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"Quantitative Models for Environmental Pollution: a Review"

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18 Quantitative Models for Environmental Pollution: a Review

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18.1 INTRODUCTION

There are, quite rightly, growing concerns world-wide about the dangers, both actual and potential, of environmental pollution. Public pressure is causing governments and their agencies, sometimes acting multinationally, to support or require the development of regulations to control and monitor the degree of such pollution. Such regulations often involve mathematical modelling. Unfortunately, turbulent diffusion, one of the basic scientific processes that controls environmental pollution, is still not understood, at least not to an extent that allows such models to be reliable or accurate. Therefore, even leaving aside scientific curiosity, there is — in the authors' opinion — an overwhelming practical case for increased public support of basic research in turbulent diffusion. This case is not at present being articulated with sufficient force by scientists nor, partly as a consequence, is it recognized by governments and their agencies. Also, what publicly funded research there is tends to have aims that are dominated by perceived market needs (e.g. to construct a mathematical model of the consequences of the accidental release of chlorine on a factory site with account taken of the buildings *or* of the effect of hot water release from a planned power station on the chemistry and biology in a complex estuarine system). The point of this comment is not that such aims are not worthwhile, but that the present state of knowledge means that they are misguided and/or unrealistic because they cannot be met except in a cosmetic sense.

In a world that is becoming 'greener', the authors believe that an extended discussion of the points made in the previous paragraph is overdue and this chapter is an attempt at that, albeit a partial and selective one. In view of Cath Allen's scientific career and philosophy, it seems particularly appropriate for such a paper to appear in a volume in her memory.

18.2 SCIENTIFIC AND MATHEMATICAL BACKGROUND

The discussion will be restricted to cases of a pollutant whose concentration $\Gamma(\boldsymbol{x},t)$ at position $\boldsymbol{x}$ and time t is determined by two processes: advection by a turbulent flow with velocity field $\boldsymbol{\Upsilon}(\boldsymbol{x},t)$, and molecular diffusion with constant diffusivity κ. The equation governing Γ

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is therefore

$$\frac{\partial \Gamma}{\partial t} + \boldsymbol{\Upsilon}.\nabla\Gamma = \kappa\nabla^2\Gamma \tag{18.1}$$

Hence cases where chemical changes occur are not included, but many of the points developed in this chapter also apply in general terms then. However, given this restriction, it is important to note that Equation 18.1 applies whether or not the pollutant is passive. This is so, in particular, when buoyancy forces (arising, for example, when a heavy gas such as chlorine is released into the atmosphere, *or* when hot water used to cool a power station is discharged into a river) have a significant influence on $\boldsymbol{\Upsilon}(\boldsymbol{x},t)$ through the body force term in the Navier–Stokes equations. In such cases, experimental evidence suggests that it is nearly always reasonable to assume that the flow is incompressible, i.e. that

$$\nabla.\boldsymbol{\Upsilon} = 0 \tag{18.2}$$

It is important first to emphasize that Γ and $\boldsymbol{\Upsilon}$ are the real concentration and velocity fields in any one realization, and that they would be the values measured by transducers in a 'perfect' world without instrument noise and smoothing. In particular Γ and $\boldsymbol{\Upsilon}$ are not average values over time or space except, of course, of the type required by the continuum hypothesis. As, however, all the flows considered are turbulent, $\boldsymbol{\Upsilon}$, and hence Γ, are random fields. This means that the values of $\boldsymbol{\Upsilon}(\boldsymbol{x},t)$ and $\Gamma(\boldsymbol{x},t)$ for any particular $\boldsymbol{x}$ and t are (i) unpredictable and (ii) vary from realization to realization.

The key theme of this chapter will be that, for both theoretical correctness and practical validity, these random fields must be described in terms of their probability structures. Attention will be focused on one probability measure, namely the probability density function (pdf) of $\Gamma(\boldsymbol{x},t)$. This will be denoted by $p(\theta;\boldsymbol{x},t)$ and is defined by

$$p(\theta;\boldsymbol{x},t) = \frac{\mathrm{d}}{\mathrm{d}\theta}[\mathrm{prob}(\Gamma(\boldsymbol{x},t) \leq \theta)] \tag{18.3}$$

for all $\theta \geq 0$. The variable θ therefore ranges over all the possible values that $\Gamma(\boldsymbol{x},t)$ can take. The probability that $\Gamma(\boldsymbol{x},t) \leq \theta$, appearing on the right-hand side of Equation 18.3, is the proportion of the realizations forming the underlying ensemble for which $\Gamma(\boldsymbol{x},t) \leq \theta$. The central concept of the underlying ensemble is discussed, for example, in Chatwin and Allen (1985a). Here it suffices to recall that the choice of the ensemble is for the investigator to decide, but that this choice must be made since it defines those conditions which are permitted to vary from realization to realization and, hence, determine $p(\theta;\boldsymbol{x},t)$ and all other probability measures. For example, when assessing the safety of a fixed storage tank containing dangerous gas, it is likely to be appropriate to consider an ensemble of releases in which the weather and the time of release are allowed to vary from realization to realization, but in which the source geometry and topography are not. If people are not in the vicinity of the storage tank at night, it is possibly more appropriate to consider another ensemble (a subensemble of the first) in which night-time releases (and hence night-time weather conditions) are excluded.

It is now convenient to state some of the more important properties of $p(\theta;\boldsymbol{x},t)$.

(i) If $\delta\theta$ is small and positive, $p(\theta;\boldsymbol{x},t)\delta\theta$ is to first order in $\delta\theta$ the probability that $\theta \leq \Gamma(\boldsymbol{x},t) < \theta + \delta\theta$.

(ii) As $p(\theta;\boldsymbol{x},t)$ is a pdf

$$\int_0^\infty p(\theta;\boldsymbol{x},t)\mathrm{d}\theta = 1 \tag{18.4}$$

(iii) In any ensemble there is a maximum possible value, $\Theta(\boldsymbol{x},t)$ say, of $\Gamma(\boldsymbol{x},t)$ so that $p(\theta;\boldsymbol{x},t)$ is identically zero for $\theta > \Theta(\boldsymbol{x},t)$ and the upper limit in the integral in Equation 18.4 could be replaced by $\Theta(\boldsymbol{x},t)$. Note that advection acting alone cannot change the concentration of pollutant in any fluid particle so that $\Theta(\boldsymbol{x},t)$ is strictly less than the source concentration θ_0 (supposed uniform) because of molecular diffusion, the only process that dilutes the pollutant. However, $\Theta(\boldsymbol{x},t)$ is unknown and is impossible to calculate except for some simple ensembles (Chatwin and Sullivan, 1979). Thus Equation 18.4 is usually a convenient form to use.

(iv) The expected value, or ensemble mean, of any function f of $\Gamma(\boldsymbol{x},t)$ is defined in the standard way by

$$E\{f[\Gamma(\boldsymbol{x},t)]\} = \int_0^\infty f(\phi)p(\phi;\boldsymbol{x},t)\mathrm{d}\phi \tag{18.5}$$

Three special choices of f in Equation 18.5 are especially relevant.

$$f[\Gamma(\boldsymbol{x},t)] = \delta[\Gamma(\boldsymbol{x},t)-\theta] \Rightarrow p(\theta;\boldsymbol{x},t) = E\{\delta[\Gamma(\boldsymbol{x},t)-\theta]\} \tag{18.6a}$$

$$f[\Gamma(\boldsymbol{x},t)] = \Gamma(\boldsymbol{x},t) \Rightarrow \mu(\boldsymbol{x},t) = E\{\Gamma(\boldsymbol{x},t)\} = \int_0^\infty \phi p(\phi;\boldsymbol{x},t)\mathrm{d}\phi \tag{18.6b}$$

$$\begin{aligned} f[\Gamma(\boldsymbol{x},t)] &= [\Gamma(\boldsymbol{x},t)-\mu(\boldsymbol{x},t)]^2 \Rightarrow \sigma^2(\boldsymbol{x},t) = E\{[\Gamma(\boldsymbol{x},t)-\mu(\boldsymbol{x},t)]^2\} \\ &= \int_0^\infty [\phi-\mu(\boldsymbol{x},t)]^2 p(\phi;\boldsymbol{x},t)\mathrm{d}\phi = \int_0^\infty \phi^2 p(\phi;\boldsymbol{x},t)\mathrm{d}\phi - \mu^2(\boldsymbol{x},t) \end{aligned} \tag{18.6c}$$

In Equation 18.6b $\mu(\boldsymbol{x},t)$ is the (ensemble) mean concentration and in Equation 18.6c $\sigma^2(\boldsymbol{x},t)$ is the concentration variance, so that $\sigma(\boldsymbol{x},t)$ is the standard deviation of the concentration. It is also common to use the term rms concentration fluctation for $\sigma(\boldsymbol{x},t)$.

(v) Use of Equations 18.5 and 18.6a in Equation 18.1 gives the evolution equation for $p(\theta;\boldsymbol{x},t)$ in the form

$$\frac{\partial p}{\partial t} + \nabla.E\{\boldsymbol{\Upsilon}\delta[\Gamma-\theta]\} = \kappa\nabla^2 p - \kappa\frac{\partial^2}{\partial\theta^2}E\{(\nabla\Gamma)^2\delta[\Gamma-\theta]\} \tag{18.7}$$

where, for brevity, the dependence of p on $\theta,\boldsymbol{x},t$ and of $\boldsymbol{\Upsilon},\Gamma$ on $\boldsymbol{x},t$ has not been shown explicitly. This will often be done in the rest of this chapter where there is no risk of confusion — for example, Equations 18.8 and 18.9 below. Use of Equations 18.6b and 18.6c with 18.7 gives equations for $\mu(\boldsymbol{x},t)$ and $\sigma^2(\boldsymbol{x},t)$

$$\frac{\partial\mu}{\partial t} + \nabla.E\{\boldsymbol{\Upsilon}\Gamma\} = \kappa\nabla^2\mu \tag{18.8}$$

$$\frac{\partial\sigma^2}{\partial t} + \nabla.\mathbf{E}\{\boldsymbol{\Upsilon}\Gamma^2\} - 2\mu\nabla.E\{\boldsymbol{\Upsilon}\Gamma\} = \kappa\nabla^2\sigma^2 - 2\kappa E\{(\nabla\Gamma-\nabla\mu)^2\} \tag{18.9}$$

See, for example, Chatwin (1990) for further details of the derivation of Equations 18.7–18.9. It is important to emphasize that Equation 18.7 and hence 18.8 and 18.9, are completely general provided Equations 18.1 and 18.2 apply throughout each realization of the underlying ensemble. The expected values in Equation 18.7 depend on the choice of this ensemble, partly through its influence on the initial and boundary conditions.

18.3 SOME COMMENTS ON TRADITIONAL APPROACHES

Neither the notation nor the methods used in Section 18.2 are yet standard in turbulent diffusion, so some explanation is perhaps in order. The symbols μ and σ^2 have been used because this is universal practice for mean and variance in statistics. More traditional notation in turbulent diffusion would be, for example, C for Γ, C for μ, c' for $(\Gamma-\mu)$ and $\overline{(c')^2}$ for σ^2. This notation is inelegant, but there is a more serious criticism. This is that overbars have a precise connotation in statistics, namely to denote arithmetic means (the same connotation is used elsewhere, e.g. for centres of mass). Suppose, for example, that a finite number n of realizations (repetitions) of an experiment are made and that $\Gamma^{(r)}(\boldsymbol{x},t)$ is the concentration at $(\boldsymbol{x},t)$ in the rth realization $(r = 1,2,\ldots,n)$. Then the standard definition of $\bar{\Gamma}_n(\boldsymbol{x},t)$ would be

$$\bar{\Gamma}_n(\boldsymbol{x},t) = \frac{1}{n}\left\{\sum_{r=1}^{n}\Gamma^{(r)}(\boldsymbol{x},t)\right\} \tag{18.10}$$

If there is no risk of confusion, $\bar{\Gamma}(\boldsymbol{x},t)$ can be used instead of $\bar{\Gamma}_n(\boldsymbol{x},t)$.

The key point is that $\bar{\Gamma}_n(\boldsymbol{x},t)$ is itself a random variable, unlike $\mu(\boldsymbol{x},t)$. The mean and variance of $\Gamma_n(\boldsymbol{x},t)$ are $\mu(\boldsymbol{x},t)$ and $\sigma^2(\boldsymbol{x},t)/n$ respectively, and $\bar{\Gamma}_n(\boldsymbol{x},t)$ is an estimate of $\mu(\boldsymbol{x},t)$. The probability that the difference between $\bar{\Gamma}_n$ and μ exceeds any given quantity tends to 0 as $n \to \infty$ and, in that sense

$$\mu(\boldsymbol{x},t) = \lim_{n\to\infty}\{\bar{\Gamma}_n(\boldsymbol{x},t)\} \tag{18.11}$$

A related point about means is more serious. Many textbooks and papers still assert (usually without comment) that the only means with which turbulence and turbulent diffusion deal are time-averages. This assertion is wrong in principle and potentially dangerous if used in practice. Suppose, for example, that $\bar{\Gamma}_T(\boldsymbol{x},t)$ is defined by

$$\bar{\Gamma}_T(\boldsymbol{x},t) = \frac{1}{T}\left\{\int_{t-T/2}^{t+T/2}\Gamma(\boldsymbol{x},s)\mathrm{d}s\right\} \tag{18.12}$$

The first point to note is that, like $\bar{\Gamma}_n$ in Equation 18.10, $\bar{\Gamma}_T$ is a random variable. The statistical properties of $\bar{\Gamma}_T$ obviously depend on T, but, unlike those of $\bar{\Gamma}_n$, cannot be expressed simply in terms of those of Γ; for one thing the values of $\Gamma(\boldsymbol{x},s)$ in the integrand of Equation 18.12 are not statistically independent. Figure 18.1 illustrates how significantly time-average estimates of one statistical property of $\Gamma(\boldsymbol{x},t)$ depend on T. It follows that measured values and mathematical models of $\bar{\Gamma}_T$, and regulations expressed in terms of time-average concentrations, that do not specify T are valueless. (Although incidental to the main thrust of this paper, it is worth commenting also that even when T is specified in regulations its choice is not always supported by convincing evidence that $\bar{\Gamma}_T$ for that value of T is the correct measure of, for example, toxic risk.)

There is only one situation where time-averages are useful and that is when the ensemble is statistically steady, i.e. when all statistical properties of Γ, including p (and therefore μ,σ^2), are independent of t. This requires the release of pollutant to be at a steady rate into a flow which is itself statistically steady. The first restriction excludes, for example, sudden releases such as those occurring in accidents, and the second restriction excludes tidal flows. In those rare situations when these conditions are met, the mean concentration $\mu(\boldsymbol{x})$ can be estimated from the record of one experiment because, by ergodicity, $\bar{\Gamma}_T(\boldsymbol{x},t)$ then tends to

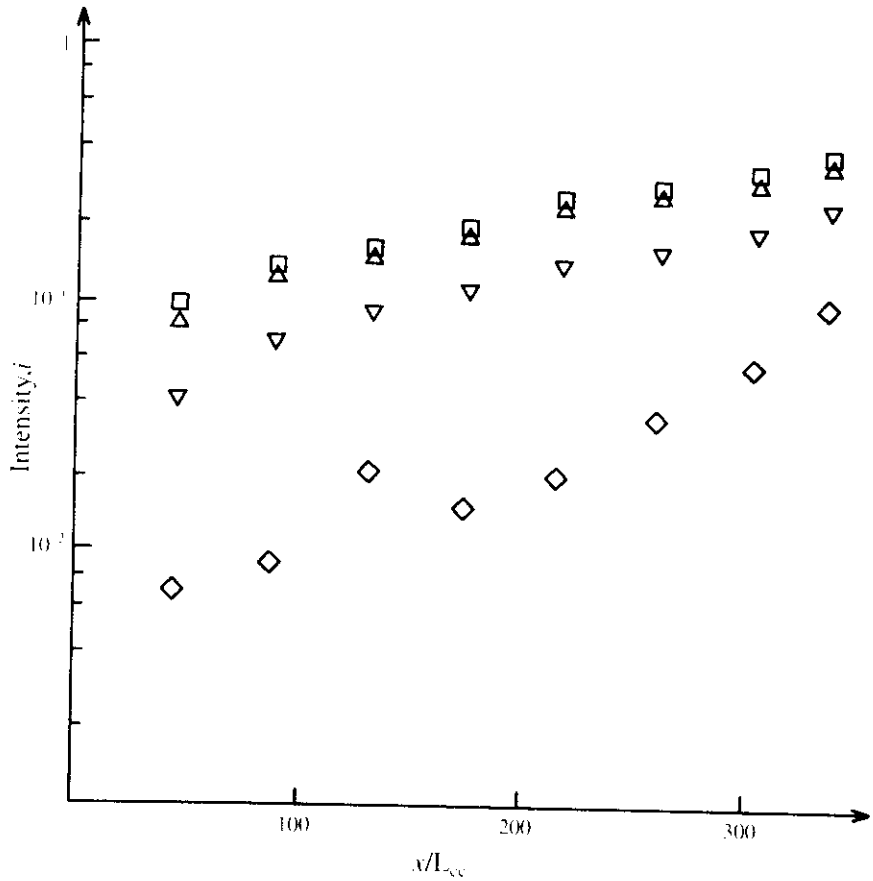

Figure 18.1 Dependence of estimate of intensity i on averaging time T for dispersion from a steady source, where $i = \sigma/\mu$, x is downwind distance and L_{cc} is a constant length scale. A running average of the time series over time T was made at each measuring station, and then μ and σ were estimated from the smoothed record (over identical segments). Symbols denote the following values of T in seconds: □ 0.07; △ 0.7; ▽ 7; and ◇ 70. Taken from Figure 14(b) of Davies (1990)

$\mu(x)$ as $T \to \infty$. When applicable, use of this result avoids costly repetitions. But, most turbulent diffusion problems, including nearly all those affecting the environment, are statistically unsteady. From the point of view of understanding, the use of time-averages is then at best an unnecessary complication and at worst a serious source of error and confusion; see Chatwin and Allen (1985b; 1985c).

Beginning in the early part of this century (Monin and Yaglom, 1971; pp. 606–676), much research on turbulent diffusion has focused on the behaviour of the mean concentration $\mu(\boldsymbol{x},t)$. The traditional approach followed that of Reynolds for the mean velocity field. If

$$\boldsymbol{U}(\boldsymbol{x},t) = E\{\boldsymbol{\Upsilon}(\boldsymbol{x},t)\} \tag{18.13}$$

is the mean velocity field and if $c(\boldsymbol{x},t)$ and $\boldsymbol{u}(\boldsymbol{x},t)$ are the concentration and velocity fluctuations defined by

$$c(\boldsymbol{x},t) = \Gamma(\boldsymbol{x},t) - \mu(\boldsymbol{x},t), \quad \boldsymbol{u}(\boldsymbol{x},t) = \boldsymbol{\Upsilon}(\boldsymbol{x},t) - \boldsymbol{U}(\boldsymbol{x},t) \tag{18.14}$$

then Equation 18.8 can be rewritten in the more familiar form

$$\frac{\partial \mu}{\partial t} + (\boldsymbol{U}.\nabla)\mu = -\nabla.E\{\boldsymbol{u}c\} + \kappa\nabla^2\mu \tag{18.15}$$

In turbulent diffusion, the term involving $E\{\boldsymbol{u}c\}$ in Equation 18.15 cannot be small. However, it is not expressed in terms of μ and $\boldsymbol{U}$, and there is no known way of doing so that is mathematically correct or even physically sound. This is the simplest manifestation of the closure problem.

For the reason given in the next paragraph, little will be written here about the many different attempts made to circumvent the closure problem for μ. Understandably, these all involve empiricism and, also understandably, none has been fully justified scientifically, either theoretically from the fundamental Equation 18.1 or by satisfactory and comprehensive validation against data. Early methods such as Gaussian plume models or those involving eddy diffusivities are now becoming obsolete. Popular current methods, which extend in practice or principle to other statistical properties, include high-order closures (many available commercially despite the lack of proper validation) and random walks. It is interesting to recall that Cath Allen (Allen, 1982) was one of the first to apply the latter technique in an environmental context by developing the ideas of Sullivan (1971).

The mean concentration μ is the simplest statistical property of $\Gamma(\boldsymbol{x},t)$ and, indisputably, one of the most important. Unfortunately, too much research over too many years has proceeded on the assumption that it is the only one. The unfortunate result of such emphasis is that, until relatively recently, little attention has been paid to the question of whether other statistical properties are important, so that the essential randomness of turbulent diffusion has largely been ignored, i.e. it has been treated almost as a deterministic phenomenon.

18.4 PRACTICAL IMPORTANCE OF A STATISTICAL DESCRIPTION

Over the last 25 years or so, experimental evidence has accelerated to show that differences between the actual concentration $\Gamma(\boldsymbol{x},t)$ and the mean concentration $\mu(\boldsymbol{x},t)$ are not small. The simplest measure of such differences is $\sigma(\boldsymbol{x},t)$ defined in Equation 18.6c, and Figure 18.1 has already shown that experimental estimates of σ/μ, the intensity, are not small. Other examples from the many possible are given in Figures 18.2 and 18.3. It will be noted that values of the estimated intensity range from $\sim 10^{-1}$ to ~ 6, ignoring the even lower values in Figure 18.1 obtained by imposing artificially large averaging times T on the raw data. Some comments on the reasons for this variation will be made later. Figures 18.4 and 18.5 show measured values of $p(\theta;\boldsymbol{x},t)$ which, again, are chosen from many possible examples and confirm that turbulent diffusion is far from being deterministic. For, were the process deterministic, the only value possible for $\Gamma(\boldsymbol{x},t)$ would be $\mu(\boldsymbol{x},t)$ so that $p(\theta;\boldsymbol{x},t)$ would have the form

$$p(\theta;\boldsymbol{x},t) = \delta[\theta - \mu(\boldsymbol{x},t)] \tag{18.16}$$

where δ denotes the Dirac delta function; according to Equation 18.16 the graph of p versus θ would have a single infinite spike — very different from those shown in Figures 18.4 and 18.5.

Given the weight of this evidence, it would be surprising if it was not now recognized that accurate models of all types of environmental impact should take into account the statistical nature of $\Gamma(\boldsymbol{x},t)$. Summaries of practical examples are given by Chatwin (1990), Sullivan (1990), Weil, Sykes and Venkatram (1992) and by many others.

However, because of the traditional, but undue, emphasis on the mean concentration noted

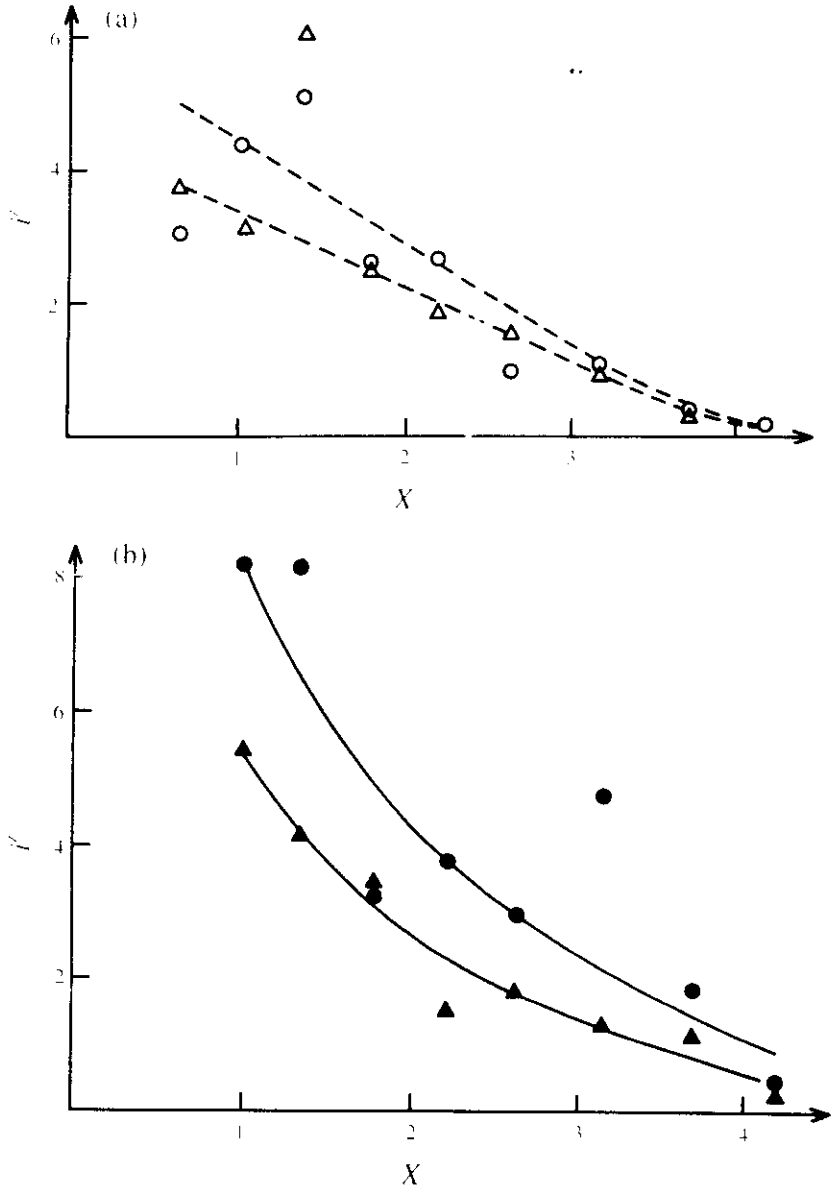

Figure 18.2 Estimates of i^2 (see caption to Figure 18.1) downstream of a steady (a) neutrally buoyant and (b) buoyant source from water tank experiments by Deardorff and Willis (1984). In each diagram the triangles and circles denote averages over the inner and outer parts of the plume, respectively. X is a dimensionless downstream distance measured from the source, as defined in the original paper (Reprinted from Deardoff and Willis, 1984, with kind permission from Pergamon Press Ltd, Headington Hall, Oxford OX3 0BW, UK)

above, most regulatory models now in use are based solely on its predicted behaviour. As turbulent diffusion is essentially a stochastic process (as evidenced by Figures 18.1–18.5), such models cannot (except perhaps for some particular types of environmental risk) ensure the protection that was presumably intended. In fairness, it should be noted that some such models attempt to recognize the existence of the large variations of actual concentrations from the mean by incorporating estimates of 'peak to mean ratio'. However, this measure is

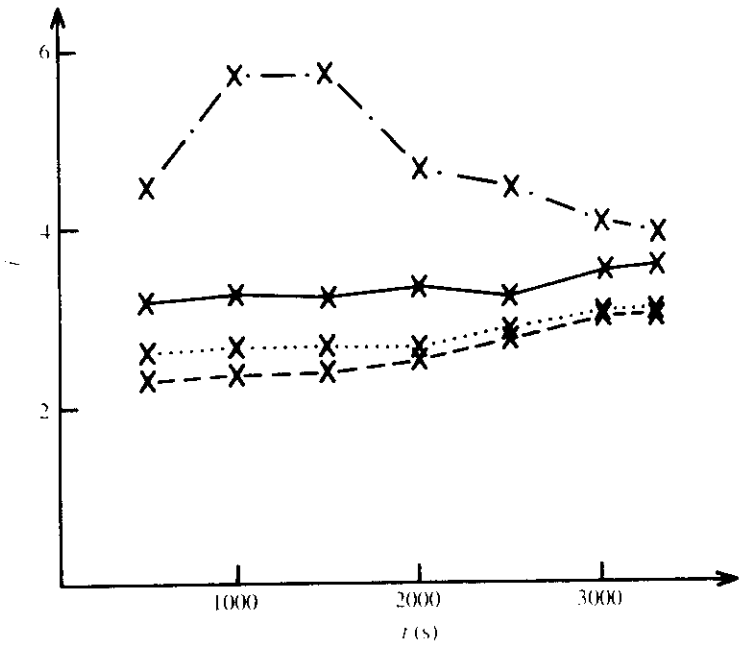

Figure 18.3 Estimates of i (see caption to Figure 18.1) for data from field trials conducted by Dr C.D. Jones under convective daytime conditions. The values of Γ were measured at four different cross-wind positions (channels) at 7.5 m downwind of the steady source and were digitized at 10 Hz. Estimates, denoted by crosses, were made for all four channels using all the data points up to the times shown to assess how statistical convergence of the estimates depended on n in Equation 18.10. The slight trends evident for the larger values of time probably indicate that the atmospheric conditions were only approximately statistically steady. Adapted from Figure C15 of Mole and Chatwin (1990)

scientifically unacceptable. Of course, there is a peak concentration Θ at each x and t as noted above in Section 18.2, but its value is unknown and very difficult, perhaps impossible, to calculate. The most serious objection to the use of the 'peak to mean ratio' is that it is almost impossible to measure Θ because its occurrence as an actual value of Γ is, by definition, an extremely rare event. This is especially true when, as is normally the case (see Figures 18.4 and 18.5), the graph of p versus θ has a long tail for the higher values of θ. It follows that it is extremely unlikely (and this conclusion can be quantified, in principle at least) that measured values of 'peak' concentration are at all close to the real value of Θ and may be orders of magnitude less. These measured values are, of course, themselves random variables, but random variables whose statistical properties depend mainly on quantities such as the length of data record and detailed characteristics of the instrumentation and data analysis. Even if Γ happens to be close to Θ for a particular value of x and t, it will be so only over a spatial region of dimensions of the order of the conduction cut-off length (Chatwin and Sullivan, 1979), typically of order 10^{-4} m in the atmosphere. This dependence of measured peak concentrations on the instruments and length of data record is well illustrated in Figure 15 of Koopman *et al.* (1982).

It is therefore clear that traditional regulatory models, based on the mean concentration μ (even when they predict its behaviour with x and t), are not satisfactory and must, eventually, be replaced by statistical models. Before discussing, in slightly more detail, the elements

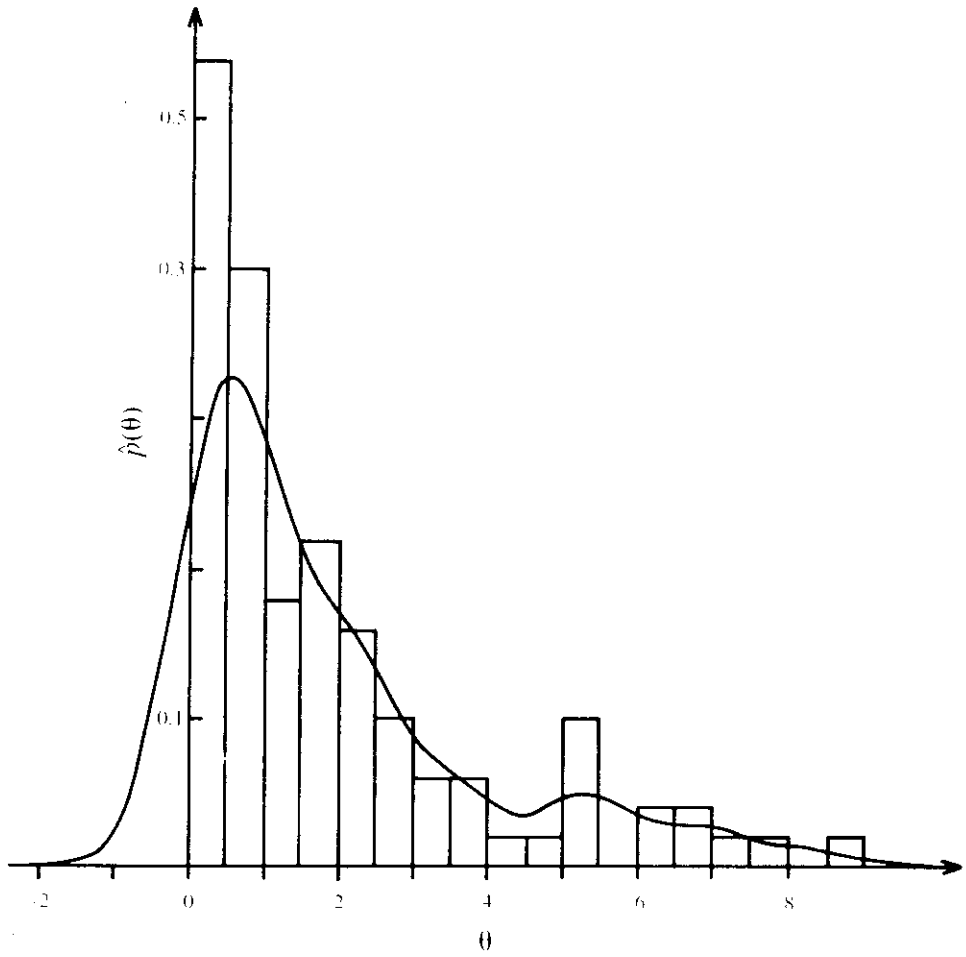

Figure 18.4 Histogram of concentrations from wind tunnel data obtained by Dr D.J. Hall, who carried out 100 repetitions of an experiment in which neutrally buoyant gas was instantaneously released. The curve shows an estimate $\hat{p}(\theta)$ of the pdf by G.W. Goodall found by a non-parametric technique. Notice the bimodality. See Chatwin and Goodall (1991) for further details of the experiments; this estimate is for the ground-level measuring station 0.70 m downwind of the source for a non-dimensional time τ after release of 9.80

of one such statistical model, it is important to reiterate a point made earlier in a different context. Any model for assessing, monitoring or regulating environmental risk, whether this arises through a potential accident or through persistent background concentrations (as is appropriate for most air and water quality control) must deal with the correct measure (or measures) of harm. Unfortunately, except perhaps for the case of local ignitability of a flammable gas, it is difficult to know what this correct measure is or, at least, whether any proposed measure (e.g. 'peak to mean ratio' again!) is supported by adequate evidence. It is understandable that this comment seems likely to be appropriate for most cases of toxicological risk.

One of the standard measures of toxicological risk is in terms of the dosage $D(x,t)$, where

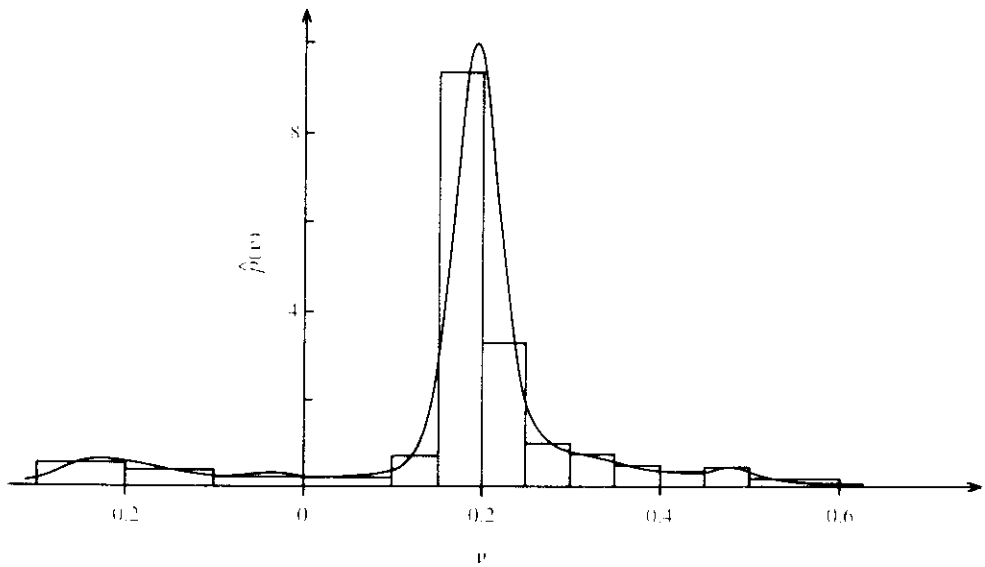

Figure 18.5 This figure has been derived by D.M. Lewis from data obtained by Dr C.D. Jones in field trials in 1991, and indicates some of the problems associated with noise. An attempt has been made to remove noise from the raw data, but about 12% of the data points still indicate negative voltages (concentrations). The curve is an estimate $\hat{p}(v)$ of the voltage pdf obtained using the same method as that in Figure 18.4; voltages are used pending final details on the calibration

$$D(\boldsymbol{x},t) = \int_{-\infty}^{t} \Gamma(\boldsymbol{x},s)\mathrm{d}s \tag{18.17}$$

In Equation 18.17 the lower limit $t = -\infty$ allows for all possibilities but, in practice, it would be replaced by the time at which the risk began by, for example, the sudden accidental release of a toxic substance. (In view of comments above, it is interesting to note that an earlier 'definition' of dosage ignored any time variation of Γ, whether random or not, and postulated, wrongly of course, the existence of a constant concentration C and a time of exposure T, with D equal to CT.) Like $\Gamma(\boldsymbol{x},t)$, $D(\boldsymbol{x},t)$ is a random variable with pdf $p_D(\psi;\boldsymbol{x},t)$, where (see Equation 18.3)

$$p_D(\psi;\boldsymbol{x},t) = \frac{\mathrm{d}}{\mathrm{d}\psi}[\mathrm{prob}\{D(\boldsymbol{x},t) \leq \psi\}] \tag{18.18}$$

Figure 18.6 shows an example of p_D estimated from experimental data.

Provided D is the appropriate measure of toxic risk, and the only one, the mortality rate $M(\boldsymbol{x},t)$ of a population at $\boldsymbol{x}$ after time t (where M is a proportion so that $0 \leq M \leq 1$) is a random variable with statistical properties dependent on those of D, i.e. on p_D. The probability $P_M(m;\boldsymbol{x},t)$ that $M(\boldsymbol{x},t) \leq m$ for any m with $0 \leq m \leq 1$ depends on toxicological response and can be expressed precisely in terms of p_d and the conditional probability distribution of $M(\boldsymbol{x},t)$ for given dosage. This latter distribution has to be estimated by toxicologists. If p_D and it were to be known, $P_M(m;\boldsymbol{x},t)$ could be determined and used to take counter-measures. For example, choose a small value m_1 (e.g. $m_1 = 0.001$) and a large value P_1 (e.g. $P_1 = 0.999$) and insist that all the population is located so that

$$P_M(m_1;\boldsymbol{x},t) > P_1 \tag{18.19}$$

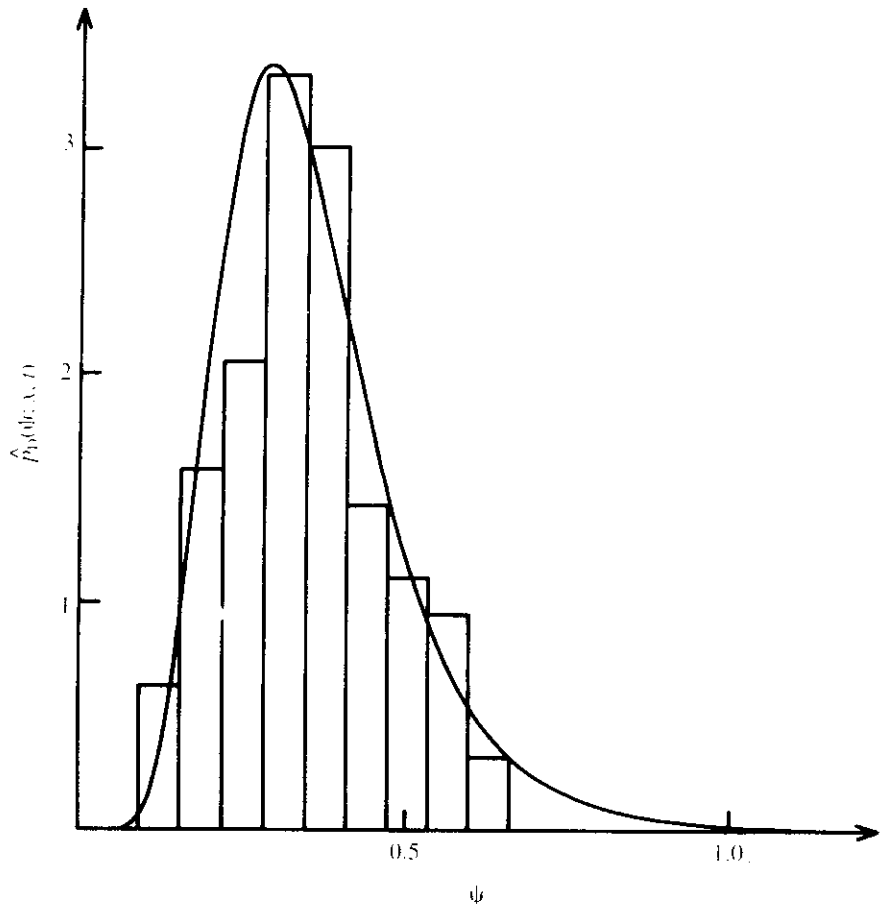

Figure 18.6 Histogram of dosages and estimate of dosage pdf $p_D(\psi;x,t)$ for Dr D.J. Hall's data. Details as in Figure 18.4 except that $\tau = 30.0$. The estimate, by W.L. Sweatman, uses a different technique from that used to obtain Figure 18.4; the curve is the log-normal pdf whose mean and variance are those obtained by maximum likelihood

With the given illustrative values for m_1 and P_1, Equation 18.19 ensures that there is a greater than 99.9% chance that less than 0.1% of the population will die; refer to Griffiths and Megson (1984), Ride (1984) and Griffiths and Harper (1985) for more detailed comments on toxicological risk.

It is clear that the approach to risk assessment just outlined can be applied in other cases, including those when Γ itself is the appropriate measure of the hazard. One such hazard is local ignitability of a mixture of air and a flammable gas such as CH_4. If the stoichiometric limits for the gas–air mixture are θ_1 and θ_2 (where $\theta_1 = 0.05$, $\theta_2 = 0.15$ for CH_4), the probability that the mixture is ignitable at x and t is determined very simply from $p(\theta;x,t)$ by

$$P_I(x,t) = \int_{\theta_1}^{\theta_2} p(\theta;x,t)\mathrm{d}\theta \tag{18.20}$$

Birch, Brown and Dodson (1980) presented convincing experimental evidence that Equation 18.20 is the correct quantification and, conversely, showed that their results could not be

correlated with the mean concentration $\mu(x,t)$. In particular, there was no apparent connection between the level surfaces of P_I (i.e. surfaces on which the probability of local ignitability was constant) and those of μ: the two sets of surfaces did not even have similar shapes.

18.5 CURRENT RESEARCH NEEDS

18.5.1 Preamble

We contend that the above arguments show that the scientific case for using probabilistic/statistical methods to assess environmental pollution, rather than traditional deterministic methods, is overwhelming. There are three important factors which tend to inhibit progress towards the much more widespread adoption of such methods in practice.

First of all is the problem of public attitudes, in which it is convenient to include, for example, those of politicians, legislators and lawyers. In many countries, including Canada and the UK, the crucial concept of a probability is not something to which most people are ever exposed, let alone understand. (It could be argued that an exception needs to be made for gambling.) For example, when a public inquiry is set up following a serious incident, it is common to hear politicians use phrases like 'to ensure complete safety' in stating aims, especially in some of the most sensitive areas, such as food and nuclear reactors. It will be clear now that 'complete safety' is unachievable and that, instead, the public must be educated to understand, and to accept, that the only type of objective that can be achieved, in principle at least, is a large probability of harm less than a specified level occurring. For example, for the case of a toxic gas in which dosage is the appropriate measure of harm, the constants m_I, level of harm, and $(1-P_I)$, the probability of this level of harm not occurring, in Equation 18.19 are legitimate targets. The precise numerical values of these two measures have to be chosen by the public, bearing in mind cost — the smaller the probability and the degree of harm, the higher will be the cost.

The second factor has already been mentioned; it is the rapid acceleration in public pressure for regulations on environmental safety due to increased perceptions of its importance. As a result of this pressure many models based on invalid premises have been developed and sold, and are now being used. In detail, such models always involve assumptions, particularly for 'resolving' the closure problem, that are, at best, inadequately validated against data and, at worst, obviously erroneous. But, additionally and more fundamentally, they postulate a deterministic process that has no connection with reality. The existence of such models (and the willingness of some people to produce new ones) reduces the pressure on users, and potential users, to demand better ones. However (Hanna *et al.* 1992), there are a few encouraging signs of more realistic perceptions.

Finally, it is important to note that producing good statistical models of turbulent diffusion is difficult. On the one hand the underlying science is not adequately understood in the sense that the methods proposed for resolving the closure problem, inevitably present in any equation for a statistical property, such as Equations 18.7–18.9, cannot be asserted to have known limits of accuracy except, sometimes, in situations for which they have been directly validated against data. Too often, the results obtained in one set of circumstances are extrapolated without justification to other (usually more complicated) situations. For example, it has sometimes been claimed (or at least implied) that values of σ/μ measured in experiments on passive tracers on flat ground in neutral atmospheres with a steady continuous release can be applied to unsteady (including instantaneous) releases of heavy gases under all atmospheric conditions

and on all terrains. The inevitable conclusion is that there must be more — much more — fundamental research. On the other hand, even if the fundamental scientific problems can be solved, the task of incorporating the new knowledge into practical models will be difficult. Particularly challenging is the need to have models that can respond to emergencies both quickly and with acceptable accuracy. Although the basic scientific problems will, in principle, be solved eventually (within one hundred years?) when computers are powerful enough for the statistical properties of Γ to be estimated from sufficiently many direct numerical solutions of Equation 18.1 and the Navier–Stokes equations (thereby avoiding the closure problem), it is almost certain that this technique will be far too slow for it to be used in, for example, a factory site in which dangerous gases are produced or stored.

18.5.2 Practical statistical models of the future

In view of the dilemma posed at the end of the last paragraph, it is proper to study the structure of statistical models that can be used in practice to assess actual or potential hazards. Such models ought to be simple enough to be applicable and useful, but not so simple that they are grossly inaccurate or misleading. Also, because of the large range of different hazards that exist (toxicity, flammability, malodour, etc.), each with its own measure, or measures, of harm (see Section 18.4), there is a need for many different models to be developed.

Given the unlikelihood, at least in the short to medium term, of being able to produce practical models (i.e. models yielding sufficiently accurate results quickly enough in real time) based on approximate solutions of equations such as 18.7–18.9 for $p(\theta;\boldsymbol{x},t)$, $\mu(\boldsymbol{x},t)$ and $\sigma^2(\boldsymbol{x},t)$, respectively, there are two promising but incipient lines of research. For illustrative purposes these will be discussed in terms of hazards for which $\Gamma(\boldsymbol{x},t)$ and $p(\theta;\boldsymbol{x},t)$ are appropriate measures; it will be clear that both approaches can be applied in other cases such as that when dosage, defined in Equation 18.7, is the relevant measure.

(1) If it is believed to be too ambitious to model p itself, the sensible goal is to predict only the magnitudes of μ and σ. As Γ cannot be negative, p cannot be the pdf of a normal distribution; see Figures 18.4 and 18.5. However, it is unlikely that the real distribution is so far from a normal distribution that probabilities obtained with the normal distribution with the same μ and σ will be dangerously different from their true values. This type of approximation can be especially useful for regulatory models expressed in terms of weekly, monthly or even annual averages, when use of the central limit theorem obviates the need for the precise form of $p(\theta;\boldsymbol{x},t)$. The principal goals in this type of research are therefore how μ and σ vary with $\boldsymbol{x}$ and t, and such factors as source geometry, flow characteristics and terrain. In some situations it may be more relevant or practical to use the same broad approach, but to replace μ and/or σ by other robust parameters such as the median concentration or cloud-average values (Sullivan, 1990; Chatwin and Sullivan, 1990b).

(2) An alternative and more ambitious line of work is to seek to model $p(\theta;\boldsymbol{x},t)$ itself in addition to μ and σ. Although successful practical use of Equation 18.7 is unlikely at the present time, the ready availability of data is now allowing the dependence of p on θ to be estimated experimentally. Figures 18.4, 18.5 and (for dosage) 18.6 were obtained in this way. The aim is to find simple two- or three-parameter model pdfs (e.g. log-normal, truncated normal, gamma, beta) that describe the data with adequate accuracy and then, on the basis of simple physical arguments and further validation, to predict how the form of the pdf varies with $\boldsymbol{x},t$ and the other key factors. If this approach is to be successful,

the variation of the form of p will be given completely by the corresponding variations in the parameters. Usually these are chosen to be μ, σ and perhaps one other, often taken to be the intermittency factor. (The intermittency factor is conventionally defined to be the probability that the concentration is greater than zero. In practice this is an extremely useful concept but, from a scientific viewpoint, the definition is not viable (Chatwin and Sullivan, 1989) as the result of applying it depends significantly on extraneous factors such as instrument characteristics. Ride (1984) discusses an important use of the idea of the intermittency factor.)

The remainder of this section summarizes some of the more specific and more important research problems that crucially affect the successful achievement of the two programmes. It is convenient to consider these in two groups: those that largely relate to data and experiments, and those that are predominantly theoretical.

18.5.3 Research problems involving data

The mathematical problems associated with turbulent diffusion are so severe that experimental data must be used in any serious research. It is convenient to distinguish two separate roles for data.

The first is the use of data to test and, hopefully, validate existing models, however obtained. In view of the fact that recent studies (Hanna, Strimaitis and Change, 1991; Britter, 1991; Hanna, 1993; see also Hanna *et al.*, 1992) have analysed the methodology of this process in some detail (in respect of hazardous gas dispersion models), little need be written here. However, three points that emerge from these studies are worth noting. The meaning of the term 'validation' is by no means clear-cut; consequently validation exercises require precise preparation and much subtlety. In general, the performance of a model is 'not related to its cost or complexity' and 'it is very difficult to demonstrate improved model performance as enhancements in model physics are added' (Hanna, 1993). Finally, the work of Hanna and co-workers uses certain measures to compare model predictions with data, and it would be worthwhile to investigate theoretically the statistical significance of the numerical values of these measures and, more generally, to assess similarly other measures of model performance.

The other role for data is in the development of models and, in particular, in research programmes like (1) and (2) above. It is first of all important to point out that the task of data analysis has changed its nature in the last 10 or 15 years principally because of the data acquisition systems that are now routinely used. Consequently, the amount of data available from nearly all experimental investigations has increased by orders of magnitude but, unfortunately, a lack of resources available to most analysts has caused many potentially valuable data sets to be inadequately analysed, or not analysed at all. Sadly, it is not always clear that this increase in the amount of data has been accompanied by a comparable rise in its quality; the studies of validation mentioned above all emphasize the magnitudes of data uncertainties. The authors believe that it is now essential that all experiments on environmental pollution be planned in conjunction with the end-users of the data and that their costs be realistically assessed. Only in this way will these mismatches be stopped.

In terms of the statistical models advocated in this review, particular attention needs to be given in advance to assessing the magnitudes of the standard deviations of experimental estimates of statistical properties such as μ, σ and p. This will lead to a choice, and costing, of the number of repetitions to be carried out or (in cases of statistically steady dispersion)

the length of the experiment. Also, in view of the high cost of field trials involving many repetitions, it is inevitable that there will be even more use of wind and water tunnels in the future so that there must be more research into modelling complex environmental flows in the laboratory.

There is now a lot of evidence that the measured properties of $\Gamma(x,t)$ can be significantly dependent on the instruments used. In particular (see Figure 18.1), the magnitudes of the real concentration fluctuations, as measured, for example, by $\sigma(x,t)$, may be substantially underestimated; see, for example, Figures 5 and 7 of Chatwin and Goodall (1991). The principal cause of this is instrument smoothing which, though largely inevitable because of the minute length and time-scales present in the structure of dispersing pollutants, has arguably received inadequate attention in the past. There is a strong case for using at least two instrumentation systems of different types and characteristics in experiments so that the measurements of Γ can be cross-validated directly.

One consequence of instrument smoothing is to underestimate the magnitudes of the highest values of Γ and their frequency of occurrence; this may clearly have serious consequences of hazard assessment. In other instances, particularly those relating to overall air and water quality, the biggest contribution to dosage comes from the lowest values of Γ because they occur so often. It is important in such circumstances to be able to estimate the behaviour of $p(\theta;x,t)$ as $\theta \to 0+$. Unfortunately, and again inevitably, such estimation involves other important instrument effects, especially noise and, often, baseline drift. In the past such effects were too often dismissed in a cavalier fasion by applying arbitrary thresholds: fortunately, this practice appears to be dying rapidly. The proper treatment of noise requires its separate measurement and consequent deconvolution from the output signal; methods are available for this. The effects considered in this paragraph and the previous one have been considered by Mole (1990a; 1990b), but much remains to be done.

18.5.4 Research problems involving theory

Some theoretical problems have recently been discussed by us (Chatwin and Sullivan, 1993). These include: (i) extending the relationship between μ and σ proposed by Chatwin and Sullivan (1990a) to more general situations such as those near a steady continuous source; (ii) use of the representation of $p(\theta;x,t)$ in terms of the new definition of the intermittency factor (Chatwin and Sullivan, 1989); and (iii) investigation of the behaviour of $p(\theta;x,t)$ as $\theta \to 0+$ using both Equation 18.7 and the representation mentioned in (ii). Further details of these problems are contained in the cited references and will not be repeated here.

Two other classes of theoretical problem also merit brief mention.

One of these is the influence of source size and geometry on the statistical properties of Γ. It has been known for some time (Chatwin and Sullivan, 1979) that source size has a potentially profound effect on the size of σ (the smaller the source the larger is σ), but many intriguing questions remain. For example, it would be valuable to quantify the effects of molecular diffusion and source characteristics on σ and p, and to obtain a stronger theoretical basis for the robust correlation between μ and σ mentioned in (i) above. Some applications require models for multiple sources and work is in progress to extend the results obtained for single sources such as those cited above. This problem also has fundamental interest and data have been presented in papers by Warhaft (e.g. Warhaft, 1984) and others.

The second class of theoretical problem is essentially statistical rather than physical. Some such problems have been mentioned earlier, especially in the subsection on research problems

involving data. Another interesting question is related to programme (1) discussed earlier, and that is to apply recent statistical research on extreme values, involving the Pareto distribution, to practical problems in which the appropriate measure of harm involves high values of concentration; see, for example, Smith (1989). Finally, there are interesting questions associated with the efficient estimation of $p(\theta;x,t)$ from data. Among these are the length of record needed to estimate statistical properties (such as μ and σ) to within acceptably small standard errors, the estimation of noise, the deconvolution of noise, and the criteria governing the use of parametric or non-parametric methods. Such problems are discussed by Mole and Jones (in press), where further references are given.

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REFERENCES

Allen, C.M. (1982) Numerical simulation of contaminant dispersion in estuary flows. *Proc. R. Soc. Lond. A* **381**, 179–194.

Birch, A.D., Brown, D.R. and Dodson, M.G. (1980) Ignition probabilities in turbulent mixing flows. *Rep. No. MRS E 374*, Midlands Research Station, British Gas, Solihull.

Britter, R.E. (1991) The evaluation of technical models used for major-accident hazard installations. *Report to Commission of the European Communities (DG XII)*.

Chatwin, P.C. (1990) Statistical methods for assessing hazards due to dispersing gases. *Environmetrics* **1**, 143–162.

Chatwin, P.C. and Allen, C.M. (1985a) Mathematical models of dispersion in rivers and estuaries. *Annu. Rev. Fluid Mech.* **17**, 119–149.

Chatwin, P.C. and Allen, C.M. (1985b) A note on time averages in turbulence with reference to geophysical applications. *Tellus* **37B**, 46–49.

Chatwin, P..C. and Allen, C.M. (1985c) Reply. *Tellus* **37B**, 315–316.

Chatwin, P.C. and Goodall, G.W. (1991) Research on continuous and instantaneous heavy gas clouds. *Report to Commission of the European Communities: Final Report on CEC Contract No. EV4T. 0025. UK(H)*. [Also Brunel University Department of Mathematics and Statistics Tech. Rep. TR/02/91.]

Chatwin, P.C. and Sullivan, P.J. (1979) The relative diffusion of a cloud of passive contaminant in incompressible turbulent flow. *J. Fluid Mech.* **91**, 337–355.

Chatwin, P.C. and Sullivan, P.J. (1989) The intermittency factor of scalars in turbulence. *Phys. Fluids A* **1**, 761–763.

Chatwin, P.C. and Sullivan, P.J. (1990a) A simple and unifying physical interpretation of scalar fluctuation measurements from many turbulent shear flows. *J. Fluid Mech.* **212**, 533–556.

Chatwin, P.C. and Sullivan, P.J. (1990b) Cloud-average concentration statistics. *Maths. & Computers in Simulation* **32**, 49–57.

Chatwin, P.C. and Sullivan, P.J. (1993) The structure and magnitude of concentration fluctuations. *Boundary-Layer Meteorol.* **62**, 269–280.

Davies, J.K.W. (1990) Research on continuous and instantaneous heavy gas clouds. *Report to Commission of the European Communities: Final Report on CEC Contract No. EV4T. 0005. UK(H)*.

Deardorff, J.W. and Willis, G.D. (1984) Groundlevel concentration fluctuations from a buoyant and a non-buoyant source within a laboratory convectively mixed layer. *Atmos. Environ.* **18**, 1297–1309.

Griffiths, R.F. and Harper, A.S. (1985) A speculation on the importance of concentration fluctuations in the estimation of toxic response to irritant gases. *J. Hazardous Mater.* **11**, 369–372.

Griffiths, R.F. and Megson, L.C. (1984) The effect of uncertainties in human toxic response on hazard range estimation for ammonia and chlorine. *Atmos. Environ.* **18**, 1195–1206.

Hanna, S.R. (1993) Uncertainties in air quality model predictions. *Boundary-Layer Meteorol.* **62**, 3–20.

Hanna, S.R., Chatwin, P.C., Chikhliwala, E., Londergan, R.J., Spicer, T.O. and Weil, J.C. (1992) Results from the model evaluation panel. *Plant/Operations Progr.* **11**, 2–5.

Hanna, S.R., Strimaitis, D.G. and Chang, J.C. (1991) Evaluation of 14 hazardous gas dispersion models with ammonia and hydrogen fluoride field data. *J. Hazardous Mater.* **26**, 127–158.

Koopman, R.P., Cederwall, R.T., Ermak, D.L., Goldwire, H.C. Jr., Hogan, W.J. McClure, J.W., McRae, T.G., Morgan, D.L., Rodean, H.C. and Shinn, J.H. (1982) Analysis of Burro series $40m^3$ LNG spill experiments *J. Hazardous Mater.* **6**, 43–83.

Mole, N. (1990a) A model of instrument smoothing and thresholding in measurements of turbulent dispersion. *Atmos. Environ.* **24A**, 1313–1323.

Mole, N. (1990b) Some intersections between turbulent dispersion and statistics. *Environmetrics* **1**, 179–194.

Mole, N. and Chatwin, P.C. (1990) Fluctuations in atmospheric contaminants. *Final Report to Chemical Defence Establishment under Agreement No. 2006/71.* [Also Brunel University Department of Mathematics and Statistics Tech. Rep. TR/16/90.]

Mole, N. and Jones, C.D. Concentration fluctuation data from dispersion experiments carried out in stable and unstable conditions. *Boundary-Layer Meteorol.*, in press.

Monin, A.S. and Yaglom, A.M. (1971) *Statistical Fluid Mechanics: Mechanics of Turbulence.* Vol. I (Ed. J.L. Lumley). MIT Press, Cambridge, Mass.

Ride, D.J. (1984) An assessment of the effects of fluctuations on the severity of poisoning by toxic vapours. *J. Hazardous Mater.* **9**, 235–240.

Smith, R.L. (1989) Extreme value analysis of environmental time series: an application to trend detection in ground-level ozone. *Stat. Sci.* **4**, 367–393.

Sullivan, P.J. (1971) Longitudinal dispersion within a two-dimensional turbulent shear flow. *J. Fluid Mech.* **49**, 551–576.

Sullivan, P.J. (1990) Physical modeling of contaminant diffusion in environmental flows. *Environmetrics* **1**, 163–177.

Warhaft, Z. (1984) The interference of thermal fields from line sources in grid turbulence. *J. Fluid Mech.* **144**, 363–387.

Weil, J.C., Sykes, R.I. and Venkatram, A. (1992) Evaluating air quality models: review and outlook. *J. Appl. Meteorol.* **31**, 1121–1145.