J. Fluid Mech. (1978), *vol.* 89, *part* 2, *pp.* 209–222
Printed in Great Britain

Stochastic properties of scalar quantities advected by a non-buoyant plume

By EDWARD E. O'BRIEN

Department of Mechanical Engineering, State University of New York, Stony Brook

(Received 3 March 1977 and in revised form 14 November 1977)

A model probability density equation is obtained by approximating the convective and diffusive terms in a single-point density formulation of homogeneous turbulent scalar transport, with first-order reaction, in a plume. The equation, which includes the intermittency factor of the scalar field explicitly, is then shown to support similarity solutions under some constraining assumptions. Comparison of the similarity solutions with field measurements of conditioned concentrations shows that they can reproduce the general features of the data for both low intermittency and high intermittency measurement regimes. On the basis of these asymptotic results a speculative modelling of the terms representing entrainment at the plume interface is proposed and a class of similarity solutions for the intermittency factor is obtained by numerical integration.

1. Introduction

In this paper we consider a continuous source of a species Γ being advected by a turbulent plume. Γ may be undergoing a linear reaction at a rate c and the species is non-existent outside the plume. An important property of such a system, as seen by a fixed sensor, is that Γ appears only intermittently, whenever the measuring point is embedded in the meandering plume. Intermittency with respect to the scalar quantity $\Gamma(\mathbf{x}, t)$ will be defined by

$$I(x,t) = \begin{cases} 1 & \text{if} \quad \Gamma(\mathbf{x},t) > 0, \\ 0 & \text{if} \quad \Gamma(\mathbf{x},t) = 0. \end{cases}$$

The use of such a characteristic function to describe a stochastic volume has been exploited in turbulence since Corrsin (1943) introduced the concept for free turbulent shear flows. Recent theoretical studies by Libby (1975) and Dopazo (1977) contain references to the subsequent development of the idea. Experimentalists have developed the art of measuring probability densities and moments conditioned by the requirement that measurements are made only while the point is in the turbulent shear layer. Such data lend themselves more readily to physical interpretation and to numerical modelling approximations (Tutu 1976; Libby 1976).

In the case of a statistically stationary non-buoyant plume in a homogeneous turbulent environment, conditioning can be based on the existence or non-existence of the species Γ such that measurements of Γ at a point $\mathbf{x}$ are made only when $\mathbf{x}$ is in the plume and subsequent statistics are computed from these conditioned measurements. It appears to us that the use of conditioned statistics is at its least complicated in the case of the plume in uniform turbulence since the turbulent advecting field itself need not then be conditioned. This is in contrast to the turbulent jet studied by Kuznetsov & Frost (1973). Both the mean velocity field and the turbulence which advects Γ inside the jet have a distinctly different character from the same quantities outside the jet. Conditioned averages of velocity are therefore perhaps more appropriate than the unconditioned values adopted by Kuznetsov & Frost. For the plume the homogeneous turbulence statistics are assumed to be uninfluenced by the presence or absence of the scalar field. They are therefore unaltered by the process of conditioning on Γ.

Some conditioned data have been obtained in field experiments on plumes and a useful summary is contained in an article by Barry (1975). In one such experiment Barry (1971) measured concentrations of the radioactive noble gas argon-41, taking averages every 6 min over several years around an isolated source whose strength was also continuously measured. Argon-40 has zero background concentration and therefore serves as a species to which intermittency arguments can easily be applied. Data from this experiment, and several others, indicate that the single-point, conditioned, probability density of the concentration is an exponential function.

More precisely, it was found that

$$F(\Gamma) = F(0)\exp\{-b\Gamma\},$$

where $F(\Gamma)$ is the proportion of the measurements for which the concentration is greater than Γ and b is a function of the measuring location.

The contribution of these conditioned measurements to the probability density is given, by definition, by

$$-\partial F(\Gamma)/\partial\Gamma = bF(0)\exp\{-b\Gamma\}.$$

Measurements taken outside the plume always exhibit the value $\Gamma = 0$ and hence the complete probability density $f_\Gamma(\Gamma)$ for Γ is of the form

$$f_\Gamma(\Gamma) = a\delta(\Gamma)+bF(0)\exp\{-b\Gamma\},$$

where a represents the proportion of the measurements for which $\Gamma = 0$. We assume that the number of locations inside the plume for which Γ is zero form a set of measure zero and that a can be interpreted as the fraction of the measurements for which the measuring point is outside the plume.

The coefficients a, b and $F(0)$ depend on the spatial location of the measurement and can be related to more common statistical quantities as follows:

$$\overline{\Gamma} = \int_0^\infty \Gamma f_\Gamma(\Gamma)\,d\Gamma = \frac{F(0)}{b}$$

and, since

$$\int_0^\infty f_\Gamma(\Gamma)\,d\Gamma = 1,$$

$$a+F(0) = 1.$$

In this case one can interpret $F(0)$ as the proportion of the measurements for which $\Gamma > 0$.

Traditional turbulence usage has assigned $\gamma = \overline{I(\mathbf{x},t)}$, the intermittency factor, to denote this quantity. Thus $b = \gamma/\overline{\Gamma}$, $a = 1-\gamma$ and

$$f_\Gamma(\Gamma) = (1-\gamma)\,\delta(\Gamma)+\frac{\gamma^2}{\overline{\Gamma}}\exp\left\{-\frac{\gamma\Gamma}{\overline{\Gamma}}\right\}.$$

In the theoretical development to follow it will be convenient to use the conditioned average $\langle\Gamma\rangle$ rather than the unconditioned mean $\bar{\Gamma}$. From the definition of γ

$$\bar{\Gamma} = \gamma\langle\Gamma\rangle$$

and
$$f_\Gamma(\Gamma) = (1-\gamma)\,\delta(\Gamma)+\frac{\gamma}{\langle\Gamma\rangle}\exp\left\{-\frac{\Gamma}{\langle\Gamma\rangle}\right\}. \tag{1}$$

Equation (1) is one form of an often used, more general relationship between the conditioned and unconditioned probability density function for an intermittent scalar field, namely
$$f_\Gamma(\Gamma) = (1-\gamma)\,\delta(\Gamma)+\gamma f_{\Gamma^*}(\Gamma), \tag{2}$$
where f_{Γ^*} refers to the probability density of the conditioned variable and may be expected to be a continuous function of Γ. Form (2) applies in general for intermittent scalar fields as can be seen from the following analysis.

The probability density for Γ can be obtained by considering the random variables

$$\Gamma(\mathbf{x},t) = I(\mathbf{x},t)\,\Gamma^*(\mathbf{x},t),$$

where $\Gamma^*(\mathbf{x},t)$ is an artificial random variable whose statistics are those of $\Gamma(\mathbf{x},t)$ when $\mathbf{x}$ is in the plume and are unchanged when $\mathbf{x}$ is outside the plume. The statistics of Γ^* are therefore those of the conditioned concentration while the statistics of Γ refer to the unconditioned measurements.

From the theorem on the probability density of the product of two random variables we have
$$f_\Gamma(\Gamma) = \int_{-\infty}^{+\infty}\frac{1}{|w|}f_{I,\Gamma^*}\left(w,\frac{\Gamma}{w}\right)dw \quad \text{(Papoulis 1965, p. 78).}$$

Since the form of the joint probability density $f_{I,\Gamma^*}(I,\Gamma^*)$ must be

$$f_{I,\Gamma^*}(I,\Gamma^*) = A(I,\Gamma^*)\,\delta(I)+B(I,\Gamma^*)\,\delta(1-I),$$

integration over I yields $\quad f_{\Gamma^*}(\Gamma^*) = A(\Gamma^*,0)+B(\Gamma^*,1)$

and further integration over Γ^* gives

$$1 = \int A(\Gamma^*,0)\,d\Gamma^*+\int B(\Gamma^*,1)\,d\Gamma^*.$$

The intermittency function γ can be defined by

$$B(\Gamma^*,1) = \gamma f_{\Gamma^*}(\Gamma^*), \quad \text{since} \quad \int_0^\infty B(\Gamma^*,1)\,d\Gamma^* = \bar{I} = \gamma,$$

and hence $A(\Gamma^*,0) = (1-\gamma)f_{\Gamma^*}(\Gamma^*)$. Thus

$$f_\Gamma(\Gamma) = \int_{-\infty}^{+\infty}\frac{1}{w}A\left(w,\frac{\Gamma}{w}\right)\delta\left(\frac{\Gamma}{w}\right)dw+\int_{-\infty}^{+\infty}\frac{1}{|w|}B\left(w,\frac{\Gamma}{w}\right)\delta\left(1-\frac{\Gamma}{w}\right)dw.$$

On changing variables from w to $y = \Gamma/w$ and carrying out the integrations, we find

$$f_\Gamma(\Gamma) = \lim_{y\to 0}\{y^{-1}A(\Gamma/y,0)\}+B(\Gamma,1),$$

or
$${}_\Gamma(\Gamma) = (1-\gamma)\lim_{y\to 0}\{y^{-1}f_{\Gamma^*}(\Gamma/y)\}+\gamma f_{\Gamma^*}(\Gamma).$$

If $\Gamma \neq 0$ the first term is zero provided that $f_{\Gamma^*}(\Gamma^*)$ approaches zero more rapidly than $1/\Gamma^*$ as $\Gamma^* \rightarrow \infty$. To evaluate the case when $\Gamma = 0$ we note first that for arbitrary fixed y

$$\int_{-\infty}^{+\infty} f_{\Gamma^*}\left(\frac{\Gamma}{y}\right) d\left(\frac{\Gamma}{y}\right) = \int_{-\infty}^{+\infty} \frac{1}{y} f_{\Gamma^*}\left(\frac{\Gamma}{y}\right) d\Gamma = 1.$$

Hence

$$\int_{-\infty}^{+\infty} \lim_{y \to 0} \frac{1}{y} f_{\Gamma^*}\left(\frac{\Gamma}{y}\right) d\Gamma = 1.$$

Therefore

$$\lim_{y \to 0} \left\{\frac{1}{y} f_{\Gamma^*}\left(\frac{\Gamma}{y}\right)\right\} = \delta(\Gamma)$$

and (2) holds.

2. Probability density equation for a plume

We consider an advected field $\Gamma(\mathbf{x}, t)$ satisfying a linear conservation equation

$$\partial\Gamma/\partial t + \nabla . \mathbf{u}\Gamma = D\nabla^2\Gamma - c\Gamma,$$

where $\mathbf{u}$ is an incompressible turbulent velocity field, D is the molecular diffusivity of Γ and $c\Gamma$ is the rate of decay of Γ which occurs, for example, if Γ undergoes a pseudo-first-order reaction in the plume.

For such a system the single-point fine-grained density $p(\hat{\Gamma})$ (Brissaud & Frisch 1974) has been shown to obey an equation of the form (O'Brien, Meyers & Benkovitz 1976)

$$\frac{\partial p(\hat{\Gamma})}{\partial t} + \nabla . \mathbf{u} p(\hat{\Gamma}) = c \frac{\partial}{\partial \hat{\Gamma}}[\hat{\Gamma} p(\hat{\Gamma})] - D \frac{\partial}{\partial \hat{\Gamma}} \lim_{\mathbf{x}' \to \mathbf{x}} \nabla^2_{\mathbf{x}'} \int \hat{\Gamma}' p(\hat{\Gamma}', \hat{\Gamma}) d\hat{\Gamma}', \qquad (3)$$

where $p(\hat{\Gamma}) = \delta[\Gamma(\mathbf{x}, t) - \hat{\Gamma}]$, $\mathbf{u}$ is the turbulent velocity and $p(\hat{\Gamma}', \hat{\Gamma})$ is the joint fine-grained density of the concentration at two points:

$$p(\hat{\Gamma}', \hat{\Gamma}) = \delta[\Gamma(\mathbf{x}', t) - \hat{\Gamma}'] \, \delta[\Gamma(\mathbf{x}, t) - \hat{\Gamma}].$$

The role of intermittency can be studied by the conditioning technique introduced by Dopazo (1977) and applied recently to two free turbulent shear flows (Dopazo & O'Brien 1977). One multiplies (3) by the intermittency function $I(\mathbf{x}, t)$ using the rules of calculus for discontinuous functions. We can summarize the procedure as follows.

If $S(\mathbf{x}, t) = 0$ is the equation of the interface, then it can be shown (Gel'fand & Shilov 1964, p. 209) that

$$\text{grad } I = \delta(S) \, \text{grad } S \qquad (4)$$

in the sense that for any 'good' function ϕ

$$(\text{grad } I, \phi) = (\delta(S) \, \text{grad } S, \phi).$$

Consequently one may write

$$Q\nabla I = Q^+ \boldsymbol{\eta} \delta(S), \quad \partial I/\partial t = -\mathbf{u}^s . \nabla I, \qquad (5)$$

where S has been so chosen that $|\text{grad } S| = 1$, $\boldsymbol{\eta}$ is the normal to the interface in the direction of the plume material, Q^+ is the value of any field quantity Q at the interface on the side in the positive direction of $\boldsymbol{\eta}$ and $\mathbf{u}^s . \boldsymbol{\eta}$ is the normal speed of the interface (Aris 1962, p. 79).

An equation describing the statistical state inside the plume is obtained by multiplying (3) by $I(\mathbf{x},t)$, using (4) and (5) to bring I inside the derivative operators and then taking the ensemble average. One finds

$$\frac{\partial}{\partial t}\overline{Ip(\hat{\Gamma})}+\overline{\nabla.\mathbf{u}Ip(\hat{\Gamma})} = c\overline{\frac{\partial}{\partial\hat{\Gamma}}[\hat{\Gamma}Ip(\hat{\Gamma})]}-D\frac{\partial}{\partial\hat{\Gamma}}\lim_{\mathbf{x}'\to\mathbf{x}}\nabla^2_{\mathbf{x}'}\int_0^\infty \overline{I(\mathbf{x},t)\,\hat{\Gamma}'p(\hat{\Gamma}',\hat{\Gamma})\,d\hat{\Gamma}'}+E_p, \tag{6}$$

where

$$E_p = \overline{p^+(\mathbf{u}-\mathbf{u}^s).\boldsymbol{\eta}\delta(S)}+2D\lim_{\mathbf{x}'\to\mathbf{x}}\nabla_{\mathbf{x}'}\frac{\partial}{\partial\hat{\Gamma}}\int\overline{\hat{\Gamma}'p(\hat{\Gamma}',\hat{\Gamma})\,d\hat{\Gamma}'.\nabla I}+D\frac{\partial}{\partial\hat{\Gamma}}\overline{[\hat{\Gamma}p(\hat{\Gamma})\,\nabla^2 I]}.$$

In (6), p^+ is the probability density at the interface on the plume side and $(\mathbf{u}-\mathbf{u}^s).\boldsymbol{\eta}$ is the normal component of the velocity of the interface relative to the fluid velocity, in other words the speed of entrainment of fluid into the plume (Phillips 1972).

An equation which describes the conditioned field outside the plume can be obtained in a similar manner, except that one begins by multiplying (3) by $1-I(\mathbf{x},t)$. Then one finds

$$\begin{aligned}\frac{\overline{\partial(1-I)\,p(\hat{\Gamma})}}{\partial t}+\overline{\nabla.\mathbf{u}(1-I)\,p} &= c\overline{\frac{\partial}{\partial\hat{\Gamma}}[\hat{\Gamma}(1-I)\,p(\hat{\Gamma})]}\\ &\quad -D\frac{\partial}{\partial\hat{\Gamma}}\lim_{\mathbf{x}'\to\mathbf{x}}\nabla^2_{\mathbf{x}'}\int_0^\infty\overline{[1-I(\mathbf{x}',t)]\,\hat{\Gamma}'p(\hat{\Gamma}',\hat{\Gamma})\,d\hat{\Gamma}'}-E_p.\end{aligned} \tag{7}$$

Since $1-I(\mathbf{x},t)$ is non-zero only when $\mathbf{x}$ is outside the plume, where $\Gamma(\mathbf{x})\equiv 0$, it is evident that the first two terms on the right-hand side of (7) are identically zero. These correspond respectively to reaction and molecular mixing in the ambient fluid. It is physically necessary, and obvious, that they must fail to contribute to the evolution of the scalar probability density function in the ambient fluid, which is devoid of the scalar. Indeed, in the absence of long-range effects such as radiative transfer, the only possible contribution to such a scalar density function can be at the interface, where molecular diffusion and turbulent straining combine to cause detrainment of the ambient fluid as it becomes incorporated into the plume through the plume boundaries. $-E_p$ can then be recognized as the detrainment term, which, both physically and mathematically, is necessarily equal and opposite to the entrainment term E_p in (6).

In moment formulations of intermittent turbulent shear flows modelling approximations have been proposed (Libby 1975, 1976; Tutu 1976) for entrainment terms similar to these. In this section we shall not need to inquire further into the nature of such terms. By adding (6) and (7), we can remove E_p and avoid a direct confrontation with the physics of entrainment.

When (7), with the diffusion term set to zero, is added to (6) one obtains

$$\frac{\partial f_\Gamma(\hat{\Gamma})}{\partial t}+\bar{\mathbf{u}}.\nabla f_\Gamma(\hat{\Gamma})+\nabla.\overline{\mathbf{u}'p(\hat{\Gamma})} = c\frac{\partial}{\partial\hat{\Gamma}}[\hat{\Gamma}f_\Gamma]-D\lim_{\mathbf{x}'\to\mathbf{x}}\nabla^2_{\mathbf{x}'}\frac{\partial}{\partial\hat{\Gamma}}\int\hat{\Gamma}'\gamma(\mathbf{x}')f_{\Gamma^*}(\hat{\Gamma}',\hat{\Gamma})\,d\hat{\Gamma}', \tag{8}$$

where $f_\Gamma(\hat{\Gamma}) = \overline{p(\hat{\Gamma})}$ is the unconditioned probability density function for the scalar field, $\bar{\mathbf{u}}$ is the divergence-free mean velocity and $\mathbf{u}'$ the fluctuating velocity defined by $\mathbf{u} = \bar{\mathbf{u}}+\mathbf{u}'$. We have also adopted the consistent definition

$$\overline{I(\mathbf{x}')\,p(\hat{\Gamma}',\hat{\Gamma})} = \gamma(\mathbf{x}')f_{\Gamma^*}(\hat{\Gamma}',\hat{\Gamma}),$$

where $f_{\Gamma^*}(\hat{\Gamma}', \hat{\Gamma})$ is the joint density of $\hat{\Gamma}(\mathbf{x}', t)$ and $\hat{\Gamma}(\mathbf{x}, t)$ conditioned by the requirement that $\mathbf{x}'$ be in the plume.

Equation (8) is not in a satisfactory form. Both the turbulent transport and the diffusion term require information not contained in f_Γ. An approximation for the turbulent transport term $\nabla . \overline{\mathbf{u}'p(\hat{\Gamma})}$ can be obtained if one also decomposes the fine-grained density $p(\hat{\Gamma})$ into a mean and fluctuating part:

$$p(\hat{\Gamma}) = \overline{p(\hat{\Gamma})} + p'(\hat{\Gamma}) = f_\Gamma(\hat{\Gamma}) + p'(\hat{\Gamma}).$$

Then the term $\overline{\mathbf{u}'p(\hat{\Gamma})} = \overline{\mathbf{u}'p'(\hat{\Gamma})}$ suggests a mean gradient diffusion analogy of the type

$$\overline{\mathbf{u}'p'(\hat{\Gamma})} = -\boldsymbol{\kappa} . \nabla f_\Gamma(\hat{\Gamma}), \tag{9}$$

where $\boldsymbol{\kappa}$ is an eddy-diffusivity tensor for the transport of probability density. We note, for example, that

$$\int_0^\infty \overline{\mathbf{u}'\hat{\Gamma}p'(\hat{\Gamma})}\, d\hat{\Gamma} = \overline{\mathbf{u}'\Gamma},$$

and an approximation of the form (9) gives

$$\int_0^\infty \overline{\mathbf{u}'\hat{\Gamma}p'(\hat{\Gamma})}\, d\hat{\Gamma} = -\boldsymbol{\kappa} \int_0^\infty \hat{\Gamma}\nabla f_\Gamma(\hat{\Gamma})\, d\hat{\Gamma} = -\boldsymbol{\kappa} . \nabla\overline{\Gamma}.$$

Hence (9) is, *inter alia*, equivalent to an eddy-diffusivity assumption for the turbulent flux of the species Γ, and it seems likely to be an adequate approximation for turbulent transport in those situations in which a mean gradient transport approximation for the turbulent flux has proved useful. An approximation of the form (9) was first used by Kuznetsov & Frost (1973) in their study of a turbulent jet. Most recently Meyers, O'Brien & Scott (1978) have proved that an eddy-diffusivity form for the turbulent transport of probability density is an exact result for the transport of reacting scalars in the absence of molecular diffusion when either (*a*) the turbulence is homogeneous and interest is restricted to time scales much larger than the Lagrangian time scale of the turbulence or (*b*) the velocity-field time correlation is very short compared with the concentration-field circulation time. Limit (*a*), which is an extension of Taylor's (1921) Lagrangian analysis of turbulent diffusion, is of interest here. Values of $\boldsymbol{\kappa}$ obtained from his Lagrangian velocity correlation analysis have long been used to represent plume development with some success (Csanady 1973, p. 236) and it seems to us that (9) is likely to be a reasonable approximation in the context of the idealized plume under investigation here. We shall adopt it for the remainder of this study.

To consider the molecular diffusion term in (8) we note that $\int \hat{\Gamma}' f_{\Gamma^*}(\hat{\Gamma}', \hat{\Gamma})\, d\hat{\Gamma}'$ can be written as $E\{\hat{\Gamma}'|\hat{\Gamma}\} f_{\Gamma^*}(\hat{\Gamma})$, where $E\{\hat{\Gamma}'|\hat{\Gamma}\}$ is the conditional expected value of $\Gamma(\mathbf{x}', t)$ given $\Gamma(\mathbf{x}, t)$. For non-intermittent flows the molecular diffusion term was first approximated by Dopazo (1973) using the assumption that $E\{\hat{\Gamma}'|\hat{\Gamma}\}$ was Gaussian. Similar results can be obtained by applying mean-square estimation theory to evaluate $E\{\hat{\Gamma}'|\hat{\Gamma}\}$ since $\iint \{\hat{\Gamma}' - g(\hat{\Gamma})\}^2 f_{\Gamma',\Gamma}(\hat{\Gamma}', \hat{\Gamma})\, d\hat{\Gamma}\, d\hat{\Gamma}'$ is minimized by the choice $g(\hat{\Gamma}) = E\{\hat{\Gamma}'|\hat{\Gamma}\}$ (Papoulis 1965, p. 163). When $g(\hat{\Gamma})$ is restricted to the class of linear functions of $\hat{\Gamma}$ one reproduces exactly the Dopazo approximation, for which some confirming experimental data now exist for homogeneous turbulence (Tavoularis 1976, private communication) and for jets (Adrian *et al.* 1976). Kuznetsov & Frost obtained a similar approximation for the molecular diffusion term in a probability density equation by postulating Langevin processes, with non-random rates, to describe small-scale mixing of both Γ and $\hat{\mathbf{u}}$. An advantage of the first two approximations is that they relate the molecular mixing coefficient to the turbulence structure.

In our case linear mean-square estimation theory applied to the scalar field inside the plume approximates $E\{\hat{\Gamma}'|\hat{\Gamma}\}$ by $\langle\hat{\Gamma}'\rangle+\alpha(\hat{\Gamma}-\langle\hat{\Gamma}\rangle)$, where $\langle\hat{\Gamma}\rangle$ is the conditioned mean value of $\hat{\Gamma}$ and α is a coefficient defined by $\alpha = \rho(\mathbf{x}',\mathbf{x})\,\sigma_{\Gamma'}/\sigma_{\Gamma}$, where $\rho(\mathbf{x}',\mathbf{x})$ is the correlation coefficient of the processes $\hat{\Gamma}'$ and $\hat{\Gamma}$ and $\sigma_{\Gamma'}$ and σ_{Γ} are, respectively, the variances of $\hat{\Gamma}'$ and $\hat{\Gamma}$.

Since one is ultimately interested in the limit $\mathbf{x}'\to\mathbf{x}$ it is appropriate to assume isotropy of the small scales and thus to obtain for α the result

$$\alpha = \rho(\mathbf{x},r), \quad \text{where} \quad r = |\mathbf{x}'-\mathbf{x}|.$$

The last term in (8) can be written in expanded form as

$$-D\frac{\partial}{\partial\hat{\Gamma}}f(\hat{\Gamma})\lim_{\mathbf{x}'\to\mathbf{x}}[2\nabla_{\mathbf{x}'}\gamma\,.\,\nabla_{\mathbf{x}'}E_c\{\hat{\Gamma}'|\hat{\Gamma}\}+\gamma(\mathbf{x}')\nabla^2_{\mathbf{x}'}E_c\{\hat{\Gamma}'|\hat{\Gamma}\}+E_c\{\hat{\Gamma}'|\hat{\Gamma}\}\nabla^2_{\mathbf{x}'}\gamma],$$

where $E_c\{\hat{\Gamma}'|\hat{\Gamma}\}$ is the conditional expected value of $\hat{\Gamma}(\mathbf{x}')$ given $\hat{\Gamma}(\mathbf{x})$ and conditioned further by the requirement that $\mathbf{x}'$ be in the plume. Denoting the scale of the plume width by L, the scale of $\hat{\Gamma}$ by $\overline{\Gamma}_0$, its centre-line mean value, and the scale of γ also by its centre-line value, which is of order unity, we can readily estimate the magnitudes of each of the bracketed terms (after taking the limit $\mathbf{x}'\to\mathbf{x}$) as

$$\Gamma_0/L^2, \quad \Gamma_0/\lambda_s^2, \quad \Gamma_0/L^2$$

respectively, where λ_s is the scalar microscale defined by

$$\lambda_s^2(x) = -2(\partial^2\rho/\partial r^2)^{-1}_{r=0}$$

and is taken, for our scaling purposes, to have its centre-line value. Similarity arguments require that $(\lambda_s/L)^2$ be proportional to the ratio of molecular to eddy diffusivity in a plume (Csanady 1973, p. 236) and the comparison between similarity predictions and mean data supports a proportionality constant whose value is of order unity. Since $|\boldsymbol{\kappa}|$ is at least several orders of magnitude larger than D in the atmosphere we can assume that the second term in the above expression will be the dominant one in describing diffusive processes. The last term in (8) is then adequately represented, using mean-square estimation theory, by

$$-\frac{6D}{\lambda_s^2(\mathbf{x})}\gamma\frac{\partial}{\partial\hat{\Gamma}}[f(\hat{\Gamma})(\hat{\Gamma}-\langle\Gamma\rangle)].$$

This dominance by the microscale process clearly becomes stronger with increasing Reynolds number and should be quite adequate for atmospheric plumes except under very stable conditions. With these approximations for turbulent transport and diffusive mixing, the closed-form equation for the probability density function $f_\Gamma(\Gamma)$ becomes

$$\frac{\partial f_\Gamma(\Gamma)}{\partial t}+\bar{\mathbf{u}}\,.\,\nabla f_\Gamma = \nabla\,.\,\kappa\nabla f_\Gamma+c\frac{\partial}{\partial\Gamma}(\Gamma f_\Gamma)+\gamma\beta\frac{\partial}{\partial\Gamma}[(\Gamma-\langle\Gamma\rangle)f_{\Gamma^*}], \tag{10}$$

where the circumflex over Γ has been dropped, $\beta = 6D/\lambda_s^2$ and κ is the eddy diffusivity, which is taken as a scalar for simplicity although in atmospheric plumes one should at least distinguish between vertical and horizontal diffusion.

In the next section we apply decomposition (2)–(10) to obtain a pair of equations for the two unknowns γ and $f_{\Gamma^*}(\Gamma)$. We then seek conditions under which similarity solutions of the equation for $f_{\Gamma^*}(\Gamma)$ are possible. In the limit of very low intermittency, $\gamma \to 0$, the one-parameter family of solutions is shown to include as its most likely member the density $f_{\Gamma^*}(\Gamma) = \langle\Gamma\rangle^{-1}\exp\{-\Gamma/\langle\Gamma\rangle\}$, which describes the measurements of Barry discussed in the introduction.

3. Similarity solutions for the conditional density

We follow Kuznetsov & Frost by introducing the Heaviside function $H(\Gamma)$ into the conditioned density:

$$f_{\Gamma^*}(\Gamma) = H(\Gamma)\,P(\Gamma),$$

where

$$H(\Gamma) = \begin{cases} 0 & \text{if} \quad \Gamma \leqslant 0, \\ 1 & \text{if} \quad \Gamma > 0 \end{cases}$$

and $P(\Gamma)$ can be assumed to be smooth and continuous at the origin.

Since $dH(\Gamma)/d\Gamma = \delta(\Gamma)$, the density function $f_\Gamma(\Gamma)$ becomes, from (2),

$$f_\Gamma(\Gamma) = (1-\gamma)\,\delta(\Gamma) + \gamma H(\Gamma)\,P(\Gamma),$$

where the dependence of f_Γ, γ and P on $\mathbf{x}$ and t is understood but not displayed. Inserting $f_\Gamma(\Gamma)$ into (10) and equating terms containing $\delta(\Gamma)$, we have

$$\frac{\partial \gamma}{\partial t} + \overline{u}_i \frac{\partial \gamma}{\partial x_i} = \frac{\partial}{\partial x_i}\left(K\frac{\partial \gamma}{\partial x_i}\right) + \gamma\beta\langle\Gamma\rangle P(0^+), \tag{11}$$

where $P(0^+)$ means $\lim_{\Gamma\to 0^+} P(\Gamma)$. Similarly the terms containing only the smooth function $P(\Gamma)$ satisfy

$$\frac{\partial P}{\partial t} + \overline{u}_i \frac{\partial P}{\partial x_i} = \frac{\partial}{\partial x_i}\left(K\frac{\partial P}{\partial x_i}\right) + \frac{2K}{\gamma}\frac{\partial P}{\partial x_i}\frac{\partial \gamma}{\partial x_i} + \beta\frac{\partial}{\partial \Gamma}[(\Gamma - \langle\Gamma\rangle)\,P] - \beta\langle\Gamma\rangle P(0)\,P + cP + c\Gamma\frac{\partial P}{\partial \Gamma}. \tag{12}$$

With the exception of a designation for β and the addition of the reaction terms, equations (11) and (12) are identical with those proposed by Kuznetsov & Frost for the probability density concentration in a turbulent jet and their method of solution seems to be applicable. We summarize it briefly here.

An equation for the mean conditioned concentration is obtained by integrating (12) over Γ to yield

$$\frac{\partial\langle\Gamma\rangle}{\partial t} + \overline{u}_i\frac{\partial\langle\Gamma\rangle}{\partial x_i} = \frac{\partial}{\partial x_i}K\frac{\partial\langle\Gamma\rangle}{\partial x_i} + \frac{2K}{\gamma}\frac{\partial\langle\Gamma\rangle}{\partial x_i}\frac{\partial\gamma}{\partial x_i} - c\langle\Gamma\rangle - \beta\langle\Gamma\rangle^2 P(0^+). \tag{13}$$

In (11)–(13) the term containing $P(0^+;\,\mathbf{x},\,t)$ represents the rate of entrainment of ambient fluid into the plume by small-scale mixing at the plume boundaries. It is physically plausible that the rate of entrainment at a point is proportional to the frequency with which fluid of very low scalar concentration is found in the plume at that point. For a point always embedded in the plume $P(0^+)$ should be zero. For flows with rapid entrainment one expects $P(0^+)$ to be relatively large when $\mathbf{x}$ is a point for which the intermittency factor $\gamma(\mathbf{x})$ is low. One effect of such entrainment is to decrease the mean concentration in the plume, in agreement with (13).

If (12) is inserted into (7) and the result compared with (11) it follows that terms representing the rate of entrainment of probability density are given by

$$E_p = \gamma\beta\langle\Gamma\rangle P(\Gamma)\,\delta(\Gamma). \tag{14}$$

A more general expression for the entrainment rate can be obtained if one makes use of (8) without any assumption to close the molecular mixing term. This takes the form

$$E_p = D \lim_{\mathbf{x}'\to\mathbf{x}} \nabla^2_{\mathbf{x}'}\gamma(\mathbf{x}')\,E_c\{\Gamma'|\Gamma\}\,P(\Gamma)\,\delta(\Gamma). \tag{15}$$

The equivalent source term for the intermittency factor γ, obtained by integrating (15) over all values of Γ, is

$$D \lim_{\mathbf{x}'\to\mathbf{x}} \nabla^2_{\mathbf{x}'}\gamma(\mathbf{x}')\,E_c\{\Gamma'|0\}\,P(0),$$

which is a generalization of the last term on the right-hand side of (11).

High-quality conditioned measurements are yet to be made in plumes, and until they are it will be necessary to use either extensive numerical computations or unverified statistical assumptions to analyse (11) and (12). In this section we consider two simple but plausible postulates concerning the nature of the density function in the limits $\gamma\to 0$ and $\gamma\to 1$ and then examine their consequences. For points in the low intermittency region, i.e. $\gamma\to 0$, we follow a proposal made by Kuznetsov & Frost for a jet: that the conditioned mean is likely to be only a weak function of position in such regions and that the conditioned density $f_{\Gamma^*}(\Gamma)$, and therefore $P(\Gamma)$, may not depend explicitly on the spatial co-ordinates but may exhibit a universal shape when scaled with the mean conditioned concentration. That is, we seek a density of the form

$$P(\Gamma) = \langle\Gamma\rangle^{-1} g(\phi), \tag{16}$$

where $\phi = \langle\Gamma\rangle^{-1}\Gamma$.

The field experiments described in § 1 were compatible with this form with the choice, from (1), $g(\phi) = e^{-\phi}$. These results apparently hold for measurements made at a number of sites with a variety of averaging times (Barry 1975). Nevertheless, in the absence of controlled laboratory measurements it is impossible to judge how satisfactory (16) might be. Its use certainly implies a rapid mixing process in the region just inside the plume boundary for otherwise no strong similarity postulate such as (16) could possibly hold. High-quality conditioned data on the density function in an entraining shear flow have been reported by LaRue & Libby (1974). These data and as yet unpublished density measurements at low γ in a half-heated grid flow (LaRue & Libby 1977, private communication) seem to show at least a qualitative agreement with (16). There is no reason to believe that the entrainment processes are of the same nature in both these flows since the large-scale structure is probably quite different. They are likely to exhibit comparably shaped density functions only if, in both flows, the small-scale mixing process is rapid enough near the interface to dominate the distribution of the scalar field in that region.

On substituting (16) into (12), using (11) and (13), and multiplying by $\langle\Gamma\rangle$, one obtains

$$\{[g(0)-1]\phi+1\}\frac{dg}{d\phi}+\{2g(0)-1\}g = B\left\{\phi^2\frac{d^2g}{d\phi^2}+\phi\frac{dg}{d\phi}+2g\right\}, \tag{17}$$

where B is the quantity

$$B = \frac{\kappa}{\beta}\left\{\frac{\partial}{\partial x_i}\log\langle\Gamma\rangle\right\}^2 \tag{18}$$

and the linear reaction term has entirely disappeared. A nonlinear reaction contributes terms which can never satisfy this kind of similarity.

Similarity solutions of (17) are possibly only when B is a constant independent of position. When $B = 0$ the solutions for $g(\phi)$ include the results (A2) (see appendix) of Barry. The case of negligibly small B may be the only one for which the similarity assumption can apply since κ, β and $\langle\Gamma\rangle$ are all functions of the spatial co-ordinates. Therefore it is of interest to estimate the size of B from the scaling relationships introduced in § 2. From (18) and the previous assumption

$$(K/D)(\lambda_s/L)^2 = O(1),$$

we find $B = O(10^{-1})$, a value which may be small enough to permit it to be ignored.

In the appendix we also provide some details of the solutions of (17) for $B \neq 0$ and for $g(0) \neq 1$. In particular an expansion of g in powers of B about the solution $g = \exp\{-\phi\}$ is displayed in (A 3). For small enough B this representation of g can be shown to preserve the following properties:

$$\int_0^\infty g\,d\phi = 1, \quad g(\phi) \geqslant 0, \quad \lim_{\phi\to\infty} g(\phi)\to 0$$

and

$$g(\phi) \text{ continuous in } \phi \text{ for } \phi \geqslant 0.$$

The limit $\gamma\to 1$, which is pertinent for locations almost always inside the plume, is often assumed to be one in which a normal distribution can be expected, since the scalar field should then be well mixed. This assumption can be examined in the light of (12) by investigating the following similarity form as a possible solution:

$$f_\Gamma(\Gamma) = P(\Gamma) = \sigma^{-1}h(\psi), \tag{19}$$

where $\psi = (\Gamma-\overline{\Gamma})/\sigma$ and $\sigma^2 = \overline{(\Gamma-\overline{\Gamma})^2}$.

One finds quite readily that reactive species, even at first order, cannot satisfy such similarity except when their kinetic rate is significantly slower than the turbulent mixing rate. Furthermore, even for non-reacting passive species, similarity will be obtained only if the following detailed balance between advection mixing and dissipation is achieved at every spatial position: $\mathbf{u}\,.\,\nabla\sigma^2+\beta\sigma^2 = \kappa(\nabla\overline{\Gamma})^2$. This situation has been examined by Csanady (1973, p. 236) and may in fact apply approximately in a limited regime of plume development. If it does, then for the non-reactive plume one finds the solution of (19) to be the normal distribution

$$h(\psi) = (2\pi)^{-\frac{1}{2}}\exp\{-\tfrac{1}{2}\psi^2\} \quad \text{or} \quad f_\Gamma(\Gamma) = (2\pi)^{-\frac{1}{2}}\exp\{-(\Gamma-\overline{\Gamma})^2/\sigma^2\}.$$

These two limiting solutions for $\gamma\to 0$ and $\gamma\to 1$ suggests a simple analytical model for the rate of entrainment of Γ probability density. We first note that the solution $g(\phi) = \exp\{-\phi\}$ exhibits the behaviour $\langle\Gamma\rangle P(0;\mathbf{x}) = 1$ and the source term for intermittency (11) is then $\beta\gamma(\mathbf{x})$ while the rate of entrainment of Γ probability density into the plume becomes, from (14),

$$E_p = \beta\gamma(\mathbf{x})\,\delta(\Gamma).$$

These results apply only to the region of the plume in which $\gamma \ll 1$.

When $\gamma \to 1$, $P(0;\mathbf{x}) \to 0$. If one assumes a grossly simplified statistical geometry of the interface such that the density function can be scaled with just the intermittency factor γ and the conditioned mean $\langle\Gamma\rangle$, dimensional analysis and the limiting solutions above suggest the form

$$\langle\Gamma\rangle P(0,\mathbf{x}) = G(\gamma), \qquad (20)$$

where $G(0) = 1$, $G(1) = 0$ and $G(\gamma)$, which is non-negative but otherwise arbitrary, is to be determined experimentally or modelled.

Under assumption (20), (11) becomes a closed equation for the intermittency factor:

$$\partial\gamma/\partial t + \bar{\mathbf{u}}\,.\,\nabla\gamma = \kappa\nabla^2\gamma + \beta\gamma G(\gamma). \qquad (21)$$

For a statistically stationary axisymmetric plume in a uniform fluid of constant mean velocity $\bar{u}_i = U\delta_{i1}$ and under the usual 'slender' plume approximation, which ignores axial diffusion, the equation for the intermittency factor becomes

$$U\frac{\partial\gamma}{\partial x} = \frac{K}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\gamma}{\partial r}\right) + \beta\gamma G(\gamma).$$

When the reaction rate is zero or negligible compared with the turbulent mixing rate, similarity solutions for γ can be investigated by defining $\xi = r/\sigma(x)$, where $\sigma(x)$ is the standard deviation of relative dispersion, a quantity commonly used in meteorology and one that is related to K (Csanady 1973, p. 236) by $K = \frac{1}{2}U d\sigma^2/dx$. We find

$$\frac{d^2\gamma}{d\xi^2} + \left(\frac{U\sigma}{K}\frac{d\sigma}{dx}\right)\left(\xi + \frac{1}{\xi}\right)\frac{d\gamma}{d\xi} + \frac{\beta\sigma^2\gamma}{K}G(\gamma) = 0.$$

Since the coefficient $(U\sigma/K)\,(d\sigma/dx)$ is unity, similarity will be possible only if $\beta\sigma^2/K$ is independent of x. This parameter is the same as that which Csanady (1973, p. 236) obtained as a condition for self-similarity of the scalar fluctuation intensity in a plume, and his evaluation of the degree with which experimental data supports self-similarity can be carried over directly to our argument here. In such a case if we let $\beta\sigma^2/K$ be α, a constant, the equation to be solved is

$$\gamma'' + (\xi + \xi^{-1})\gamma' + \alpha\gamma G(\gamma) = 0, \qquad (22)$$

where $\gamma'(0) = 0$ and $\gamma(\infty) = 0$.

One form of G which satisfies both boundary conditions on (22) and introduces no arbitrary parameters is $G = 1-\gamma$. Both Libby (1975) and Tutu (1976) have proposed modelling momentum entrainment in a shear flow by a term proportional to $\gamma(1-\gamma)$.

Csanady proposed for α a value near 3·0. In figure 1 we display numerical solutions of (22) for $G = 1-\gamma$ and $\alpha = 3$. All solutions for which $\gamma'(0) = 0$ and $0 < \gamma(0) < 1$ automatically satisfy the boundary condition $\gamma(\infty) \to 0$. When $\gamma(0) < 0{\cdot}625$ such solutions have negative regimes and must be discarded. They have already violated

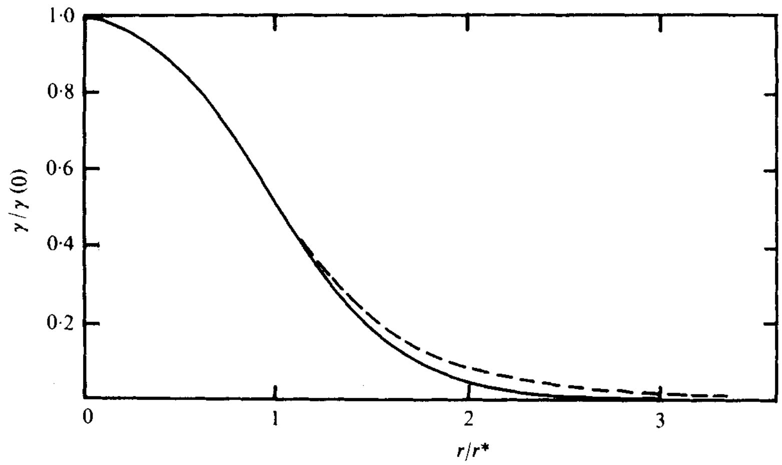

FIGURE 1. Reduced intermittency factor $\gamma/\gamma(0)$ *vs.* reduced radial position r/r^*, where r^* is the radial position at which $\gamma/\gamma(0) = \frac{1}{2}$. ——, $\gamma(0) = 0{\cdot}7$, $r^* = 1{\cdot}65$; ---, $\gamma(0) = 0{\cdot}9$, $r^* = 2{\cdot}68$.

the assumption that the centre-line intermittency is near unity. The cases $\gamma(0) = 0{\cdot}7$ and 0·9 are displayed in figure 1 and show at least qualitatively similar behaviour to measured distributions of γ in wakes and jets.

4. Conclusion

Equation (10), which is proposed to be adequate to describe the evolution of the single-point probability density function in a plume, is a result of two approximations. Turbulent transport of the density is represented by an eddy diffusivity in a way that conforms to the traditional use of these coefficients in meteorology. Small-scale mixing of the scalar field in the plume due to the interaction of turbulence and molecular diffusion has been represented by linear mean-square estimation theory with a time scale determined from the turbulence microscale and the scalar diffusivity. There is no way to verify the accuracy of these approximations in the absence of laboratory measurements but they seem to provide the correct qualitative features of entrainment and subsequent mixing and reaction.

The similarity postulates which led in § 3 to solutions for the conditioned density and the intermittency factor, under various added constraints, must be considered speculative as they are not at present supported by density or intermittency measurements in plumes. It is feasible to bypass such postulates by seeking numerical solutions of (10), combined with assumptions about either the statistical geometry of the interface or the entrainment rate or the intermittency factor, but that seems to be a substantial undertaking, the worth of which is also compromised by the present lack of experimental data. It has been found possible to understand a great deal about fast, diffusion-limited reactions with low heat release in the light of knowledge of the one-point density function for a passive scalar in the same flow (Lin & O'Brien 1974; Bilger 1976). The application of the present theory to the case of photochemical smog production from nitric-oxide-rich emissions into ozone-containing ambient air and the qualitative comparison of its predictions with measured data may be an appropriate test of the modelling introduced here since, under some conditions, the evolution of reactive and product species will be mostly determined by the entrainment process and by small-scale mixing inside the plume. A study of this application is in progress.

Professor R. Chevray and Dr C. Dopazo stimulated the author's interest in this area through many valuable discussions. The work was supported in part by the National Science Foundation through grant no. KO40738 and in part by the U.S. Energy Research and Development Administration under constract no. E(30-1)-16 to the Atmospheric Science Division of Brookhaven National Laboratory.

Appendix. Similarity solutions

Kuznetsov & Frost have examined (17) in the limit in which B is zero. Their solution can be written as

$$g = \begin{cases} g(0)\{1-[1-g(0)]\,\phi\}^{-[1-2g(0)]/[1-g(0)]} & \text{if} \quad g \leqslant [1-g(0)]^{-1}, \\ 0 & \text{if} \quad g > [1-g(0)]^{-1}. \end{cases} \tag{A 1}$$

In the special case $g(0) = 1$, (A 1) reduces to

$$g = \exp\{-\phi\},$$

or

$$f_{\Gamma^*}(\Gamma) = \frac{1}{\langle\Gamma\rangle}\exp\left\{-\frac{\Gamma}{\langle\Gamma\rangle}\right\}$$

and

$$f_{\Gamma}(\Gamma) = (1-\gamma)\,\delta(\Gamma)+\frac{\gamma}{\langle\Gamma\rangle}\exp\left\{-\frac{\Gamma}{\langle\Gamma\rangle}\right\}, \tag{A 2}$$

in agreement with (1) and the measurements reported by Barry. $g = \exp\{-\phi\}$ is the only member of (A 1) which has finite and non-zero probability densities for all ϕ and whose moments of all orders are finite.

When $B \neq 0$ the case $g(0) = 1$ becomes

$$B\,\frac{d^2}{d\phi^2}\{\phi^2 g\}-\frac{dg}{d\phi}-g = 0.$$

A series solution in powers of B can be constructed and is of the form

$$g = \exp\{-\phi\}\left(1+B\int_0^{\phi} f(\phi_1)\,d\phi_1+\ldots+B^n\int_0^{\phi}\ldots\int_0^{\phi} f(\phi)\,d\phi\ldots\,d\phi_1+\right),$$

where $f(\phi) = \phi^2-4\phi+2$. Using a result from iterated integrals (Goursat 1964) gives

$$g = \exp\{-\phi\}\left(1+\sum_{n=1}^{\infty}\frac{B^n}{(n-1)!}\int_0^{\phi}(\phi-t)^{n-1}(t^2-4t+2)\,dt\right). \tag{A 3}$$

The series converges rapidly for all finite B and one can easily show that $\int g\,d\phi \equiv 1$. However, for B large enough, it fails to guarantee $g \geqslant 0$ near $\phi = 2$. We have not computed an upper bound on B for which g as given by (A 3) has all the desired properties of a probability density.

More generally, by using the following form equivalent to (17),

$$B(\phi^2 g')' + [2B - (g(0)-1)](\phi g)' - g' - g(0)g = 0,$$

we can easily show that

$$\int_0^\phi g\,d\phi \equiv 1$$

if $g(0)$ and $g'(0)$ are bounded and $\lim_{\phi\to\infty} g = O(1/\phi)$.

Equation (17) has two parameters $g(0)$ and B. The special case $g(0) > \frac{1}{2}$, $B = g(0) - \frac{1}{2}$ can also be readily integrated by rewriting (A 1) in the form

$$g' = y, \quad B\phi^2 y' + [(2A + g(0))\phi - 1]y = 0.$$

Hence $g' = y = \phi^{-b}\exp\{c_1 - 1/B\phi\}$, where $b = 2 + g(0)/B$ and c_1 is a constant of integration.

Finally, by one more integration, letting $B\phi = 1/z$, we find

$$g = g(0) + (\exp c_1)B^{b-1}\int_{1/B\phi}^{\infty} z^{b-2}e^{-z}\,dz.$$

In this case g is everywhere positive but the integral, which is related to the incomplete gamma function $\gamma(b-1,\ 1/B\phi)$ (Abramowitz & Stegun 1965, p. 260), increases monotonically with ϕ and g can be normalized only by truncation at some finite $\phi < g(0)^{-1}$.

REFERENCES

ABRAMOWITZ, M. & STEGUN, I. 1965 *Handbook of Mathematical Functions.* Dover.

ADRIAN, R. J., JONES, B. G., CHUNG, M. K & NTHIANANDAN, C. K. 1975 *Bull. Am. Phys. Soc.* **21**, 1233.

ARIS, R. 1962 *Vectors, Tensors and the Basic Equations of Fluid Mechanics.* Prentice-Hall.

BARRY, P. J. 1971 *Nuclear Techniques in Environmental Pollution*, p. 241. Vienna: International Atomic Energy Agency.

BARRY, P J 1975 *Chalk River Nuclear Labs., Ontario, Rep.* AECL-5012.

BILGER, R. W. 1976 *Comb. Sci. Tech.* **13**, 155.

BRISSAUD, A. & FRISCH, U. 1974 *J. Math. Phys.* **38**, 665.

CORRSIN, S. 1943 *N.A.C.A. Wartime Rep.* W-94.

CSANADY, G. 1973 *Turbulent Diffusion in the Environment.* Reidel.

DOPAZO, C. 1973 Ph.D. dissertation, State University of New York at Stony Brook.

DOPAZO, C. 1977 *J. Fluid Mech.* **81**, 433.

DOPAZO, C. & O'BRIEN, E. E. 1977 *Symp. Turbulent Shear Flows, Penn. State Univ.* vol. 1, p. 1.

GEL'FAND, I. M. & SHILOV, G. E. 1964 *Generalized Functions*, vol. 1, chap. 3. Academic Press.

GOURSAT, E. 1964 *A Course in Mathematical Analysis*, vol. 2, part 2. Dover.

KUZNETSOV, V. R. & FROST, V. A. 1973 *Izv. Akad. Nauk SSSR, Mekh. Zh. i Gaza* **2**, 58.

LARUE, J. C. & LIBBY, P. A. 1974 *Phys. Fluids* **17**, 1956.

LIBBY, P. A. 1975 *J. Fluid Mech.* **68**, 273.

LIBBY, P. A. 1976 *Phys. Fluids* **19**, 494.

LIN, C. H. & O'BRIEN, E. E. 1974 *J. Fluid Mech.* **64**, 195.

MEYERS, R. E., O'BRIEN, E. E. & SCOTT, L. R. 1978 *J. Fluid Mech.* **85**, 233.

O'BRIEN, E. E., MEYERS, R. E. & BENKOVITZ, C. 1976 *3rd Symp. Atmos. Turb., Diffusion Air Quality, Am. Met. Soc., Raleigh, North Carolina*, p. 160.

PAPOULIS, A. 1965 *Probability, Random Variables, and Stochastic Processes.* McGraw-Hill.

PHILLIPS, O. M. 1972 *J. Fluid Mech.* **51**, 97.

TAYLOR, G. I. 1921 *Proc. Lond. Math. Soc.* (2), **20**, 196.

TUTU, N. K. 1976 Ph.D. dissertation, State University of New York at Stony Brook.