THE STRUCTURE AND MAGNITUDE OF CONCENTRATION FLUCTUATIONS

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Abstract. This paper is concerned with the science of turbulent diffusion and not, except incidentally, with its numerous practical applications. It discusses some recent research, particularly that by the authors and their collaborators. Among the topics considered are (i) the intermittency factor, (ii) the relationship between the mean of the concentration and its variance, and (iii) the interpretation of data. The principal aim of the paper is to draw attention to some outstanding basic questions which would seem promising targets for future research. Without progress on these questions (and others), regulatory models of air quality will continue - inevitably - to be unreliable and hardly worth using.

1. Introduction

The increased importance accorded by the public worldwide, and their governments, to air quality, and to the assessment of hazards associated with the accidental release of dangerous gases into the atmosphere, is resulting in a larger demand for mathematical models for regulatory (and associated) purposes. The number of such models - and their developers - is increasing; so moreover, are quasi-political pressures for the international "harmonisation" of such models. Although less advanced, a similar process is taking place in respect of water pollution.

From this point of view, it is unfortunate that the science of turbulent diffusion, which underpins these important practical problems, is still not understood to enough depth to allow (in general) such models to be well founded and reliable, as well as practically useful. While undeniable, this fact is undoubtedly surprising to most non-experts; nor, regrettably, has it inhibited many people from producing and selling software which, however attractively packaged, cannot fulfil the purposes for which it was purchased since it is based on inadequate, often wrong, science.

Therefore it continues to be important and timely, to conduct research into turbulent diffusion. (Indeed, proper recognition of the real situation ought to lead to vastly increased financial support for such work, but that is another story!) In this paper we summarise some of our recent results on the fundamental science, and consider what we believe to be promising developments. Throughout, the emphasis is on a better understanding of the underlying physics and mathematics. We have given complementary, more practical viewpoints elsewhere (e.g., Chatwin and Sullivan, 1990b; Chatwin, 1991).

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2. Basic Background

We denote by $\Gamma = \Gamma(x, t)$ the concentration (in arbitrary units) of a dispersing contaminant, and we shall suppose throughout this paper that the dependence of Γ on x and t is determined by advection by the ambient fluid, with velocity fluid $Y = Y(x, t)$, and by molecular diffusion, with diffusivity κ. In particular we ignore chemical changes. The equation governing Γ is

$$\frac{\partial \Gamma}{\partial t} + Y \cdot \nabla \Gamma = \kappa \nabla^2 \Gamma . \tag{1}$$

Since the ambient flow is turbulent, Y is a random vector field (satisfying the Navier-Stokes equations); so therefore is Γ. The correct mathematical description of Γ (and Y) must therefore be a statistical one. (This simple truth is largely ignored by the present generation of model developers.) A statistical description requires a definition of the underlying ensemble (Chatwin & Sullivan, 1979; Sullivan, 1990). Since Γ is a random variable it has, in any ensemble, a probability density function $p(\theta; x, t)$ defined in the normal way by

$$p(\theta; x, t) = \frac{d}{d\theta}[prob\{\Gamma(x, t) \leq \theta\}] . \tag{2}$$

The equation governing the evolution of p can be derived from (1) (Chatwin, 1990) and is

$$\frac{\partial p}{\partial t} + \nabla \cdot E\{Y\delta[\Gamma(x, t) - \theta]\} = \kappa \nabla^2 p - \kappa \frac{\partial^2}{\partial \theta^2} E\{\nabla \Gamma)^2 \delta[\Gamma(x, t) - \theta]\} \tag{3}$$

where the symbol E denotes "expected value" in the technical statistical sense.

Equations for some of the simplest and most commonly used statistical properties of Γ can be derived from (3). The mean concentration $\mu(x, t)$, where

$$\mu(x, t) = \int_0^\infty \theta p(\theta; x, t) d\theta = E\{\Gamma(x, t\}, \tag{4}$$

satisfies

$$\frac{\partial \mu}{\partial t} + U \cdot \nabla \mu + \nabla \cdot E\{uc\} = \kappa \nabla^2 \mu, \tag{5}$$

in (5), $U = U(x, t) = E\{Y(x, t)\}$ is the mean velocity field, and $u = u(x, t)$, $c = c(x, t)$ are the "fluctuations" defined by

$$u = Y - U, \qquad c = \Gamma - \mu . \tag{6}$$

The variance $\sigma^2(x, t)$ of the concentration, often termed the mean-square fluctuation, satisfies

$$\sigma^2(x, t) = \int_o^\infty (\theta - \mu)^2 p(\theta; x, t) d\theta = E\{\Gamma^2(x, t)\} - \mu^2(x, t), \tag{7}$$

and its governing equation is:

$$\frac{\partial \sigma^2}{\partial t} + U \cdot \nabla \sigma^2 + \nabla \cdot E\{uc^2\} + 2 \nabla \mu \cdot E\{uc\} = \kappa \nabla^2 \sigma^2 - 2\kappa E\{(\nabla c)^2\}. \quad (8)$$

Details of the derivation of the standard equations (5) and (8) from (3) are given in Chatwin (1989). Justification for the use of the standard statistical notations μ and σ^2 (rather than symbols using overbars and dashes that are still more conventional in turbulence and turbulent diffusion research) is given by Chatwin (1990).

Except in very rare cases (e.g., Chatwin & Sullivan, 1979), exact results cannot be obtained from (5) and (8), let alone from (3). This is because of the "closure problem" which, in (3) is evident in the last (i.e., second) terms on each side of the equation. The expected values in these terms are defined by equations like (4) and (7), but with the crucial difference that the relevant probability density functions are in neither case $p(\theta; x, t)$, but are more complicated. For example, the last term on the left-hand side of (3) involves the joint probability density function of velocity and concentration. Therefore equation (3) for p is not "closed", and neither, consequently, are the equations for μ and σ^2.

Many attempts have been made to solve, or avoid, the closure problem among the earliest of which are Gaussian plume models, and models using the long-discredited concept of eddy diffusivity. These models, and later ones, such as most of those considered in the papers by Hanna and Klug in this volume, are usually of the mean concentration μ. They do not (except in very rare instances) consider the statistics of the deviations between μ and what is observed, that is of the concentration fluctuations; indeed they are not capable of doing so. This is despite the fact that the magnitude of these deviations is known to be at least of the order of μ itself and sometimes much larger. An interesting development in recent years which does account for the fluctuations is the use of random walk models (see Sullivan, 1971; Allen, 1982). Latest work in this technique is described in papers in this volume by van Dop, by Kaplan & Dinar, and by Sawford. It is now known that such models, as currently structured, have inevitable inconsistencies (Thomson, 1990). While the consequences of these appear to be numerically small, the inconsistencies are fundamental and cannot be removed without a radical change in model structure. Real mastery of the closure problem will probably occur only when a future generation of computers is large enough and fast enough to allow full 3D and unsteady solutions of (1) to be directly calculated to adequate accuracy, which requires, in particular, a satisfactory resolution of all length scales down to the conduction cut-off length ($\approx 10^{-4}m$), and of all time scales. In addition, enough solutions must be generated for each ensemble of velocity fields to permit direct estimation of the statistical properties of $\Gamma(x, t)$ to within acceptable limits which will, of course, require a large enough sample size. What evidence there is (Thomson, 1990) suggests that the number of solutions will need to be at least 10^4 (and perhaps larger than 10^5) for the estimation of statistical properties as simple as $\sigma^2(x, t)$. Such power is unlikely to be available soon.

In a series of papers (Chatwin & Sullivan, 1979, 1980, 1989a, 1990a) we have adopted a different approach, using physical reasoning to extend simple results for idealised cases to real situations. One success of this approach (Chatwin & Sullivan, 1979) was the demonstration that the magnitude of σ^2 depends significantly on source size and geometry, and much more so than μ. Small sources generate large fluctuations. In the present paper we focus on our later results and some of their implications.

3. The Intermittency Factor

A statistical property of $\Gamma(x,t)$ that has not yet been considered but which is prominent in many research papers is the intermittency factor $\gamma = \gamma(x,t)$, conventionally defined by

$$\gamma(x,t) = prob\{\Gamma(x,t) > 0\}\,. \tag{9}$$

(Some workers - and not without linguistic justification - use a complementary definition in which the right-hand side of (9) is 1-γ.) But, because of molecular diffusivity, solutions of the basic equation (1) for Γ have $\Gamma(x,t) > 0$ everywhere for all times after release of the contaminant. It then follows from (9) that γ(x,t)=1 everywhere after release and, therefore, that the definition (9) is theoretically meaningless.

The only logical way in which (9) can be made meaningful is to reject (1) and, in particular, to insist that its replacement does not permit instantaneous diffusion of matter, i.e. diffusion with "infinite velocity". Since molecular velocities are not infinite, such amendments to (1) are physically reasonable (and were, indeed, considered by Russian scientists over 35 years ago). However, there is no experimental evidence whatsoever that (1) is not an entirely satisfactory description of the evolution of $\Gamma(x,t)$ on the continuum scale. (It is pertinent to note that if (1) is to be rejected because it predicts the instantaneous diffusion of matter then so, logically, should be the Navier-Stokes equations because they predict the instantaneous diffusion of vorticity). Rejecting (1) would therefore be too drastic a resolution of the dilemma on present evidence, and this possibility will not be considered further here.

It follows that it is the definition (9) that must be discarded. Before considering replacements, it is important to note that reported values of γ less than 1 (but greater than 0) must occur because of instrumentation characteristics (almost all inevitable) or because of signal processing strategy such as thresholding. In other words, reported values of γ between 0 and 1 have no relevance at all to turbulent diffusion.

However the concept of intermittency of the velocity field is so useful that it is important to seek a new definition of $\gamma(x,t)$ that
(i) represents all relevant properties of the velocity field Y, and
(ii) is meangingful.

In particular, (ii) implies that a new definition of γ should be one that can, in principle at least, be a legitimate goal of mathematical modellers.

Such a definition was proposed by Chatwin & Sullivan (1989a), in a paper on which this section is largely based. It is convenient in what follows to use a zero subscript. e.g. γ_o, μ_o..., to denote properties in a hypothetical ensemble of releases in which the velocity field and geometry are identical to those in the real situation, but in which there is no molecular diffusion i.e. $\kappa = 0$. Suppose that, at $t = 0$, there is a release of contaminant of uniform concentration θ_1. In the hypothetical ensemble of releases there is no molecular diffusion; it follows that $p_o(\theta; x, t)$, the probability density function of concentration in this hypothetical ensemble, must have the form

$$p_o(\theta; x, t) = \gamma_o(x, t)\delta(\theta - \theta_1) + \{1 - \gamma_o(x, t)\}\delta(\theta)\,. \tag{10}$$

Equation (10) indicates that in this hypothetical ensemble the only values of Γ_o that occur are θ_1 and 0; moreover, without molecular diffusion, the logical objection to (9) disappears, so that $\gamma_o = \gamma_o(x, t)$ in (10) is defined by (9) but with Γ_o replacing Γ. Use of (4) and (10) gives

$$\mu_o(x, t) = \theta_1 \gamma_o(x, t) \to \gamma_o(x, t) = \frac{\mu_o(x, t)}{\theta_1}. \tag{11}$$

In (10) and (11), the properties γ_o and μ_o are determined by the velocity field and geometry in the real situation. It is very likely, and commonly supposed, that $\mu(x, t)$, the real mean concentration, is insensitively dependent on κ, i.e. that $\mu(x, t) \approx \mu_o(x, t)$. We therefore propose that the definition (9) be replaced by $\gamma(x, t) = \gamma_o(x, t)$, where $\gamma_o(x, t)$ is as in (11), and that, in practice, γ be estimated from data by

$$\gamma(x, t) \approx \frac{\mu(x, t)}{\theta_1}\,. \tag{12}$$

One of the merits of (12) is that μ and θ_1 are two properties of the real concentration field that are most straightforward to measure reliably.

Because measurements of γ between 0 and 1 that are claimed to be obtained using the conventional definition (9) cannot satisfy (9), and, instead, reflect only characteristics of the instrumentation and signal processing strategy, little, if any, support is provided for the proposed new definition (12) by the fact that most graphs of γ reported in the research literature are at least qualitatively similar to the corresponding graphs of μ, and sometimes very close. Similar remarks apply to the agreement between profiles of γ and μ obtained by numerical simulation - see, for example, Figure 3 of Kaplan and Dinar (1988). Nevertheless the new definition of γ does appear to have the required properties. Further developments are discussed in our cited paper and also in Chatwin & Sullivan (1989b).

4. Relationship Between μ And σ

The hypothetical ensemble, identical to the real one except that κ=0 has been successfully exploited in another way by Chatwin & Sullivan (1990a). Use of (7) and (10) gives

$$\sigma_o^2(x,t) = \theta_1^2\gamma_o(x,t) - \theta_1^2\gamma_o^2(x,t), \tag{13}$$

and elimination of θ_1 from (11) and (13) gives

$$\sigma_o^2 = \mu_o(\theta_1 - \mu_o), \tag{14}$$

for all x and t. As in the previous section, this relationship incorporates the real velocity field completely (except insofar as this enhances the effects of κ in the real case by, for example, stretching).

Because molecular diffusion is a "weak" process compared with advection, we thought that (14) could be adapted to the real case by relatively simple changes. In particular we proposed that for dispersion in self-similar situations, such as jets and wakes (in which all data available to us had been measured), we allowed for molecular diffusion by

- (i) replacing the source (and maximum) concentration θ_1 by $\alpha\mu_*$, where μ_* is the maximum value of μ at any cross section (and so depends on position downstream of the source) and, in view of the application to self-similar flows, α is a constant;
- (ii) introducing a constant of proportionality β.

We therefore proposed the following relationship between σ and μ for the real situation:

$$\sigma^2 = \beta\mu(\alpha\mu_* - \mu). \tag{15}$$

In brief, our reasoning was that the term involving α allowed for the reduction of the maximum concentration by κ, and that the constant β represented the effects of dissipation of σ^2. We also had in mind that instrument smoothing could cause measured values of σ^2 to be less than the real values, and we recognised that this could affect the value of β.

The agreement between (15) and the data available to us was remarkably good; full details are given in Chatwin and Sullivan (1990a). In all cases examined, data from the self-similar dispersion region obeyed (15) to within normal experimental errors, bearing in mind also the uncertainty in the measurements of σ^2 due to statistical noise. With one exception, the value of the constant α was between 1 and 2 (but depended on the particular flow), and it can be shown easily that such values ensure that the maximum of σ^2 occurs at an off-axis location, different from that at which $\mu = \mu_*$. This phenomenon is of course well-known. In the one exception (Nakamura, et al., 1987), the maxima of μ and σ^2 coincided and the value of α

was 3; nevertheless these measurements also satisfied (15). We discuss further measurements from this group later. The values of the constant β also varied from flow to flow but satisfied $0 < \beta \leq 1$ in all cases.

We also discussed extensions of the ideas to other statistical properties (higher moments and the probability density function). Although there were substantially fewer measurements of such properties available to us, the comparisons that were made were reasonably encouraging given, especially, the greatly enhanced statistical noise.

Many workers have been interested in the asymptotic (far downstream of the source) value of σ/μ, the concentrations intensity. Although we are not convinced that this is an important measure from the point of view of basic understanding, it is interesting to record that (15) gives, on the axis,

$$\frac{\sigma}{\mu} = \sqrt{\{\beta(\alpha - 1)\}}, \tag{16}$$

which is a constant whose value varied from 0.1 to (approximate) 1 for the data that we examined.

The success of the comparison of (15) with data from self-similar regimes naturally led to attempts to apply it more generally. Preliminary ideas were discussed in Chatwin et al., (1990), and led to a model (Moseley, 1991; Moseley and Sullivan, 1991) which showed good agreement with data from grid turbulence for all distances dowstream of the grid including those prior to the establishment of the self-similar dispersion regime. Figure 1, taken from Moseley and Sullivan (1991), is typical; further comparisons are given in Moseley (1991).

In brief, the extended model retains (15) but recognizes that α and β cannot be constant throughout the dispersion regime. On the basis of a simple hypothesis - in essence a closure hypothesis - evolution equations for α and β as functions, for example, of downstream distance are obtained and solved. These equations (coupled ordinary differential equations) incorporate source size and geometry (not explicity included in our first model for the self-similar regime) and involve the growth rate of the dispersing contaminant plume. The solutions of these evolution equations tend to the constant values in the first version as downstream distance tends to infinity.

The generic problem in turbulent diffusion is arguably the one arising from instantaneous release of a finite quantity of contaminant, and therefore intrinsically both (statistically) non-stationary and inhomogeneous. In view of the success of the model based on (15), it seems likely that further extensions to these more difficult (but more realistic-in practice at least) situations would be worth attempting. Unfortunately, but understandably, very few measurements are available.

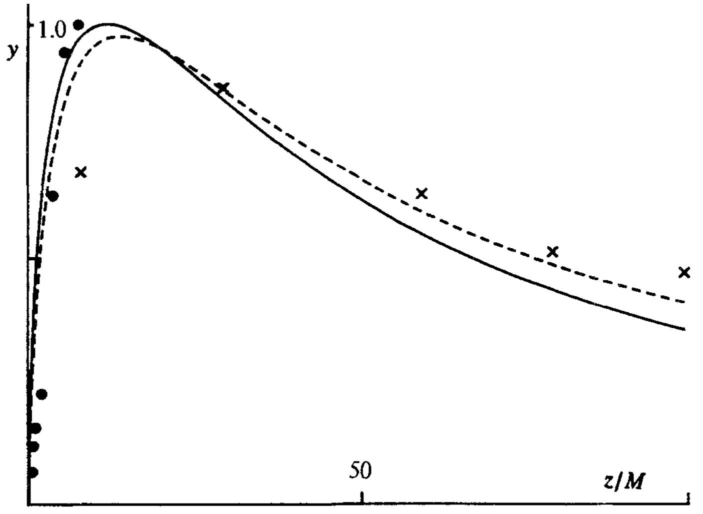

Fig. 1. Comparison of extension of (15) from Moseley & Sullivan (1991) with data of Warhaft (1984) from grid turbulence. The ordinate y is the centre-line value of σ^2 normalized by its absolute maximum, z is downstream distance and M is the grid mesh length. The solid circles and crosses are data from the 0.025mm and 0.127mm sources respectively, and the solid and dashed curves are the corresponding predictions from the extended theory.

5. Some Unanswered Problems

Research in turbulent diffusion is so difficult that any success in achieving increased understanding requires the intimate interplay of theory with experiment; that is one of its most attractive features. But recent developments seem to us to have consequences for research and future research priorities that are more important than seems to have been generally realized.

The three most important such developments are perhaps:

(i) Increasing power and availability of computers.

(ii) Improved measurements techniques.

(iii) Growing public demand for the control and monitoring of air and water quality, including the assessment of potential dangerous accidents involving the release of harmful substances into the atmosphere or natural water bodies.

In themselves these developments are welcome, but that is not necessarily true of their immediate consequences.

The sophistication of present data collection and acquisition system has led to the generation of enormous quantities of data, orders of magnitude greater than some data analysts and theoreticians (including at least one of us!) have been accustomed to. Consequently much data are not being examined or analysed or interpreted to the extent which is merited, or which the experimenter would wish. For us, the implication is not only that data analysts should show more foresight but that, given the inevitable resource limitations, all experiments must now be planned with the data analysis regarded as an integral part. Otherwise much data will continue to be ignored. It is obvious that the experiments should be designed with a clear purpose in mind, particularly the use(s) to which the data analysis will be put by the theoreticians on the project. Less obvious perhaps is the need to be clear about the underlying ensemble; many potential valuable experimental projects have been corrupted, sometimes beyond redemption, by arbitrary changes in the ensemble. One example is provided by investigations on the effects of buildings, where the demands of sponsors have often led to so many (apparently random) changes in the building(s) configuration during the experimental series being made that no quantitative results of value can be obtained for any ensemble. Even when this problem does not arise, it is necessary to assess in advance whether the uncertainty in the estimated statistical properties, due to limited sample size or length of record, will be acceptably small. In many cases of importance, the cost of obtaining such acceptably small uncertainty in full-scale (or field) trials will be too great. It will therefore be necessary to continue to use wind or water tunnels, and the demands of sponsors for "answers" to increasingly more sophisticated questions (such as effects of atmospheric stability or buildings) then require increased research into the capability of these facilities to model full-scale conditions.

Associated with the points above is the treatment of raw data before it is "validated" for transmission to the analysts. Such points as the signal noise and its deconvolution, thresholding strategy (if any), and baseline drift, are so crucial to the interpretation of experiments that they need more attention than has been customary (Mole, 1989, 1990a,b).

One of the key theoretical problems which such experimental phenomena influence is the behaviour of $p(\theta; x, t)$ as $\theta \to 0+$. The theoretical arguments used earlier to discredit the conventional definition (9) of the intermittency factor would seem to suggest $p(\theta; x, t)$ should tend to zero as $\theta \to 0+$, but this is not observed in experiments. An obvious explanation is that noise and the other factors mentioned in the previous paragraph make it extremely difficult, if not impossible, to measure very small concentrations with reliability or discrimination.

An associated, but different, problem connected with instrumentation is that, even with vastly improved modern measurement techniques, it is not likely that the small-scale dynamics of the fluctuating concentration field can be accurately resolved in all three space dimensions and in time. (It was noted earlier that significant dynamics occurs at length scales down to $O(10^{-4}m)$ in the atmosphere). Instrument smoothing is therefore inevitable, but its degree and type will depend on the characteristics of the instrumentation system. Figure 2, due to Sakai et al.,(1990), appears categorically to show that instrument smoothing can be significant. The measured value of the constant β in the relationship (15), but not that of α, is shown to depend strongly on probe size d_o. Details are given in Table I. It will be noted that (15) describes the data well in all four cases but that as d_o decreases (increased resolution), the measured value of β, i.e. of σ^2, increases.

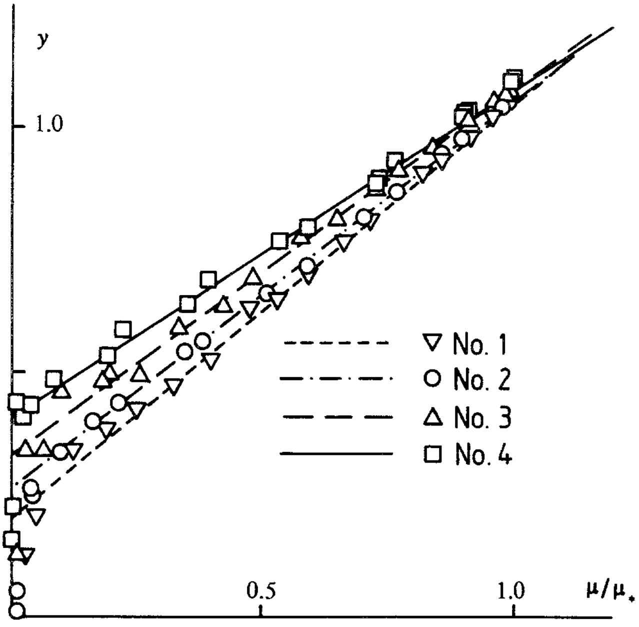

Fig. 2. The influence of probe size on the constants α and β in (15). Data from Sakai et al. (1990). The ordinate y is $(\mu^2 + \sigma^2)(\mu\mu_*)$ and rearrangement of (15) gives $y = \alpha\beta + 1(1 - \beta)(\mu/\mu_*)$. Numerical values of α and β are given in Table 1.

Probe no.	d_o/mm	α	β
1	0.54	1.31	0.16
2	0.30	1.25	0.22
3	0.13	1.33	0.25
4	0.10	1.25	0.33

Although some theoretical investigations of instrument effects have been undertaken by Mole (loc.cit.) and others, we believe that increased understanding of turbulent diffusion in general, and concentration fluctuations in particular, is being severely inhibited because insufficient attention is being placed on them by the research community. As examples, we record our opinion that there is a strong case (supported by data like that in Figure 2) for measurements of concentration fluctuations to be carried out by two (or more) transducer systems operating simultaneously, and that attempts should be made to see whether calibration procedures are valid for the actual experimental conditions.

Except indirectly, we have deliberately not discussed purely theoretical problems in this section.

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