

Chapter 1

TURBULENT DISPERSION: HOW RESULTS FOR THE ZERO MOLECULAR DIFFUSIVITY CASE CAN BE USED IN THE REAL WORLD

NILS MOLE*, PHILIP CHRISTOPHER CHATWIN[†] and
PAUL J. SULLIVAN[‡]

We consider the dispersion of contaminants in turbulent flows at high Péclet number. Except at the smallest scales, molecular diffusion acts slowly by comparison with turbulent advection. Molecular diffusion is still important because it is the only means by which the concentration in a fluid particle can be changed. Exact solution of the full problem, including both turbulent advection and molecular diffusion, is not possible because of the well-known turbulence closure problem. But exact results for moments and the probability density function (pdf) of concentration can be derived for the hypothetical case of zero diffusivity. For high Péclet number these results can be expected to hold in certain ranges of space and time with only slight modification. We outline the results in the absence of molecular diffusion, and consider how the results for the moments can be modified to account for the presence of slowly acting diffusion. The model expressions for the moments involve the mean concentration, and a set of parameters which vary slowly with distance from the source, or with time since release. The corresponding form of the pdf is considered, and results for large concentrations are presented. It is shown that many of the results obtained with this approach agree well with experimental observations. Areas needing further work are also suggested.

Keywords: Concentration Fluctuations; Concentration Moments; Intermittency; Maximum Concentration; Molecular Diffusion; Probability Density Function; Turbulent Dispersion; Generalised Pareto Distribution

*Department of Applied Mathematics, University of Sheffield, Hicks Building, Sheffield S3 7RH, U.K., Email: n.mole@sheffield.ac.uk

[†]Department of Applied Mathematics, University of Sheffield, Hicks Building, Sheffield S3 7RH, U.K., Email: p.chatwin@sheffield.ac.uk

[‡]Department of Applied Mathematics, The University of Western Ontario, London N6A 5B7, Ontario, Canada, Email: pjsul@uwo.ca

1. Introduction

We present a review of a novel approach to the modelling of turbulent dispersion which has been developed over the last twenty years or so. Rather than attempting to solve the Navier-Stokes and advection-diffusion equations for the full problem, or applying closures directly to those equations, this approach involves deriving exact results for the hypothetical case of no molecular diffusion, and then making physical arguments for how these results should be modified in the presence of diffusion.

Two fundamental physical processes govern the dispersion of a passive conserved scalar in a turbulent flow: advection by the turbulent velocity field, and molecular diffusion. The relative importance of these processes can be roughly quantified using the Péclet number $Pe = ul/\kappa$, where u and l are appropriate velocity and length scales for the turbulent fluctuations, and κ is the molecular diffusivity. In most environmental and engineering applications the Péclet number is very large^a. In such cases turbulent advection acts on a much shorter timescale than molecular diffusion, the ratio of the timescales being given by Pe^{-1} if the length scales for the velocity and concentration fields are comparable. Advection acts to stretch the scalar cloud or plume out into thin sheets and strands [4], as observed experimentally [2, 5–7]. Molecular diffusion is a much slower process, but it is nevertheless an important one, since it is the only means by which the scalar concentration in a fluid particle can be altered, and it limits the smallest scales which can be present in the scalar field. For Schmidt number ν/κ of order 1 or greater, where ν is the kinematic viscosity, this smallest scale is of the order of the conduction cut-off length $\lambda_c = (\nu\kappa^2/\epsilon)^{1/4}$ [8], where ϵ is the turbulent energy dissipation rate per unit mass. Molecular diffusion also has the effect of dissipating the variance and higher moments of the concentration [8–10].

Nevertheless, it seems likely that at high Pe , and for times t and positions \mathbf{x} not too far from the source values, the effect of molecular diffusion can be described by appropriate modification of the results when there is

^aFor example, for heat, water vapour, carbon monoxide, methane or propane dispersing in air, κ is of order $10^{-5} \text{ m}^2\text{s}^{-1}$ [1–3], so even values as small as $u = 0.1 \text{ ms}^{-1}$ and $l = 0.1 \text{ m}$ would give Pe of order 10^3 . For salt, methane, ethanol, toluene or urea dispersing in water, κ is of order $10^{-9} \text{ m}^2\text{s}^{-1}$, and for heat dispersing in water it is of order $10^{-7} \text{ m}^2\text{s}^{-1}$. So even for scales as small as $u = 10^{-3} \text{ ms}^{-1}$ and $l = 0.1 \text{ m}$ we would have Pe of order 10^5 and 10^3 , respectively. In many applications the typical velocity and length scales would be larger than the values given here, leading to even larger values of Pe .

no molecular diffusion. This approach was initiated in [11], and taken further in many papers, especially [12–14]. This article is an appraisal of the present position.

In Sec 2 we derive results for the moments and probability density function (pdf) of concentration for the case of no molecular diffusion. In Sec 3 we show how the results for the moments can be modified to include the effects of diffusion, and in Sec 4 we discuss the implications for the pdf, including its behaviour at high concentrations. Finally, in Sec 5, we discuss the successful aspects of this approach, and suggest areas which require further work.

2. The case of no molecular diffusion

2.1. Uniform source

In [11] the following results for the case of no molecular diffusion and a uniform concentration source were derived. The only possible concentrations are then zero and the source concentration θ_1 . The pdf, $p(\theta; \mathbf{x}, t)$, of concentration $\Gamma(\mathbf{x}, t)$ is defined by

$$p(\theta; \mathbf{x}, t) = \frac{d}{d\theta} \text{Prob}(\Gamma(\mathbf{x}, t) \leq \theta),$$

and can then be written as

$$p(\theta) = (1 - \pi)\delta(\theta) + \pi\delta(\theta - \theta_1), \quad (1)$$

where $\pi(\mathbf{x}, t)$ is the probability that (\mathbf{x}, t) is in fluid which originated in the source, and δ is the Dirac delta-function. Note that $\pi(\mathbf{x}, t)$ is completely determined by the statistical properties of the turbulent velocity field.

The mean concentration $C(\mathbf{x}, t) = E\{\Gamma(\mathbf{x}, t)\}$, where $E\{\cdot\}$ denotes the expected value, or ensemble mean, is then given by

$$C = (1 - \pi) \int \theta \delta(\theta) d\theta + \pi \int \theta \delta(\theta - \theta_1) d\theta = \pi\theta_1, \quad (2)$$

and the n th central moment

$$\mu_n(\mathbf{x}, t) = E\{[\Gamma(\mathbf{x}, t) - C(\mathbf{x}, t)]^n\}$$

is given by

$$\begin{aligned} \mu_n &= (1 - \pi) \int (\theta - C)^n \delta(\theta) d\theta + \pi \int (\theta - C)^n \delta(\theta - \theta_1) d\theta \\ &= (1 - \pi)(-C)^n + \pi(\theta_1 - C)^n. \end{aligned}$$

This can be rewritten, using Eq. (2), as

$$\frac{\mu_n}{\theta_1^n} = \frac{C}{\theta_1} \left(1 - \frac{C}{\theta_1}\right)^n + (-1)^n \left(1 - \frac{C}{\theta_1}\right) \left(\frac{C}{\theta_1}\right)^n. \quad (3)$$

2.2. Non-uniform source

We now consider the more general case of random, non-uniform, source concentration. [15] included random, but stationary and homogeneous, source concentration, while [12] derived results for a non-random, spatially varying instantaneous source. Here we generalise to the case of random source concentration whose distribution may vary in space and time. A smokestack is a practical example of a case where the concentration of effluent at the source will be a random function of spatial position (within the stack exit cross-section) and of time. The development given below follows that of [12], but is applied to these more general source conditions.

Let the source concentration be $\Gamma_S(\mathbf{x}, t)$, with Γ_S random, and let the probability that fluid at (\mathbf{x}, t) came from a volume-time element $dV(\mathbf{y}) d\tau$ about (\mathbf{y}, τ) be $P(\mathbf{y}, \tau; \mathbf{x}, t) dV(\mathbf{y}) d\tau$. Then the pdf of $\Gamma(\mathbf{x}, t)$ can be written as

$$p(\theta; \mathbf{x}, t) = \int_{a.s.t.} p_S(\theta; \mathbf{y}, \tau) P(\mathbf{y}, \tau; \mathbf{x}, t) dV(\mathbf{y}) d\tau,$$

where *a.s.t.* denotes that the integration takes place over all space and for all times satisfying $\tau \leq t$, and $p_S(\theta)$ is the pdf of Γ_S . This is essentially equation (10.5) of [16], applied to the pdf of concentration rather than to the concentration itself.

We assume that the source occupies a finite volume V_0 . Then

$$\begin{aligned} p(\theta; \mathbf{x}, t) &= \int_{\mathbf{y} \notin V_0, \tau \leq t} \delta(\theta) P(\mathbf{y}, \tau; \mathbf{x}, t) dV(\mathbf{y}) d\tau \\ &\quad + \int_{\mathbf{y} \in V_0, \tau \leq t} p_S(\theta; \mathbf{y}, \tau) P(\mathbf{y}, \tau; \mathbf{x}, t) dV(\mathbf{y}) d\tau \\ &= [1 - \pi(\mathbf{x}, t)] \delta(\theta) + \int_{\mathbf{y} \in V_0, \tau \leq t} p_S(\theta; \mathbf{y}, \tau) P(\mathbf{y}, \tau; \mathbf{x}, t) dV(\mathbf{y}) d\tau, \end{aligned} \quad (4)$$

where

$$\pi(\mathbf{x}, t) = \int_{\mathbf{y} \in V_0, \tau \leq t} P(\mathbf{y}, \tau; \mathbf{x}, t) dV(\mathbf{y}) d\tau \quad (5)$$

is the probability that fluid at (\mathbf{x}, t) came from the source.

Far from the source, the initial location of a fluid particle within the source is “forgotten”. If we take the origin to be within V_0 , we then have

$$P(\mathbf{y}, \tau; \mathbf{x}, t) \approx P(\mathbf{0}, \tau; \mathbf{x}, t)$$

for $\mathbf{y} \in V_0$, and

$$\pi(\mathbf{x}, t) \approx V_0 \int_{\tau \leq t} P(\mathbf{0}, \tau; \mathbf{x}, t) d\tau. \quad (6)$$

This gives

$$p(\theta; \mathbf{x}, t) \approx [1 - \pi(\mathbf{x}, t)] \delta(\theta) + \int_{\tau \leq t} d\tau P(\mathbf{0}, \tau; \mathbf{x}, t) \int_{\mathbf{y} \in V_0} dV(\mathbf{y}) p_S(\theta; \mathbf{y}, \tau). \quad (7)$$

If $p_S(\theta; \mathbf{y}, \tau)$ does not depend on τ then the time and space integrals decouple, and we are able to derive a result analogous to Eq. (1). This is the case if either (a) the source concentration distribution is stationary, or (b) the release is instantaneous. In both of these cases we find, using Eq. (6), that

$$p(\theta; \mathbf{x}, t) \approx [1 - \pi(\mathbf{x}, t)] \delta(\theta) + \frac{\pi(\mathbf{x}, t)}{V_0} \int_{\mathbf{y} \in V_0} dV(\mathbf{y}) p_S(\theta; \mathbf{y}). \quad (8)$$

In this formulation, all (\mathbf{x}, t) dependence is accounted for by the variation of $\pi(\mathbf{x}, t)$. Thus, for a wide class of source conditions, very simple results can be derived in the zero molecular diffusivity case.

For a uniform source, $p_S(\theta; \mathbf{y}) = \delta(\theta - \theta_1)$ and Eq. (1) is recovered from Eq. (8). It can also be shown that Eq. (8) is consistent with results in [12, 15] – for details see Appendix A.

Here we consider the case of a random source concentration $\Gamma_S(\mathbf{x}, t)$ which varies in space and time, but has a pdf p_S which does not depend on time, so that Eq. (8) applies. The absolute moments m_n of Γ therefore satisfy

$$m_n(\mathbf{x}, t) = \int \theta^n p(\theta; \mathbf{x}, t) d\theta = \frac{\pi(\mathbf{x}, t)}{V_0} \int_{\mathbf{y} \in V_0} m_{S_n}(\mathbf{y}) dV(\mathbf{y}), \quad (9)$$

where

$$m_{S_n}(\mathbf{x}) = \int \theta^n p_S(\theta; \mathbf{x}) d\theta$$

is the n th absolute moment of $\Gamma_S(\mathbf{x}, t)$. If we let the mean source concentration be $\theta_1(\mathbf{x}) = m_{S1}(\mathbf{x})$, then the mean concentration $C(\mathbf{x}, t)$ is given by

$$C(\mathbf{x}, t) = m_1(\mathbf{x}, t) = \frac{\pi(\mathbf{x}, t)}{V_0} \int_{\mathbf{y} \in V_0} \theta_1(\mathbf{y}) \, dV(\mathbf{y}). \quad (10)$$

Define the constant concentration scale θ_0 by

$$\theta_0 = \frac{\int_{\mathbf{y} \in V_0} m_{S2}(\mathbf{y}) \, dV(\mathbf{y})}{\int_{\mathbf{y} \in V_0} \theta_1(\mathbf{y}) \, dV(\mathbf{y})}. \quad (11)$$

Then the central moments $\mu_n = \mu_n(\mathbf{x}, t)$ for $n \geq 2$ can easily be shown to be given by

$$\begin{cases} \mu_2(\mathbf{x}, t) = C(\mathbf{x}, t) \{ \theta_0 - C(\mathbf{x}, t) \} \\ \mu_3(\mathbf{x}, t) = C(\mathbf{x}, t) \{ \lambda_3^2 \theta_0^2 - 3\theta_0 C(\mathbf{x}, t) + 2C(\mathbf{x}, t)^2 \} \\ \mu_4(\mathbf{x}, t) = C(\mathbf{x}, t) \{ \lambda_4^3 \theta_0^3 - 4\lambda_3^2 \theta_0^2 C(\mathbf{x}, t) + 6\theta_0 C(\mathbf{x}, t)^2 - 3C(\mathbf{x}, t)^3 \} \\ \vdots \end{cases} \quad (12)$$

where the non-dimensional constants λ_n are defined for $n \geq 3$ by

$$(\lambda_n \theta_0)^{n-1} = \frac{\int_{\mathbf{y} \in V_0} m_{Sn}(\mathbf{y}) \, dV(\mathbf{y})}{\int_{\mathbf{y} \in V_0} \theta_1(\mathbf{y}) \, dV(\mathbf{y})}. \quad (13)$$

(Note that Eq. (11) and Eq. (13) for $n = 2$ are consistent with taking $\lambda_2 = 1$.) Apart from trivial changes of notation, Eq. (12) is the same as equation (10) of [12], and Eq. (13) is the generalisation of equation (11) of [12].

3. The effects of molecular diffusion

For the real case with molecular diffusion, we consider the analogue of Eq. (1) and Eq. (4). Suppose we let $f_s(\theta; \mathbf{x}, t)$ be the pdf of $\Gamma(\mathbf{x}, t)$ conditional on being in fluid that came from the source, and $f_a(\theta; \mathbf{x}, t)$ be the pdf of $\Gamma(\mathbf{x}, t)$ conditional on being in ambient fluid that did not come from

the source. As outlined by [15], it follows from the law of total probability that for any source the pdf of concentration can then be written as

$$p(\theta; \mathbf{x}, t) = \pi(\mathbf{x}, t)f_s(\theta; \mathbf{x}, t) + \{1 - \pi(\mathbf{x}, t)\} f_a(\theta; \mathbf{x}, t), \quad (14)$$

where $\pi(\mathbf{x}, t)$, as before, is the probability that fluid at (\mathbf{x}, t) came from the source. This probability is not affected by molecular diffusion, but depends only on the velocity field, so Eq. (5) still applies.

3.1. Intermittency

In turbulent dispersion applications the concept of intermittency is used by many authors^b. The aim is to characterise the contrast between concentrations close to zero (found mainly in ambient, non-source, fluid) and relatively large concentrations (which can often be many standard deviations larger than the mean) found mainly in fluid originating from the source. Such authors usually define intermittency to be the probability of non-zero concentration (see, for example, [17]). There are two problems associated with this definition. Firstly, [15] pointed out that the solution of the advection-diffusion equation for Γ will have non-zero values for all \mathbf{x} for all times after release, so this definition of intermittency would strictly give a value of 1 everywhere. Secondly, small measured concentrations will be contaminated with noise. The most usual methods for dealing with this have been the use of a threshold, or the fitting of a Gaussian distribution to small concentrations. The measured value of intermittency depends strongly on the arbitrary choice of threshold, or on the method chosen to fit the Gaussian.

[15] proposed instead that intermittency be defined as $\pi(\mathbf{x}, t)$, the probability that fluid at (\mathbf{x}, t) comes from the source. Intermittency is then well-defined, and conveys valuable information about the effect of the velocity field. To measure π experimentally one can make the reasonable assumption that the mean concentration is little affected by molecular diffusion. Eq. (10) then gives

$$\pi(\mathbf{x}, t) \approx \frac{V_0 C(\mathbf{x}, t)}{\int_{\mathbf{y} \in V_0} \theta_1(\mathbf{y}) dV(\mathbf{y})}. \quad (15)$$

^bIn the theory of turbulence itself, “intermittency” is used in a different sense, to denote the spatial patchiness of dissipation.

This is consistent with equation (15) of [12], which applies to a homogeneous source for which $\theta_1(\mathbf{x})$ is a constant. A practical advantage of this (albeit approximate) method of estimating the intermittency $\pi(\mathbf{x}, t)$ is that it will be little affected by measurement errors associated with noise and/or lack of resolution.

3.2. Concentration moments

[11] argued that, since molecular diffusion is a slow process, the structure of the moments will be close to that in the absence of molecular diffusion. They proposed that for self-similar dispersion in stationary turbulent flows, downwind of a uniform source Eq. (3) could be modified in two ways to take account of the effect of molecular diffusion.

Firstly, they replaced the constant source concentration θ_1 by a representative local value αC_0 , where α is a constant and C_0 is the mean concentration on the plume centreline for a steady release (or at the cloud centre for an instantaneous release [12]). Secondly, constants of proportionality were introduced to take account of dissipation of concentration moments and of the increased background concentration resulting from diffusion out of the sheets and strands of source fluid with high concentrations:

$$\frac{\mu_n}{(\alpha C_0)^n} = \beta^n \left\{ \hat{C}(1 - \hat{C})^n + (-1)^n(1 - \hat{C})\hat{C}^n \right\}, \quad (16)$$

where

$$\hat{C} = \frac{C}{\alpha C_0}.$$

(Note that [11] used $\beta^{1/2}$ where we use β in Eq. (16), *i.e.* we follow the more convenient usage of [12].) Because of dissipation, β would be expected to be less than or equal to 1 and, to avoid negative variance, we need $\alpha \geq 1$. [11] found that observations from a number of experiments could be fitted well by Eq. (16) with a constant value of α , and β approximately constant in the crosswind direction.

Normalised moments, in particular the skewness K_3 , kurtosis K_4 and higher order equivalents K_n , defined by

$$K_n = \frac{\mu_n}{\mu_2^{n/2}} \quad \text{for } n = 3, 4, \dots, \quad (17)$$

are useful in many contexts. [13] showed that Eq. (16) implies that

$$\begin{cases} K_4 = K_3^2 + 1 \\ K_5 = K_3^3 + 2K_3, \end{cases} \quad (18)$$

and gave a general expression for K_n as a function of K_3 . Analysis of experimental data from a steady release close to the ground in the field suggested that Eq. (18) ought to be replaced by

$$\begin{cases} K_4 = a_4 K_3^2 + b_4 \\ K_5 = a_5 K_3^3 + b_5 K_3, \end{cases} \quad (19)$$

where a_4 , b_4 , a_5 and b_5 are constants. Eq. (19) has been shown to be a good approximation in a variety of experiments, including plumes in atmospheric boundary layers under various stability classes [13, 18], clouds in the wind tunnel with varying density and with different forms of fence [19], and plumes in wind tunnels [20, 21]. Figure 1 shows an example for the steady line source, grid turbulence, releases of [22], which were analysed further by [12].

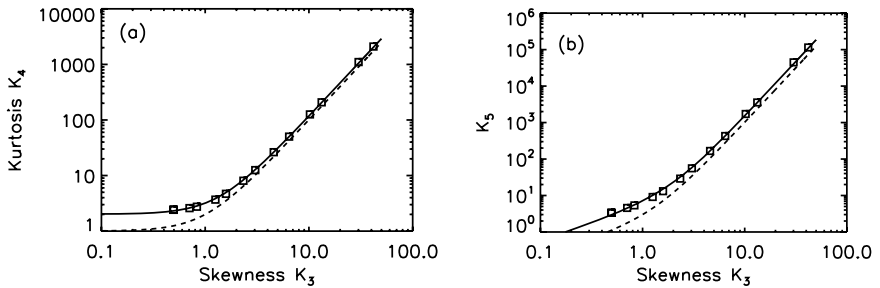


Fig. 1. An example of the fit of Eq. (19) to data from the steady line source, grid turbulence, releases of [22] at downwind distance 50 mm from the source. The fitted values are $a_4 = 1.161$, $b_4 = 2.025$, $a_5 = 1.485$, and $b_5 = 5.647$. The squares are the measured values, and the solid curves are the fits of Eq. (19). (a) Kurtosis K_4 against skewness K_3 , (b) K_5 against K_3 . The dashed curves show the relationships given by Eq. (18).

[12] argued that Eq. (16) would describe the lateral moment structure, with α and β varying with time for an instantaneous release, or with downwind distance for a steady release. For the case of non-uniform source concentration they applied the same argument that led to Eq. (12), to

obtain

$$\begin{cases} \frac{\mu_2}{(\alpha\beta C_0)^2} = \hat{C}(1 - \hat{C}) \\ \frac{\mu_3}{(\alpha\beta C_0)^3} = \hat{C}(\lambda_3^2 - 3\hat{C} + 2\hat{C}^2) \\ \frac{\mu_4}{(\alpha\beta C_0)^4} = \hat{C}(\lambda_4^3 - 4\lambda_3^2\hat{C} + 6\hat{C}^2 - 3\hat{C}^3) \\ \vdots \end{cases} \quad (20)$$

for some parameters $\lambda_3, \lambda_4, \dots$. If $\lambda_n = 1$ for all n then Eq. (20) reduces to the uniform source case Eq. (16). Since we expect a non-uniform source to increase the spread of concentration values by comparison with a uniform source, and since $\hat{C} \leq 1$, we expect $\lambda_n \geq 1$ in general. This is supported by the fitted values given in Table 1 of [21]. The λ_n would be expected to vary in time for an instantaneous release, or with downwind distance for a steady release, in the same way as for α and β . [12] fitted these parameters to data from a steady line source experiment in wind tunnel grid turbulence [22], finding that they varied very slowly with downwind distance (see Table 1 of [21]). We would also expect the parameters a_4, b_4, a_5, b_5 and higher order equivalents to be approximately constant in the crosswind direction, but to vary slowly with time or downwind distance. This has been confirmed for the experiments of [22] by [20, 21]. [20] also showed that the expression for K_4 in Eq. (19) followed approximately from Eq. (20). The relationships between the moments proposed by [11, 12] have been found to agree reasonably well with measurements from a range of experiments, including jets, wakes, plumes, uniformly sheared flow and buoyant jets [11, 12, 23, 24]. Figure 2 shows an example from the steady line source releases of [22].

[14] argued that β is mainly a measure of the dissipation accomplished by molecular diffusion, so that β tends to zero in the limit of large diffusion time. Therefore β would not be expected to be constant across the whole plume cross-section. Any source material arriving far from the centreline will have taken a long time to get there, because of the large distance involved. So it will have been acted on by diffusion for a long time, implying that β tends to zero far from the centreline. But the distance from the centreline at which β becomes small compared with its centreline value will be much greater than the downwind distance from the source and, hence, the plume width, which is the distance at which C becomes small. Where C is very small, it is usually difficult to obtain reliable measurements, so we

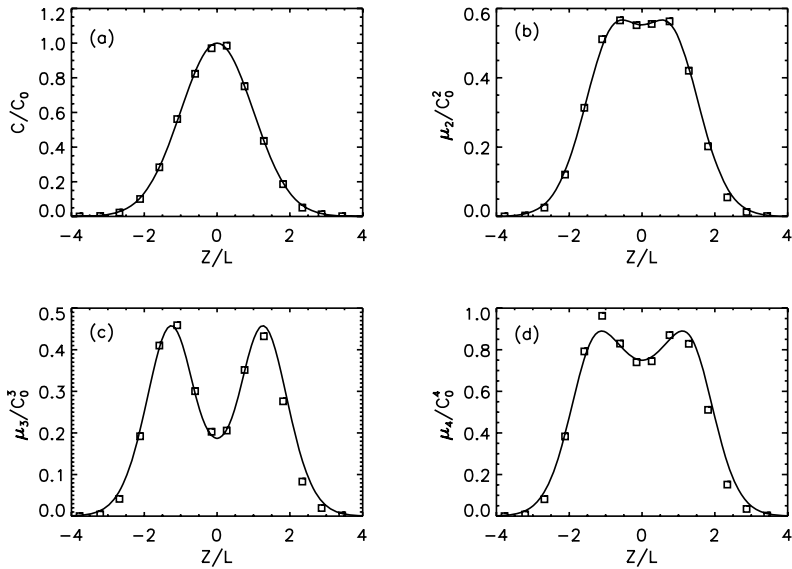


Fig. 2. Fits of Eq. (20) to data from [22] at 50 mm downwind of the source. The fitted values are $\alpha = 1.729$, $\beta = 0.871$, $\lambda_3^2 = 1.161$ and $\lambda_4^3 = 1.511$. The horizontal axis is the distance Z from the mean plume centreline divided by the mean plume width L . The squares are the measurements, and the curves are the fits. (a) Gaussian fit to C/C_0 , (b) Second moment μ_2/C_0^2 , (c) Third moment μ_3/C_0^3 , (d) Fourth moment μ_4/C_0^4 .

expect *measured* values of β to be approximately constant in the crosswind direction over the range of distances for which measurements are usually made, consistent with the results described above.

The successful agreement of Eq. (19) and Eq. (20) with data from a wide variety of experiments encourages us to develop the argument further, especially to the pdf itself.

4. The probability density function of concentration

4.1. The overall pdf

The pdf is the principal function describing the distribution of concentration in turbulent diffusion. This fact is now, but belatedly, well recognised (and there is also recognition that, in itself, the mean concentration is of little value, theoretically and practically). The pdf is also practically important; for example, if the lower and upper flammable limits of a flammable gas

are θ_L and θ_U , the probability that the mixture at (\mathbf{x}, t) is flammable is

$$\int_{\theta_L}^{\theta_U} p(\theta; \mathbf{x}, t) d\theta.$$

Many other examples can be given.

For a uniform source the exact pdf of concentration in the absence of molecular diffusion is given by Eq. (1), and consists of delta-functions located at zero concentration (corresponding to non-source fluid), and at the source concentration θ_1 . The corresponding model moments when diffusion is included, given by Eq. (16), satisfy the first of Eq. (18). But the theory of probability distributions shows that this implies that the pdf must consist of two delta-functions (see, for example, the appendix of [13]). [24, 25] showed that this pdf, with moments given by Eq. (16), is

$$p(\theta) = (1 - \hat{C})\delta(\theta - \theta_{\min}) + \hat{C}\delta(\theta - \theta_{\max}), \quad (21)$$

where

$$\theta_{\min} = (1 - \beta)C \geq 0 \quad \text{and} \quad \theta_{\max} = (1 - \beta)C + \alpha\beta C_0 \leq \alpha C_0, \quad (22)$$

with equality in both cases when $\beta = 1$, which corresponds to being at the source, where we also have $\alpha = 1$ and $C_0 = \theta_1$. In the absence of diffusion, $\alpha C_0 = \theta_1$, $\hat{C} = C/\theta_1 = \pi$ and $\beta = 1$, so Eq. (1) is recovered. (It takes some time for the cumulative effects of molecular diffusion to become important, so at small times after release, or at small downwind distances from the source, the $\kappa = 0$ results will still give a good approximation.)

This model captures the effect of molecular diffusion in reducing the largest concentrations and increasing the smallest concentrations. Since it only has two possible concentrations, θ_{\min} and θ_{\max} , it fails to represent the continuous distribution of concentrations in real turbulent dispersion. However, Eq. (21) can give a good indication of the structure of the real pdf. [24, 25] analysed experimental data for line and point sources in grid turbulence, and for a buoyant jet in a boundary-layer cross flow, and showed that in cases when the pdf had 2 peaks, the location and height of the peaks corresponded quite well to the location and size of the delta-functions in Eq. (21).

If we non-dimensionalise the concentration by $\hat{\Gamma} = \Gamma/(\alpha\beta C_0)$ then we can rewrite Eq. (21) for a uniform source as

$$\hat{p}(\hat{\theta}) = (1 - \hat{C})\delta(\hat{\theta} - D) + \hat{C}\delta(\hat{\theta} - D - 1), \quad (23)$$

where \hat{p} is the pdf of $\hat{\Gamma}$ and

$$D = \left(\frac{1}{\beta} - 1 \right) \hat{C}.$$

For a non-uniform source we can write the pdf corresponding to the moments Eq. (20), and analogous to Eq. (23), in the form

$$\hat{p}(\hat{\theta}) = \hat{p}_D(\hat{\theta} - D),$$

where $\hat{p}_D(\hat{\theta})$ is a pdf with mean \hat{C} , second absolute moment \hat{C} , and n th absolute moment $\lambda_n^{n-1} \hat{C}$ for $n = 3, 4, \dots$ (these results are a consequence of the choice of θ_0 in Eq. (11)). Details of the derivation of these results are given in Appendix B. At first sight it might appear that \hat{p}_D should be given by the appropriately normalised version of p in Eq. (8). In that case the normalisation $\hat{\theta} = \theta/\theta_0$ and $\hat{C} = C/\theta_0$ gives first and second absolute moments equal to \hat{C} , and higher absolute moments equal to $\lambda_n^{n-1} \hat{C}$. But this is inconsistent with our present normalisation, which is $\hat{\theta} = \theta/(\alpha\beta C_0)$ and $\hat{C} = C/(\alpha C_0)$. This means that, in general, \hat{p}_D cannot be obtained from Eq. (8).

If $\lambda_n = 1$ for all n , then $\hat{p}_D(\hat{\theta} - D) = (1 - \hat{C})\delta(\hat{\theta} - D) + \hat{C}\delta(\hat{\theta} - D - 1)$ and Eq. (23) is recovered. Given the dependence of the moments of \hat{p}_D on \hat{C} , it would be natural to look for a general solution in the form $\hat{p}_D(\hat{\theta} - D) = (1 - \hat{C})\delta(\hat{\theta} - D) + \hat{C}\hat{f}(\hat{\theta} - D)$ for some pdf \hat{f} , so that \hat{C} was scaled out of the moments. Thus \hat{f} would have first and second absolute moments equal to 1, and higher moments equal to λ_n^{n-1} . However, this gives zero variance for \hat{f} , so \hat{f} would have to be a single delta-function, and this form is only possible if $\lambda_n = 1$ for all n , i.e. the uniform source case. The fitted values of λ_n in [12, 21] are close to 1, so we would expect \hat{p}_D to be bimodal, with molecular diffusion having the effect of smoothing out the delta-functions of the no-diffusion case.

4.2. Large concentrations

For a general, non-uniform, source, while we are unable to produce a simple exact closed form expression for \hat{p}_D , and hence for \hat{p} or p , there are some questions of practical interest for which we do not need this. One such is applications to toxic and malodorous gases where we want to know the distribution of large concentration values, but we do not necessarily need to know the form of the distribution for smaller concentrations.

Statistical extreme value theory provides a framework for analysing the distribution of large concentration values. Of particular use is the work

of [26] on values above a high threshold. Let $P_c(\theta; \theta_T)$ be the distribution function of the concentration Γ , conditional on Γ being greater than a threshold θ_T , i.e.

$$P_c(\theta; \theta_T) = \text{Prob}(\Gamma < \theta | \Gamma > \theta_T) = \frac{\int_{\theta_T}^{\theta} p(\phi) d\phi}{1 - \int_0^{\theta_T} p(\phi) d\phi} \quad \text{for } \theta \geq \theta_T.$$

The pdf $p_c(\theta; \theta_T)$ of Γ , conditional on $\Gamma \geq \theta_T$, is then

$$p_c(\theta; \theta_T) = \frac{d}{d\theta} P_c(\theta; \theta_T) = \frac{p(\theta)}{1 - \int_0^{\theta_T} p(\phi) d\phi} \quad \text{for } \theta \geq \theta_T.$$

For large θ_T , [26] showed that, subject to some conditions which are satisfied in most cases,

$$p_c(\theta; \theta_T) \approx g(\theta - \theta_T; k, a), \quad (24)$$

where $g(\theta; k, a)$ is the pdf of the generalised Pareto distribution (GPD) and is given by

$$g(\theta; k, a) = \frac{1}{a} \left(1 - \frac{k\theta}{a} \right)^{1/k-1}. \quad (25)$$

Here k is the shape parameter and $a (> 0)$ is the scale parameter. In the present application the maximum possible concentration θ_{\max} is finite (less than or equal to the largest possible source concentration), in which case we expect $k > 0$ so that $g(\theta; k, a)$ has a finite upper endpoint a/k . We then have

$$\theta_{\max} = \theta_T + \frac{a}{k}. \quad (26)$$

It is straightforward to show from Eq. (25) that increasing the threshold θ_T does not change the form of the asymptotic distribution nor the value of the shape parameter k , but does change the value of the scale parameter a . Eq. (26) shows that this change can be calculated by $\Delta a = -k\Delta\theta_T$, where Δa and $\Delta\theta_T$ are the changes in a and θ_T . Thus a higher threshold gives a smaller value of the scale parameter.

The standard statistical approach is to fit Eq. (25) to excesses over a high threshold, using maximum likelihood (for examples of applications to turbulent dispersion see [27–30]). This does not, however, lend itself to modelling based on expressions for concentration moments, like Eq. (20). [14] presented an alternative method, which allows k , a and, hence, θ_{\max} to be derived from the moments. In [14] the overall pdf was expressed as

$$p(\theta) = (1 - \eta)f(\theta) + \eta g(\theta; k, a),$$

for some function f and parameter $\eta (> 0)$, with f assumed to make a negligible contribution for large θ . The upper endpoint of the distribution given by $p(\theta)$ is then a/k , so

$$\theta_{\max} = \frac{a}{k}.$$

If $p(\theta) \approx \eta g(\theta; k, a)$ for all θ above some threshold θ_T , then it is straightforward to show that

$$p_c(\theta; \theta_T) \approx \frac{\left(1 - \frac{k\theta}{a}\right)^{1/k-1}}{a \left(1 - \frac{k\theta_T}{a}\right)^{1/k}} = g(\theta - \theta_T; k, a - k\theta_T) \quad \text{for } \theta \geq \theta_T,$$

consistent with Eq. (24). [14] found for a line source experiment that the upper 50% or more of the concentration range is well-approximated by the GPD.

For large n , the absolute moment m_n could be expected to be dominated by the contribution from large concentrations, so that

$$m_n \approx \eta \int_0^{\theta_{\max}} \theta^n g(\theta; k, a) d\theta. \tag{27}$$

[14] showed that this implied that, for sufficiently large n ,

$$\frac{m_{n-1}}{m_n} \approx \frac{1}{a} \left(\frac{1}{n}\right) + \frac{k}{a}. \tag{28}$$

Thus, the parameters k and a can be identified from the linear relationship between the ratio of successive moments, m_{n-1}/m_n , and $1/n$. This method shares with the standard statistical approach the advantage that the fits are hardly affected by baseline and/or noise problems at low measured concentrations. It also has the advantage that it can be used when only moments are known, as in the model described in Sec. 3.2. Figure 3 shows examples of some fits of the GPD to concentration tails, carried out in this way, for the data of [22].

[14] also considered the behaviour in Eq. (20) far from the plume centreline, for a steady line source release (the same arguments would also apply for a steady point source). They deduced that

$$\lambda_n^{n-1} = a_n \lambda_3^{2(n-2)}, \tag{29}$$

where the a_n are defined by Eq. (19). By combining this result with Eq. (28) far from the centreline, they derived the relationship

$$\frac{a_{n-1}}{a_n} = \frac{1}{r} \left\{ 1 - \frac{20}{n} \left(\frac{a_4^2 - a_5}{5a_4^2 - 4a_5} \right) \right\}, \tag{30}$$

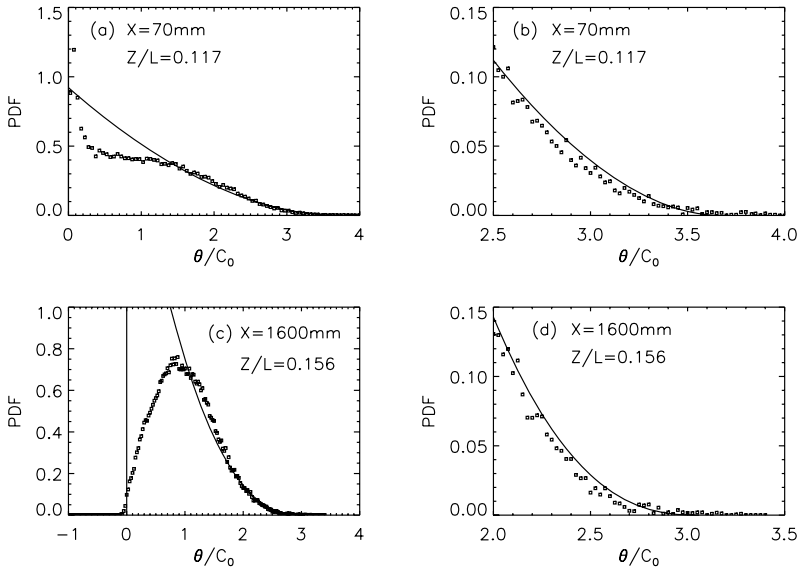


Fig. 3. Examples of the measured pdf of θ/C_0 (points), and GPD (curves) for fitted k and a values, for the data of [22]. The right-hand panels show blow-ups of the tails. X is the downwind distance from the source.

where

$$r = \frac{a_4 a_5}{5a_4^2 - 4a_5},$$

and also showed that

$$\frac{\theta_{\max}}{C_0} \approx \alpha \left\{ \beta \lambda_3^2 r + (1 - \beta) \hat{C} \right\} = \alpha \beta (\lambda_3^2 r + D). \tag{31}$$

Note that for a uniform source we have $\lambda_n = 1$ and $a_n = 1$ so that, as desired, Eq. (31) reduces to Eq. (22). Equation (31) has strictly only been shown to be valid far from the centreline, *i.e.* as $\hat{C} \rightarrow 0$, but since α , β , λ_3 , a_4 and a_5 are expected to be roughly constant over the distances over which \hat{C} becomes small, it seems likely that Eq. (31) can also be applied near the centreline. The results of [14] support this.

Equation (31) suggests that as we go very far from the centreline, ultimately $\theta_{\max}/C_0 \rightarrow 0$, since we expect that $\beta \rightarrow 0$ in that limit. Since, as noted earlier, $\beta \rightarrow 0$ more slowly than \hat{C} , being controlled by diffusion rather than advection, the first term in Eq. (31) will give the leading or-

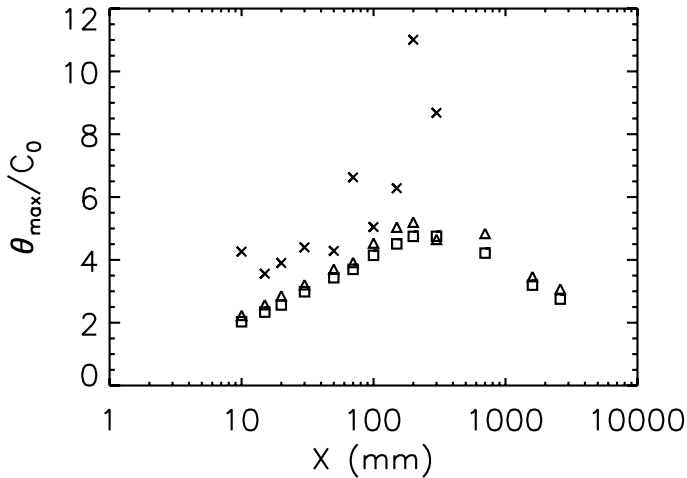


Fig. 4. Downwind variation of θ_{\max}/C_0 for the data of [22]. X is the downwind distance from the source. The crosses are the model values calculated from Eq. (31); the squares are calculated directly from the measurements by least-squares fits to the moment ratios for $n = 4$ to $n = 8$; and the triangles are calculated directly from the measurements by a linear fit to the moment ratios for $n = 7$ and $n = 8$.

der behaviour. Far downwind we expect $\beta \rightarrow 0$, since diffusion has had a long time to act, so Eq. (31) suggests that $\theta_{\max}/C_0 \rightarrow \alpha \hat{C} = C/C_0$, *i.e.* $\theta_{\max} \rightarrow C$. [14] argued that both of these results would be expected on physical grounds.

Figure 4 compares the downwind variation of θ_{\max}/C_0 estimated from Eq. (31) using fitted values of the parameters, with estimates obtained directly from the concentration data, for the data of [22]. Results of two different estimation methods are shown — one where Eq. (28) was fitted by least-squares to the points for $n = 4$ to $n = 8$, and one which used a straight line fit through the points for $n = 7$ and $n = 8$. The model values are larger than the direct estimates, and are not shown at the furthest downwind distances (where they become unreliable, because the denominator in r becomes close to zero). The direct estimate using only $n = 7$ and $n = 8$ is a little larger than that using $n = 4$ to $n = 8$, suggesting that n is not yet large enough to have attained the asymptotic result Eq. (27), and that θ_{\max}/C_0 may well be underestimated. So the model may be in better agreement with the true values of θ_{\max}/C_0 than indicated by this figure. Given the difficulty of making accurate estimates of the upper endpoint

of a distribution, the degree of agreement here is very promising. Further investigation into the optimal fitting of Eq. (28) is required, and also of the standard errors and biases of the estimates.

5. Discussion

The models described here were derived by considering the hypothetical case of no molecular diffusion, for which many exact results can be obtained, and then taking account of the physical effects of molecular diffusion. These models have proved highly successful at reproducing a number of aspects of turbulent dispersion, thus allowing results for the zero molecular diffusivity case to be used in the real world.

The models, given by Eq. (16) and Eq. (20), for the crosswind variation of concentration moments have been shown to fit experimental data very well. For example, excellent fits were found by [11] for point, jet and wall sources in boundary layers and grid turbulence, and by [12] for a line source in grid turbulence. One feature of the latter experiment which is captured particularly well is the downwind evolution of the cross-plume structure of the variance. Near the source the variance is bimodal (corresponding to $\alpha < 2$), with peaks located symmetrically about the mean plume centreline. Further downwind the variance becomes unimodal (corresponding to $\alpha > 2$), with a single peak on the centreline, and far downwind it becomes bimodal again. The relationship between skewness and kurtosis (and also higher order equivalents), given by Eq. (19), approximates a variety of experiments well, including steady sources and clouds, and cases with and without fences (further details were given in Sec. 3.2). The most recent work, in [14], shows very promising results when these models are extended to tackle the highly challenging problem of large concentrations, in particular that of estimating the largest possible concentration.

There are a number of pieces of work which would improve these models and make them even more useful for real world problems. The first is to develop better models for the evolution (with time since release or with distance from the source) of the parameters involved in the models. This will require further investigation of suitable closure schemes, similar to those discussed by [31–35]. The second is to carry out a thorough analysis of the moment-based method (i.e. that using Eq. (28)) for fitting the generalised Pareto distribution (GPD) for large concentrations. This will aim to answer questions about the optimum choice of moments to use, and about biases and standard errors of the resulting estimates. Finally, further work on

identifying the form of the pdf \hat{p} corresponding to the moments Eq. (20) would give valuable insight into the underlying structure of the model.

Acknowledgements

We are grateful for the financial support we have received for this work over the years, from Canada NSERC, UK Research Councils (in particular SERC, now EPSRC), the Royal Society, the UK Ministry of Defence, the UK Health and Safety Executive, the European Union and NATO.

Appendix A.

We show here that Eq. (8) is consistent with the results obtained previously by [12, 15].

In [15] it was assumed that the source was stationary and homogeneous, i.e. that the probability distribution of the source concentration $\Gamma_S(\mathbf{x}, t)$ was independent of \mathbf{x} and t . This case corresponds to applying Eq. (8) with $p_S(\theta; \mathbf{y}) = p_S(\theta)$. Thus

$$\int_{\mathbf{y} \in V_0} d\mathbf{y} p_S(\theta; \mathbf{y}) = V_0 p_S(\theta),$$

so Eq. (8) becomes

$$p(\theta; \mathbf{x}, t) \approx [1 - \pi(\mathbf{x}, t)] \delta(\theta) + \pi(\mathbf{x}, t) p_S(\theta),$$

which is equation (14) of [15].

In [12] it was assumed that the release occurred instantaneously at $t = 0$, with spatially-varying but non-random concentration $\Gamma_S(\mathbf{x})$. This corresponds to taking $p_S(\theta; \mathbf{y}) = \delta(\theta - \Gamma_S(\mathbf{y}))$, so that Eq. (8) immediately gives equation (8) of [12].

Appendix B.

Let us define a pdf $q(\hat{\theta})$ for $0 \leq \hat{\theta} \leq \infty$ by $q(\hat{\theta}) = \hat{p}_D(\hat{\theta} - D)$, where $D = \left(\frac{1}{\beta} - 1\right) \hat{C}$ and $\hat{p}_D(\hat{\theta})$ has mean \hat{C} , second absolute moment \hat{C} , and n th absolute moment $\lambda_n^{n-1} \hat{C}$ for $n = 2, 3, \dots$. Let $q(\hat{\theta})$ have mean μ_q , and n th central moment μ_{qn} for $n = 2, 3, \dots$. Then

$$\mu_q = \hat{C} + D = \frac{1}{\beta} \hat{C} = \frac{C}{\alpha \beta C_0}$$

and

$$\begin{aligned}\mu_{qn} &= \int_0^\infty (\hat{\theta} - \mu_q)^n q(\hat{\theta}) d\hat{\theta} = \int_{-D}^\infty (s - \hat{C})^n \hat{p}_D(s) ds \\ &= \sum_{i=0}^n \binom{n}{i} (-\hat{C})^{n-i} \hat{m}_{Di},\end{aligned}$$

where \hat{m}_{Di} is the i th absolute moment of $\hat{p}_D(\hat{\theta})$.

Thus we have

$$\mu_{q2} = \hat{C}(1 - \hat{C}),$$

and for $n \geq 3$

$$\mu_{qn} = \hat{C} \left\{ (n-1)(-\hat{C})^{n-1} + \binom{n}{2}(-\hat{C})^{n-2} + \sum_{i=3}^n \binom{n}{i} \lambda_i^{i-1} (-\hat{C})^{n-i} \right\}.$$

Thus the central moments of $q(\hat{\theta})$ are just the right-hand sides of Eq. (20). The left-hand sides of Eq. (20) are the central moments of $\hat{p}(\hat{\theta})$ (since the normalisation is $\hat{\Gamma} = \Gamma/(\alpha\beta C_0)$). Since $\hat{p}(\hat{\theta})$ also has mean $C/(\alpha\beta C_0) = \mu_q$, we can identify \hat{p} with q , and hence obtain

$$\hat{p}(\hat{\theta}) = \hat{p}_D(\hat{\theta} - D).$$

References

- [1] G. K. Batchelor, *An Introduction to Fluid Dynamics*. (Cambridge University Press, Cambridge, 1967).
- [2] K. A. Buch and W. J. A. Dahm, Experimental study of the fine-scale structure of conserved scalar mixing in turbulent shear flows. Part 2. $Sc \approx 1$, *J. Fluid Mech.* **364**, 1–29, (1998).
- [3] D. R. Lide, Ed., *CRC Handbook of Chemistry and Physics*. (Taylor & Francis, Boca Raton, Florida, 2005), 86th edition.
- [4] G. K. Batchelor, The effect of homogeneous turbulence on material lines and surfaces, *Proc. R. Soc. Lond. A.* **213**, 349–366, (1952).
- [5] W. J. A. Dahm, K. B. Southerland, and K. A. Buch, Direct, high resolution, four-dimensional measurements of the fine scale structure of $Sc \gg 1$ molecular mixing in turbulent flows, *Phys. Fluids A.* **3**, 1115–1127, (1991).
- [6] A. F. Corriveau and W. D. Baines, Diffusive mixing in turbulent jets as revealed by a pH indicator, *Exps. Fluids.* **16**, 129–136, (1993).
- [7] K. A. Buch and W. J. A. Dahm, Experimental study of the fine scale structure of conserved scalar mixing in turbulent shear flows. Part 1. $Sc \gg 1$, *J. Fluid Mech.* **317**, 21–71, (1996).

- [8] G. K. Batchelor, Small-scale variation of convected quantities like temperature in turbulent fluid. Part 1. General discussion and the case of small conductivity, *J. Fluid Mech.* **5**, 113–133, (1959).
- [9] P. C. Chatwin and P. J. Sullivan, The relative diffusion of a cloud of passive contaminant in incompressible turbulent flow, *J. Fluid Mech.* **91**, 337–355, (1979).
- [10] P. C. Chatwin and P. J. Sullivan, Cloud-average concentration statistics, *Math. Computers Sim.* **32**, 49–57, (1990).
- [11] P. C. Chatwin and P. J. Sullivan, A simple and unifying physical interpretation of scalar fluctuation measurements from many turbulent shear flows, *J. Fluid Mech.* **212**, 533–556, (1990).
- [12] B. L. Sawford and P. J. Sullivan, A simple representation of a developing contaminant concentration field, *J. Fluid Mech.* **289**, 141–157, (1995).
- [13] N. Mole and E. D. Clarke, Relationships between higher moments of concentration and of dose in turbulent dispersion, *Boundary-Layer Met.* **73**, 35–52, (1995).
- [14] N. Mole, T. P. Schopflicher, and P. J. Sullivan, High concentrations of a passive scalar in turbulent dispersion, *J. Fluid Mech.* **604**, 447–474, (2008).
- [15] P. C. Chatwin and P. J. Sullivan, The intermittency factor of scalars in turbulence, *Phys. Fluids A*, **1**, 761–763, (1989).
- [16] A. S. Monin and A. M. Yaglom, *Statistical Fluid Mechanics, Volume 1*. (The MIT Press, Cambridge, 1971).
- [17] D. J. Wilson, A. G. Robins, and J. E. Fackrell, Intermittency and conditionally-averaged concentration fluctuation statistics in plumes, *Atmos. Environ.* **19**, 1053–1064, (1985).
- [18] D. M. Lewis, P. C. Chatwin, and N. Mole, Investigation of the collapse of the skewness and kurtosis exhibited in atmospheric dispersion data, *Il Nuovo Cimento*. **20 C**, 385–398, (1997).
- [19] P. C. Chatwin and C. Robinson, The moments of the pdf of concentration for gas clouds in the presence of fences, *Il Nuovo Cimento*. **20 C**, 361–383, (1997).
- [20] T. P. Schopflicher and P. J. Sullivan, The relationship between skewness and kurtosis of a diffusing scalar, *Boundary-Layer Met.* **115**, 341–358, (2005).
- [21] T. P. Schopflicher, C. J. Smith, and P. J. Sullivan, Scalar concentration reduction in a contaminant cloud, *Boundary-Layer Met.* **122**, 683–700, (2007).
- [22] B. L. Sawford and C. M. Tivendale. Measurements of concentration statistics downstream of a line source in grid turbulence. In *Proc. 11th Australasian Fluid Mechanics Conf.*, Hobart, Australia (Dec., 1992).
- [23] D. J. Moseley. A closure hypothesis for contaminant fluctuations in turbulent flow. Master's thesis, University of Western Ontario, Canada, (1991).
- [24] H. Ye. *A New Statistic For The Contaminant Dilution Process In Turbulent Flows*. PhD thesis, University of Western Ontario, London, Ontario, Canada, (1995).
- [25] P. J. Sullivan and H. Ye, Moment inversion for contaminant concentration in turbulent flows, *Canadian Applied Mathematics Quarterly*. **4**, 301–310, (1996).

- [26] J. Pickands, Statistical inference using extreme order statistics, *Ann. Statist.* **3**, 119–131, (1975).
- [27] N. Mole, C. W. Anderson, S. Nadarajah, and C. Wright, A generalized Pareto distribution model for high concentrations in short-range atmospheric dispersion, *Environmetrics.* **6**, 595–606, (1995).
- [28] C. W. Anderson, N. Mole, and S. Nadarajah, A switching poisson process model for high concentrations in short-range atmospheric dispersion, *Atmos. Environ.* **31**, 813–824, (1997).
- [29] T. P. Schopflocher, An examination of the right-tail of the PDF of a diffusing scalar in a turbulent flow, *Environmetrics.* **12**, 131–145, (2001).
- [30] R. J. Munro, P. C. Chatwin, and N. Mole, The high concentration tails of the probability density function of a dispersing scalar in the atmosphere, *Boundary-Layer Met.* **98**, 315–339, (2001).
- [31] L. Clarke and N. Mole, Modelling the evolution of moments of contaminant concentration in turbulent flows, *Environmetrics.* **6**, 607–617, (1995).
- [32] F. Labropulu and P. J. Sullivan, Mean-square values of concentration in a contaminant cloud, *Environmetrics.* **6**, 619–625, (1995).
- [33] N. Mole, The α - β model for concentration moments in turbulent flows, *Environmetrics.* **6**, 559–569, (1995).
- [34] N. Mole, The large time behaviour in a model for concentration fluctuations in turbulent dispersion, *Atmos. Environ.* **35**, 833–844, (2001).
- [35] P. J. Sullivan, The influence of molecular diffusion on the distributed moments of a scalar PDF, *Environmetrics.* **15**, 173–191, (2004).