INTERNATIONAL ATOMIC ENERGY AGENCY
UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION
INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
I.C.T.P., P.O. BOX 586, 34100 TRIESTE, ITALY, CABLE: CENTRATOM TRIESTE

UNITED NATIONS INDUSTRIAL DEVELOPMENT ORGANIZATION
INTERNATIONAL CENTRE FOR SCIENCE AND HIGH TECHNOLOGY
c/o INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS 34100 TRIESTE (ITALY) VIA GRIGNANO, 9 (ADRIATICO PALACE) P.O. BOX 586 TELEPHONE 040-224572 TELEFAX 040-224575 TELEX 460449 APH I

SMR/760-16

"College on Atmospheric Boundary Layer
and Air Pollution Modelling"
16 May - 3 June 1994

Lecture 1: "What Are Concentration Fluctuations & Why Are They Important"
Lecture 2: "How Should We Quantify Air Pollution"?
Lecture 3: "Mathematical Models and Data"
Lecture 4: "Simple Probabilistic Models and Recent Development"

P.C. CHATWIN
University of Sheffield
School of Mathematics and Statistics
Sheffield, United Kingdom

Please note: These notes are intended for internal distribution only.

Lectures by

PROFESSOR P C CHATWIN

ICTP, Trieste

May 1994

Lecture 1

What are concentration fluctuations and why are they important?

§1.1 *TURBULENCE AND ATMOSPHERIC DISPERSION*

Turbulence is very important in practical problems, but it is also still impossible to explain it properly in terms of mathematics and physics. By "properly" is meant the accurate quantification that is the goal of any serious scientific investigation. For these reasons, there are still lots of different approaches being pursued to try to model phenomena involving turbulence; many of these are interesting and appear potentially useful. In these lectures I am not going to attempt a comprehensive review; for one thing there is not enough time. Rather my focus will be on what I believe to be a promising approach towards increased **scientific** understanding of how fluids in turbulent motion (and, in particular, the atmosphere) transport and mix pollutants. This approach is one that Nils Mole, Paul Sullivan and I have been developing with our coworkers over several years at Brunel University (until the end of 1990), the University of Sheffield (from the beginning of 1991) and the University of Western Ontario (for ever - or so it seems!)

The Earth's atmosphere is always in **turbulent** motion. This means that the air velocity $\mathbf{\Upsilon}(\boldsymbol{x},t)$ at every point $\boldsymbol{x}$ and every time t is a **random variable.** This fact is a matter of everyday experience. However, I want to emphasize first that it is not in contradiction with the further fact that the evolution of $\mathbf{\Upsilon}(\boldsymbol{x},t)$ with $\boldsymbol{x}$ and t is governed by the laws of mechanics including, in particular, mass conservation and Newton's laws. The first of these is almost always adequately approximated in the atmosphere by the **continuity equation**

$$\nabla.\mathbf{\Upsilon}=0, \qquad (1)$$

and the second by the **Navier-Stokes equations**, often in a form like

$$\frac{\partial\mathbf{\Upsilon}}{\partial t}+(\mathbf{\Upsilon}.\nabla)\mathbf{\Upsilon}=-\frac{1}{\rho}\nabla p+\boldsymbol{F}+\nu\nabla^2\mathbf{\Upsilon}, \qquad (2)$$

where p,ρ,ν and $\boldsymbol{F}$ are the pressure, density, kinematic viscosity and body force (e.g. Coriolis force and/or gravity) respectively. I shall not need to use (2) in detail in these lectures, so I shall not discuss the relative importance of the various terms in different circumstances. But what I do want to emphasize is that much work in atmospheric turbulence focusses, sometimes exclusively, on the **mean velocity**, which I shall denote by $\boldsymbol{U}(\boldsymbol{x},t)$, and the mean velocity does **not** satisfy (2). Rather, $\boldsymbol{U}(\boldsymbol{x},t)$ satisfies **Reynolds equations**, which are derived from (2) by a process like that often used to derive the equation for the mean concentration.

As a consequence of the randomness of the velocity field, the concentration $\Gamma(\boldsymbol{x},t)$ of a pollutant dispersing in the atmosphere is also random. The pollutants considered in these lectures will be those, a substantial proportion in practice (if not a vast majority), whose concentrations are determined by two processes, namely the random transport (advection) by the air, and molecular diffusion. I shall not, in particular, consider chemical processes. The equation governing $\Gamma(\boldsymbol{x},t)$ is then

$$\frac{\partial\Gamma}{\partial t}+(\mathbf{\Upsilon}.\nabla)\Gamma=\kappa\,\nabla^2\Gamma, \qquad (3)$$

where κ is the **molecular diffusivity**. Associated with (3) in each application will be boundary and initial conditions (in general).

Some typical data records of concentrations are given in *Figure 1*. While there are interesting and important differences between these records, some of which will be discussed later, each of them clearly indicates the randomness of $\Gamma(\boldsymbol{x},t)$ caused by the term involving $\mathbf{\Upsilon}$ in (3). Given that the concentration of a pollutant in the atmosphere is random, *i.e.* **unpredictable, non-deterministic,** it is necessary to consider whether the randomness is important, both scientifically and practically, and, if so, how it can be dealt with.

§ 1.2 *MATHEMATICAL FRAMEWORK*

Whether or not the randomness of $\Gamma(\boldsymbol{x},t)$ is important, the use of probabilistic concepts is obviously essential in any satisfactory quantitative description. Such concepts can be applied only in relation to an underlying population, or **ensemble.** For the situation of atmospheric dispersion that we are considering, the ensemble is a precisely defined set of **releases** or "experiments". The definition serves only to determine unambiguously whether or not any particular release is within the ensemble. From the scientific point of view, what the definition is does not matter; from the practical point of view, it is crucial. In the latter case the definition will specify obvious points like the source location(s), relevant local geography *etc.* but it must also make clear whether, for example, the population includes releases at night time, when it is raining *etc.* Further discussion of the ensemble concept is given by Chatwin (1982) and Carn and Chatwin (1985).

For a given ensemble, there is a **probability density function** (pdf) for $\Gamma(\boldsymbol{x},t)$. The pdf will be denoted by $p(\theta;\boldsymbol{x},t)$, where the variable θ ranges over all the values that Γ can take. In the standard manner $p(\theta;\boldsymbol{x},t)$ is defined by

$$p(\theta;\boldsymbol{x},t) = \frac{d}{d\theta}\{\text{prob}\,[\Gamma(\boldsymbol{x},t) \le \theta]\}, \tag{4}$$

and it follows that $p(\theta;\boldsymbol{x},t)\delta\theta$ is, for $\delta\theta$ small and positive, the probability that $\theta \le \Gamma(\boldsymbol{x},t) < \theta + \delta\theta$. There are some obvious properties that $p(\theta;\boldsymbol{x},t)$ must satisfy. Since concentrations are non-negative:

$$p(\theta;\boldsymbol{x},t) = 0 \;\; \forall \theta < 0, \tag{5}$$

and, since p is a pdf (i.e. since $\Gamma(\boldsymbol{x},t)$ must have some value):

$$\int_0^\infty p(\theta;\boldsymbol{x},t)\,d\theta = 1. \tag{6}$$

In fact there is, of course, for each ensemble and for each $\boldsymbol{x}$ and t, a maximum possible value Θ that Γ can take, and the upper limit in (6) can be replaced by Θ. But this is not usually useful since Θ is unknown. (However, note that for a given ensemble, $\Theta = \Theta(\boldsymbol{x},t)$ is determined by (3) and its boundary conditions and by the statistical properties of $\mathbf{Y}$; the problem is that, like all other statistical properties of Γ, it is not known **how** to determine it.)

In these lectures (unlike the corresponding ones I gave here in Trieste in 1990), I shall use standard statistical symbols for the most important properties of the concentration. This is unconventional, as yet, in turbulence and turbulent diffusion but is justified for many reasons, including elegance; see p356 of Chatwin and Sullivan (1994). Thus the **mean concentration** will be denoted by $\mu = \mu(\boldsymbol{x},t)$ and, by definition, is related to p by

$$\mu(\boldsymbol{x},t) = \int_0^\infty \theta p(\theta;\boldsymbol{x},t)\,d\theta. \tag{7}$$

The **concentration fluctuation** $c(\boldsymbol{x},t)$ is then defined by

$$c(\boldsymbol{x},t) = \Gamma(\boldsymbol{x},t) - \mu(\boldsymbol{x},t). \tag{8}$$

The **variance** of $\Gamma(\boldsymbol{x},t)$ will be denoted by $\sigma^2 = \sigma^2(\boldsymbol{x},t)$, and is the mean square of $c(\boldsymbol{x},t)$. Thus

$$\sigma^2(\boldsymbol{x},t) = \int_0^\infty [\theta - \mu(\boldsymbol{x},t)]^2 p(\theta;\boldsymbol{x},t)\,d\theta = \left\{\int_0^\infty \theta^2 p(\theta;\boldsymbol{x},t)\,d\theta\right\} - \mu^2(\boldsymbol{x},t). \tag{9}$$

(More conventional symbols for Γ, μ and σ^2 in research papers on turbulent diffusion are, for example, C, $\overline{C}$ and $\overline{c'^2}$ respectively.)

§ 1.3 *REMARKS ON TIME AVERAGES ETC.*

It requires emphasis first that, as the notation demonstrates, $p(\theta;\boldsymbol{x},t)$ does depend explicitly on $\boldsymbol{x}$ and t for most ensembles; so therefore do statistical properties like $\mu(\boldsymbol{x},t)$ and $\sigma^2(\boldsymbol{x},t)$. This is true, for instance, when the ensemble is concerned with dispersion following the sudden release (caused, for example, by an accident) of a finite quantity of pollutant into the atmosphere. In such a case the pollutant cloud spreads (but randomly!) and therefore dilutes as the time since release increases. This is evident from *Figures 2 (a) & 2 (b)*.

Estimates of statistical properties like $\mu(\boldsymbol{x},t)$ and $\sigma^2(\boldsymbol{x},t)$ must in general be made by taking appropriate averages of the results of many repeat experiments, and this is how the graphs in *Figures 2 (c) & 2 (d)* were obtained. It is clear that both μ and σ are strongly dependent on t (for a fixed $\boldsymbol{x}$).

Nevertheless there are some ensembles where $p(\theta;\boldsymbol{x},t)$ does not depend explicitly on t, or where this is a reasonable practical approximation. This has to be regarded as an exceptional circumstance, but may be appropriate, for example, in considering the environmental consequences of a continuous release of material like smoke from a factory chimney. Clearly the weather characteristics and the rate of release of material must not, on average, change with time during the dispersion period that is of interest. Such ensembles are called **statistically stationary**. Although statistical properties like p, μ and σ^2 can still be estimated by averages over repeat experiments, it is much more convenient (and much cheaper) to obtain such estimates then by exploiting a property that holds for statistically stationary ensembles, namely that means can be estimated from the results of one experiment by appropriate time averaging. For then, $p(\theta;\boldsymbol{x},t) = p(\theta;\boldsymbol{x})$, $\mu(\boldsymbol{x},t) = \mu(\boldsymbol{x})$ etc., and $\mu(\boldsymbol{x})$, for example, can be estimated by $\hat{\mu}(\boldsymbol{x};T)$, where

$$\hat{\mu}(\boldsymbol{x};T) = \frac{1}{2T}\int_{t-T}^{t+T}\Gamma(\boldsymbol{x},s)ds. \qquad (10)$$

Because statistically stationary ensembles are the simplest type conceptually, and because they are the cheapest to investigate experimentally, there has been undue emphasis on their properties. For example, all the data records in *Figure 1* are from such ensembles. Partly for this reason, it is often assumed that all means considered in turbulence and turbulent diffusion are time averages. Such an assumption is wrong, and potentially dangerous.

The points above were dealt with at greater length in my earlier lectures here at Trieste (Chatwin 1990, p3); see also pp 356-7 of Chatwin and Sullivan (1994).

§ 1.4 *THE IMPORTANCE OF CONCENTRATION FLUCTUATIONS*

All experimental evidence known to me shows that the degree of unpredictability in Γ is not small. One measure of unpredictability is the **intensity** $I(\boldsymbol{x},t)$, where

$$I(\boldsymbol{x},t) = \sigma(\boldsymbol{x},t)/\mu(\boldsymbol{x},t), \qquad (11)$$

and μ & σ are defined above in (7) & (9) respectively. Values of I are typically of order unity and greater. Some typical results from experiments were shown in *Figures 3, 4 & 5* of my previous lectures here (Chatwin 1990), and *Figure 3* of the present lectures shows estimates of I for the experiments leading to *Figure 2*.

Another way of recognizing that the degree of unpredictability in Γ is significant is to look at estimates of $p(\theta;\boldsymbol{x},t)$. Were the process purely deterministic, Γ would always equal μ, σ would be zero and p would have the form

$$p(\theta;\boldsymbol{x},t) = \delta[\theta - \mu(\boldsymbol{x},t)]. \qquad (12)$$

i.e. p would have an (infinite) spike at $\theta = \mu$ and would be zero everywhere else. Experimental estimates of p known to me never have anything like this form and, in particular, they are spread out over a substantial range of values of θ. Examples are given in *Figures 4, 5 & 6*.

It follows that, scientifically, the study of concentration fluctuations is interesting and important.

More relevant for present purposes, perhaps, is that concentration fluctuations are practically important. The evidence both for flammable gases and for toxic gases given here in 1990 (Chatwin 1990, pp 4 - 5) will not be repeated. Reference may also be made to Mole, Chatwin and Sullivan (1993, pp 318 - 322) and to Chatwin and Sullivan (1994, pp 358 - 364) for more recent discussions. There is, in summary, no doubt that a scientifically satisfactory assessment of any hazard involving a dangerous gas dispersing in the atmosphere should involve proper consideration of concentration fluctuations. Conversely, I believe strongly that there is no hazard for which satisfactory assessments of danger can be made on the basis of the mean concentration $\mu(\boldsymbol{x},t)$ alone. Unfortunately this does not yet seem to be understood, especially by governments and their agents!

It also seems, however, that not enough is known about what measure or measures of the dispersing gas concentration determine the degree of harm in any particular case. For toxic gases, for example, a conventional measure is the **dosage** $D(\boldsymbol{x},t)$, defined by

$$D(\boldsymbol{x},t) = \int_{-\infty}^{t} \Gamma(\boldsymbol{x},s)ds. \tag{13}$$

Yet there appears to be little hard medical evidence that this is the correct measure. See p320 of Mole, Chatwin and Sullivan (1993) for further discussion of this point, and consideration of other related measures. (In view of the earlier part of the lecture, it is relevant to note that D is itself a random variable. This is illustrated in *Figure 7*, derived from the data shown earlier in *Figures 2 & 3*.)

§ 1.5 *FURTHER READING*

Many of the themes in this lecture are developed further in Mole, Chatwin and Sullivan (1993), and Chatwin and Sullivan (1994). The former review discuss several other approaches to modelling concentration fluctuations than that which will be emphasized here.

Lecture 2

How should we quantify air pollution?

§ 2.1 *INTRODUCTION*

By now it will perhaps be clear that I believe that eventually atmospheric dispersion (and, more generally, all phenomena involving turbulence and turbulent diffusion) will be described in terms of probabilities since not to do so would be scientifically misleading, if not wrong. This implies that, one day, regulatory models will be estimating probabilities (not concentrations) in response to questions like "What is the probability that following an accidental release of CH_4 (*say*), the gas-air mixture is ignitable at **50m** (*say*) downwind?" In order for such a view to be acceptable, there is obviously a need for re-education of involved professions like politicians and lawyers and of the general public; but this would be no bad thing since we live in a stochastic world, not a deterministic one.

Unfortunately there is also a need for re-educating some scientists and engineers who, it must be said, should know better. One problem is that the massive and continual increase in computing power has enabled many hitherto intractable problems to be solved; it is my contention that, unfortunately, many of the problems being solved have little scientific or practical value. A second problem is, perhaps, deeper (even cultural), and this is that some scientists and engineers appear incapable of accepting the stochastic nature of atmospheric dispersion; perhaps this is due to faults in their training in terms, exclusively, of deterministic models. But there certainly seems to be a mental block in the minds of such people in that they cannot conceive that science is other than deterministic and that (therefore) anything involving probability or statistics must be unscientific. I am going to spend much of this lecture demonstrating that this view is, quite simply, wrong.

§ 2.2 *THE EQUATION FOR $p(\theta;\boldsymbol{x},t)$ AND SOME CONSEQUENCES*

It is necessary, first, to introduce the concept of **expected value** which generalises the idea of a mean. Let $f[\Gamma(x,t)]$ be any function of $\Gamma(x,t)$. Then the expected value of f, denoted by $E\{f\} = E\{f[\Gamma(x,t)]\}$, is defined by

$$E\{f\} = \int_0^\infty f(\phi)p(\phi;x,t)d\phi, \tag{14}$$

where $p(\phi;x,t)$ is the pdf of $\Gamma(x,t)$ introduced in *Lecture 1* (and the use of ϕ rather than θ is for later mathematical convenience). It follows directly from (14) that the mean concentration $\mu(x,t)$ and its variance $\sigma^2(x,t)$, defined in (7) and (9) respectively, satisfy

$$\mu(x,t) = E\{\Gamma(x,t)\};\ \sigma^2(x,t) = E\{[\Gamma(x,t) - \mu(x,t)]^2\}. \tag{15}$$

Moreover, the choice of $f[\Gamma(x,t)] = \delta[\Gamma(x,t) - \theta]$ in (14) shows that

$$p(\theta;x,t) = E\{\delta[\Gamma(x,t) - \theta]\}. \tag{16}$$

Equation (16) provides my starting-point in obtaining the equation satisfied by $p(\theta;x,t)$ From (16)

$$\frac{\partial p}{\partial t} = E\left\{\frac{\partial\Gamma}{\partial t}\delta'[\Gamma(x,t) - \theta]\right\}, \tag{17}$$

so that, from (3),

$$\frac{\partial p}{\partial t} = -E\left\{Y_j\frac{\partial\Gamma}{\partial x_j}\delta'[\Gamma(x,t)-\theta]\right\} + E\left\{\kappa\frac{\partial}{\partial x_j}\left(\frac{\partial\Gamma}{\partial x_j}\right)\delta'[\Gamma(x,t)-\theta]\right\}. \tag{18}$$

(It is worth noting, for clarity, that the expected values on the right hand side of (18) are defined by equations like (14) but involving pdfs other than p; for example, the first term involves the **joint pdf** of **Y** and Γ.) After integration by parts, (18) becomes

$$\frac{\partial p}{\partial t} = -E\left\{Y_j\frac{\partial\Gamma}{\partial x_j}\delta'[\Gamma(x,t)-\theta]\right\} + E\{\kappa\nabla^2\delta[\Gamma(x,t)-\theta]\} - E\{\kappa(\nabla\Gamma)^2\delta''[\Gamma(x,t)-\theta]\}. \tag{19}$$

Use of (1) and (16) then gives

$$\frac{\partial p}{\partial t} + \nabla.E\{\mathbf{Y}\delta[\Gamma(x,t)-\theta]\} = \kappa\nabla^2 p - \kappa\frac{\partial^2}{\partial\theta^2}E\{(\nabla\Gamma)^2\delta[\Gamma(x,t)-\theta]\}. \tag{20}$$

This equation may also be written in terms of conditional probabilities (Mole, Chatwin and Sullivan 1993). Partly because p is a non-linear function of θ, and partly because of the term $(\mathbf{Y}.\nabla)\Gamma$ in (3) involving the product of two random variables, (20) exhibits the notorious **closure problem**, namely that this equation for $p(\theta;x,t)$ involves higher-order unknowns (such as the joint pdf of **Y** and Γ), and that the equations for these involve further unknowns, and so on.

But this formidable complication is a familiar one to all who work with phenomena involving turbulence. It does not weaken the fact that (2), and its derivation, show that $p(\theta;x,t)$ is determined by the basic physics (i.e. equations (1), (2) and (3), and the associated boundary conditions) just as surely as $\mu(x,t)$ and $\sigma^2(x,t)$ are.

It is therefore instructive to note that the familiar equations for $\mu(x,t)$ and $\sigma^2(x,t)$ can be derived directly from (20), using (7) and (9) and some algebra. The results are

$$\frac{\partial\mu}{\partial t} + (U.\nabla)\mu + \nabla.E\{uc\} = \kappa\nabla^2\mu, \tag{21}$$

and

$$\frac{\partial\sigma^2}{\partial t} + (U.\nabla)\sigma^2 + \nabla.E\{uc^2\} + 2\nabla\mu.E\{uc\} = \kappa\nabla^2\sigma^2 - 2\kappa E\{(\nabla c)^2\}, \tag{22}$$

where c is the concentration fluctuation defined in (8), $U = U(x,t) = E\{\mathbf{Y}(x,t)\}$ is the mean velocity, and $u(x,t) = \mathbf{Y}(x,t) - U(x,t)$ is the **velocity fluctuation.** (The standard derivation of (21) and (22) uses **Reynolds decomposition,** *viz.* $\Gamma = \mu + c$, $\mathbf{Y} = U + u$, directly in (3).)

In large measure, the history of atmospheric dispersion research consists of attempts to "resolve" the closure problem by developing acceptably accurate models of the "difficult" terms in (21) and (22), *viz*. those involving $E\{\mathbf{u}c\}$, $E\{\mathbf{u}c^2\}$ and $E\{(\nabla c)^2\}$. By far the most attention has been on $E\{\mathbf{u}c\}$, and I am sure that other lecturers will discuss some of these models, beginning with eddy diffusivity concepts and progressing to more complicated ideas involving high-order closures. So I am not going to follow this route any further for reasons given at the beginning of §2.4 below.

§2.3 *DIRECT METHODS*

At some time in the future, the closure problem will be circumvented because it will be possible to solve the governing equations (1), (2) and (3) directly (**DNS** = direct numerical simulation) using powerful computers. Because the flow is turbulent, each solution will be (significantly) different, the differences being generated for example by minute differences in input data or by rounding errors in the computer arithmetic. Enough solutions will have to be obtained to allow reliable statistical estimates to be obtained, and experience suggest that acceptable estimates of $p(\theta;\mathbf{x},t)$ may require several hundred solutions. Moreover, each solution must resolve length scales extending from $0(10^{-4}\text{m})$ – the scale of the conduction cut-off length and the Kolmogoroff microscale – to $0(10^4\text{m})$ – the scale of the energy - containing eddies. As well as spatial resolution in all three dimensions, each solution must provide adequate temporal resolution. Estimates suggest that economically feasible DNS will require computers many orders of magnitude (greater than 10^7) faster than the most powerful computers available today (Mole, Chatwin and Sullivan 1993, p323), and it seems unlikely that such computers will be available within the lifetime of anyone here. In addition, the outcomes of such massive computing power will not, in themselves, constitute understanding. This will require a further massive use of resources both for interpretation and for presentation in "digestible" form.

§ 2.4 *MODELS OF* $p(\theta;\mathbf{x},t)$

In line with the philosophy that I have tried to explain, the rest of these lectures are going to deal with models of $p(\theta;\mathbf{x},t)$. I shall be considering $\mu(\mathbf{x},t)$ and $\sigma^2(\mathbf{x},t)$ because they are two of the most important properties of p, and not because they have independent significance. In this connection, it is worth pointing out that the mean concentration $\mu(\mathbf{x},t)$ satisfies (21), but **not** (3) - it is not a concentration distribution that can ever be observed.

Other lecturers are likely to discuss some of the closure hypotheses adopted for (21) and (22), particularly the former. Some similar approaches have been used for equation (20) for $p(\theta;\mathbf{x},t)$ itself; see *e.g.* Pope (1985). I do not believe that anyone would claim that these have been totally successful (or even moderately successful for (20) and (22)). Moreover the use of such hypotheses leads to heavy computing and/or rather complicated algebra.

The rapid and continual increase in computing power tends to encourage the development of mathematically complicated models. Recent emphasis on the problems associated with assessing the performance of mathematical models is therefore timely and welcome. Notable contributions have been made by two of next week's lecturers, Steve Hanna (Hanna *et al* 1991, Hanna *et al* 1992, Hanna 1993) and Rex Britter (Britter 1991). I hope they will refer to this work but, in case they don't, I cannot resist including some quotes which indicate a degree of scepticism about some models of atmospheric dispersion (and in many other research fields) which I share.

> (1) *"In a broad sense, there is generally an over-confidence in the ability of mathematical models to provide accurate prediction. In particular, there is significant over-confidence in the ability of very large numerical models to provide accurate prediction"* (Britter 1991);
>
> (2) The performance of a model is *"not related to its cost or complexity"* (Hanna 1993);

(3) *"It is very difficult to demonstrate improved model performance as enhancements in model physics are added"* (Hanna 1993).

§ 2.5 *SIMPLE PHYSICAL MODELS OF* $p(\theta;\boldsymbol{x},t)$.

I am therefore going to consider simple models in the remainder of these lectures based not on (20) but on elementary physics. Before describing these models, it is worth noting that simple models are **quick** models - an important consideration for practical response to real accidents.

For clarity and simplicity, suppose that the **source** concentration is **uniform** (the same at all places in the source) and equal to θ_1. If the release is not instantaneous it will also be assumed that the source concentration is θ_1 for all times. This is nearly always a good approximation in practice; moreover the results below can be generalised fairly easily for non-uniform sources (Chatwin and Sullivan 1989, Sawford and Sullivan 1994). In these circumstances, the concentration within an element of fluid (air) **immediately** following release is either θ_1 (source fluid) or 0 (ambient fluid). Subsequently advection - the term $(\mathbf{Y}.\nabla)\Gamma$ in (3) - transports and distorts each fluid element but does not change the concentration within it. Changes in concentration occur only through **molecular diffusion** - the term $\kappa\nabla^2\Gamma$ in (3).

However molecular diffusion is, in some respects, a weak process and a slow process; for example the order of magnitude of the term $\kappa\nabla^2\mu$ in (21) is nearly everywhere much less than that of the term $\nabla.E(\boldsymbol{u}c)$. It is therefore of interest to consider the **hypothetical** situation in which there is no molecular diffusion *i.e.* $\kappa=0$ in (3). It can be anticipated that the results obtained will be relevant in real situations sufficiently close to the source, even, perhaps, elsewhere. When $\kappa=0$, the concentration in a fluid element is (for all $\boldsymbol{x}$ and t) either θ_1 or 0. Hence $p(\theta;\boldsymbol{x},t)$ is equal to $p_0(\theta;\boldsymbol{x},t)$, where

$$p_0(\theta;\boldsymbol{x},t)=\pi_0(\boldsymbol{x},t)\,\delta(\theta-\theta_1)+\{1-\pi_0(\boldsymbol{x},t)\}\,\delta(\theta). \tag{23}$$

(In (23), and the sequel, the zero subscript always denotes the case of zero molecular diffusivity.) The function $\pi_0(\boldsymbol{x},t)$ is determined by the statistical properties of the velocity field $\mathbf{Y}$ and by factors like geometry. Evidently

$$\pi_0(\boldsymbol{x},t)=\mathrm{prob}\,[\Gamma_0(\boldsymbol{x},t)>0], \tag{24}$$

(where Γ_0 is the value of Γ, in accordance with the convention noted above) and so π_0 is the **intermittency factor** in this hypothetical situation.

It follows from (23), using (7) and (9), that

$$\mu_0=\theta_1\pi_0\ ,\ \sigma_0^2=\theta_1^2\pi_0(1-\pi_0), \tag{25}$$

and hence that (Chatwin and Sullivan 1990)

$$\sigma_0^2=\mu_0(\theta_1-\mu_0)=\left(\tfrac{1}{2}\theta_1\right)^2-\left(\mu_0-\tfrac{1}{2}\theta_1\right)^2. \tag{26}$$

Figure 8 illustrates some of the consequences of these formulae schematically. They will be developed further in the next two lectures, and it will be shown in particular that, when generalised, (26) has wide relevance.

§ 2.6 *FURTHER READING*

I think it is important to read either Britter (1991) or Hanna (1993) to appreciate the difficulties involved in evaluating models of atmospheric dispersion, and the strength of the evidence for the quotations given above at the end of § 2.4. For more detail on the mathematics in § 2.2, see Pope (1985).

Lecture 3

Mathematical models and data

§ 3.1 *THE EFFECTS OF MOLECULAR DIFFUSION: I BASIC PHYSICS*

Molecular diffusion has several interrelated effects that invalidate the results in § 2.5. Most obviously it reduces the concentration in a fluid element originating in the source from θ_1 to a value that is (a) random (since different fluid elements are distorted differently by the turbulent velocity field and this distortion affects the magnitude of $\kappa\nabla\Gamma$), and (b) tends to decrease with time and with distance from the source. This process increases the concentration in fluid elements not originating in the source from zero to positive values. Also molecular diffusion **dissipates** concentration fluctuations, by which is meant that σ^2 decreases with t because the term $-2\kappa E\{(\nabla c)^2\}$ on the right hand side of (22) is essentially negative.

It is known (Batchelor 1959) that the characteristic length scale on which these processes take place is of order $(\nu\kappa^2/\varepsilon)^{1/4}$, where ε is the local rate of dissipation of mechanical energy per unit mass into heat. This length scale is known as the **conduction cut-off length** and is very small, of $O(10^{-4}\mathrm{m})$ in the atmosphere.

While these processes are fundamental, the facts that the conduction cut-off length is much less than the length scales of the energy - containing eddies which, of course, dominate the changes in concentration at a fixed point, and that molecular diffusion is (as already noted in § 2.5) a slow weak process, suggest that the **structure** of (23), (25) and (26) may well be applicable even in real situations when $\kappa \neq 0$. Consider (26), *viz.*

$$\sigma_0^2 = \mu_0(\theta_1 - \mu_0). \tag{26}$$

The reduction in the source concentration suggests replacing θ_1 by $\alpha(x,t)\mu^{(0)}$, where $\mu^{(0)}$ is a local scale for the mean concentration - see § 3.2 for an example of how it can be precisely defined in particular cases. The natural way to model the dissipation process

is by multiplying the right hand side of (26) by $\beta^2(x,t)$. The result is the proposal:

$$\sigma^2 = \beta^2(x,t)\mu\{\alpha(x,t)\mu^{(0)} - \mu\}, \tag{27}$$

where $\mu = \mu(x,t)$ and $\sigma^2 = \sigma^2(x,t)$ are the **real** $(\kappa \neq 0)$ mean and variance of the concentration. (27) is a generalisation of the result in Chatwin and Sullivan (1990) for the situations that will be considered further in § 3.2 (but note that, for algebraic convenience, I now use β^2 instead of the original β).

To be sure, (27) can be useful only if the dimensionless functions α and β are "simple". Although they were introduced to model the effects of molecular diffusion, it is not expected (for the normal reasons) that they will depend significantly on κ for the high Reynolds numbers and Peclet numbers that characterise atmospheric turbulence. Finally, I want to note that measurements of Γ tend to lead to underestimates of σ^2 because of **instrument smoothing**, and it is reasonable to conceive that this effect, where present, can also be modelled in the size of β.

§ 3.2 *THE EFFECTS OF MOLECULAR DIFFUSION: II SELF-SIMILAR REGIMES.*

As noted in § 1.3, most turbulent diffusion data are taken in statistically steady conditions, usually achieved through steady continuous releases in wind (or water) tunnels. Sometimes the experiments of this type are specifically designed to simulate atmospheric dispersion (*e.g.* Fackrell and Robins 1982). In these circumstances, μ and σ are independent of t. Moreover, in the datasets to be examined here, μ and σ are **self-similar** functions of position, *i.e.* if x denotes downwind distance from the source, and $\mu^{(0)} = \mu^{(0)}(x)$ is the **maximum** value of μ in the cross-section at a distance x downwind, there is a function f such that

$$\mu(x) = \mu^{(0)}(x)f(\eta) \tag{28}$$

where η is a non-dimensional transverse coordinate; there is an analogous expression for σ^2. In practice (see *e.g. Figure 9)*, the function f in (28) is close to *Gaussian*, whereas the corresponding function for σ^2 has its maximum not at the place in the cross-section where $\mu = \mu^{(0)}$ but at an offset position. If (27) holds, elementary algebra as in (26) shows that σ^2 has its maximum where $\mu = \frac{1}{2}\alpha\,\mu^{(0)}$, *i.e.* where f in (28) is equal to $\frac{1}{2}\alpha$. Thus, since $0<f(\eta)\le 1$, an offset maximum in the profile of σ^2 occurs if $1<\alpha<2$. (Note that $\alpha>1$ is necessary in (27) when (28) holds to ensure that $\sigma^2>0$ for all x.)

For such data, α and β in (27) must be independent of t. Although the observed self-similar structure, exemplified by (28), requires only that $\alpha = \alpha(\eta)$, $\beta = \beta^{(0)}(x)\,\beta^{(1)}(\eta)$, Chatwin and Sullivan (1990) took the more drastic step of proposing that α and β were both **constant**. Remarkably, it was found that within experimental error all nine datasets examined were consistent with this proposal. *Figure 10* indicates the degree of consistency, and the accompanying table shows the wide variety of flows, pollutants and measuring techniques covered in the different experiments. Although the values of α and β differed from experiment to experiment, they all satisfied

$$1<\alpha<2;\quad 0<\beta<1. \tag{29}$$

The minimum value of β was about 0.35 (for (5) in *Figure 10*) and the maximum value was about 0.85 (for (8) in *Figure 10*). *Figure 11* shows, for one particular experiment ((8) in *Figure 10*), that the use of (27) with α,β constant gave a better fit to the measured profile of σ^2 than a complicated formula (with three adjustable constants) obtained by curve-fitting this specific dataset. The formula is due to Wilson, Robins and Fackrell (1982).

Subsequent examination of other datasets has confirmed the applicability of (27) with α,β constant. Data on the dispersion of dye in grid-generated water turbulence taken with a light absorption probe by Nakamura, Sakai and Miyata (1987) were particularly interesting in that the observed values were $\alpha = 3$ and $\beta = 1$. The value of α is

particularly interesting since (a) it is greater than any of the values for the experiments in *Figure 10* – see (29) – and (b) as noted above, a value of $\alpha>2$ means that the maximum in the σ^2 profile is not offset, but occurs where $\mu = \mu^{(0)}$. In this context, it is of interest to note that these experiments were very similar (except that dye replaced smoke particles as the pollutant) to those of Gad-el-Hak and Morton (shown as (7) in *Figure 10*). The difference in the results was attributed by Nakamura *et al* to the fact that their measured fluctuating concentration field was everywhere highly intermittent unlike that recorded by Gad-el-Hak and Morton. It seems unlikely that this difference could be due to the differences in the ambient fluid (water for Nakamura *et al*; air for Gad-el-Hak and Morton) or even to the different pollutants. The most likely cause is differences in the degree of instrument smoothing.

A further indication of the likely importance of instrument smoothing is provided by the data in *Figure 12* taken in a turbulent water jet by Sakai *et al* (1989) using probes of four different sizes. Although the values of α were the same (within normal experimental tolerances) for the four different probes, the value of β **increased** as the probe size **decreased**. This observation is consistent with remarks in the last paragraph of § 3.1 earlier in this lecture.

§ 3.3 *THE EFFECTS OF MOLECULAR DIFFUSION: II EXTENSIONS.*

Given the simplicity of (27), and its agreement with data from self-similar flows with α,β constant, several further avenues of research are obvious. Some of these are being actively investigated by us, so most of what follows is necessarily rather speculative (though up-to-date!).

Several datasets from steady sources taken at distances x downwind of the source where the profiles of μ and σ^2 are not self-similar have been examined. In these cases too, the data has been consistent with (27) with, naturally, α and β being functions of downwind distance x. *Figure 13* shows some results from a comprehensive set of experiments for a line source (of heat) in grid-generated turbulence in a wind tunnel

(Sawford and Tivendale 1994). The analysis is from Sawford and Sullivan (1994). The data for the σ^2 versus $\eta\,(=z/\sigma_z)$ profiles for the three cases shown is consistent with (27), as indeed it is at all twenty-one downwind positions at which measurements were made. *Figure 14* (solid symbols) shows the downwind evolution of α and β, together with values from two other sets of experiments (open symbols). It will be seen that the values of α and β in the "far-field" (*Figure 13 (c)*: $x=2.6\mathrm{m}$, $x/x_0=8.39$) are consistent with those for the self-similar data shown in *Figure 10* (although self-similarity was not "clearly reached" for the data shown in *Figure 13*). Near the source (*Figure 13 (a)*: $x=0.002\mathrm{m}$, $x/x_0=6.45\times10^{-3}$) the value of α approaches unity, consistent with the theory in § 2.5 (and shown schematically in *Figure 8*) for the case when molecular diffusion has not had time to have a significant effect. (Note that (26) is (27) with $\mu^{(0)}=\theta_1$ and $\alpha=\beta=1$, and that (26) gives $\sigma^2=0$ at the source.) The highest value of α is indistinguishable from 2 (*Figure 13 (b)*: $x=0.1\mathrm{m}$, $x/x_0=0.323$), and $\alpha=2$ gives the maximum of σ^2 at the same location as that of μ.

All data that have been examined are consistent with (27), with α and β being **slowly-varying** functions of downwind distance x, and with α rising from a value of 1 at the source to a maximum of 2 (*Figure 13 (b)*) or greater (open symbols in *Figure 14*; Nakamura *et al* 1987), before gradually reducing to an asymptotic value between 1 and 2. Work is in hand to develop **simple** evolution equations for α and β, and this appears likely to be successful. The values of α and β will, of course, depend on the **source geometry** (theory: Chatwin and Sullivan 1979; experiments: Fackrell and Robins 1982, Nakamura *et al* 1987).

Figure 15 shows data from a more complicated flow in which again the offset maximum of σ^2 is present near the source ($-1<\alpha<2$), disappears in mid-field ($\alpha>2$) and then reappears in the far-field. *Figure 16* is more ambitious in that it shows two plots of σ^2 versus μ for the instantaneous release data illustrated in *Figure 2*; thus here μ and σ^2 are both explicit functions of t, and the closed curves are traversed as t increases (and behaves as a parameter). It is less clear how (27) can be used in such cases but some progress has been made (Zimmerman and Chatwin 1994).

§ 3.4 *IMPORTANT INTERLUDE: DATA ANALYSIS*

The phenomenon of instrument smoothing has been mentioned above, and was also considered in my earlier lectures here (Chatwin 1990, p8). This important problem has been considered theoretically by Mole (1990a, 1990b). See also Mole and Jones (1993).

Here, before going on to consider models of the pdf, I want to discuss briefly two other problems connected with the interpretation of concentration measurements that are often ignored. These are (a) the treatment of **noise**, and (b) the shortcomings of **thresholding**.

It is inevitable that concentration measurements are contaminated by noise from a variety of sources (mechanical vibration, electronic *etc.*) If no attempt is made to remove the noise the resulting **histogram** of concentration frequencies will inevitably contain spurious negative concentrations as in *Figure 17*. These arise because genuine positive (but small) concentrations are corrupted by negative noise of larger amplitude.

A common, but crude, method of dealing with noise is to specify a **threshold concentration** θ_T, with $\theta_T\geq0$, and to set arbitrarily to zero all concentration readings less than θ_T. This procedure is unsatisfactory because, as in *Figure 17*, many histograms are dominated by low concentrations and it ought to be important to try to assess the behaviour of $p(\theta;x,t)$ for small θ as accurately as possible, and certainly not to let such assessment depend (and rather critically so) on the choice of θ_T which has nothing to do with real concentrations.

Mole was the first to consider this problem from a fundamental viewpoint. Recently his work has been taken substantially further by my colleague David Lewis (Lewis and Chatwin 1994). Lewis has successfully adapted the method of "**maximum entropy inversion**" (MEI) from other branches of signal processing to datasets like that shown in

Figure 17. This method ensures that all estimated concentrations are non-negative and has much theoretical support. Its application to the data shown in *Figure 17* results in *Figure 6* earlier. We are confident that this method is the best presently available for dealing with real noisy datasets. By contrast, *Table 1* (adapted from Robinson, Lewis and Chatwin 1994) illustrates, for two other datasets, the significant errors that can be caused by thresholding. (For the data in *Table 1*, $\theta_T = 0$.)

UVIC 1				UVIC 2			
Parameter	MEI	Thresh.	% error	Parameter	MEI	Thresh.	% error
$\mu/10^{-6}V$	39.4	38.7	-1.7	$\mu/10^{-6}V$	27.2	31.8	+16.9
$\sigma/10^{-6}V$	98.9	86.3	-12.7	$\sigma/10^{-6}V$	79.7	65.4	-17.9
S	5.4	3.1	-42.6	S	7.0	3.3	-52.9
K	53.0	15.0	-71.7	K	91.2	17.0	-81.4

Table 1 Comparison of estimates obtained for two datasets by (i) maximum entropy inversion (MEI), and (ii) thresholding (Thresh.) Here μ, σ are as in text (NB: The readings are taken in microvolts and have not been converted to units of Γ), and S (skewness) and K (kurtosis) are other parameters of the pdf to be discussed further in *Lecture 4.*

In case anyone is still not convinced of the importance of accurate data analysis and interpretation, a glance back at *Figure 1 (a)* should suffice. The four graphs are the results of four different attempts (using three instruments) to measure the same concentration field!

§ 3.5 *FURTHER READING*

Some material in this lecture is a summary of that in Chatwin and Sullivan (1990). It would also be profitable to read some of the papers describing the experimental work, especially perhaps Fackrell and Robins (1982), since their investigation was concerned with simulating atmospheric dispersion.

Lecture 4

Simple probabilistic models and recent development

§ 4.1 *INTRODUCTION*

As noted at the beginning of § 2.4, my principal theme is modelling $p(\theta;\boldsymbol{x},t)$. The work described in *Lecture 3* on μ and σ^2, particularly (27), will be of interest to those whose main concern is only with these parameters (and their evolution), but I want mainly to understand how that work constrains and influences the form of $p(\theta;\boldsymbol{x},t)$. In pursuit of that aim, I shall first present work which gives a representation that $p(\theta;\boldsymbol{x},t)$ **must** have, and then I shall consider some extensions of the work of *Lecture 3* that are quite remarkable. Then I shall give a speculative discussion of how these results may be linked; again, I shall be describing work in progress which is motivated by the reasonable hope that there are **simple** models of $p(\theta;\boldsymbol{x},t)$ that are both scientifically sensible and practically accurate.

§ 4.2 *THE STRUCTURE OF THE PDF & THE INTERMITTENCY FACTOR*

When $\kappa = 0$, the exact form of the pdf has been given in (23). It has been fairly common to attempt to represent the pdf when $\kappa \neq 0$ by an extension of this, such as

$$p(\theta;\boldsymbol{x},t) = \pi(\boldsymbol{x},t)p'(\theta;\boldsymbol{x},t) + \{1 - \pi(\boldsymbol{x},t)\}\delta(\theta), \tag{30}$$

where $\pi(\boldsymbol{x},t)$ has been defined by

$$\pi(\boldsymbol{x},t) = \mathrm{prob}\{\gamma(\boldsymbol{x},t) > 0\} \tag{31}$$

by analogy with (24), and $p'(\theta;\boldsymbol{x},t)$ is the pdf of Γ **conditional** on $\Gamma(\boldsymbol{x},t) > 0$. (In practice, (31) has often been replaced by the probability that $\Gamma > \theta_T$, where θ_T is a threshold concentration, but this means, as discussed in § 3.4, that π, p and p' depend critically on the choice of θ_T and, since θ_T has no connection with $\Gamma(\boldsymbol{x},t)$ or $p(\theta;\boldsymbol{x},t)$ but is determined entirely by the experimenter and the instrumentation, this convention is finally disappearing – and not before time.) However, (30) and (31) are unacceptable also, since they do not represent reality. The simple point is that when $\kappa \neq 0$, the solution of equation (3) for Γ has $\Gamma(\boldsymbol{x},t)>0$ for **all** $\boldsymbol{x}$ at all times t following release, and therefore (31) gives $\pi(\boldsymbol{x},t)=1$ for all such $\boldsymbol{x}$ and t. Then (30) gives $p(\theta;\boldsymbol{x},t)=p'(\theta;\boldsymbol{x},t)$ and is devoid of interest.

This objection to (30) and (31) arises because (3) is itself an approximate equation; molecular velocities are not infinite. However there is no experimental evidence whatsoever that (3) is not a totally acceptable model of reality on the continuum scale for all practical purposes. Given this viewpoint, it is **scientifically inconsistent** to use (30) and (31), and alternatives should be sought.

The concepts of **intermittency** and **intermittency factor** that motivated (23) and (30) are important ones in turbulence and turbulent diffusion since they represent the effects of the turbulent velocity field. Chatwin and Sullivan (1989) proposed that (24) be adopted as the definition of intermittency factor even when $\kappa \neq 0$. This has the merit, unlike (31), of being meaningful and of depending only on those factors, namely the velocity and geometry, that are desirable in any definition of intermittency factor. Superficially, however, this definition, while theoretically sensible, is unacceptable because $\pi_0(\boldsymbol{x},t)$ cannot be **measured** since κ is never zero (leaving aside the controversial question of whether (3) with $\kappa=0$ governs the dispersion of pollutants like smoke that involve solid particles). However this objection appears to be removable because all available evidence suggests that the mean concentration $\mu(\boldsymbol{x},t)$ is not detectably dependent on the value of κ, and that, therefore, $\mu(\boldsymbol{x},t) \approx \mu_0(\boldsymbol{x},t)$, where $\mu_0(\boldsymbol{x},t)$ is the mean concentration in the hypothetical situation when $\kappa=0$. Use of the first result in (25) then gives

$$\pi_0(\boldsymbol{x},t)=\mu_0(\boldsymbol{x},t)/\theta_1 \approx \mu(\boldsymbol{x},t)/\theta_1, \tag{32}$$

where θ_1 is the source concentration, supposed uniform. (Chatwin and Sullivan (1989) showed that these ideas could be generalised to cases where the source concentration is not uniform.) Thus (32) enables $\pi_0(\boldsymbol{x},t)$ to be estimated by measuring the real concentration.

An important consequence of this new definition of the intermittency factor is that the pdf in real flows ($\kappa \neq 0$) can be represented **exactly** in terms of it. Chatwin and Sullivan (1989) showed that

$$p(\theta;\boldsymbol{x},t)=\pi_0(\boldsymbol{x},t)\,p_s(\theta;\boldsymbol{x},t)+\{1-\pi_0(\boldsymbol{x},t)\}p_a(\theta;\boldsymbol{x},t), \tag{33}$$

where p_s, and p_a, are the pdfs of Γ conditional, respectively, on $(\boldsymbol{x},t)$ being occupied by a fluid element from the source, and not from the source (*i.e.* from ambient). An alternative way of stating these conditions is $\Gamma_0(\boldsymbol{x},t)=\theta_1$ and $\Gamma_0(\boldsymbol{x},t)=0$ respectively. Equation (33) is an extension of (23) to real flows that does not have the flaws of (30). It should be noted that p_s and p_a differ from delta functions only because of the effects of molecular diffusion.

§ 4.3 *HIGHER MOMENTS*

Define $\mu_n(\boldsymbol{x},t)$ for $n \geq 2$ by

$$\mu_n(\boldsymbol{x},t)=E\{[\Gamma(\boldsymbol{x},t)-\mu(\boldsymbol{x},t)]^n\}=\int_0^\infty[\theta-\mu(\boldsymbol{x},t)]^n p(\theta;\boldsymbol{x},t)\,d\theta. \tag{34}$$

Then μ_n is the ***nth* central moment** of $p(\theta;\boldsymbol{x},t)$ with $\mu_2(\boldsymbol{x},t)=\sigma^2(\boldsymbol{x},t)$ by (9) and (15). In principle, the pdf is determined uniquely by μ and by the set of all $\mu_n(n=2,3,4,\ldots)$; see Derksen and Sullivan (1990). In § 3.2, it was seen that the relationship (27), *viz*

$$\mu_2(\boldsymbol{x},t)=\sigma^2(\boldsymbol{x},t)=\beta^2\mu(\boldsymbol{x},t)\{\alpha\mu^{(0)}-\mu(\boldsymbol{x},t)\}, \tag{35}$$

was in good agreement with data from many different types of experiment. This relationship was derived from the $\kappa=0$ model for $p(\theta;\boldsymbol{x},t)$ in § 2.5, and Chatwin and Sullivan (1990) considered the application of these ideas to the higher moments. They showed first that (23) gives

$$\mu_{on} = \frac{1}{\theta_1}[\mu_0(\theta_1 - \mu_0)^n + (-1)^n(\theta_1 - \mu_0)\mu_0^n], \qquad (36)$$

for $n = 2,3,4,\ldots$, where μ_{on} is the *nth* central moment when $\kappa = 0$. (Note that (36) is the same as (26) when $n = 2$.) They further suggested that, when $\kappa = 0$, the ideas leading to (27) and (35) could be extended to the higher moments to give

$$\mu_n = \frac{\beta_n^n}{\alpha\mu^{(0)}}\{\mu(\alpha\mu^{(0)} - \mu)^n + (-1)^n(\alpha\mu^{(0)} - \mu)\mu^n\}, \qquad (37)$$

where α is as in § 2.5 and $\beta_2 = \beta$. This was in reasonable agreement with the limited datasets examined for $n = 3,4$ and, moreover, it was suggested that $\beta = \beta_2 = \beta_3 = \beta_4 = \ldots$ was consistent with these data and with theoretical arguments. For $n = 3,4$, this further suggestion gives, from (37),

$$\mu_3 = \beta^3\mu(\alpha\mu^{(0)} - \mu)(\alpha\mu^{(0)} - 2\mu); \quad \mu_4 = \beta^4\mu(\alpha\mu^{(0)} - \mu)\{\alpha^2\mu^{(0)2} - 3\alpha\mu^{(0)}\mu + 3\mu^2\}. \qquad (38)$$

(Before proceeding, it is interesting to note that Sawford and Sullivan (1994) have shown that a different extension of (36) from that in (37), but equivalent in its algebraic simplicity, was in good agreement with the data from Sawford and Tivendale (1994).)

Instead of μ_3 and μ_4, it is common to use non-dimensional measures derived from them, namely the **skewness** S and the **kurtosis** K, where

$$S = \frac{\mu_3}{\sigma^3} \quad , \quad K = \frac{\mu_4}{\sigma^4}. \qquad (39)$$

From (35) and (38) it follows that

$$S = \frac{(\alpha - 2x)}{[x(\alpha - x)]^{1/2}} \; , \; K = \frac{(\alpha^2 - 3\alpha x + 3x^2)}{x(\alpha - x)} \; , \; x = \frac{\mu}{\mu^{(0)}}. \qquad (40)$$

Mole and Clarke (1994) eliminated α from the two equations in (40). The result is extremely simple:

$$K = S^2 + 1. \qquad (41)$$

Since the equations for S and K in (40) contain α, plots of S and K against $\mu/\mu^{(0)} = x$ can be expected to vary with experimental conditions, reflecting changes in α. But (41) predicts that a plot of K versus S should be independent of experimental conditions. *Figures 18* and *19* largely verify these predictions, although the K versus S plot in *Figure 19* shows collapse to a curve lying **above** that in (41), *i.e.* the data suggest that there is a "universal" relationship between K and S but that, for this relationship,

$$K > S^2 + 1. \qquad (42)$$

Examination of other datasets has confirmed the same remarkable tendency for the (K,S) plot to collapse. A severe test is provided by the instantaneous release data for the experiments illustrated in *Figure 2*, and *Figure 20* shows a typical result.

Before discussing the implications of these results for modelling the pdf, I want briefly to mention two further points. Mole and Clarke (1994) extended the ideas above to central moments with $n > 4$. For example, they showed from (38) that

$$K_5 = S^3 + 2S \; , \quad K_5 = \frac{\mu_5}{\sigma^5}, \qquad (43)$$

and the data used in *Figures 18* and *19* showed again a collapse, this time for the (K_5,S) plot. But again the collapse was to a curve somewhat above that in (43). Perhaps even more remarkable, but not directly relevant to the main theme of these lectures, is that Mole and Clarke (1994) used the same data to estimate **generalised dosages** $D_{pT}(x,t)$, where (*cf* (10) and (13))

$$D_{pT}(x,t) = \frac{1}{2T}\int_{t-T}^{t+T}\{\Gamma(x,s)/\theta_1\}^p ds, \qquad (44)$$

and *Figure 21* shows the extraordinary collapse of the (K,S) plot. Note that *Figure 21* covers 12 different values of T (for $p=1$) and 12 different values of p (for $T=5s$); note also, however, that the theory given above to justify equations like (41) certainly does not apply, at least not directly, to the statistical properties of D_{PT}.

§ 4.4 *DISCUSSION*

I want to note first a set of results from probability theory that were discussed by Mole and Clarke (1994). It can be shown that for any pdf whatsoever

$$K \geq S^2 + 1, \tag{45}$$

with corresponding results for higher moments. Moreover, equality in (45) occurs only when the pdf represents a **two-state process**, *i.e.* the pdf is a weighted sum of two delta functions. This result is consistent with the theory presented in these lectures (§§ 3.2, 4.3 especially) which was based on extending the "two-delta-function" pdf in (23). It is therefore of particular interest to note that the pdf $p(\theta;x,t)$ that is consistent with (35) and (41) is (Sullivan and Ye 1993):

$$p(\theta;x,t) = \pi_0\,\delta(\theta-\mu_s) + \{1-\pi_0\}\,\delta(\theta-\mu_a), \tag{46}$$

where

$$\pi_0 = \frac{\mu}{\alpha\mu^{(0)}}\ ,\ \mu_s = (1-\beta)\mu + \alpha\beta\mu^{(0)}\ ,\ \mu_a = (1-\beta)\mu \tag{47}$$

and

$$\mu = \pi_0\mu_s + (1-\pi_0)\mu_a. \tag{48}$$

But, as noted at the end of § 4.2, the real pdf has the structure (33), where, because of molecular diffusion, p_s and p_a cannot be delta functions. However, the implications of the theory and the data comparisons shown are (a) that p_s and p_a are, in some sense, "close" to delta functions, and (b) that because of the collapses of the data like those shown in *Figures 19* and *20*, the pdf $p(\theta;x,t)$ is determined (at least to a good approximation) by **three** parameters only, *e.g.* μ, σ and S. (Conclusion (b) follows from the data comparisons independent of any theory.)

How can these conclusions be reconciled with (33)? What are the structural forms of p_s and p_a?

These are questions to which I do not yet know the answers, but I am convinced that the work I have summarized makes their investigation worthwhile in the sense that real progress can soon be expected. One line of attack that we are currently using can be summarised as follows. Let μ_a^2, σ_a^2, S_a and K_a be the mean, variance, skewness and kurtosis of $p_a(\theta;x,t)$ in (33), with a corresponding notation for the properties of $p_s(\theta;x,t)$. It is easy to derive expressions from (33) for the overall mean, variance, skewness and kurtosis. The results for μ and σ^2 are

$$\mu = \mu_a + \pi_0\Delta,\ \ \sigma^2 = \pi_0(1-\pi_0)\Delta^2 + \pi_0\sigma_s^2 + (1-\pi_0)\sigma_a^2, \tag{49}$$

where

$$\Delta = \mu_s - \mu_a. \tag{50}$$

(The first of (49) is the same as (48), as it must be.) Sufficiently close to the source $\mu_a = \sigma_a = \sigma_s \approx 0, \mu_s \approx \theta_1$, and the results in § 2.5 are recovered. But this cannot remain true. As a result of molecular diffusion, σ_s and σ_a become non-zero, and both $\Delta = \mu_s - \mu_a$ and π_0 must approach zero, as x and t become large enough. Eventually the dominant term in the expression for σ^2 in (49) will be σ_a^2. Likewise the dominant terms in the corresponding expressions to (49) for S and K are S_a (unless $S_a = 0$) and K_a. Hence, away from the source,

$$(K - S^2) \approx (K_a - S_a^2). \tag{51}$$

To explain the experimental results discussed in *Lectures 3* and *4*, it therefore seems to be necessary to find a family of pdfs p_a with the following properties:

(1) $\sigma_a^2 \approx \beta^2 \mu_a(\alpha \mu_a^{(0)} - \mu_a)$ – to satisfy (27) and (35);

(2) $(K_a - S_a^2)$ is (approximately) independent of μ_a – to ensure the collapse of the (K,S) plots like those shown in *Figures 19* and *20*;

(3) as the parameters of p_a vary, a large spread of S_a and K_a values must be generated since such a spread has been found for all datasets examined.

Requirement (3) rules out "simple" distributions (*e.g.* uniform, normal, exponential, ...) for which S_a and K_a are universal constants, but there are other candidate distributions which are now being examined. Provided this exercise is successful, $p(\theta;\mathbf{x},t)$ will be approximately equal to $p_a(\theta;\mathbf{x},t)$ except very near the source and, in practice, the statistical properties of Γ can then be determined in terms of μ_a – and this is (relatively) easy to predict.

§ 4.5 *FURTHER READING*

Except for the work in § 4.2 which is in Chatwin and Sullivan (1989), most of the material in this Lecture is ongoing research which, so far, appears in conference presentations *etc.* Be patient!

References

Batchelor, GK 1959 *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.

Britter, RE 1991 *The evaluation of technical models used for major-accident hazard installations.* Report to Commission of the European Communities, DGXII, August 1991.

Carn, KK & Chatwin, PC 1985 *Variability and heavy gas dispersion.* J. Haz. Materials **11**, 281-300.

Chatwin, PC 1982 *The use of statistics in describing and predicting the effects of dispersing gas clouds.* J. Haz. Materials **6**, 213-230.

Chatwin, PC 1990 *Air pollution modelling for environmental impact assessment.* Lectures to College on Atmospheric Boundary Layer Physics, ICTP, Trieste, June 1990. (Also: Brunel University Department of Mathematics and Statistics Technical Report TR/06/90.)

Chatwin, PC & Sullivan, PJ 1979 *The relative diffusion of a cloud of passive contaminant in incompressible turbulent flow* J. Fluid Mech. **91**, 337-355.

Chatwin, PC & Sullivan, PJ 1989 *The intermittency factor of scalars in turbulence.* Phys. Fluids A **1**, 761-763.

Chatwin, PC & Sullivan, PJ 1990 *A simple and unifying physical interpretation of scalar fluctuation measurements from many turbulent shear flows* J. Fluid Mech. **212**, 533-556.

Chatwin, PC & Sullivan, PJ 1994 *Quantitative models for environmental pollution: a review.* In **Mixing and Transport in the Environment**(edited by KJ Beven, PC Chatwin and J H Millbank), John Wiley & Sons Ltd., 353-369.

Derksen, RW & Sullivan, PJ 1990 *Moment approximations for probability density functions.* Combustion and Flame, **81**, 378-391.

Fackrell, JE & Robins, AG 1982 *Concentration fluctuations and fluxes in plumes from point sources in a turbulent boundary layer.* J. Fluid Mech. **117**, 1-26.

Gad-el-Hak, M & Morton, JB 1979 *Experiments on the diffusion of smoke in isotropic turbulent flow.* AIAA J. **17**, 558-562.

Hanna, SR, 1993 *Uncertainties in air quality model predictions.* Boundary-Layer Meteorol. **62**, 3-20.

Hanna, SR, Chatwin, PC, Chikhliwala, E, Londergan, RJ, Spicer, TO & Weil, JC 1992 *Results from the model evaluation panel.* Plant/Operations Progress **11**, 2-5.

Hanna, SR, Strimaitis, DG & Chang, JC 1991 *Evaluation of 14 hazardous gas dispersion models with ammonia and hydrogen fluoride field data.* J. Haz. Materials **26**, 127-158.

Lewis, DM & Chatwin, PC 1994 *The treatment of atmospheric dispersion data in the presence of noise and baseline drift.* To appear in Boundary-Layer Meteorol.

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, Chatwin, PC & Sullivan, PJ 1993 *Modelling concentration fluctuations in air pollution.* In **Modelling Change in Environmental Systems** (edited by AJ Jakeman, MB Beck and MJ McAleer), John Wiley & Sons Ltd., 317-340.

Mole, N & Clarke, ED 1994 *Simple extensions to the Chatwin and Sullivan theory.* Manuscript to be submitted for publication.

Mole, N & Jones, CD 1994 *Concentration fluctuation data from dispersion experiments carried out in stable and unstable conditions.* Boundary-Layer Meteorol. **67**, 41-74.

Nakamura, I, Sakai, Y & Miyata, M 1987 *Diffusion of matter by a non-buoyant plume in grid-generated turbulence.* J. Fluid Mech. **178**, 379-403.

Pope, SB 1985 *PDF methods for turbulent reactive flows.* Progress Energy Combust, Sci. **11**, 119-192.

Robinson, C, Lewis, DM & Chatwin, PC 1994 *The pitfalls of thresholding atmospheric dispersion data.* Manuscript to be submitted for publication.

Sakai, Y, Nakamura, I, Tsunoda, H & Shengian, L 1990 *Private communication* (and lecture presented at Euromech 253, Brunel University, August 1989).

Sawford, BL & Sullivan, PJ 1994 *A simple representation of a developing contaminant concentration field.* Submitted to J. Fluid Mech.

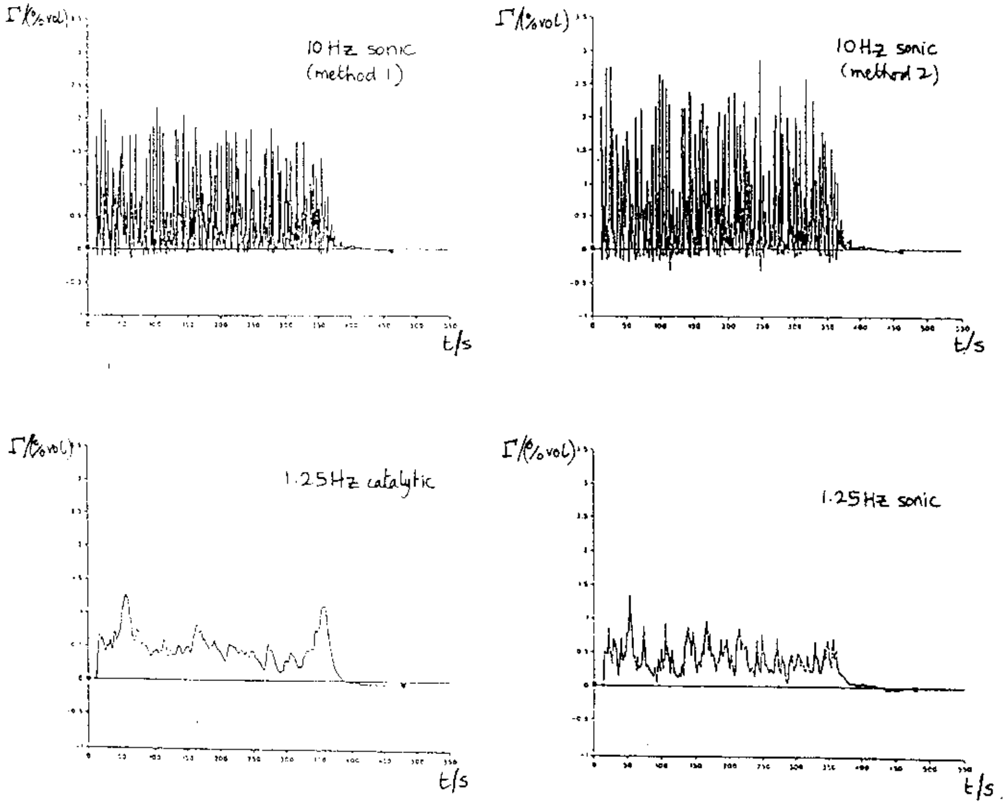
Sawford, BL & Tivendale, CM 1994 *Measurements of concentration statistics downstream of a line source in grid turbulence.* Manuscript in preparation.

Sullivan, PJ & Ye, H 1993 *Private communication.*

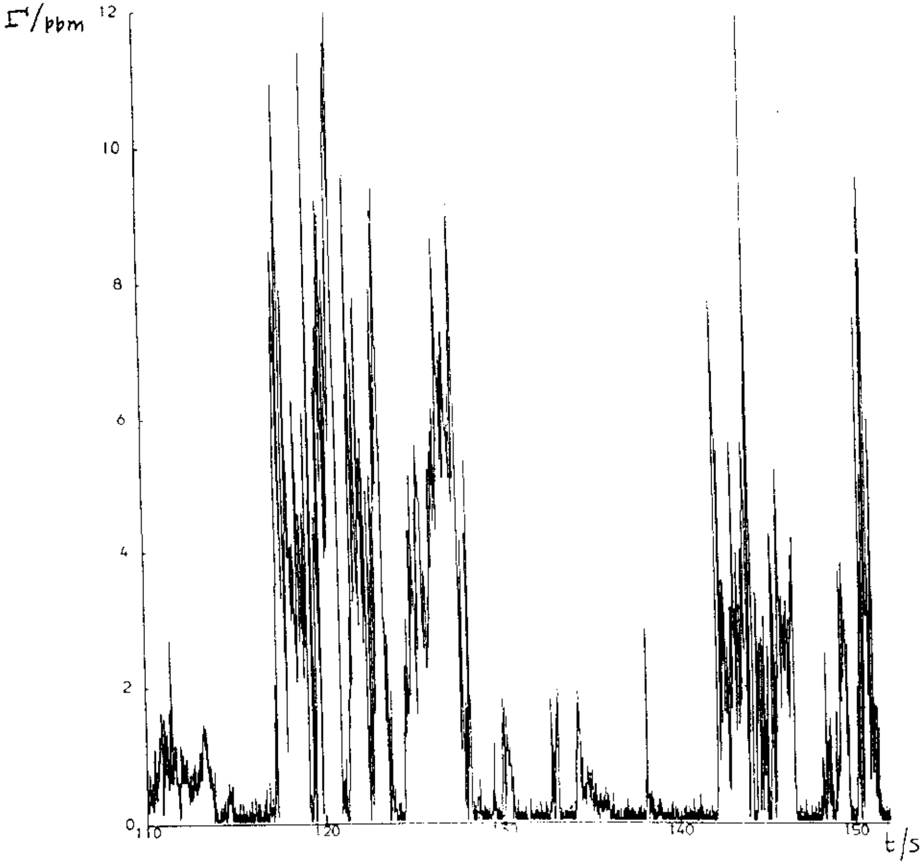
Wilson, DJ, Robins, AG & Fackrell, JE 1982 *Predicting the spatial distribution of concentration fluctuations from a ground level source.* Atmos. Environ. **16**, 497-504.

Zimmerman, WB & Chatwin, PC 1994 Manuscript in preparation.

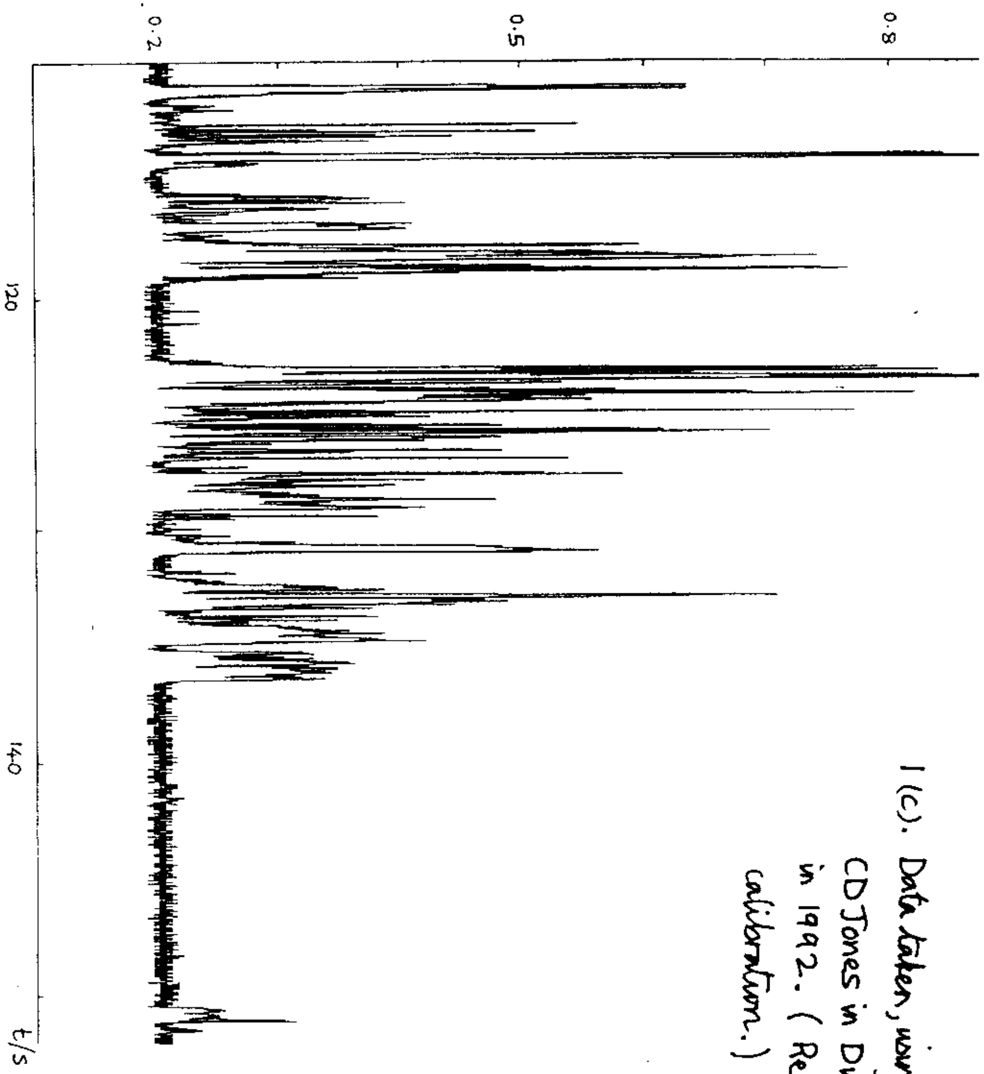
FIGURE 1. Examples of time series of concentration for atmospheric dispersion.



1(a). Records taken in Lathen, Germany (TüV & Risø) using 4 different methods for the same $\Gamma(\underset{\sim}{x},t)$.

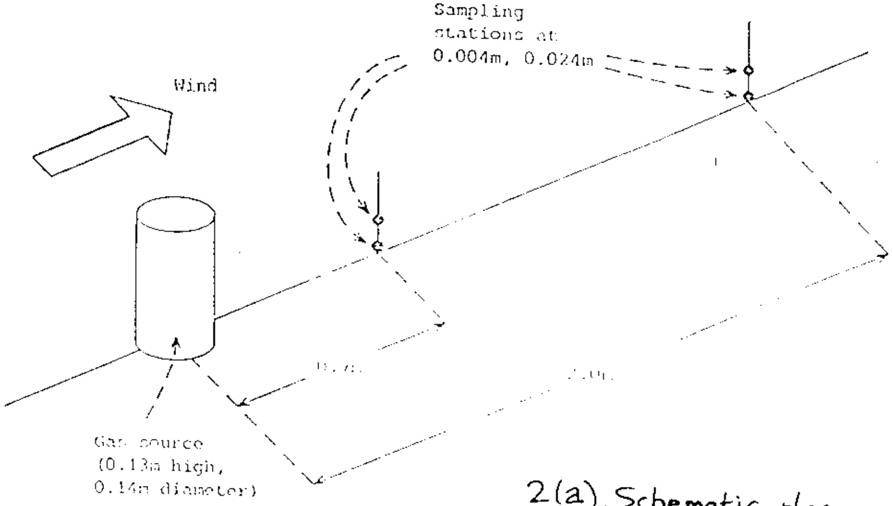


1(b). Data taken, using FIDs, by CD Jones in Dugway, Utah in 1992

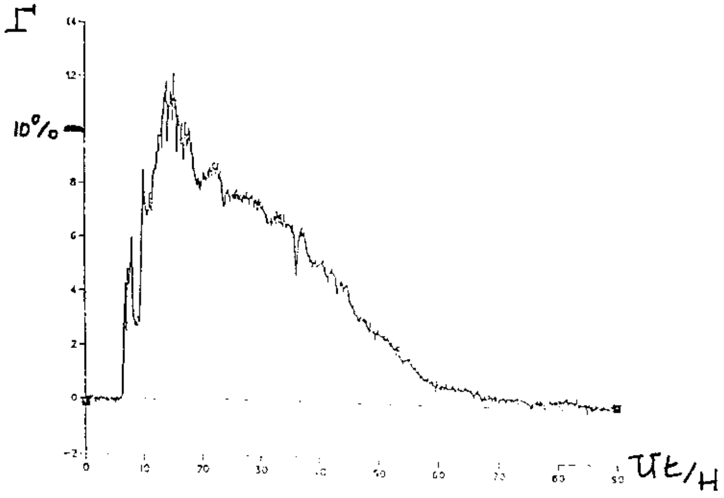


1(c). Data taken, using UVICs, by CD Jones in Dugway, Utah in 1992. (Readings before calibration.)

FIGURE 2 Instantaneous release experiments at Warren Spring Laboratory by D J Hall.



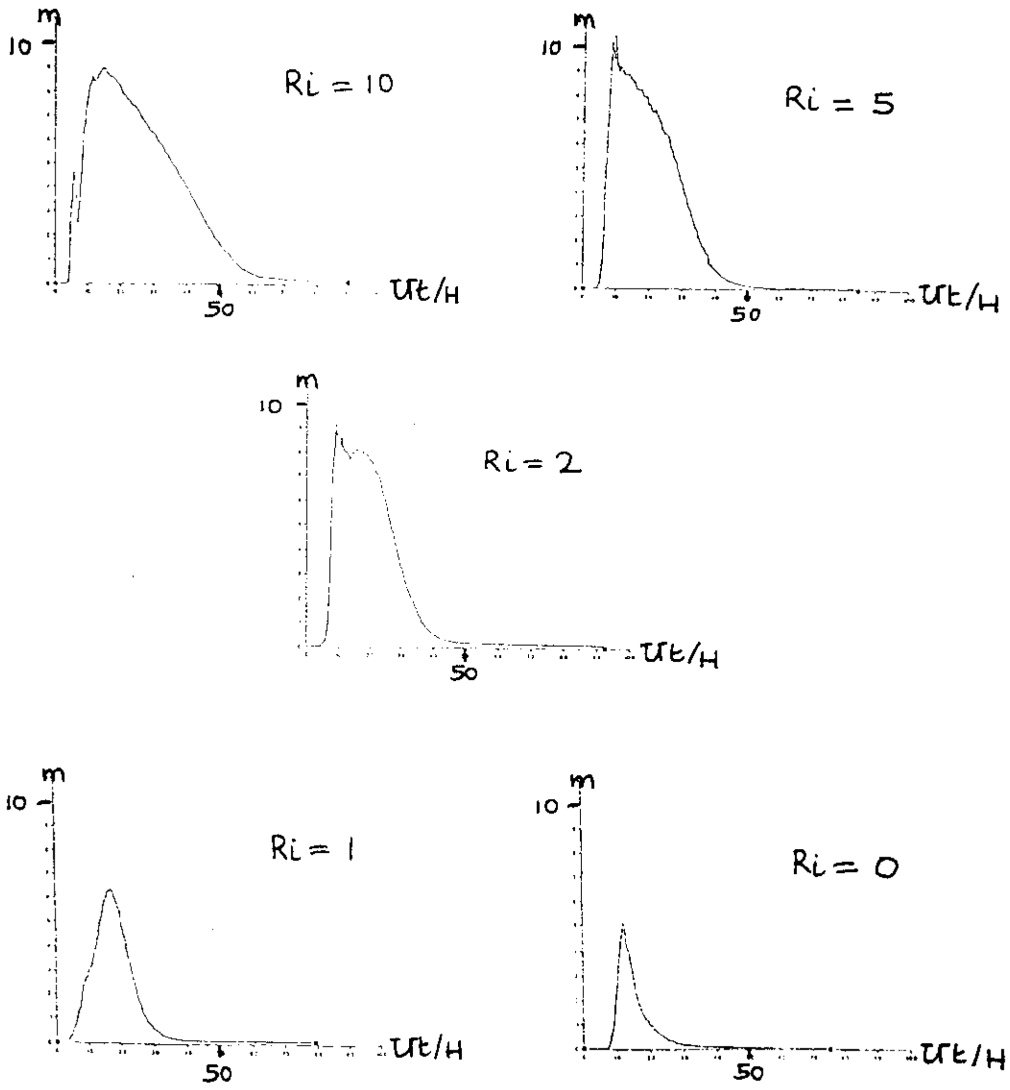
2(a). Schematic plan



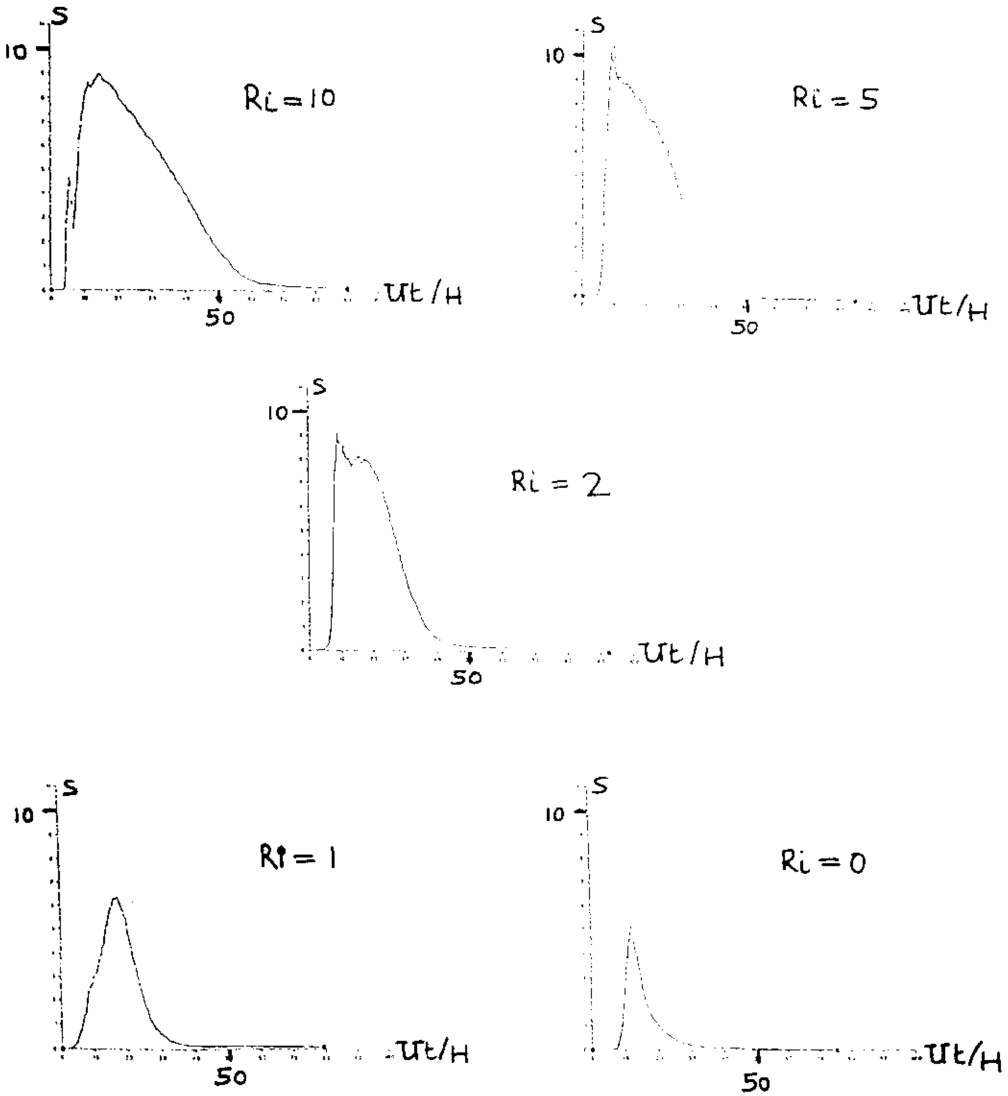
2(b). Typical time series.
(Ri = 10)

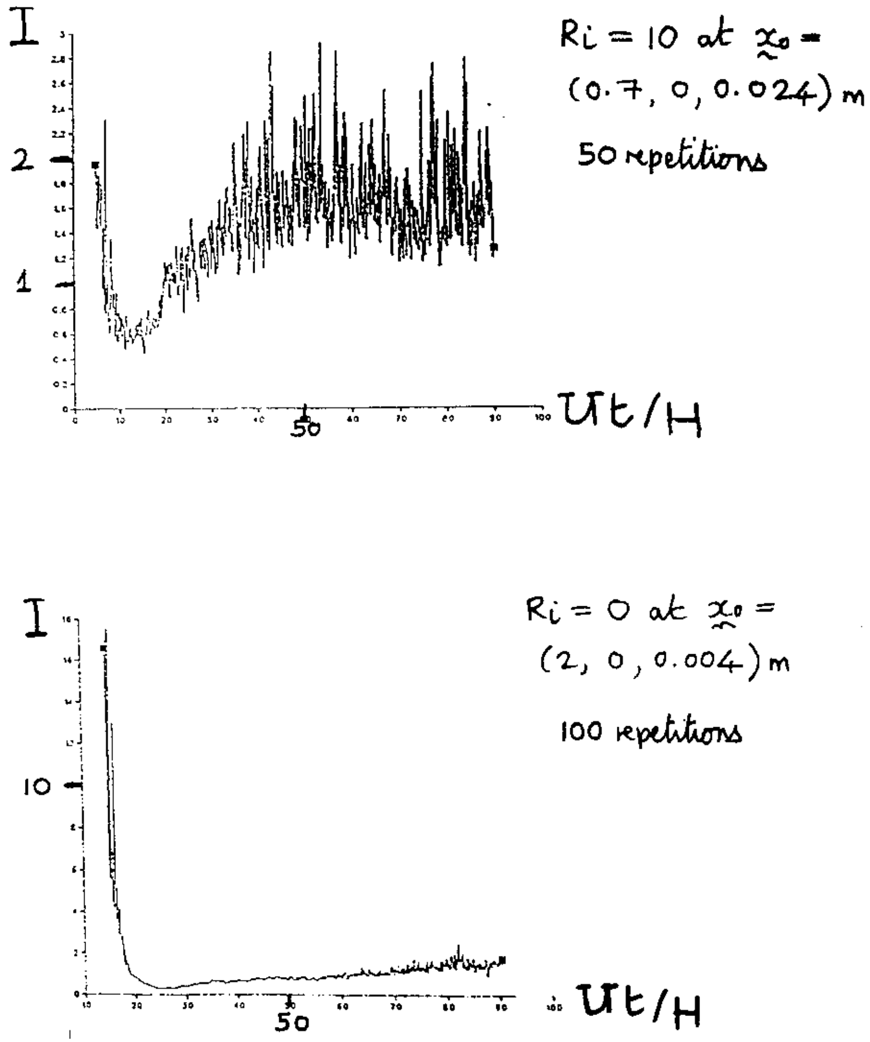
NB. $Ri = \frac{gH}{\mathcal{U}^2}\frac{\Delta\rho}{\rho_0}$ (H = source height = 0.13m)

2(c) Estimates (= m(t)) of $\mu(\underset{\sim}{x}_0, t)$ for five Ri, where $\underset{\sim}{x}_0$ = (0.7, 0, 0.004)m. (Units are %)



2 (d) Estimates (= s(t)) of $\sigma(\underset{\sim}{x}_0, t)$ for five Ri, where $\underset{\sim}{x}_0$ = (0.7, 0, 0.004)m. (Units are %.)





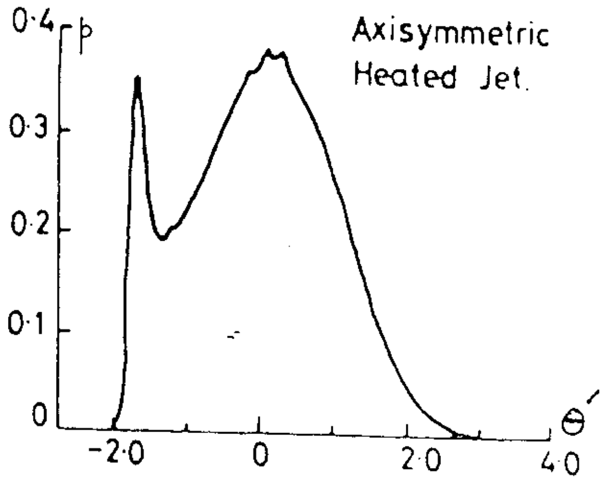
Ri = 10 at $\underset{\sim}{x}_0$ = (0.7, 0, 0.024) m

50 repetitions

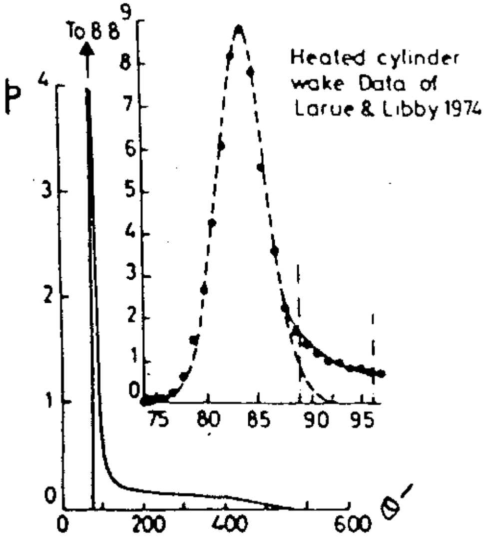
Ri = 0 at $\underset{\sim}{x}_0$ = (2, 0, 0.004) m

100 repetitions

FIGURE 3 Two estimates of I($\underset{\sim}{x}_0$, t) for the experiments of Figure 2. (Note that the vertical scales differ, and that the scatter is "statistical noise" due to the limited number of repetitions.)

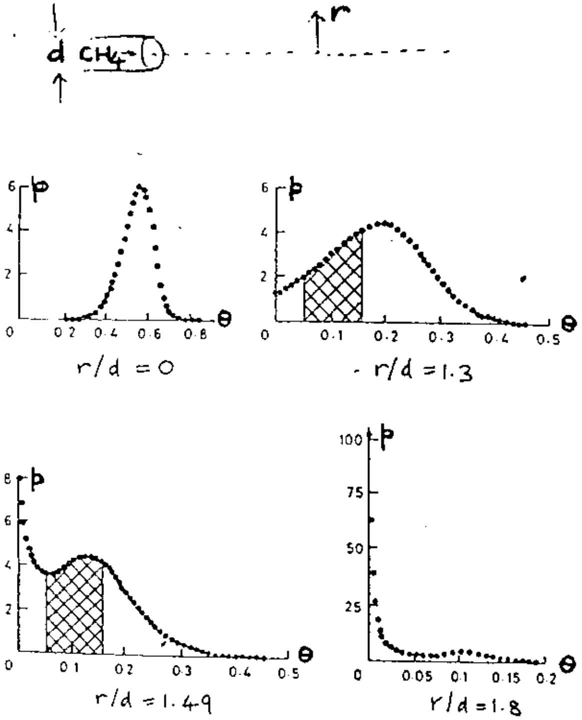


From: Bilger, Antonia & Sreenivasan 1976



From Bilger, Antonia & Sreenivasan 1976.

FIGURE 4. Some estimated pdfs for temperature from wind-tunnel experiments, where θ′ is a non-dimensional measure of temperature. [Phys. Fluids 19, 1976, 1471 – 1474.]



From: Birch, A.D., Brown, D.R. and Dodson M.G. 1980 Ignition probabilities in turbulent mixing flows. *18th International Symposium of the Combustion Institute*, Waterloo, Canada.

NOTE

For each diagram the value of r/d is given, where r denotes distance from the jet centre-line and d is the jet diameter. Each set of data is taken at 10d downwind of the source.

FIGURE 5 Estimates of $p(\theta; \underset{\sim}{x}, t)$ at various positions in a methane jet

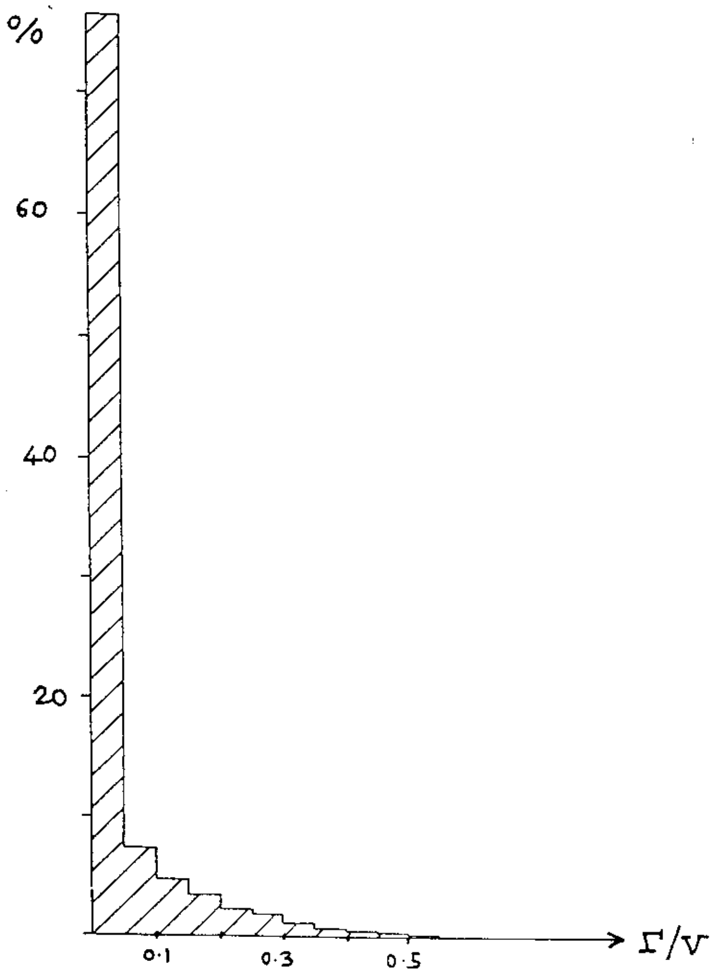
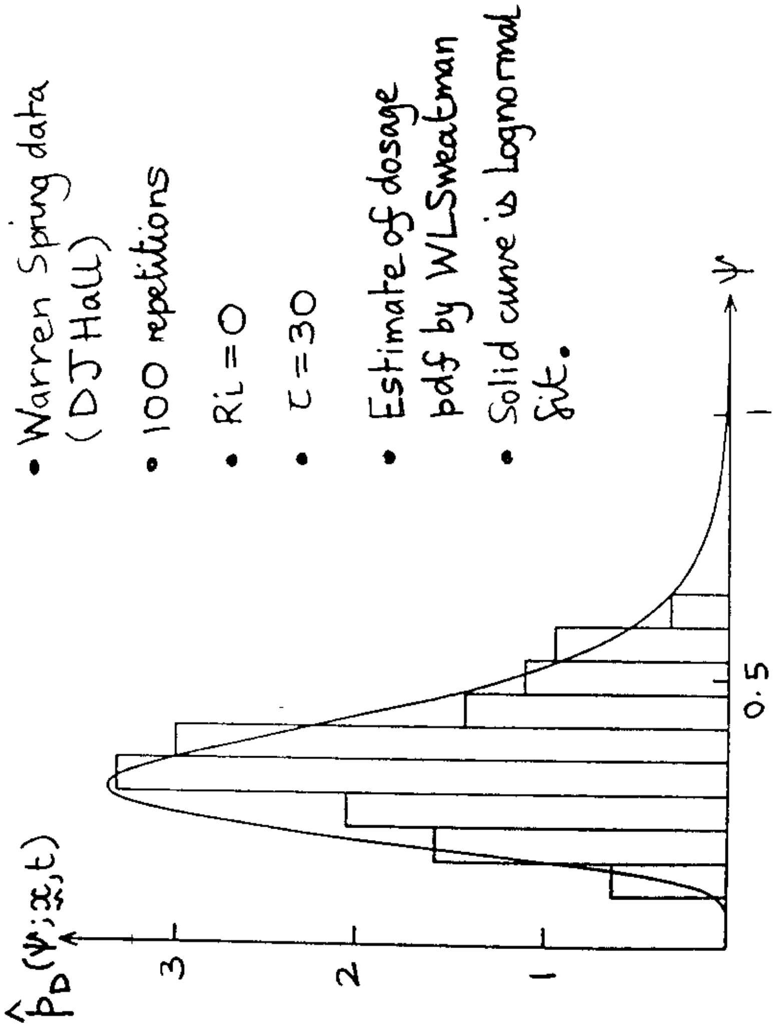


FIGURE 6 Histogram of concentration data after treatment by method of maximum entropy inversion. From Lewis and Chatwin (1994). See Figure 17.



- Warren Spring data (DJ Hall)
- 100 repetitions
- $R_i = 0$
- $\tau = 30$
- Estimate of dosage pdf by WLSweatman
- Solid curve is Lognormal fit.

FIGURE 7 Estimate of a dosage pdf. (see Figure 2(a))

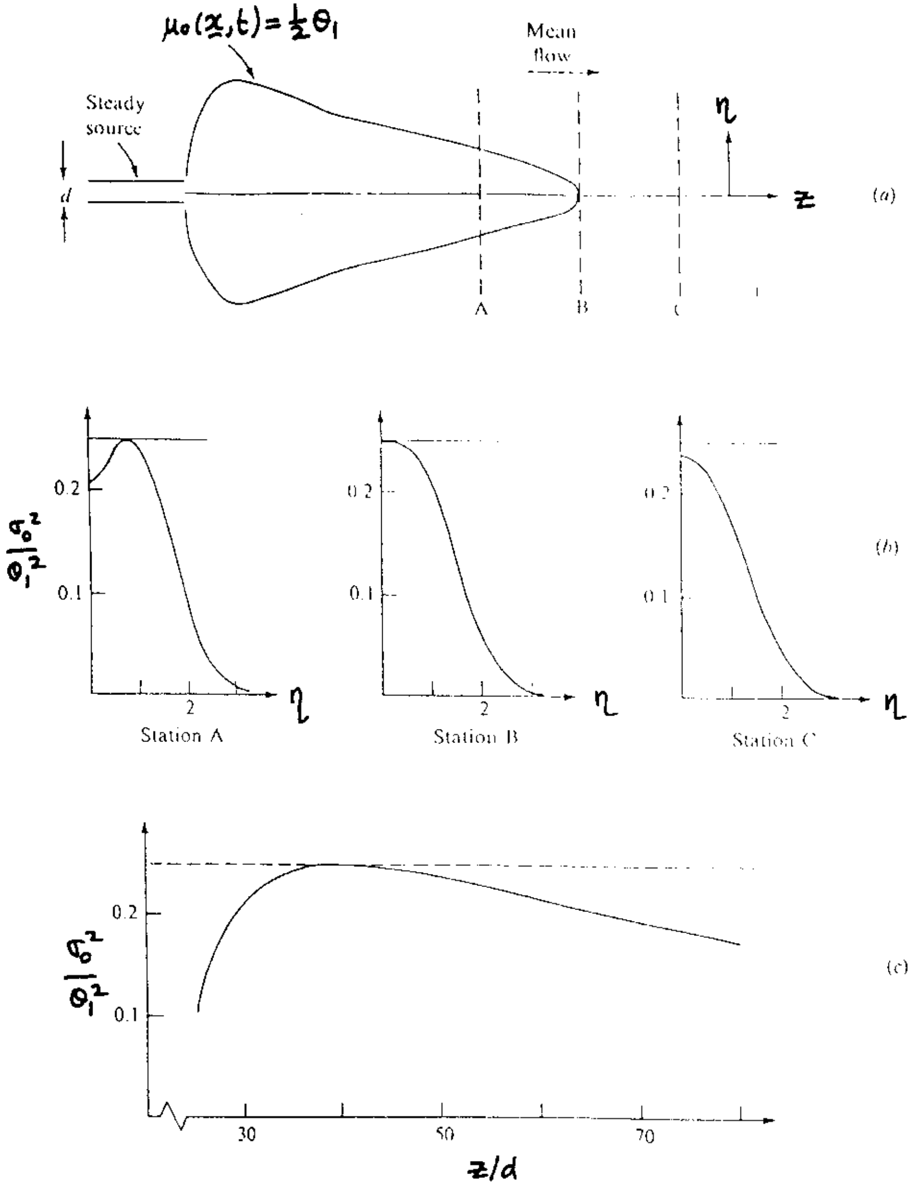


FIGURE 8. Schematic illustration of (26): (a) geometry ; (b) transverse profiles ; (c) axial profile ($\eta = 0$). Note that $\sigma_0^2/\theta_1^2 < \frac{1}{4}$ everywhere except on the surface indicated in (a), and that maxima in the profiles occur on crossing this surface.

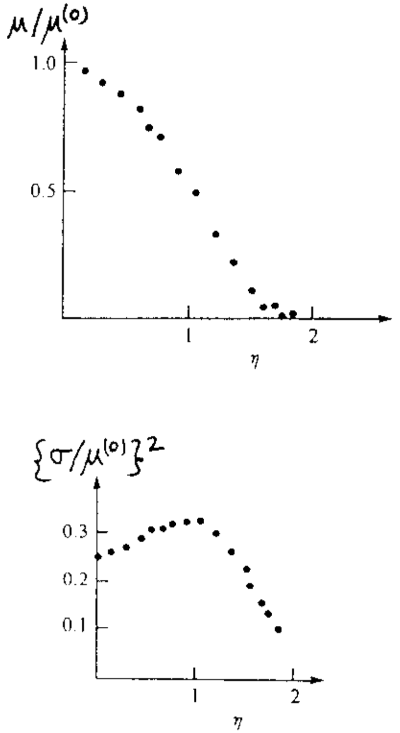
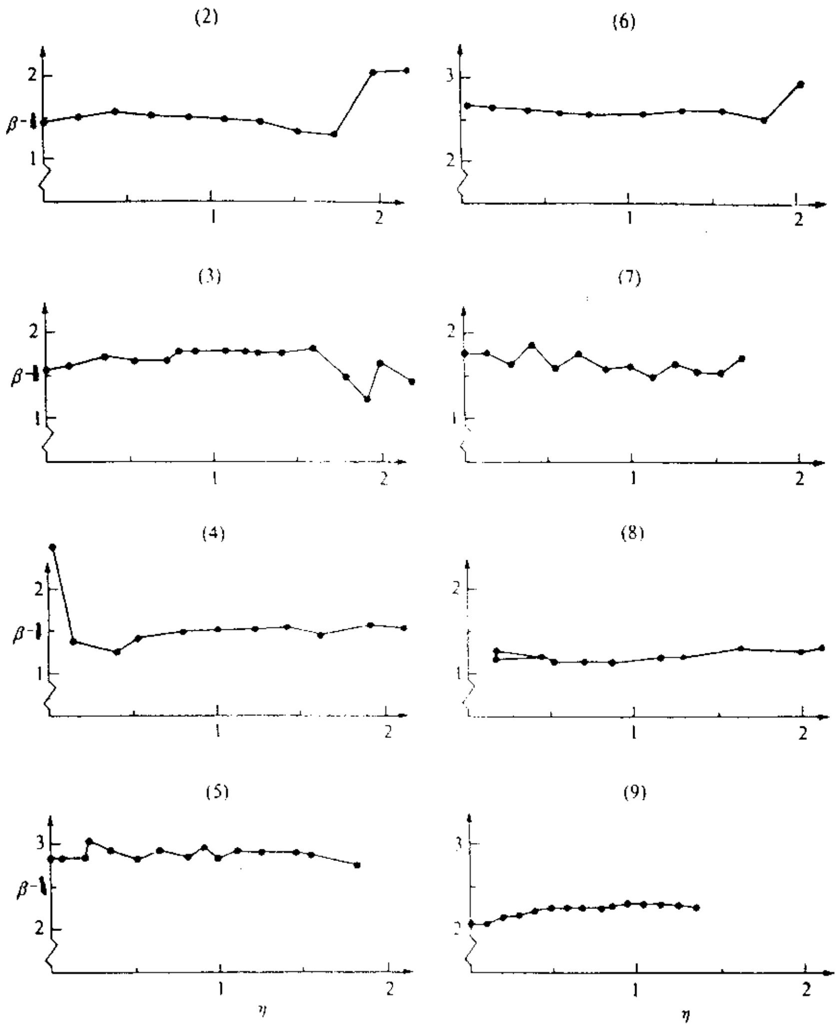


FIGURE 9 Typical profiles of μ and σ^2. Data from Antonia *et al* (1975) JFM **72**, 455-480.

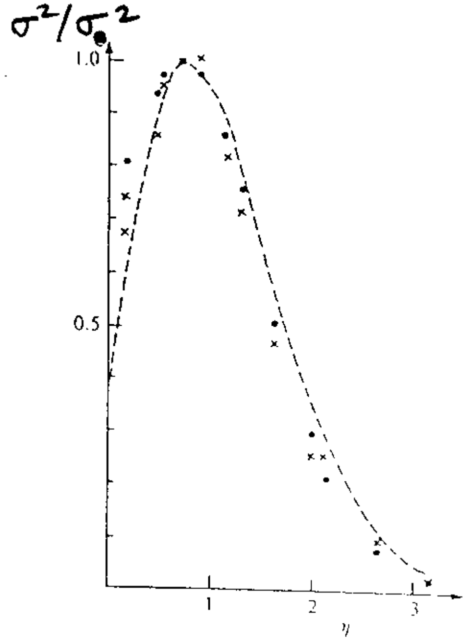


NO.	WORKERS	FLOW	SCALAR	MEASUREMENT TECHNIQUE
(1)	Becker *et al.* (1967)	Round jet	Oil smoke	Light-scatter
(2)	LaRue & Libby (1974)	Plane wake	Heat	Platinum wire resistance thermometer
(3)	Antonia *et al.* (1975)	Round jet with coflowing stream	Heat	Platinum/10% rhodium wire
(4)	Sreenivasan, Danh & Antonia (1976)	Smooth-walled boundary layer	Heat	Wollaston wire
(5)	Shaughnessy & Morton (1977)	Round jet exhausting into a secondary air flow	Smoke particles	Light scatter
(6)	Birch *et al.* (1978)	Round methane jet	Methane	Raman scattering of laser light
(7)	Gad-el-Hak & Morton (1979)	Grid turbulence	Smoke particles	Laser anemometer
(8)	Fackrell & Robins (1982)	Rough-walled boundary layer	Propane/helium mixture	Flame ionisation detector system
(9)	Antonia *et al.* (1983*a*)	Plane jet	Heat	Wollaston wire

NO.	α	β^2	NOTES
(1)	1.31	0.16	Values from empirical relationship in paper
(2)	1.17	0.46	
(3)	1.16	0.34	
(4)	1.09	0.46	Data for $z/d = 102$
(5)	1.24	0.12	
(6)	1.27	0.14	Data for $z/d = 40$
(7)	1.52	0.37	
(8)	1.35	0.72	Data for $z/H = 5.00, 5.92$
(9)	1.15	0.20	

FIGURE 10 Graphs of $\{\mu(\alpha\mu^{(0)}-\mu)/\sigma^2\}^{\frac{1}{2}}$ versus η for datasets (2) to (9) in table. If (27) holds, this should be constant and equal to β^{-1}, as is verified in each case (within experimental error). The data in (1) were shown to follow (27) by Becker *et al*.

FIGURE 11



σ_0 is maximum of σ.

× Data from Fackrell and Robins (1982)
Propane-Helium mixture in rough-walled boundary layer

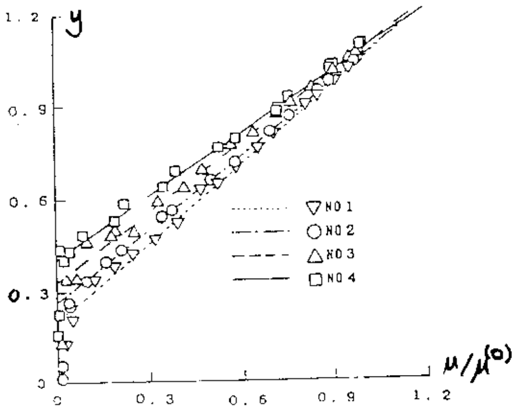
• Proposed new formula $\sigma^2 = 0.72\mu\,(1.35\mu^{(0)} - \mu)$

- - - Curve-fit for these data by Wilson, Robins and Fackrell (1982):

$$\sigma^2 = 0.784\left\{\exp\left[-\ln 2\,|\eta - 0.6|^{1.7}\right] - 0.6\exp\left[-\ln 2\,|\eta + 0.6|^{1.7}\right]\right\}$$

FIGURE 12

Data taken by SAKAI, NAKAMURA, TSUNODA & SHENGIAN in a turbulent water jet (1st version at EUROMECH 253 1989; 2nd (this) version privately in 1990.)

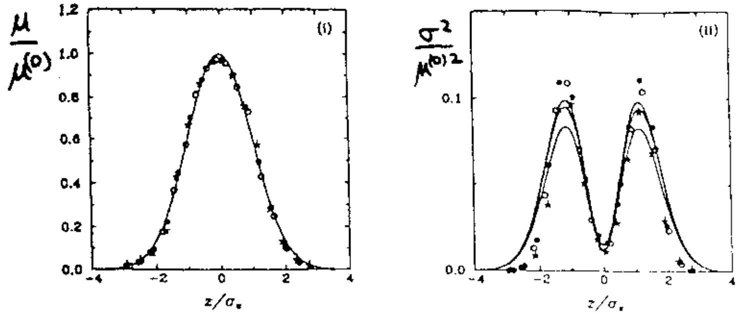


$$\sigma^2 = \beta^2\mu\,(\alpha\mu^{(0)} - \mu) \Rightarrow \underbrace{\frac{\mu}{\mu^{(0)}} + \frac{\sigma^2}{\mu\,\mu^{(0)}}}_{y} = \alpha\beta^2 + (1-\beta^2)\frac{\mu}{\mu^{(0)}}$$

PROBE NO.	d_0^*(mm)	α	β^2
1	0.54	1.31	0.16
2	0.30	1.25	0.22
3	0.13	1.33	0.25
4	0.10	1.25	0.33

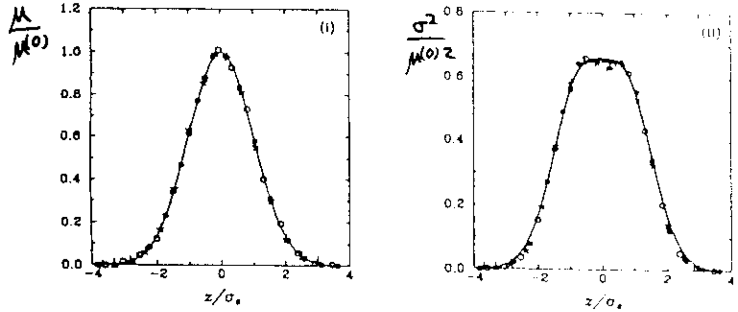
* $d_0 = (\text{sample volume})^{1/3}$

FIGURE 13



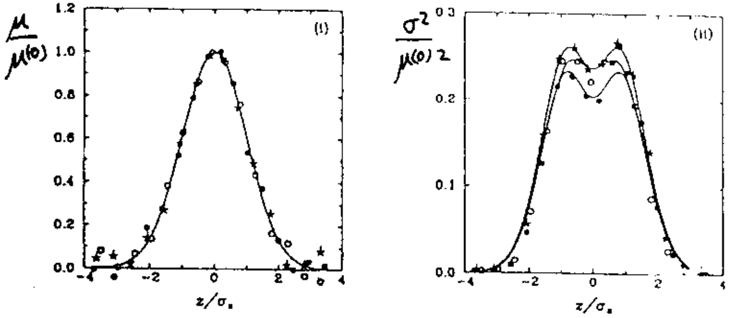
13(a)

$x/x_0 = 6.45 \times 10^{-3}$



13(b)

$x/x_0 = 0.323$



13(c)

$x/x_0 = 8.39$

Analysis of data by Sawford and Sullivan (1994). (Experiments of Sawford and Tivendale (1994). NB $x_0 = 0.31$m

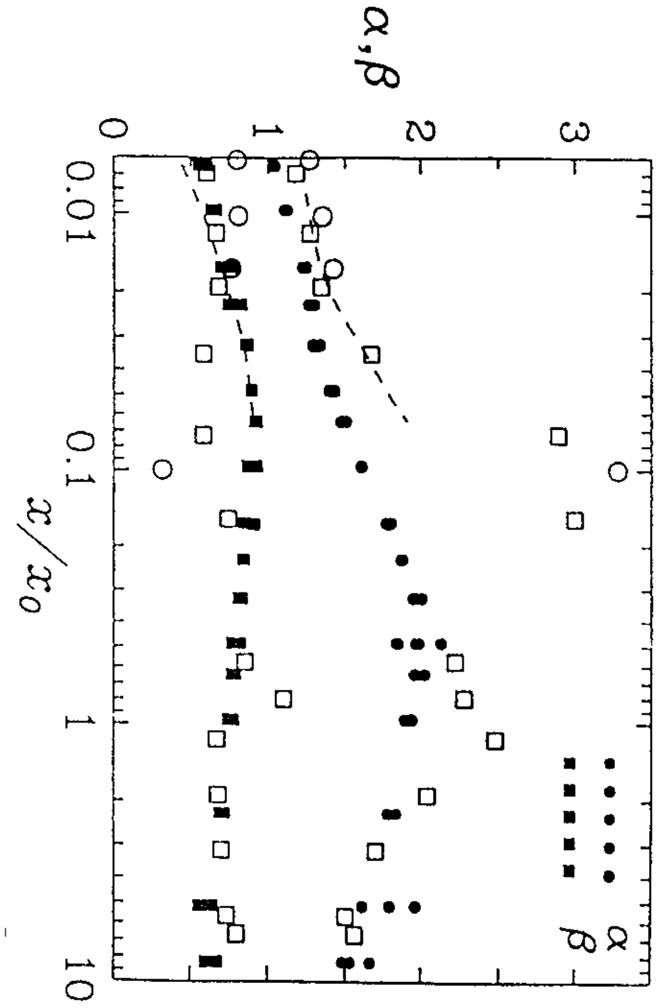
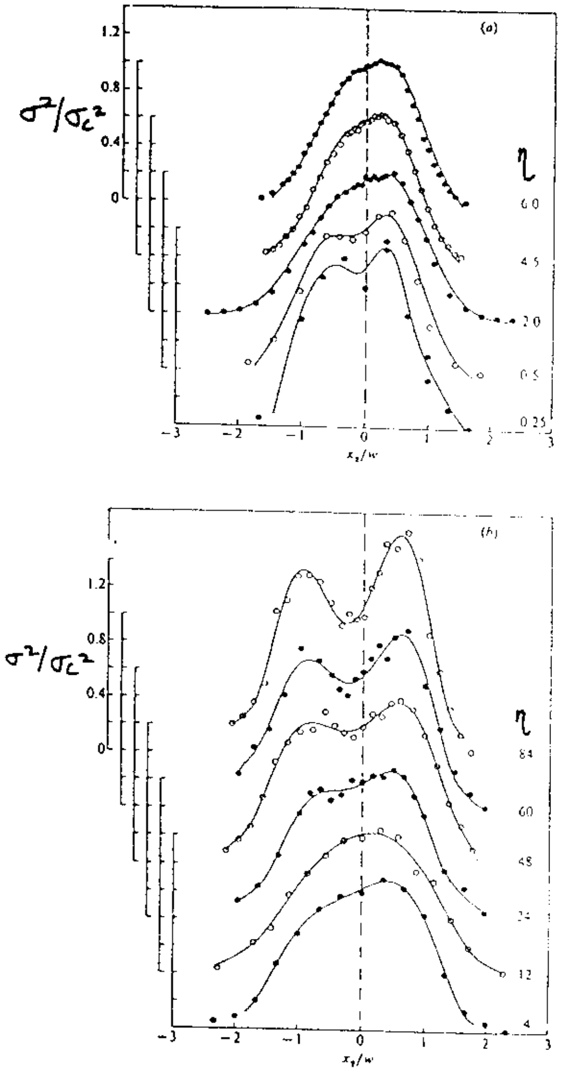


FIGURE 14 Variation of α and β with downwind distance.



Transverse distribution of the mean-square temperature fluctuations, downstream of (a) the 0.051 mm diameter wire, and (b) the ribbon; ——, fitted curves.

FIGURE 15. Analysis of data by U. Karnik and S. Tavoularis (JFM 202, 1989, 233–261) (Continuous line source in uniformly mean sheared turbulent flow)

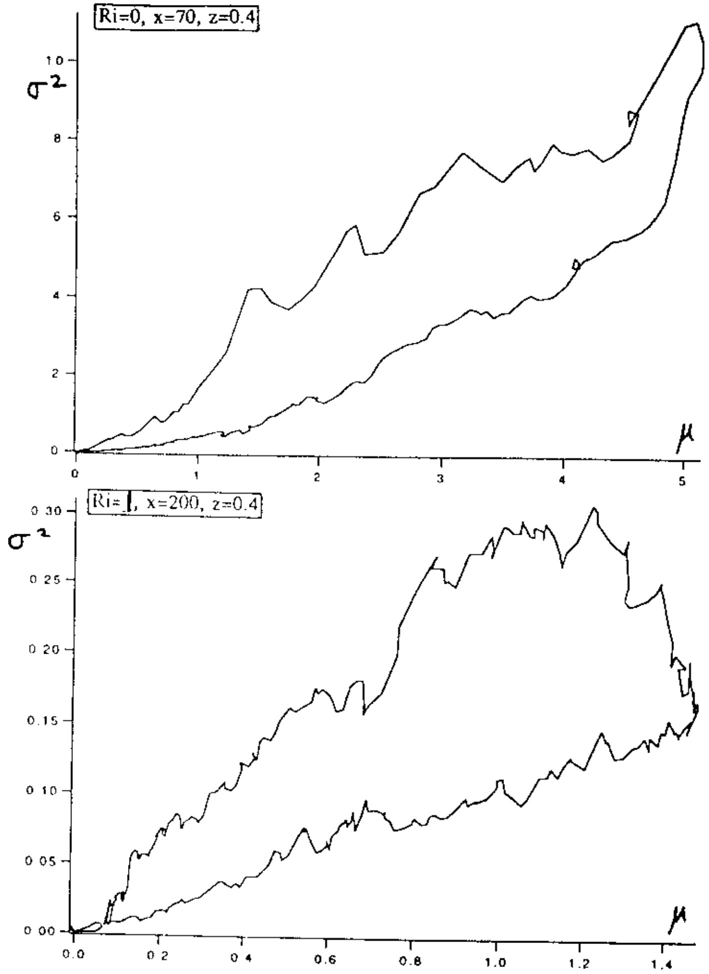


FIGURE 16 Two examples of σ^2 versus μ plots for the experiments shown in Figure 2. Analysis by WB Zimmerman (Zimmerman and Chatwin 1994).

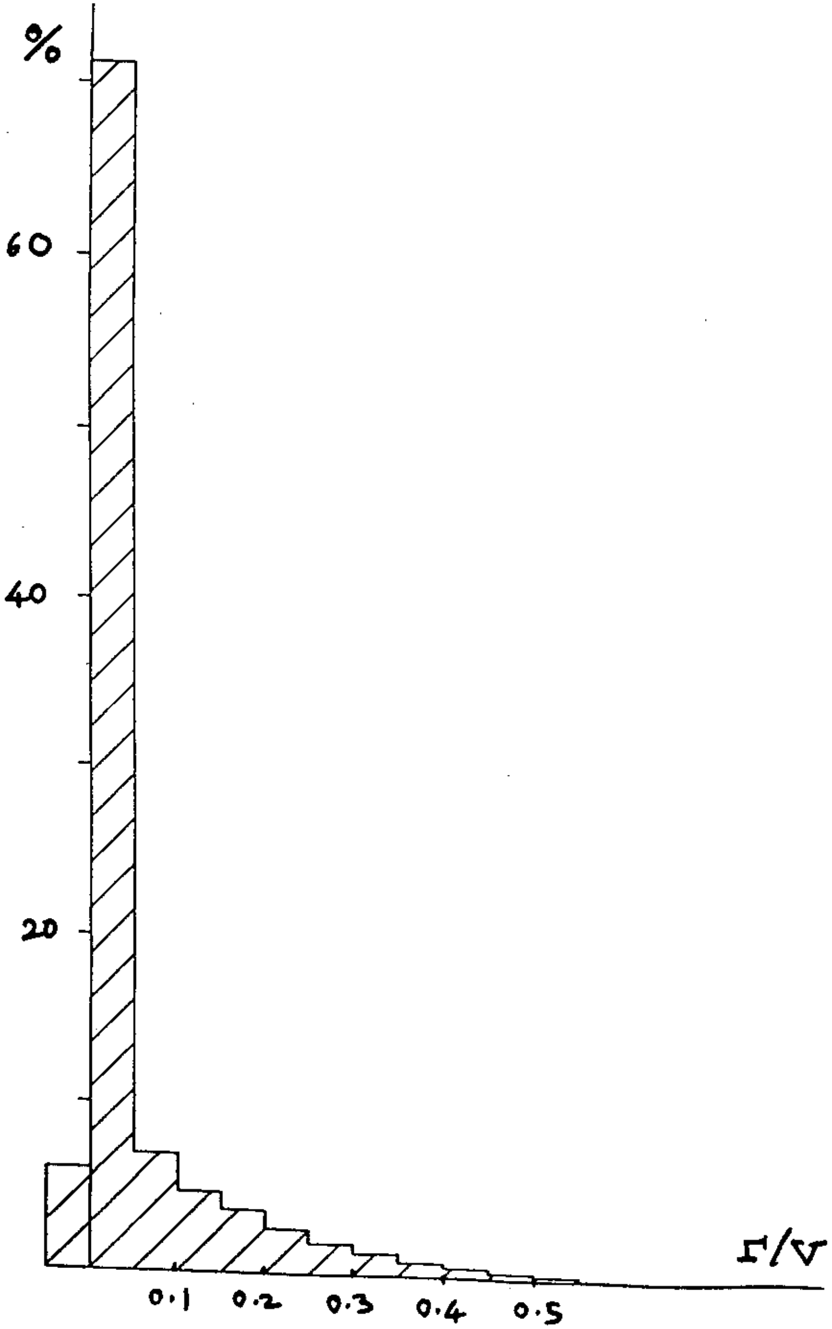


FIGURE 17 Raw data (≈ 6% of "concentration" readings negative) from which Figure 6 was derived. (Lewis and Chatwin 1994.)

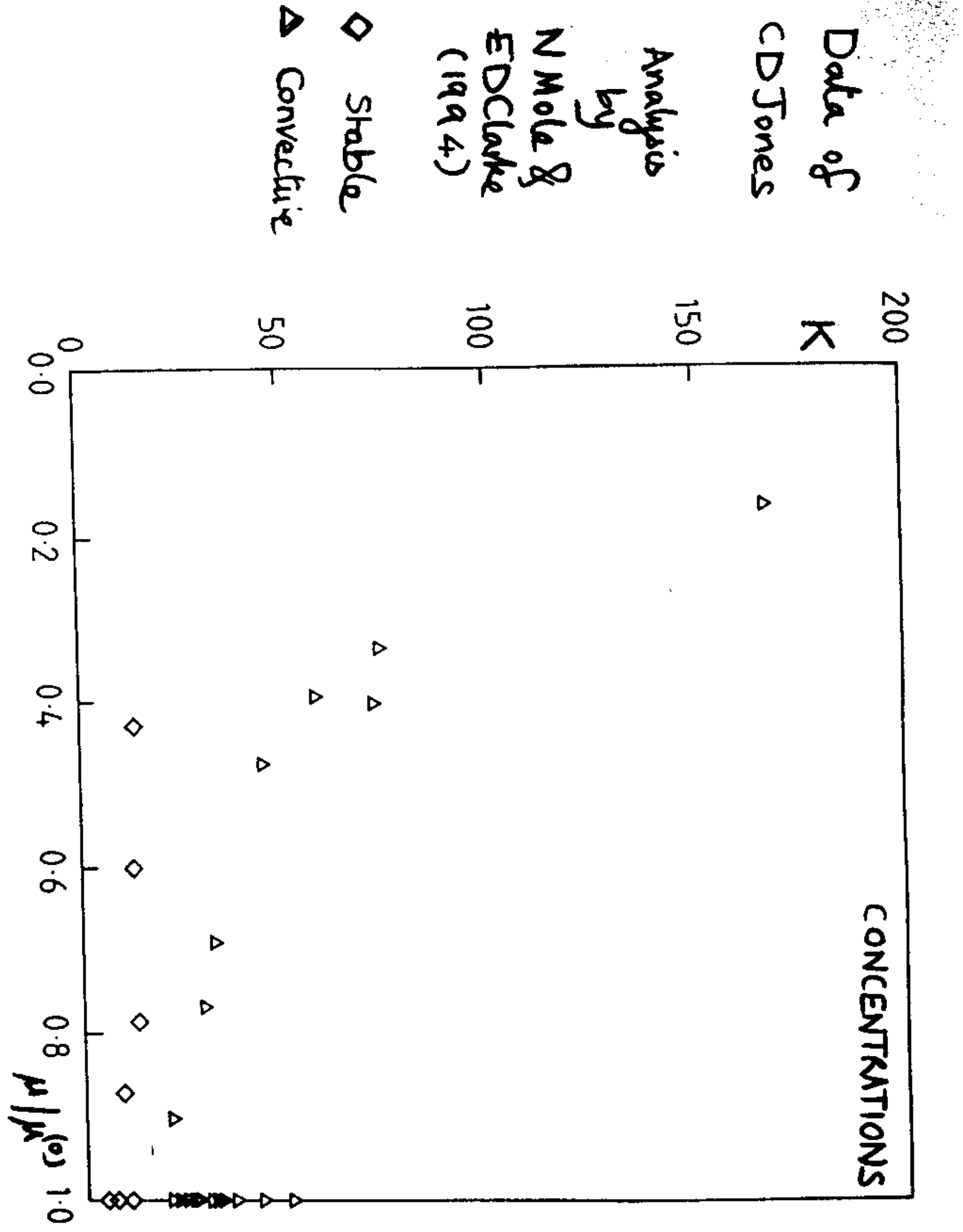


FIGURE 18

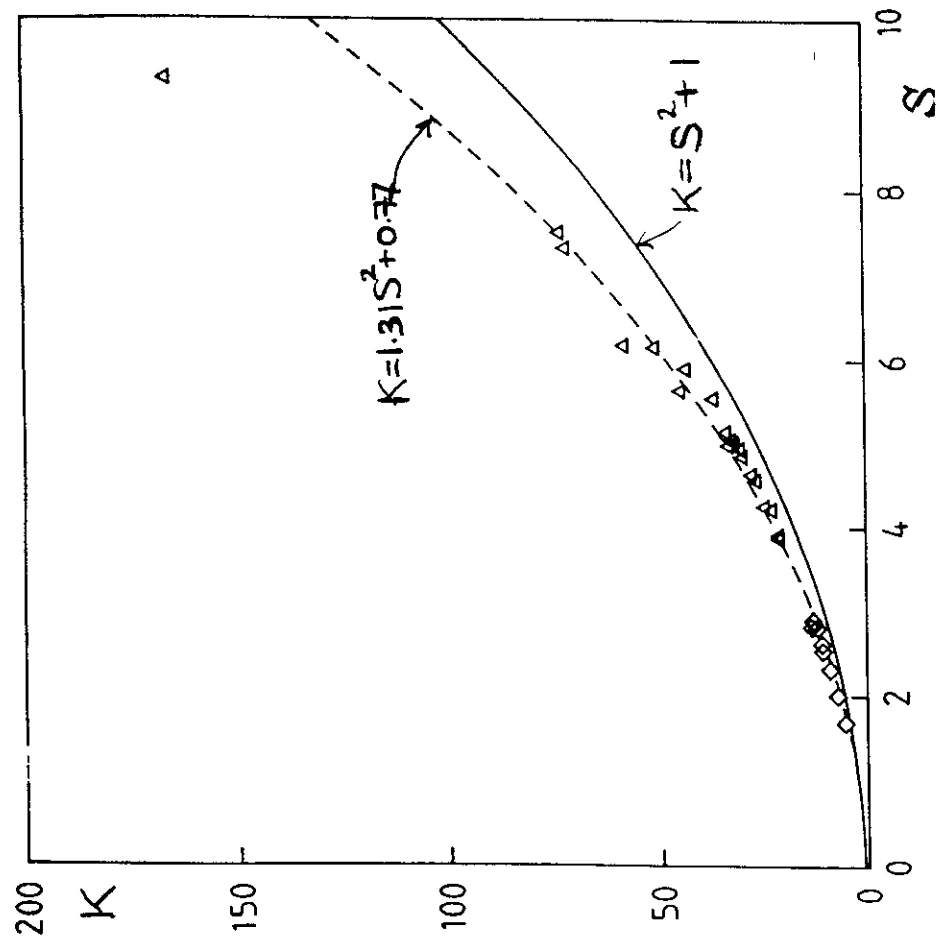


FIGURE 19

Data of CD Jones (same as Figure 18)

Analysis by N Mole & E D Clarke (1994)

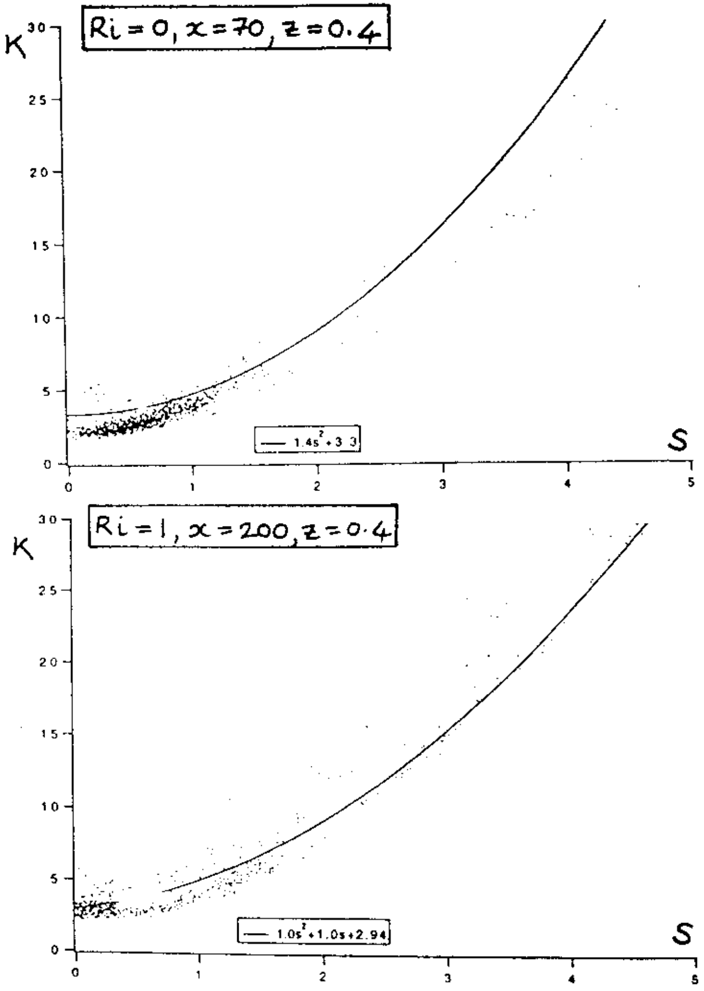


FIGURE 20 (K, S) plots for the two data sets for instantaneous releases that yield FIGURE 16. (Set-up as in FIGURE 2)

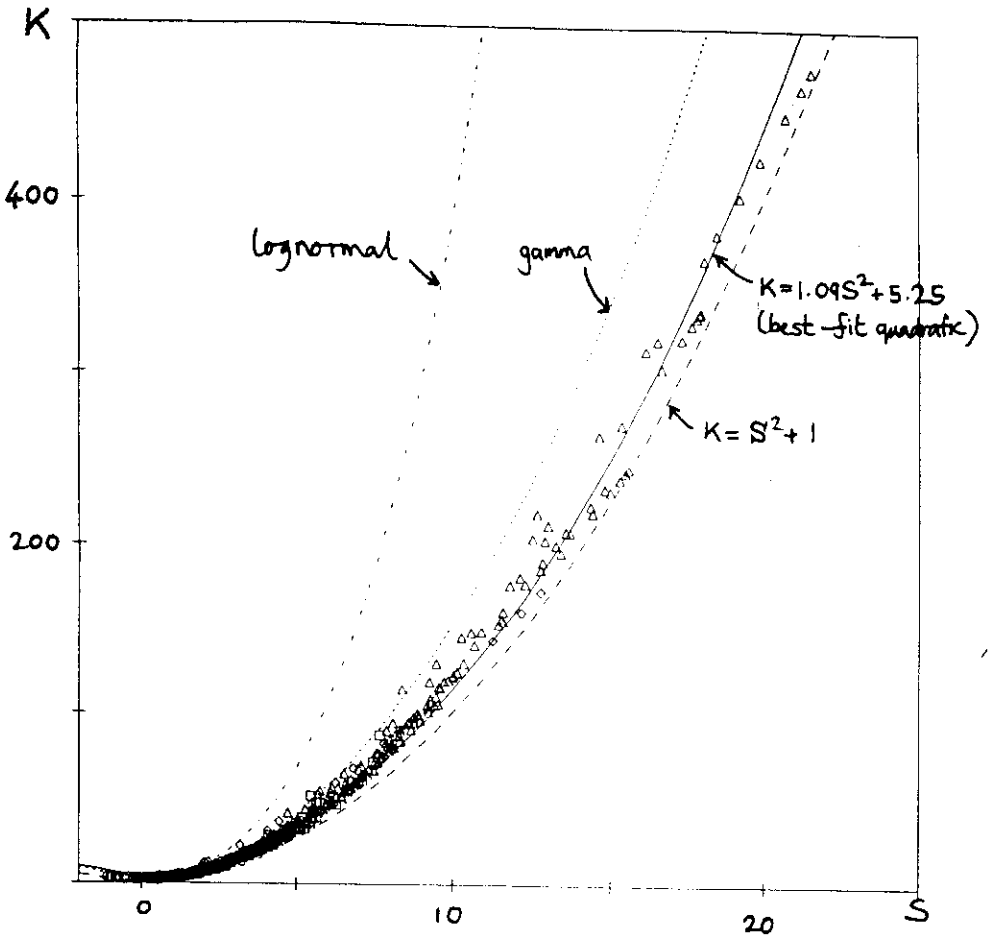


FIGURE 21 (K, S) plots for D_{PT} defined in (44) for data shown in Figures 18, 19. From Mole and Clarke (1994).