c^2}$ is, in general, too superficial. In the exact solution the centre of mass, and points in a small volume of decreasing size surrounding it, lie in marked fluid with probability 1, i.e. for all realizations of the turbulence. It is only for points in the bulk of the cloud that the probability of lying in marked fluid is of the order assumed in the probability argument, viz. L_0^3/L^3.

Does a similar argument, leading to a core–bulk structure, apply in flows in which $\mathbf{\Upsilon}$ is not given by (3.2)? It seems certain that in many flows the answer to this question is yes; all that is necessary is that in the early stages of most realizations of the dispersion the initial cloud of marked fluid is deformed into an elongated shape (which,

† Note that $\overline{\Gamma} = Q/L_0^3$ and $\overline{\Gamma^2} = Q^2/L_0$ when $|\mathbf{x}| \leqslant a_1$ for the special case when the initial marking is uniform.

unlike in the exact solution, may also be distorted and fibrous at the edges) with maximum dimension of order $L(t)$, and that the centre of mass lies in marked fluid not necessarily certainly but in a fixed proportion of these realizations. In such circumstances C and $\overline{c^2}$ still have magnitudes of order Q/L_0^3 and Q^2/L_0^6 in a small volume whose linear dimension is of the order of the minimum diameter of the elongated cloud at time t, while in the bulk of the cloud C and $\overline{c^2}$ have magnitudes of order Q/L^3 and $Q^2/L_0^3L^3$. (Indeed the above conditions are too restrictive for the existence of a core–bulk structure. Even if the probability that the centre of mass lies in marked fluid is not fixed, but decreases with time less rapidly than L_0^3/L^3, cores exist in the distributions of C and $\overline{c^2}$.)

It is not possible at the moment to give a precise description of those turbulent velocity fields which satisfy the above conditions, and further investigation of this question would be valuable (though difficult) in view, especially, of the crucial role of the core on the effect of κ (to be described in §4). Granted though that the velocity field considered in the exact solution, which certainly yields a core–bulk structure, has given accurate predictions over length-scales larger than those over which it can be expected to hold (Townsend 1951; Batchelor 1959; Saffman 1963), and granted the common observation that clouds of contaminant do tend to become more elongated with time, it is natural now to propose the existence of a core–bulk structure in a wide range of cases, and to investigate the consequences of this proposal further. Obviously the details of the core structure, of the transition from the core to the bulk, and of the bulk region itself, depend on the detailed statistical structure of the velocity field.

In the argument so far in this paper no use has been made of the equations satisfied by the statistical properties of Γ, in particular of those satisfied by C and $\overline{c^2}$. Indeed one of the most important themes has been, in fact, that these statistical properties can never be observed in one realization. No core or bulk region is proposed (or indeed exists, except for very special initial distributions) for the distribution of Γ in one realization, and, when such structure exists in the distributions of the statistical properties of Γ, it does so entirely as a result of the process of taking the ensemble average. Nevertheless, some interpretation of the proposed structure in terms of hypothetical processes such as production and transfer of $\overline{c^2}$ is useful, if only to emphasize the fundamental differences between it and that often put forward, and described e.g. in Csanady (1973).

The equations governing the distributions of C and $\overline{c^2}$ are obtained from (1.1) with $\kappa = 0$, (1.2) and (1.3), and are:

$$\partial C/\partial t + \nabla \,.\, (\mathbf{U}C + \overline{\mathbf{u}c}) = 0; \tag{3.11}$$

$$\frac{\partial \overline{c^2}}{\partial t} + \nabla \,.\, (\mathbf{U}\overline{c^2} + \overline{\mathbf{u}c^2}) + 2\overline{\mathbf{u}c} \,.\, \nabla C = 0. \tag{3.12}$$

In (3.12) the divergence term has zero integral over all space and, using conventional language, represents the transfer of $\overline{c^2}$ from place to place. On the other hand the term $2\overline{\mathbf{u}c}\,.\,\nabla C$ has a non-zero integral over the flow field and is conventionally described as the production of $\overline{c^2}$ (by feeding from the distribution of C through the mechanism described by the term in (3.11) involving $\overline{\mathbf{u}c}$). To illustrate the consequences of the proposed structure on the interpretation of (3.11) and (3.12) consider a special case when $\mathbf{U} = 0$ and both the turbulence and the initial distribution of contaminant are spherically symmetric, conditions satisfied, for example, in the exact solution considered above. Suppose further that the distributions of C and $\overline{c^2}$ are self-similar in the bulk region, so that they satisfy (2.10) there. On substituting $C = (Q/L^3)\,F(R)$ in (3.11), where

$$\mathbf{R} = \mathbf{x}/L, \quad R = |\mathbf{R}|, \tag{3.13}$$

it follows that

$$\overline{\mathbf{u}c} = (Q\dot{L}/L^3)\,\mathbf{R}F. \tag{3.14}$$

Thus the production term in (3.12), viz. $2\overline{\mathbf{u}c}\,.\,\nabla C$, is of order $Q^2\dot{L}/L^7$ and is negligible compared with the term $\partial\overline{c^2}/\partial t$, which, using $\overline{c^2} = (Q^2/L_0^3\,L^3)\,J(R)$ from (2.10), is of order $Q^2\dot{L}/L_0^3\,L^4$. Since the production term is of no consequence in the bulk, the rate of change of $\overline{c^2}$ at any point there is determined entirely by transfer. It is then immediate, on substituting into (3.12), that

$$\overline{\mathbf{u}c^2} = (Q^2\dot{L}/L_0^3\,L^3)\,\mathbf{R}J, \tag{3.15}$$

so that, in the bulk, the transfer of $\overline{c^2}$ is everywhere outwards.

In contrast with what happens in the bulk region, the production of $\overline{c^2}$ is not negligible in the core region. Consider for example the case when the core region has dimension of order $L_1(t)$, and C and $\overline{c^2}$ have magnitudes of order Q/L_0^3 and Q^2/L_0^6 there – their values in the exact solution. Then, from (3.11), $\overline{\mathbf{u}c}$ is of order $Q\dot{L}_1/L_0^3$ in the core, so that all terms in (3.12) are of order $Q^2\dot{L}_1/L_0^6\,L_1$. The $\overline{c^2}$ produced in the core is transferred outwards across the cloud, at a rate given by (3.15) far enough away from the core. As a result of production in the core and the consequent transfer outwards, the core radius decreases, but at no time is the spreading taking place uniformly over the whole cloud.

As explained earlier, the detailed shapes of the distributions of C and $\overline{c^2}$ depend on the detailed structure of the velocity field. It will be shown in a later paper that in many cases it is possible to match the distributions in the core directly onto those in the bulk. To illustrate what is involved, one special case is discussed briefly. Suppose that, in the exact solution defined by (3.2), (3.3) and (3.4),

$$\alpha_1 < 0, \quad \alpha_2 = \alpha_3 > 0. \tag{3.16}$$

Then an initial sphere of marked fluid is deformed into a flat ellipsoid of revolution, and the core thickness, say $L_1(t)$, is of the order of the minimum diameter of this ellipsoid. Hence, by conservation of mass,

$$L_1 = O(L_0^3/L^2). \tag{3.17}$$

If the distributions of C and $\overline{c^2}$ are self-similar in the core, they have the forms

$$C = (Q/L_0^3) f(\rho), \quad \overline{c^2} = (Q^2/L_0^6)\, j(\rho), \tag{3.18}$$

where

$$\boldsymbol{\rho} = \mathbf{x}/L_1, \quad \rho = |\boldsymbol{\rho}|. \tag{3.19}$$

It then follows from (3.11) that

$$\overline{\mathbf{u}c} = (Q\dot{L}_1/L_0^3)\,(\boldsymbol{\rho}/\rho^3) \int_0^\rho z^3 f'(z)\,dz, \tag{3.20}$$

and thus from (3.12) that

$$\overline{\mathbf{u}c^2} = -(Q^2\dot{L}_1/L_0^6)\,\boldsymbol{\rho}h(\rho), \quad h(\rho) = \rho^{-3}\int_0^\rho \left[2f'(s)\int_0^s z^3 f'(z)dz - s^3 j'(s)\right] ds. \quad (3.21)$$

Since both $f'(s)$ and $j'(s)$ are expected to be negative, $h(\rho)$ is positive. Now it can be shown that, in this case, the two expressions (3.15) and (3.21) for $\overline{\mathbf{u}c^2}$ are mutually consistent in an overlap region satisfying $L_1 \ll |\mathbf{x}| \ll L$ provided their orders of magnitude are equal. Thus $\dot{L}_1$ is negative as expected, and further

$$\dot{L}_1/L_0^6 \propto -\dot{L}/L_0^3 L^3 \Rightarrow L_1 \propto L_0^3/L^2, \quad (3.22)$$

which agrees with (3.17), obtained by an independent argument. But it must be stressed that this is only one of many possible examples.

4. The effects of molecular diffusion

The important transfer processes represented by the terms $\nabla.\overline{(\mathbf{u}c)}$ and $\nabla.\overline{(\mathbf{u}c^2)}$ in (3.11) and (3.12) respectively are conventionally modelled using eddy diffusivities, but in the arguments above proper account has been taken of these terms, without empiricism. If the proposed core–bulk structure for a cloud of marked fluid particles does not fully describe a real cloud of passive contaminant, it follows that the differences are due entirely to the single process so far ignored, viz. molecular diffusion. As will be shown, there are such differences, and these are not just minor perturbations, but differences in fundamental structure. It then follows further that molecular diffusion is an essentially different physical process, both fundamentally and in its practical effects, from those represented by $\nabla.\overline{(\mathbf{u}c)}$ and $\nabla.\overline{(\mathbf{u}c^2)}$. Thus the concept of an eddy diffusivity must be physically incorrect and, in the end, practically inadequate.

The differences between a real cloud and a cloud of marked fluid particles

As a result of molecular diffusion, molecules of contaminant cross the boundaries of fluid particles. When this process has had a significant cumulative effect it becomes meaningless to refer to marked fluid particles and, *a fortiori*, to consider the mass of contaminant contained within one marked fluid particle. It is certain that ultimately the smoothing effect of molecular diffusion will cause the values of C and $\overline{c^2}$ in the core, and elsewhere, to decay eventually to zero. Molecular diffusion also affects the total amount of $\overline{\Gamma^2}$-stuff, for, on multiplying (1.1) by $\frac{1}{2}\Gamma$, integrating over all space, and taking the ensemble mean it follows that

$$\frac{d}{dt}\int \overline{\Gamma^2}dV = \frac{d}{dt}\int C^2 dV + \frac{d}{dt}\int \overline{c^2}dV = -2\kappa \int \overline{(\nabla\Gamma)^2}dV. \quad (4.1)$$

The term on the right-hand side is always negative so that, inevitably,

$$\int \overline{\Gamma^2}dV \to 0 \quad \text{as} \quad t\to\infty. \quad (4.2)$$

Since, from (2.8),

$$\int C^2 dV \to 0 \quad \text{as} \quad t\to\infty,$$

for a cloud of marked fluid particles, and therefore for a cloud of contaminant molecules, it follows from (4.1) and (4.2) that

$$\int \overline{c^2}\, dV \to 0 \quad \text{as} \quad t \to \infty, \tag{4.3}$$

in contrast with the result in (2.8) for a cloud of marked fluid particles.

Obviously the results just derived show that the core–bulk structure in a cloud of marked fluid particles is ultimately, at least in the form presented above, an inadequate description of the structure of a real cloud. The remainder of this paper aims to investigate the physics leading to these results and to present a modified structure consistent with them.

As a result of advection the minimum thickness of all parts of a cloud of marked fluid particles shrinks to zero as $t \to \infty$, resulting in a continual increase in the gradients of Γ across the thinnest part of the cloud and thus, in a real cloud, in a continual increase in the effect of molecular diffusion tending to widen the distance over which the contaminant is spread. As Batchelor (1952*b*) points out, these two competing effects are eventually in balance when the minimum thickness of all parts of the cloud is of order λ_c, where

$$\lambda_c = (\nu\kappa^2/\epsilon)^{\frac{1}{4}}, \tag{4.4}$$

is the so-called conduction cut-off length. Subsequently the minimum thickness of the cloud remains constant but Γ gradually decays to zero as a result of the diffusion of molecules down its gradient.

On the other hand the largest dimension of a cloud of marked fluid particles, which is of order $L(t)$ defined in (1.6), grows at an increasing rate as $t \to \infty$, resulting in a continual decrease in the gradients of Γ along the parts of the cloud which are being stretched and thus in the flux of contaminant molecules in this direction. Hence there is little effect of molecular diffusion on the rate of increase of L.

An analysis of some exact solutions

Consider once again the class of flows for which an exact solution is available when $\kappa = 0$, i.e. those flows for which axes can be chosen in each realization so that the velocity field is given by (3.2). As shown by the authors referred to earlier, the equation governing Γ, viz. (1.1), can be solved exactly even when $\kappa \neq 0$. The details given in the appendix show that when the initial distribution of contaminant has spherical symmetry, then it remains true when $\kappa \neq 0$ that surfaces of constant Γ are ellipsoids. But these ellipsoids differ from the equivalent ones when $\kappa = 0$ in the following ways. When $\kappa = 0$, the ellipsoid containing a constant fraction f of the contaminant has a constant volume of order fL_0^3 (and equal to fL_0^3 if the initial distribution of contaminant is uniform) but, when $\kappa \neq 0$, the ellipsoid containing the same constant fraction of the contaminant has an increasing volume. Let the semi-axes of the ellipsoid be

$$A_i(f,t) \quad (i = 1,2,3),\dagger$$

where, without loss of generality,

$$A_1 \leqslant A_2 \leqslant A_3. \tag{4.5}$$

It follows from the results in § 3 of this paper that, when $\kappa = 0$, $A_1 \to 0$ as $t \to \infty$, $A_3 = O(L) \to \infty$ as $t \to \infty$ and $A_1 A_2 A_3$ is a constant of order fL_0^3. But, when $\kappa \neq 0$, the

† When $\kappa = 0$, $A_i(1, t)$ is of course equal to a_i, defined immediately before (3.5).

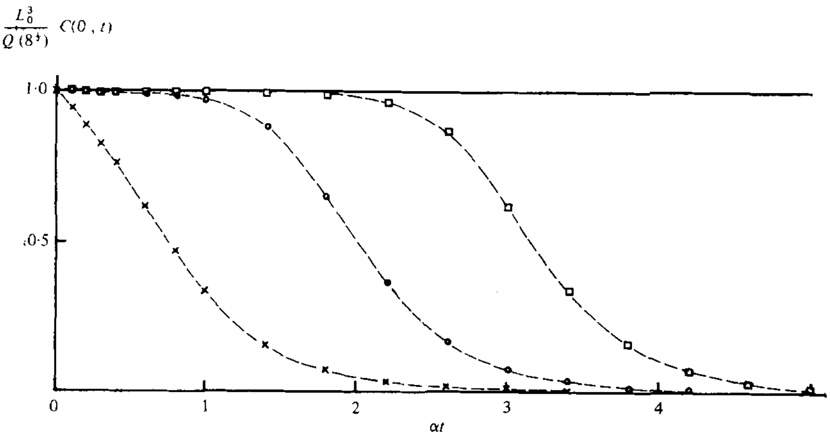

FIGURE 2. The effect of molecular diffusion on the variation of $(L_0^3/Q(8^{\frac{1}{2}}))\,C(0,t)$ according to (A 18). ——, $\kappa = 0$; ×, $2\pi\kappa/\alpha_3 L_0^2 = 10^{-1}$; ○, $2\pi\kappa/\alpha_3 L_0^2 = 10^{-3}$; □, $2\pi\kappa/\alpha_3 L_0^2 = 10^{-5}$.

mathematics in the appendix confirm the arguments earlier in this section that A_1 tends to a constant of order λ_c, A_3 is negligibly affected by κ and $A_1 A_2 A_3 \to \infty$ as $t \to \infty$.

Consider now the special case of (3.2) when

$$\alpha_1 < 0, \quad \alpha_2 = \alpha_3 > 0. \tag{4.6}$$

Then, in each realization, each of the ellipsoids containing a given constant fraction of the contaminant is a flat ellipsoid of revolution of thickness of order λ_c and of increasing radius L. Thus $A_1 A_2 A_3$ is of order $L^2\lambda_c$, and in each realization Γ is of order $Q/L^2\lambda_c$ within the ellipsoid. On taking the ensemble mean it follows, exactly as with a cloud of marked fluid particles, that, ultimately,

$$C = O(Q/L^2\lambda_c), \quad \overline{c^2} = O(Q^2/L^4\lambda_c^2) \quad \text{for} \quad |\mathbf{x}| \lesssim \lambda_c, \tag{4.7}$$

and that

$$C = O(Q/L^3), \quad \overline{c^2} = O(Q^2/L^5\lambda_c) \quad \text{for} \quad |\mathbf{x}| = O(L). \tag{4.8}$$

These are to be compared with the earlier results (3.9) and (3.10) for a cloud of marked fluid particles. There is still a core-bulk structure, but now the core has constant thickness of order λ_c and the magnitudes of C and $\overline{c^2}$ within it decay to zero, albeit at slower rates than in the bulk. In each realization the largest derivatives of Γ are across the ellipsoid and thus of order $Q/L^2\lambda_c^2$. Since $-2\kappa\int\overline{(\nabla\Gamma)^2}dV$, the right-hand side of (4.1), is of the same order as $-2\kappa\int(\nabla\Gamma)^2 dV$, it now follows from (4.1) that, since ultimately $\overline{c^2} \gg C^2$ in the bulk according to (4.8),

$$\frac{d}{dt}\int\overline{c^2}dV = -O\left\{\kappa\left(\frac{Q}{L^2\lambda_c^2}\right)^2 L^2\lambda_c\right\} = -O\left(\frac{\kappa Q^2}{L^2\lambda_c^3}\right). \tag{4.9}$$

The exact results in the appendix are consistent with (4.7)–(4.9). Figures 2 and 3 show how, according to these exact results for a particular initial distribution, $C(0,t)$ and $\int\overline{c^2}dV$ vary with t.

Another special case of (3.2) can be analysed in a similar way, viz. that when

$$\alpha_1 = \alpha_2 < 0, \quad \alpha_3 > 0. \tag{4.10}$$

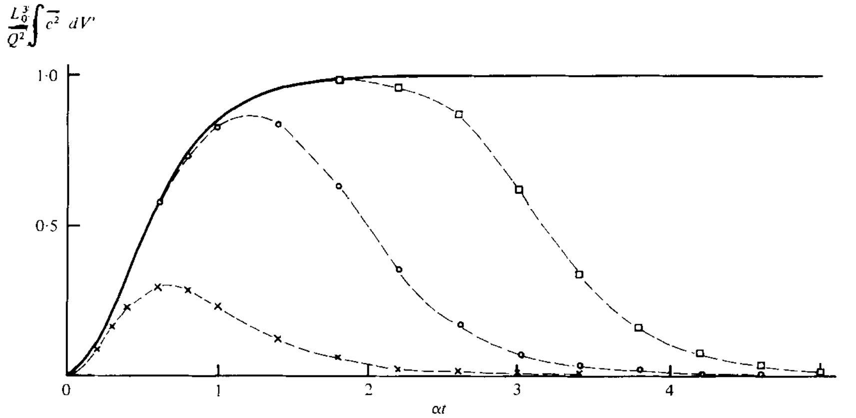

FIGURE 3. The effect of molecular diffusion on the variation of $(L_0^3/Q^2)\int\overline{c^2}dV$ according to (A 19). ——, $\kappa = 0$; ×, $2\pi\kappa/\alpha_3 L_0^2 = 10^{-1}$; ○, $2\pi\kappa/\alpha_3 L_0^2 = 10^{-3}$; □, $2\pi\kappa/\alpha_3 L_0^2 = 10^{-5}$.

In this case a constant fraction of contaminant is contained in an ellipsoid of revolution which is cigar-shaped (rather than disk-shaped when (4.5) holds). Both small semi-axes of the ellipsoid eventually have constant lengths of order λ_c with only the long axis having a length of order L. Thus $A_1A_2A_3$ is now of order $L\lambda_c^2$ so that the results corresponding to (4.7)–(4.9) are:

$$C = O(Q/L\lambda_c^2), \quad \overline{c^2} = O(Q^2/L^2\lambda_c^4) \quad \text{for} \quad |\mathbf{x}| \lesssim \lambda_c; \tag{4.11}$$

$$C = O(Q/L^3), \quad \overline{c^2} = O(Q^2/L^4\lambda_c^2) \quad \text{for} \quad |\mathbf{x}| = O(L); \tag{4.12}$$

$$\frac{d}{dt}\int \overline{c^2}dV = -O\left(\frac{\kappa Q^2}{L\lambda_c^4}\right). \tag{4.13}$$

For values of α_1, α_2, α_3 not satisfying (4.5) or (4.10), the results corresponding to (4.7)–(4.9) and (4.11)–(4.13) are in between them.

The final stages of decay of a general cloud

The linear velocity field given by (3.2) is in theory a good approximation only if L is of order no bigger than $(\nu^3/\epsilon)^{\frac{1}{4}}$, the viscous cut-off length. But Townsend (1951) showed that in one case (the decaying isotropic turbulence behind a grid, at grid Reynolds numbers ranging from 2700 to 10820) this velocity field correctly predicted the decay of $C(0,t)$ with t up to values of L greater than $15(\nu^3/\epsilon)^{\frac{1}{4}}$†. Although (3.2) does not describe the velocity field for such (relatively) large clouds, Townsend concluded that its use, nevertheless, predicted correctly the gradients of Γ (except perhaps when the cloud has become so extended that substantial parts of it are folded back on themselves, when the results described in Kraichnan (1974) will certainly be relevant). Since it is the size of these gradients that alone determine the effects of molecular diffusion it seems reasonable to propose that, for a cloud of arbitrary size,

† Note that, to account for the decay of the turbulence, Townsend prescribed α_1, α_2, α_3 in (3.2) to be inversely proportional to t. Although such variation is not explicitly considered in the present paper, results like (4.7)–(4.9) are unaffected since they are expressed in terms of L, and are therefore independent of rate-of-strain history (except insofar as this determines L).

the magnitudes of C and $\overline{c^2}$ in both the core and the bulk, and the rate at which $\int \overline{c^2} dV$ decays, are given ultimately by results between those in (4.7)–(4.9) and those in (4.11)–(4.13). Furthermore, since the significant effects of molecular diffusion occur largely, in effect, near the boundaries of the core and are then transferred outwards, it seems possible that the *shapes* of the distributions of C and $\overline{c^2}$ in the bulk are not greatly affected by molecular diffusion, irrespective of the size of the cloud. Practical formulae consistent with the above suggestions are, for example:

$$C = O(Q/L^n\lambda_c^{3-n}), \quad \overline{c^2} = O(Q^2/L^{2n}\lambda_c^{6-2n}) \quad \text{for} \quad |\mathbf{x}| \lesssim \lambda_c; \tag{4.14}$$

$$C = (Q/L^3)\, F(|\mathbf{x}|/L), \quad \overline{c^2} = (Q^2/L^{3+n}\lambda_c^{3-n})\, J(|\mathbf{x}|/L) \quad \text{for} \quad |\mathbf{x}| = O(L); \tag{4.15}$$

$$\frac{d}{dt}\int \overline{c^2} dV = -O\left(\frac{\kappa Q^2}{L^n \lambda_c^{5-n}}\right), \tag{4.16}$$

where F and J are the functions defined originally in (2.10), and n is a number between 1 and 2, which can be a slowly varying function of the variables describing the statistics of the velocity field, and those like λ_c/L_0, which describe the initial state of the cloud. It is hoped that these proposals, presented here without full justification, may be examined further, both theoretically and experimentally.

The effect of the initial distribution of contaminant on the final decay

All the discussion in this section, including that leading to (4.14)–(4.16), has been concerned with the final period of decay when the core thickness has reached its constant asymptotic value of order λ_c. But another important question is how the transition to this final period depends on the initial distribution of contaminant, particularly on L_0. Evidently L_0 influences the size of L, but not, according to (1.7), the rate at which L ultimately increases with t. But there is a more interesting possibility if L_0 is sufficiently large, which is that a real cloud may disperse as a cloud of marked fluid particles for a substantial period. This will occur when the minimum thickness of the dispersing cloud in each realization is much less than L but also much greater than λ_c. For then the arguments in this section show that the effects of molecular diffusion are still practically insignificant, while the arguments in § 3 show that there is already a core–bulk structure satisfying (3.9) and (3.10). For the special case when (3.2) and (4.6) hold, this is confirmed by figures 2 and 3 which show that $C(0,t)$ and $\int \overline{c^2} dV$ behave as for a cloud of marked fluid particles for a period of time which increases with $\alpha_3 L_0^2/\kappa$, which, by (4.4), is of order L_0^2/λ_c^2 since α_3 is of order $(\epsilon/\nu)^{\frac{1}{2}}$ (Townsend 1951). Since typical values of λ_c in the surface layers of the atmosphere and in the ocean are 10^{-3} m and 10^{-5} m respectively (based on values of ϵ quoted in Sullivan, 1971, and on p. 480 of Monin & Yaglom, 1975) it can be anticipated that, in most practical cases, the effects of molecular diffusion will take some time to become important.

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Appendix. Mathematical details for the class of exact solutions

This appendix justifies some results in the body of the paper for the case when Υ is given by (3.2).

Suppose first that there is no molecular diffusion, and that the initial distribution of contaminant is given by

$$\Gamma(\mathbf{x}, 0) = \begin{cases} 3Q/4\pi a^3 & \text{for} \quad |\mathbf{x}| \leqslant a, \\ 0 \quad \text{for} & |\mathbf{x}| > a, \end{cases} \tag{A 1}$$

where, according to the definition in (2.5),

$$a^3 = (3/4\pi) L_0^3. \tag{A 2}$$

Then, when (3.2) holds, the subsequent distribution of contaminant satisfies

$$\Gamma(\mathbf{x}, t) = \begin{cases} 3Q/4\pi a^3 & \text{for} \quad (x_1/a_1)^2 + (x_2/a_2)^2 + (x_3/a_3)^2 \leqslant 1, \\ 0 & \text{otherwise}, \end{cases} \tag{A 3}$$

where, for $i = 1, 2, 3$,

$$a_i = a \exp(\alpha_i t). \tag{A 4}$$

The function $L_0^6\, q(\mathbf{y}, t)$ defined in (3.6) is, as explained in the text and illustrated in figure 1, the volume common to the ellipsoids

$$(x_1/a_1)^2 + (x_2/a_2)^2 + (x_3/a_3)^2 = 1$$

and

$$(x_1+y_1)^2/a_1^2 + (x_2+y_2)^2/a_2^2 + (x_3+y_3)^2/a_3^2 = 1.$$

Now under the transformation of co-ordinates $x_i = a_i X_i$, $y_i = a_i Y_i$ ($i = 1, 2, 3$ and no summation) the ellipsoids become the spheres $|\mathbf{X}| = 1$, $|\mathbf{X}+\mathbf{Y}| = 1$. These spheres intersect only if $|\mathbf{Y}| \leqslant 2$, i.e. only if $0 \leqslant d \leqslant 1$, where $|\mathbf{Y}| = 2d$, consistent with (3.7). Then the common volume in $\mathbf{X}$ space is

$$2\pi \int_d^1 (1-z^2)\, dz = \tfrac{2}{3}\pi(2 - 3d + d^3),$$

so that $L_0^6\, q(\mathbf{y}, t)$, the common volume in real space, is

$$\tfrac{2}{3}\pi a_1 a_2 a_3 (2 - 3d + d^3).$$

Since $a_1 a_2 a_3 = a^3 = (3/4\pi)\, L_0^3$, this is the result in (3.8). The distance-neighbour function $p(\mathbf{y}, t)$ is the ensemble mean of $q(\mathbf{y}, t)$, obtained by averaging over all possible positions of the axes with respect to which (3.2) holds. Thus, assuming isotropic turbulence,

$$p(\mathbf{y}, t) = p(y, t) = \frac{1}{4\pi y^2} \iint q(\mathbf{y}, t)\, dS. \tag{A 5}$$

where $y = |\mathbf{y}|$ and the integral is over the sphere of radius y. The result is expressible in closed form only for special values of α_1, α_2, α_3, of which that when (using incompressibility which requires $\alpha_1 + \alpha_2 + \alpha_3 = 0$)

$$\alpha_2 = \alpha_3 > 0, \quad \alpha_1 = -2\alpha_3 < 0 \tag{A 6}$$

will be the only one considered here. It is typical in all important respects. The result of the integration is

$$L_0^3 p = \begin{cases} \left(1 - \dfrac{3NZ}{8} + \dfrac{N^3Z^3}{64} + \dfrac{3NZ^3}{128} + E^{-1}\left[\dfrac{3Z^3}{128} - \dfrac{3Z}{8}\right]\ln\left[E+N\right]\right) & \text{for} \quad Z \leqslant 2N^{-1}, \\ E^{-1}\left(\dfrac{3(4-Z^2)^{\frac{1}{2}}}{8Z} + \dfrac{3Z(4-Z^2)^{\frac{1}{2}}}{64} + \left[\dfrac{3Z^3}{128} - \dfrac{3Z}{8}\right]\ln\left[\dfrac{(4-Z^2)^{\frac{1}{2}}+2}{Z}\right]\right) & \text{for} \quad 2N^{-1} \leqslant Z \leqslant 2, \\ 0 \quad \text{for} \quad Z \geqslant 2, \end{cases} \tag{A 7}$$

where
$$Z = y/a_3, \quad N = a_3/a_1, \quad E = (N^2-1)^{\frac{1}{2}}. \tag{A 8}$$

Noting that
$$L(t) = O(a_3) \quad \text{for} \quad N \gg 1 \tag{A 9}$$

(i.e. for $\exp(\alpha_3 t) \gg 1$ from (A 6) and (A 8)), it is straightforward to verify that $p(y,t)$ satisfies all the conditions stated in the body of the paper.

When the conditions necessary for (A 7) hold (i.e. when the initial distribution is uniform over a sphere of radius a, when there is no molecular diffusion and when (A 6) holds), it is easy to show by taking the ensemble mean of (A 3) that, writing $r = |\mathbf{x}|$,

$$C(\mathbf{x},t) = (Q/L_0^3)f(r,t), \quad \overline{c^2}(\mathbf{x},t) = (Q^2/L_0^6)\{f(r,t) - f^2(r,t)\}, \tag{A 10}$$

where
$$f(r,t) = \begin{cases} 1 \quad \text{for} \quad r \leqslant a_1; \\ \dfrac{a_1}{r}\left(\dfrac{a_3^2 - r^2}{a_3^2 - a_1^2}\right)^{\frac{1}{2}} \quad \text{for} \quad a_1 \leqslant r \leqslant a_3; \\ 0 \quad \text{for} \quad r \geqslant a_3. \end{cases} \tag{A 11}$$

Thus (3.9) is satisfied immediately, while (3.10) follows almost as quickly by noting that, from (A 11), f is of order a_1/r for $r \gg a_1$, and that, by incompressibility,

$$a_1/r = a_1 a_3^2/r a_3^2 = (3/4\pi) \times (L_0^3/L^3) \times (L/r).$$

The case of a uniform initial distribution without molecular diffusion is very special since the governing equation (1.1) is satisfied trivially. Full details of the exact general solution of (1.1) for arbitrary values of α_i in (3.2), and for an arbitrary initial distribution are given in Saffman (1963). To avoid complicated algebra adding nothing to the arguments in the present paper, the only results given here are for the simple (though essentially typical) case when (A 6) holds, and when the initial distribution of contaminant satisfies, consistent with (2.6),

$$\Gamma(\mathbf{x},0) = \frac{2^{\frac{3}{2}}Q}{L_0^3}\exp\left[-\frac{2\pi r^2}{L_0^2}\right], \tag{A 12}$$

where, once more, $r = |\mathbf{x}|$. It can be verified by direct substitution in (1.1), using (3.2) and (A 6), that

$$\Gamma(\mathbf{x},t) = \frac{2^{\frac{3}{2}}Q}{ab^2}\exp\left[-2\pi\left(\frac{x_1^2}{a^2} + \frac{x_2^2+x_3^2}{b^2}\right)\right], \tag{A 13}$$

where $a(t)$ and $b(t)$ satisfy

$$\left.\begin{aligned} a^2 &= \left(L_0^2 - \frac{2\pi\kappa}{\alpha_3}\right)\exp(-4\alpha_3 t) + \frac{2\pi\kappa}{\alpha_3}, \\ b^2 &= \left(L_0^2 + \frac{4\pi\kappa}{\alpha_3}\right)\exp(2\alpha_3 t) - \frac{4\pi\kappa}{\alpha_3}. \end{aligned}\right\} \tag{A 14}$$

To determine $C(\mathbf{r}, t) = \overline{\Gamma}$ and $\overline{c^2}(\mathbf{r}, t) = \overline{\Gamma^2} - (\overline{\Gamma})^2$, the appropriate function of Γ in (A 13) has to be integrated over all possible directions of the axes. The results are:

$$C(\mathbf{r}, t) = \frac{(2\pi)^{\frac{1}{2}} Q}{ab^2}\exp\left[-\frac{2\pi r^2}{b^2}\right]\frac{\operatorname{erf}(r/d)}{(r/d)}, \tag{A 15}$$

$$\overline{c^2}(\mathbf{r}, t) = \frac{Q^2}{a^2 b^4}\exp\left[-\frac{4\pi r^2}{b^2}\right]\left\{4\pi^{\frac{1}{2}}\frac{\operatorname{erf}(2^{\frac{1}{2}} r/d)}{(2^{\frac{1}{2}} r/d)} - 2\pi\left[\frac{\operatorname{erf}(r/d)}{(r/d)}\right]^2\right\}, \tag{A 16}$$

where

$$d(t) = \frac{ab}{[2\pi(b^2 - a^2)]^{\frac{1}{2}}}. \tag{A 17}$$

From (A 15) and (A 16) are obtained

$$C(0, t) = \frac{8^{\frac{1}{2}} Q}{ab^2}; \tag{A 18}$$

$$\int \overline{c^2}\, dV = \frac{Q^2}{ab^2}\left\{1 - \frac{4ab}{(b^2 - a^2)}\arctan\left(\frac{b-a}{b+a}\right)\right\}. \tag{A 19}$$

From these expressions the graphs in figures 2 and 3 were drawn. The fundamental length $L(t)$, defined in (1.6), is found to satisfy

$$L^2(t) = \frac{1}{4\pi}(a^2 + 2b^2). \tag{A 20}$$

Consider first the case when $\kappa = 0$. Then for $\alpha_3 t \gtrsim 1$,

$$L \approx \frac{b}{(2\pi)^{\frac{1}{2}}} \approx \frac{L_0}{(2\pi)^{\frac{1}{2}}}\exp(\alpha_3 t) \gg L_0, \tag{A 21}$$

$$d \approx \frac{a}{(2\pi)^{\frac{1}{2}}} \approx \frac{L_0}{(2\pi)^{\frac{1}{2}}}\exp(-2\alpha_3 t) \approx \frac{1}{(2\pi)^{\frac{3}{2}}}\frac{L_0^3}{L^2} \ll L_0. \tag{A 22}$$

From (A 15) and (A 16) it now follows that for $\rho = r/d \lesssim 1$:

$$\left.\begin{aligned} C(\mathbf{r}, t) &\approx \frac{Q}{L_0^3}\frac{(2\pi)^{\frac{1}{2}}\operatorname{erf}\rho}{\rho}; \\ \overline{c^2}(\mathbf{r}, t) &\approx \frac{Q^2}{L_0^6}\left\{\frac{4\pi^{\frac{1}{2}}\operatorname{erf}[2^{\frac{1}{2}}\rho]}{[2^{\frac{1}{2}}\rho]} - \frac{2\pi\operatorname{erf}^2\rho}{\rho^2}\right\}. \end{aligned}\right\} \tag{A 23}$$

These are entirely consistent with (3.9) and (3.18). Note that $d(t)$ is a measure of the core thickness, and that it is of the same order as $L_1(t)$ defined in (3.17). On the other hand when $\rho = r/d \gg 1$, it follows from (A 15) and (A 16) that, writing $R = r/L$,

$$\left.\begin{aligned} C(\mathbf{r}, t) &\approx \frac{Q}{L^3}\frac{\exp(-R^2)}{2\pi R}; \\ \overline{c^2}(\mathbf{r}, t) &\approx \frac{Q^2}{L_0^3 L^3}\frac{\exp(-2R^2)}{\pi R}. \end{aligned}\right\} \tag{A 24}$$

These are entirely consistent with (3.10) and other results in the body of the paper.

Mathematically one of the keys to the above results is that, when $\kappa = 0$, $ab^2 = L_0^3$ for all t. The physical significance of this is, of course, that the volume of any definite portion of marked fluid remains constant as t changes.

But now consider what happens when $\kappa \neq 0$. Noting that α_3 is of order $(\epsilon/\nu)^{\frac{1}{2}}$, it follows from (A 14) that, as $t \to \infty$, a tends to a constant of order λ_c, the conduction cut-off length defined in (4.4). On the other hand, provided $L_0^2 \gg \lambda_c^2$, $b \approx L_0 \exp(\alpha_3 t)$ which is the value given in (A 21) when $\kappa = 0$. Thus ab^2 is of order $L^2\lambda_c$ for large t. This is consistent with the results in § 4, in particular with (4.7) and (4.8). It is to be noted also that the *shapes* of the distributions of C and $\overline{c^2}$ in the bulk (and in the core) are as in (A 24), the results when $\kappa = 0$. Once more this is consistent with the results in § 4.

For cases when the velocity field is given by (3.2) without (A 6) holding, it can be shown from the exact solution that the magnitudes of C and $\overline{c^2}$ when $\kappa = 0$ are as in (A 23) and (A 24) since these depend only on the constant volume property, not on the details of the velocity field. But, when $\kappa \neq 0$, the magnitudes are affected by the details of the velocity field in the way, and for the reasons, described in § 4. However the shapes of the distributions in both cases do depend on the values of $\alpha_1, \alpha_2, \alpha_3$.

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