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Some remarks on modelling the PDF of the concentration of a dispersing scalar in turbulence

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The paper deals with the probability density function (PDF) of the concentration of a scalar within a turbulent flow. Following some comments about the overall structure of the PDF, and its approach to a limit at large times, attention focusses on the so-called small scale mixing term in the evolution equation for the PDF. This represents the effect of molecular diffusion in reducing concentration fluctuations, eventually to zero. Arguments are presented which suggest that this quantity could, in certain circumstances, depend inversely upon the PDF, and a particular example of this leads to a new closure hypothesis. Consequences of this, especially similarity solutions, are explored for the case when the concentration field is statistically homogeneous.

1 Introduction

One of the most important features of turbulent flows is how they disperse scalars (e.g. a dissolved dye, an aerosol in the atmosphere, heat). The concentration of a dispersing scalar (or temperature in the case of heat) is inevitably a random function of position $\mathbf{x}$ and time t, but a random function whose statistical properties are determined by physics. Historically research tended to focus on one or two of the simplest statistical properties, especially the (ensemble) mean and variance of the concentration. But, though important, these two properties are not able, in themselves, to provide an adequate (either physical or practical) description of the random concentration field. Thus, there has naturally been increasing focus on other statistical properties including the Probability Density Function (PDF) of concentration.

It has been recognised for many years [23] that a proper description of turbulent combustion requires consideration of PDFs. More recently, but belatedly, there has developed acceptance (now widespread) that PDFs are needed to quantify hazards, such as toxicity, flammability and malodour, associated with gases dispersing in the atmosphere. Measurements of the PDF of increasing reliability are now being obtained not only in the laboratory but also in the field [21, 16]. Pioneering theoretical work on the PDF is described by Borghi [2], Dopazo [9], Kuznetsov & Sabel'nikov [15], Pope [23, 24] and Sullivan & Ye [26]. However, theoretical research is, of course, hampered by the ubiquitous closure problem and while this may be circumvented one day using DNS (Direct Numerical Simulation), estimates in Mole *et al.* [18] suggest that this will not be feasible for most realistic flows and geometries for many years. It is reasonable to claim that theoretical research on the PDF of a dispersing scalar is a relatively new topic with inadequate understanding, and that there is still considerable scope for simple mathematical models based on good physical insight. The present paper is a contribution with this philosophy.

2 The evolution equation for the PDF

Consider a dispersing scalar in a turbulent flow, subject only to advection and molecular diffusion. More precisely, it will be supposed throughout that the concentration $C(\mathbf{x}, t)$ of the dispersing scalar (or temperature in the case of heat) obeys the advection-diffusion equation

$$\frac{\partial C}{\partial t} + \mathbf{U}.\nabla C = \kappa \nabla^2 C, \tag{2.1}$$

where $\mathbf{U}=\mathbf{U}(\mathbf{x},t)$ is the random turbulent velocity field, satisfying mass conservation and the Navier–Stokes equations, and the positive constant κ is the molecular diffusivity. The randomness of $\mathbf{U}$ enforces randomness of C. The PDF of C is denoted by $p_C(q; \mathbf{x}, t)$ where (except perhaps at isolated points):

$$p_C(q; \mathbf{x}, t) = \frac{d}{dq}\left[\text{prob}\left\{C(\mathbf{x}, t) \leqslant q\right\}\right]; \tag{2.2}$$

alternatively,

$$p_C(q; \mathbf{x}, t)\ \delta q \ \approx\ \text{prob}\left\{q \leqslant C(\mathbf{x}, t) < q + \delta q\right\}, \tag{2.3}$$

where δq is positive and small. Since p_C is a PDF, it follows that

$$\int_0^\infty p_C(q; \mathbf{x}, t) dq = 1, \tag{2.4}$$

and the (ensemble) mean concentration $E\{C\}$, where $E\{\cdot\}$ denotes the expected value, is defined by

$$E\{C\} = \int_0^\infty q\ p_C(q; \mathbf{x}, t) dq, \tag{2.5}$$

with analogous equations for the variance and higher moments – see Mole *et al.* [18]. The PDF $p_C(q; \mathbf{x}, t)$ obeys an evolution equation determined from equation (2.1). This equation can be written in many equivalent forms, of which the most convenient for present purposes is

$$\frac{\partial p_C}{\partial t} + \nabla.\left[p_C\ E\left\{\mathbf{U} \mid C = q\right\}\right] = \kappa \nabla^2 p_C - \kappa \frac{\partial^2}{\partial q^2}\left[p_C\ E\left\{(\nabla C)^2 \mid C = q\right\}\right]. \tag{2.6}$$

In equation (2.6), $E\{A|B\}$ denotes the expected value of event A conditional upon the occurrence of the event B. The second terms on each side of equation (2.6) are not closed, i.e. they are not expressible in terms of p_C and the independent variables. Thus, for example, the second term on the right-hand side of equation (2.6), called the Small Scale Mixing Term (SSMT) by Pope [24], depends also upon the joint PDF of C and ∇C. Much of the present paper deals with the SSMT.

3 Some limiting properties of the PDF

In this paper, it is useful to fix attention on a particular type of situation. Suppose that, at $t = 0$, a finite quantity M of scalar of uniform concentration C_m is released into a turbulent flow in which there is no scalar. This statistically unsteady dispersion situation is a reasonable approximation, at worst, to many real accidents, and was investigated experimentally by Hall *et al.* [11].

Thus there are regions Σ_1 and Σ_2, with volumes V_1 and V_2 respectively, where the union of Σ_1 and Σ_2 is the total region Σ, with volume V, available for dispersion, such that

$$C(\mathbf{x},0) = \begin{cases} C_m & (\mathbf{x} \subset \Sigma_1), \\ 0 & (\mathbf{x} \subset \Sigma_2). \end{cases} \tag{3.1}$$

Note that V and V_2 may be infinite. Since subsequent dispersion is governed by equation (2.1), it follows from a well-known property of this equation that, for all times $t > 0$ and for all finite $\mathbf{x} \subset \Sigma$:

$$C(\mathbf{x},t) < C_m \quad \text{and} \quad C(\mathbf{x},t) > 0. \tag{3.2}$$

Thus there are concentrations $q_1 = q_1(\mathbf{x},t)$ and $q_2 = q_2(\mathbf{x},t)$, with $q_1 > 0$ and $q_2 < C_m$, such that

$$p_C(\mathbf{x},t) = 0 \quad \text{for} \quad q < q_1\,,\, q > q_2. \tag{3.3}$$

It merits emphasis that q_1 and q_2 are determined by the governing equations and by the choice of ensemble.

Equation (3.2) and its consequence, equation (3.3), arise because equation (2.1) is parabolic, thereby allowing, in effect, some pollutant molecules to have infinite velocities. This is obviously wrong and has led some authors to consider replacing (2.1) by, for example, a generalized telegraph equation (see Monin & Yaglom [19, pp. 676–693]). Nevertheless there is no experimental evidence that (2.1) does not describe turbulent dispersion adequately for all practical purposes, and (3.2) and (3.3) then have to be accepted.

Estimates of the *a priori* unknown quantities q_1 and q_2, and the behaviour of p_C near q_1 and q_2, can in principle be made from data using the statistical theory of extremes – see Mole *et al.* [17] and Munro *et al.* [20] – but there is considerable uncertainty in such estimates.

The constraints in (3.2) and (3.3) have often been overlooked, particularly that q_2 is finite. The latter constraint rules out the power law decay predicted by Sinai & Yakhot [25], the clipped Normal fitted by Mylne & Mason [21] and the clipped gamma derived by Yee & Chan [32], as well as many others.

For the case when V is finite, the SSMT ensures that the concentration will tend to the uniform value M/V as $t \to \infty$, i.e.

$$p_C(q;\mathbf{x},t) \to \delta\left(q - \frac{M}{V}\right) \quad \text{as} \quad t \to \infty. \tag{3.4}$$

In the limit given by (3.4)

$$\mu = E\{C\} = \frac{M}{V}; \quad \sigma^2 = Var\{C\} = E\{(C-\mu)^2\} = 0, \tag{3.5}$$

where $Var\{.\}$ denotes the variance and σ is the standard deviation of C. Thus

$$\frac{\sigma}{\mu} \to 0 \quad \text{as} \quad t \to \infty. \tag{3.6}$$

In view of controversy about the limiting value of σ/μ in other circumstances (e.g. see Thomson [29]) it might be worth adding as an aside that whenever it can be deduced that $p_C \to \delta(q - q_0)$ with $q_0 > 0$, σ/μ must tend to zero. One such example occurs a long way downwind of a steady source in a windtunnel or pipe of constant cross-section; in this case the limit is approached as downwind distance, rather than time, increases. But in many other cases, such as some pollution plumes, lateral spreading is unconfined so that V in (3.4) is effectively infinite. Then $p_C \to \delta(q)$ so both μ and σ in (3.5) approach zero and (3.6) no longer necessarily holds.

4 The basis of a new closure hypothesis for the SSMT

For the situation described at the beginning of the previous section, the initial structure of p_C is given by

$$p_C(q; \mathbf{x}, 0) = \pi(\mathbf{x})\delta(q - C_m) + [1 - \pi(\mathbf{x})]\delta(q), \tag{4.1}$$

where $\pi(\mathbf{x})$ is determined by the selected ensemble. Two examples out of many possibilities are:

(a) The region Σ_1 has fixed shape and location. Then

$$\pi(\mathbf{x}) = \begin{cases} 1 & (\mathbf{x} \subset \Sigma_1), \\ 0 & (\mathbf{x} \subset \Sigma_2). \end{cases} \tag{4.2}$$

Recall that Σ_2 is the complement of Σ_1 within the total region Σ.

(b) The region Σ is finite with volume V, but the scalar-containing region Σ_1 of volume V_1 has random shape and location, with the randomness such that

$$\pi(\mathbf{x}) = \frac{V_1}{V} \quad \text{for all } \mathbf{x} \subset \Sigma. \tag{4.3}$$

The condition (4.2) applies to the experiments of Hall *et al.* [11].

By contrast, when (4.3) holds, $p_C(q; \mathbf{x}, 0)$ is homogeneous, i.e. independent of $\mathbf{x}$. In common with many other studies (e.g. Eswaran & Pope [10] and Jaberi *et al.* [12]), this paper considers the simplest possible circumstances when $p_C(q; \mathbf{x}, t)$ is homogeneous for all t. Then equation (2.6) reduces to

$$\frac{\partial p_C}{\partial t} = -\kappa \frac{\partial^2 g_C}{\partial q^2}, \tag{4.4}$$

where

$$g_C = p_C \, E\left\{(\nabla C)^2 \mid C = q\right\}. \tag{4.5}$$

By this choice it is hoped to gain some understanding. But, since homogeneous turbulence can never be exactly realised in practice, the results can, at best, only describe approximately the local behaviour of p_C, i.e. on length scales small compared with those characteristic of changes in the statistical properties of the velocity field. This could occur in the interiors of pollution plumes downwind of industrial chimney stacks. Alternatively, (4.4) and (4.5) could model p_C within a box which is continually and randomly stirred with initial conditions given by (4.1) and (4.3).

In summary, therefore, a major problem is to study the way in which p_C evolves via (4.4) and (4.5) from the double-delta-function distribution in (4.1) and (4.3) to the single-delta-function distribution in (3.4).

Two distinct types of theoretical method have dominated previous work on this problem. The first, also used in the present paper, is to express the SSMT on the right-hand side of (4.4) in terms, undoubtedly approximate, of p_C; this is known as a 'closure hypothesis' and has a long history in turbulence, and turbulent dispersion, research beginning with the use of eddy viscosities and diffusivities. Those closure hypotheses that have been applied to the SSMT are discussed by many authors (e.g. Pope [24, pp. 158–163], Borghi [2, pp. 259–261], Kuznetsov & Sabel'nikov [15, pp. 51–53] and Dopazo [9, pp. 409–421]. A popular alternative calculation method for predicting p_C is DNS, i.e. equation (2.1) is solved numerically many times sufficient both to (a) include an adequate sample of the possible velocity fields, although the ways in which these are modelled vary greatly, and (b) allow reasonably robust estimations of p_C. Papers using DNS include Eswaran & Pope [10], Jaberi *et al.* [12] and Zimmerman [34]. Use of a closure hypothesis has the great potential advantage over DNS of speed of calculation, but it must be physically reasonable and, for practical use, of known accuracy in specified circumstances.

In laboratory experiments, Tavoularis & Corrsin [27] observed that p_C was approximately Gaussian (i.e. a Normal distribution) when there was a constant and non-zero spatial gradient of the mean concentration μ. Pope [24, p. 157] cites this in claiming that an "aim of the modelling is, therefore, to produce Gaussian PDFs whose first and second moments evolve correctly" and later (*loc. cit.*, p. 159) writes "A satisfactory modelled PDF equation should admit this Gaussian PDF as a solution". In introducing their DNS experiments with spatially homogeneous statistics, Eswaran & Pope [10, p. 515] state that "It is generally assumed that the scalar PDF, starting from a double-delta distribution, evolves towards a Gaussian", and this claim is repeated by other authors, e.g. Dopazo [9, p. 410]. The following comments may be made immediately:

- The experimental conditions used by Tavoularis & Corrsin [27] are not consistent with an initial double-delta-function distribution.
- A Gaussian PDF, i.e. one for which
$$p_C = \frac{1}{\sigma\sqrt{2\pi}} \exp\left\{-\frac{(q-\mu)^2}{2\sigma^2}\right\} \quad (-\infty < q < \infty), \tag{4.6}$$
where $\mu = \mu(t)$, $\sigma = \sigma(t)$, can at best be an approximation, and an unphysical one, since it does not satisfy (3.3) above.
- Measured PDFs in field experiments with an initial double-delta-function distribution are very frequently of the strongly non-Gaussian form typified by Figure 1. (Further examples can be found in Lewis and Chatwin [16] and many other papers.) Characteristic features include a pronounced maximum of p_C for a low value of q[1] and a long tail which, by (3.3), terminates at a finite value of q.

[1] Because of binning in data acquisition, it is usually impossible to distinguish this value of q from zero, although (3.3) shows that it must be strictly positive.

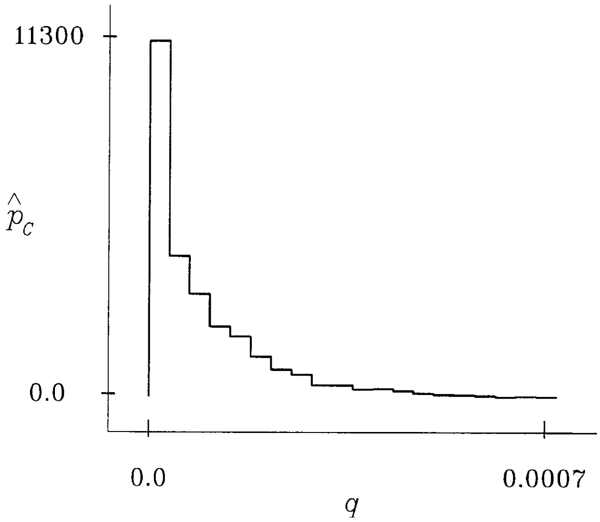

FIGURE 1. Measured values of p_C, denoted by $\hat{p}_C$, against q from field experiments conducted by Risø National Laboratory, Denmark (BORRIS campaign 1994).

Some evidence for the statement by Eswaran & Pope [10], quoted above, may be found in their paper. Figures 14, 15 and 16 of that paper show that as time evolves the two initial peaks of p_C (used in the simulation to model the initial double-delta-function) shrink in magnitude with a concomitant rise in the values of p_C between the peaks. Eventually a single peak, with growing magnitude, emerges. While the final shapes of the graphs of p_C against q after four eddy-turnover times appear qualitatively similar to Gaussians, Eswaran & Pope [10, pp. 517–518] note that the flatness (i.e. kurtosis) shows "little sign of levelling off (let alone returning to 3)", refer to the "lack of Gaussianity of p_C", and state "Thus Figs. 20 and 24 consistently indicate the lack of Gaussianity in p_C at the end of the simulations". However a single simulation for the extended period of twelve eddy-turnover times does suggest that the skewness and kurtosis are approaching the Gaussian values although there is non-negligible statistical variability.

Subsequent to the work of Eswaran & Pope [10], other workers have indeed noted that a Gaussian form of p_C "is only one of many possible outcomes" (Jaberi *et al.* [12, p. 244]). Later (*loc. cit.*, p. 277) the same authors state that "The primary observation made here is to verify that the long-time PDF of a passive scalar in homogeneous turbulent flows is not necessarily Gaussian (or of any particular form)... ". Using a variety of DNS techniques, Jaberi *et al.* [12] obtain PDFs, some of which are Gaussian and some of which are exponential (the latter, like (4.6), can only be approximate since it too does not satisfy (3.3) above). Kimura and Kraichnan [13] also obtain non-Gaussian PDFs using both DNS and an idealized analytical model.

Another interesting difference between the work of Eswaran and Pope [10] and that of Jaberi *et al.* [12] is that the former note that "the evolution of PDF shapes ... appears to be independent of the initial conditions" (Eswaran and Pope [10, p. 515]) whereas the latter state "that both Gaussian and exponential scalar PDFs emerge depending on the parameters of the simulations and the initial conditions of the scalar field" (Jaberi *et al.* [12, p. 241).

The last quote indicates the potential importance of the precise DNS technique used. It also merits emphasis that in the DNS methods used in the papers referred to, in Zimmerman [34], and in many others, periodic boundary conditions were imposed and the resulting PDFs may, at least in part, be influenced by this strong and unreal constraint. Tsinober [31, p. 423] notes that because of periodic boundary conditions: " . . . the correlation coefficient between two values of any quantity at . . . opposing boundaries, i.e. the points separated at maximal distance in the flow domain, is precisely equal to unity . . . . On the contrary, in any real flow the correlation coefficient becomes very small for points separated by a distance of the order of, and larger than, the integral scale of turbulent flow."

In summary, the present modelling situation is rather confused, with different numerical techniques giving results that differ strongly from one another in some fundamental respects; nor are they generally consistent with experimental measurements or basic theoretical results like (3.3).

The remainder of the present paper is based on a new idea for closing (4.4). It is claimed merely that this idea may provide fresh insight, not that it will resolve decisively the problems indicated in the previous paragraph.

First, suppose that the initial volume V_1 of the scalar-containing region is small compared with the total volume V; this is a condition satisfied in nearly all real situations. It has been assumed for many years, following such papers as Batchelor [1] and Corrsin [6], and has relatively recently been directly confirmed in beautiful experiments by Dahm *et al.* [7], that scalar-containing regions are drawn out into thin threads or sheets. These become widely separated, and the threads or sheets regenerate (albeit with lower concentrations within them). The thinness ensures large values of $|\nabla C|$ where C is large, i.e. $E\{(\nabla C)^2|C = q\}$ is large when q is large. Since V_1/V is small, large values of q are associated with small values of p_C as in Figure 1. Thus g_C/p_C is large where p_C is small, where g_C is defined in (4.5). Scalar diffuses by molecular diffusion across the boundaries of these scalar-containing regions into the ambient fluid in which, because $V_1/V \ll 1$, concentrations are low and thorough mixing ensures that $|\nabla C|$ is small there, except very near the boundaries of the thin threads or sheets containing most of the scalar. Thus the converse to the above holds: $E\{(\nabla C)^2|C = q\}$ is small where q is small and, because V_1/V is small, small values of q tend to be associated with large values of p_C. Thus g_C/p_C is small where p_C is large.

These arguments suggest the qualitative, and new, conclusion that in the postulated circumstances:

$$p_C \text{ and } \frac{g_C}{p_C} \text{ depend inversely upon one another.} \tag{4.7}$$

This is a principal result in the present paper, arguably the principal result.

It is interesting to compare it with the well-studied proposal in Dopazo (1975), which is that

$$\frac{\partial g_C}{\partial q} = -\frac{3(\nu/\kappa)}{\lambda^2}(q - \mu)p_C, \tag{4.8}$$

where λ is the Taylor microscale and ν is the kinematic viscosity. Pope [24, p. 159] and Dopazo [9, p. 411] note that use of (4.8) in (4.4) preserves the shape of p_C so that (4.8) does not enable the initial double-delta-function distribution (4.1) to evolve via (4.4) towards, for example, the Gaussian distribution (4.6). While this is a serious drawback, more relevant to present purposes is that, as already noted, both Dopazo and Pope suppose that a closure hypothesis for (4.4) should lead to a p_C which is Gaussian. In this case, i.e. when (4.6) holds, it is easy to show that (4.8) gives

$$g_C = \frac{3(\nu/\kappa)}{\lambda^2}\sigma^2 p_C, \tag{4.9}$$

i.e. g_C/p_C is independent of p_C, thus contradicting (4.7).

5 A specific closure hypothesis for the SSMT

This paper has tried to emphasize the difficulty of modelling p_C, including the likelihood that many different functional forms occur in practice and therefore that different closure hypotheses for the SSMT may hold in different circumstances. However, a particular closure hypothesis consistent with (4.7) arises from a PDF obtained in earlier work by the author with Zimmerman (Zimmerman & Chatwin [35]; Chatwin & Zimmerman [5]), and it is natural to consider this further. In the first of these papers, turbulent dispersion was modelled simplistically by supposing that the scalar disperses deterministically with no velocity field and the sole stochastic feature is that the sensor measuring C is located randomly within a domain of finite volume. The calculations in the two papers are for a one-dimensional domain $-L \leqslant x \leqslant L$, but there is no difficulty in extending the ideas to three dimensions. It is noted that an alternative interpretation of the random feature is that the domain is randomly translated; this is analogous to the random stirring discussed above. The PDF p_C can be obtained from first principles by elementary methods. For large t, it is well approximated by

$$p_C(q) = \frac{1}{\pi C_0 \Theta}\left\{1 - \frac{(q-\mu)^2}{C_0^2\Theta^2}\right\}^{-\frac{1}{2}} \quad (q_1 < q < q_2), \tag{5.1}$$

where C_0 and μ are positive constants (with μ being the mean concentration as usual), and

$$q_1 = \mu - C_0\Theta\ ,\ q_2 = \mu + C_0\Theta\ ,\ \Theta = e^{-\pi^2 T}\ ,\ T = \frac{\kappa t}{l^2}. \tag{5.2}$$

This PDF has integrable singularities at q_1 and q_2, and is symmetric about μ. In view of the discussion above, it is pertinent to note that as $t \to \infty$ (i.e. $T \to \infty$), $q_1 \to \mu$ from below and $q_2 \to \mu$ from above. Thus, as $t \to \infty$, p_C in equation (5.1) approaches $\delta(q-\mu)$ consistent with (3.4). But the approach occurs because the singularities at q_1 and q_2 move closer to one another and eventually coalesce. At no stage is p_C approximated, even crudely, by the Normal distribution in (4.6). It may also be noted that it is easy to show from (5.1) that $\sigma = C_0\Theta/\sqrt{2}$ so that equation (3.6) is satisfied. Finally, since (5.1) may be rewritten

$$p_C(q) = \frac{1}{\pi}(q-q_1)^{-\frac{1}{2}}(q_2-q)^{-\frac{1}{2}} \quad (q_1 < q < q_2), \tag{5.3}$$

it is a member of the beta distribution family which has been used frequently in turbulent dispersion (e.g. see Chatwin *et al.* [4]).

Now p_C in (5.1) must satisfy (4.4) for $q_1 < q < q_2$. It is then straightforward to show from (4.4) that

$$g_C p_C = \frac{1}{L^2} \quad (q_1 < q < q_2). \tag{5.4}$$

and this is equivalent to

$$E\left\{(\nabla C)^2 \mid C = q\right\} = \frac{g_C}{p_C} = \left(\frac{1}{p_C L}\right)^2 \quad (q_1 < q < q_2), \tag{5.5}$$

consistent with (4.7) above. The results in (5.4) and (5.5) can also be obtained easily and directly from the model equations in Chatwin & Zimmerman [5].

It will also be shown – see (6.17) and the remarks following – that (5.4) applies to an exact solution for the PDF derived and used by Kowe & Chatwin [14].

In both this exact solution, and in (5.1), there are singularities in p_C at $q = q_1$ and $q = q_2$, and (5.4) applies in both cases only in the limits as $q \to q_1$ from above and as $q \to q_2$ from below.

However it is clear that (5.4) cannot apply at q_1 and q_2 when there are no singularities, for then both p_C and ∇C (hence g_C/p_C) are zero at q_1 and q_2. Moreover the PDF in (5.1) is very different in shape from that in Figure 1, and from that underlying the argument leading to (4.7). (It could be argued that each side of the symmetric expression in (5.1) is qualitatively similar to the left-hand side of the experimental curve in Figure 1 so that part of the argument leading to (4.7) still applies.) Thus (5.4) cannot apply universally, but this has nowhere been suggested. Like any other simple closure hypothesis, its range of applicability must be limited and so therefore are the results in the penultimate section of this paper when similarity solutions satisfying (5.4) are briefly investigated.

Before this, it is of interest and importance to consider timescales. From the purely mathematical viewpoint, the length L in the Zimmerman & Chatwin [35] model appearing in (5.4) is arbitrary (although the title of the paper makes it clear that it is a microscale in real applications). In the work leading to (5.1), the timescale is of order L^2/κ, whereas it is well-known that in most circumstances the timescale associated with the evolution of p_C is of order l/u, where l is proportional to the size of the energy-containing eddies and u is a velocity of the order of the size of the velocity fluctuations. Now [28, p. 67], u^3/l is of order of $\nu u^2/\lambda^2$ (where λ is the Taylor microscale introduced in (4.8) and (4.9) above), so that l/u is of order L^2/κ provided

$$\frac{L^2}{\kappa} \propto \frac{\lambda^2}{\nu} \quad \Rightarrow \quad L \propto \frac{\lambda}{(\nu/\kappa)^{1/2}} = \lambda_C, \tag{5.6}$$

where λ_C is the Taylor microscale of the concentration field [8]. This result is entirely consistent with (4.9) and (5.4). Thus (4.9) gives $g_C p_C \propto (\nu/\kappa)\lambda^{-2}(\sigma p_C)^2$ and (5.4) gives $g_C p_C \propto L^{-2}$. In general, σp_C is of order unity so that the *magnitudes* of (4.9) and (5.4) are of the same order provided $L^{-2} \propto (\nu/\kappa)\lambda^{-2}$, i.e. $L \propto \lambda_C$ in agreement with (5.6). (These remarks do not weaken the earlier criticism of (4.8) and (4.9) above.)

6 Some consequences of the new closure hypothesis: similarity solutions

This section shows that the closure hypothesis (5.4) is consistent with two families of similarity solutions for p_C. As usual, similarity solutions can be expected to apply in the later stages of the dispersion process when the influence of the initial conditions has weakened. Thus the solutions will not necessarily be expected to satisfy the initial double-delta-function distribution in (4.1), but to have plausibly developed from it in the way, for example, suggested after (5.1) above.

Substitution of (5.4) into (4.4) gives

$$\frac{\partial p_C}{\partial t} = \frac{\kappa}{L^2}\frac{\partial}{\partial q}\left(p_C^{-2}\frac{\partial p_C}{\partial q}\right), \tag{6.1}$$

which is reminiscent of the porous medium equation

$$\frac{\partial u}{\partial t} = \nabla.(u^n \nabla u), \tag{6.2}$$

except that n is not positive (see Ockendon *et al.* [22, p. 257]). For similarity solutions, write

$$T = \frac{\kappa t}{L^2}\ ,\ \eta = \frac{(q - q_0)}{C_0 \Theta_1(T)}\ ,\ p_C = \frac{f(\eta)}{C_0 \Theta_2(T)}, \tag{6.3}$$

where q_0 and C_0 are positive constants, and Θ_1 , Θ_2 and f are positive functions to be determined. Substitution in equation (6.1) gives

$$\frac{d^2}{d\eta^2}\left(\frac{1}{f}\right) = \frac{\dot{\Theta}_2 \Theta_1^2}{\Theta_2^3} f + \frac{\dot{\Theta}_1 \Theta_1}{\Theta_2^2}\eta\frac{df}{d\eta}, \tag{6.4}$$

where a dot denotes differentiation with respect to T. It follows that for similarity solutions there must be constants α and β such that

$$\frac{\dot{\Theta}_2 \Theta_1^2}{\Theta_2^3} = -\alpha\ ,\ \frac{\dot{\Theta}_1 \Theta_1}{\Theta_2^2} = -\beta, \tag{6.5}$$

where the minus signs are chosen because, then, positive values of α and β allow Θ_1 and Θ_2 to tend to zero as $T \to \infty$ to attain the limiting delta-function distribution in equation (3.4). Then equation (6.4) becomes

$$\frac{d^2}{d\eta^2}\left(\frac{1}{f}\right) = -\alpha f - \beta\eta\frac{df}{d\eta}. \tag{6.6}$$

It follows from equation (6.5) that

$$\frac{\dot{\Theta}_2}{\Theta_2} = \frac{\alpha}{\beta}\frac{\dot{\Theta}_1}{\Theta_1}, \tag{6.7}$$

and hence that

$$\Theta_2 = A\Theta_1^{\alpha/\beta}, \tag{6.8}$$

where A is an arbitrary positive constant. Thus

$$\dot{\Theta}_1 = -\beta A^2 \Theta_1^{(2\alpha-\beta)/\beta}, \tag{6.9}$$

and there are two cases to consider.

If $\alpha \neq \beta$

$$\begin{aligned}\Theta_1 &= \left[2(\alpha-\beta)A^2T + \Theta_{10}^{-2(\alpha-\beta)/\beta}\right]^{-\beta/2(\alpha-\beta)} \\ \Theta_2 &= A\left[2(\alpha-\beta)A^2T + \Theta_{10}^{-2(\alpha-\beta)/\beta}\right]^{-\alpha/2(\alpha-\beta)} ,\end{aligned} \tag{6.10}$$

where $\Theta_{10} = \Theta_1(0)$. It follows from equation (6.10) that $\alpha > \beta$ because, if $\alpha < \beta$, equation (6.10) predicts that $\Theta_1 = \Theta_2 = 0$ in finite time. For large values of T, the dependence of Θ_1 and Θ_2 on Θ_{10} weakens, and the results in equation (6.10) can be approximated by

$$\Theta_1 = \left[2(\alpha-\beta)A^2T\right]^{-\beta/2(\alpha-\beta)} \ , \ \Theta_2 = A\left[2(\alpha-\beta)A^2T\right]^{-\alpha/2(\alpha-\beta)} \ , \tag{6.11}$$

or, more concisely,

$$\Theta_1 = B_1T^{-p} \ , \ \Theta_2 = B_2T^{-p-\frac{1}{2}} \ , \ p = \frac{\beta}{2(\alpha-\beta)} > 0 \ , \ \frac{B_1^2}{B_2^2} = 2(\alpha-\beta). \tag{6.12}$$

The second case is when $\alpha = \beta$. Direct integration then gives

$$\Theta_1 = \Theta_{10}e^{-\alpha A^2T} \ , \ \Theta_2 = A\Theta_{10}e^{-\alpha A^2T} \ , \tag{6.13}$$

and equation (6.6) becomes

$$\frac{d^2}{d\eta^2}\left(\frac{1}{f}\right) = -\alpha\frac{d}{d\eta}(\eta f). \tag{6.14}$$

Equation (6.14) can be integrated once trivially, and the substitution $\eta f = g^{-1}$ then enables it to be integrated completely. A special case of this family of solutions is equation (5.1) for which

$$\alpha = 1 \ , \ q_0 = \mu \ , \ A = \pi \ , \ f = (1-\eta^2)^{-\frac{1}{2}}. \tag{6.15}$$

Returning now to the general case given by equation (6.6), an exact solution has been found when $\alpha = 2\beta$ so that, from equation (6.12),

$$p = \frac{1}{2} \ , \ \Theta_1 = B_1T^{-\frac{1}{2}} \ , \ \Theta_2 = B_2T^{-1} \ , \ \frac{B_1}{B_2} = \alpha^{\frac{1}{2}}. \tag{6.16}$$

This solution is

$$f(\eta) = d\eta^{-1}(c\log\eta)^{-\frac{1}{2}} \tag{6.17}$$

for $\eta_1 < \eta < \eta_2$ where $c = -\alpha d^2$, and d is an arbitrary positive constant. Thus $c < 0$ and $\eta < 1$ for the realistic case when α is positive. The expression in equation (6.17) gives a PDF satisfying equation (2.4) provided

$$\left[\log(1/\eta_1)\right]^{\frac{1}{2}} - \left[\log(1/\eta_2)\right]^{\frac{1}{2}} = \frac{1}{2}T^{-\frac{1}{2}}, \tag{6.18}$$

independently of the values of α and d. A special case of this exact solution is equation (30) in Chatwin & Zimmerman [5] when

$$\eta_1 = \exp\left(-\frac{1}{4T}\right) \ , \ \eta_2 = 1. \tag{6.19}$$

This PDF is bimodal like that in equation (5.1) above, with modes at the end points η_1 and η_2. As $T \to \infty$, η_1 tends to η_2 so that the limiting case of the delta-function distributions in equation (3.4) is again approached by the modes coalescing and not via the Gaussian (Normal) PDF in equation (4.6). As well as arising in the model of Zimmerman & Chatwin [35], it is remarkable to note that essentially the same PDF was shown by Kowe and Chatwin [14] to occur in certain cases when the velocity field is the linear rate-of-strain field considered in a classical paper describing 'hot spots' by Townsend [30]. (The random feature is then that the principal axes of rate-of-strain are oriented in random directions in space.)

Solutions of equation (6.6) when α and β have arbitrary values must presumably be found numerically; in all cases there will be an equation analogous to equation (6.18).

There is an interesting generalisation of equation (5.4), the closure hypothesis proposed in this paper and investigated above. This is to replace equation (5.4) by

$$g_C p_C = \frac{h(T)}{L^2}, \tag{6.20}$$

where $h(T)$ is an arbitrary (non-dimensional) function taking positive values. Physically, this could represent random stirring at a time-varying intensity. Use of the variables in equation (6.3) in equation (4.4), but with equation (6.20) replacing equation (5.4), yields

$$h\frac{d^2}{d\eta^2}\left(\frac{1}{f}\right) = \frac{\dot{\Theta}_2\Theta_1^2}{\Theta_2^3}f + \frac{\dot{\Theta}_1\Theta_1}{\Theta_2^2}\eta\frac{df}{d\eta}, \tag{6.21}$$

instead of equation (6.4). Thus

$$\frac{\dot{\Theta}_2\Theta_1^2}{\Theta_2^3} = -\alpha h\,,\ \frac{\dot{\Theta}_1\Theta_1}{\Theta_2^2} = -\beta h, \tag{6.22}$$

are the generalisations of the results in equation (6.5). Hence equations (6.6) and (6.8) remain valid without change. Once more there are two cases to consider. If $\alpha \neq \beta$

$$h(T) = \frac{1}{2A^2(\alpha-\beta)}\frac{d}{dT}\left\{\Theta_1^{-2(\alpha-\beta)/\beta}\right\}, \tag{6.23}$$

whereas, if $\alpha = \beta$,

$$h(T) = -\frac{1}{\alpha A^2}\frac{d}{dT}\{\log\Theta_1\}. \tag{6.24}$$

7 Some concluding remarks

The new closure hypothesis in equation (5.4) has been investigated very recently, using numerical simulations, by Yeun [33] and Zimmerman [34]. Their results are so far preliminary, and do not allow firm conclusions.

As noted near the beginning of this paper, theoretical research on the PDF of a dispersing scalar is still a relatively new topic. While, ultimately, all models must be validated against data, the assumptions made in many papers, and this one, that render the equations to some extent tractable are such that direct experimental comparisons are then not possible. This applies particularly to the simplifications that the scalar distribution and the velocity field are homogeneous. Moreover, costs make experimental investigations of unsteady phenomena in turbulent dispersion unattractive.

It is well-known that turbulence, and turbulent diffusion, are stochastic processes. The present contribution emphasises that the details of the stochasticity are controlled by physics. But many, if not most, real world processes are also stochastic and it is hoped that this work may encourage applied mathematicians to undertake more stochastic modelling while, at the same time, seeking to understand, and involve, the laws controlling the stochasticity.

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