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

"Air Pollution Modelling for Environmental Impact Assessment"

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AIR POLLUTION MODELLING FOR ENVIRONMENTAL IMPACT ASSESSMENT

P.C.Chatwin.

ICTP TRIESTE

College on Atmospheric Boundary Layer Physics

21 May - 15 June 1990

WORKSHOP ON

AIR POLLUTION MODELLING FOR ENVIRONMENTAL IMPACT ASSESSMENT

4 - 15 June 1990

Lectures by

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Lecture 1. Monday 4 June 14.30 - 15.30

CONCENTRATION FLUCTUATIONS IN THE ATMOSPHERE

Lecture 2. Tuesday 5 June 14.30 - 15.30

AIR POLLUTION PROBABILITY DENSITY FUNCTIONS

Summary

The aims of the lectures are:

- (i) to explain what concentration fluctuations are;
- (ii) to illustrate their importance in environmental impact assessment;
- (iii) to discuss some factors relevant to the quantitative description of concentration fluctuations;
- (iv) to describe a framework for this description.

It will be clear from the lectures, and from others later in the Workshop, that there is rapidly increasing awareness of the importance of concentration fluctuations and, consequently, much research activity into their properties. Not surprisingly there are still many unsolved problems, and a by-product of the lectures will be to highlight one or two of the most important.

Lecture 1. Concentration Fluctuations in the Atmosphere

1. *TURBULENCE AND ATMOSPHERIC DISPERSION*

The Earth's atmosphere is in turbulent motion. This means that the velocity **T(x,t)** of the air at every point **x** and every time t is a **random variable.** This fact is a matter of common experience, and there is no contradiction between it and the further fact that the evolution of T(x,t) with **x** and t is governed by the laws of mechanics. For the atmosphere the most important of these laws are those of **conservation of mass** almost always adequately represented by

$$\nabla . T = 0 \qquad (1)$$

in the atmosphere (equation of continuity), and **Newton's Second Law** (which becomes the Navier-Stokes equations for fluids like air).

In these two lectures it will not be necessary to use the Navier-Stokes equations. (But note that they are necessary in a full description of an air pollution problem.) From our point of view, it is necessary only to recognise that the randomness of the velocity field causes the **concentration** Γ(**x**,t) of a pollutant in the atmosphere to be random also. The pollutants considered in these lectures will be those - the vast majority in practice - whose concentrations are determined by two processes, namely the random transport (**advection**) by the air that has already been mentioned, and **molecular diffusion.** I do not, in particular, consider processes like chemical reactions. The equation governing Γ(**x**,t) is then

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

where κ is the **molecular diffusivity.** In addition there are boundary and initial conditions.

A selection of some typical data records of concentrations is given in Figures 1(a) to 1(f). While there are interesting and important differences between these records, of which some will be discussed later, each of them clearly indicates the randomness of Γ(**x**,t). This randomness is driven by the term involving T in (2).

Given that the concentration of a pollutant in the atmosphere is not **predictable,** it is necessary to consider (a) whether the unpredictability is important and, if so, (b) how it can be incorporated into quantitative work on environmental impact assessment.

Before considering these questions it will be useful to introduce some notation and to make some fundamental points.

2. *BASIC CONCEPTS AND NOTATION*

Whether or not the unpredictability of Γ(**x**,t) is **practically** important, its quantitative scientific description clearly requires the use of **statistics.** Statistical ideas and techniques are applicable 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 "experiments". The definition serves only to determine unambiguously whether, or not, any particular release is a realisation within the ensemble (and therefore covered by the statistical description). From the scientific point of view, what the definition is does not matter; from the practical point of view it is crucial. The definition is then likely to include, apart from obvious points like the location(s) of the sources and the relevant local geography, details of the releases (nature, duration etc.) and the meteorological conditions. Further discussion of the ensemble concept is given by Chatwin (1982) [*J. Haz. Mat.* 6, 213-230] and Carn and Chatwin (1985) [*J. Haz. Mat.* 11, 281-300].

Given the ensemble, there is a definite (albeit unknown) probability distribution describing the possible values of $\Gamma(\mathbf{x},t)$; in particular there is a probability density function (pdf), denoted by $p(\theta;\mathbf{x},t)$, and defined for each $\theta \geq 0$ by

$$p(\theta;x,t) = \frac{d}{d\theta}\left\{\mathrm{prob}[\Gamma(x,t)\leq\theta]\right\} . \tag{3}$$

Another, perhaps more useful, interpretation of $p(\theta;\mathbf{x},t)$ is obtained by noting that $p(\theta;\mathbf{x},t)\delta\theta$ is the probability that $\theta \leq \Gamma(\mathbf{x},t) < \theta + \delta\theta$ (for $\delta\theta$ small and positive). I shall discuss many aspects of the pdf in my second lecture, and it is only necessary to note some obvious properties and definitions here. Since concentrations cannot be negative

$$p(\theta;x,t) = 0 \text{ for all } \theta < 0 ; \tag{4}$$

since $p(\theta;\mathbf{x},t)$ is a pdf

$$\int_0^\infty p(\theta;x,t)d\theta = 1 . \tag{5}$$

In practice there will be a maximum possible concentration $\theta_{\max}(\mathbf{x},t)$ but, because equation (2) cannot be solved for general $T(\mathbf{x},t)$, its precise value is unknown. Clearly $p(\theta;\mathbf{x},t) = 0$ for all $\theta > \theta_{\max}$ so the upper limit in (5) can be replaced by $\theta_{\max}$ if required, but this is not usually useful. The **ensemble mean** (or simply **mean**) concentration will be denoted by $C(\mathbf{x},t)$ and is obtained from $p(\theta;\mathbf{x},t)$ in the standard way:

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

The **concentration fluctuation** $c(\mathbf{x},t)$ is defined (following the classical definitions of Reynolds for the turbulent velocity field) by:

$$\Gamma(x,t) = C(x,t) + c(x,t) . \tag{7}$$

The **mean square fluctuation** will be denoted by $\overline{c^2}(\mathbf{x},t)$ and is obtained from $p(\theta;\mathbf{x},t)$ by the equation

$$\overline{c^2}(x,t) = \int_0^\infty [\theta - C(x,t)]^2 p(\theta;x,t)d\theta = \int_0^\infty \theta^2 p(\theta;x,t)d\theta - C^2(x,t) . \tag{8}$$

Of course $\overline{c^2}(\mathbf{x},t)$ is the variance of $\Gamma(\mathbf{x},t)$ and $\surd\{\overline{c^2}(\mathbf{x},t)\}$ – the <u>rms</u> concentration – is its **standard deviation.** The ordinary statistical symbols for C and $\overline{c^2}$ would be μ and σ^2 respectively; unfortunately this usage is not yet conventional in work on turbulent diffusion, including atmospheric dispersion.

It requires emphasis that, as the notation suggests, $p(\theta;\mathbf{x},t)$ does depend **explicitly** on **x** <u>and</u> t for most ensembles; so therefore do statistical properties of $\Gamma(\mathbf{x},t)$ like $C(\mathbf{x},t)$ and $\overline{c^2}(\mathbf{x},t)$. This is true, for example, when the ensemble is concerned with dispersion following sudden release of a finite quantity of pollutant into the atmosphere. Consideration of this type of ensemble is relevant in assessing the environmental consequences of the accidental release of flammable materials (like liquid natural gas) from either a fixed container (like a storage tank) or a mobile one (like a ship or a lorry). In such a case the pollutant cloud tends to spread, and therefore dilute, as it is dispersed by the atmospheric turbulence and the pdf must change with t at each point $\mathbf{x}$ (and with $\mathbf{x}$ at each time time t). Figure 2 shows estimates of $C(\mathbf{x}_0,t)$ and $\overline{c^2}(\mathbf{x}_0,t)$ at one location $\mathbf{x}_0$ for Thorney Island; the fact that the estimates depend on t illustrates the comments above. For such an ensemble estimates of statistical properties like C and $\overline{c^2}$ can be obtained only by taking appropriate averages of the results of many repeat experiments; in essence this was the method used to obtain the estimates shown in Figure 2.

There are some ensembles where $p(\theta;\mathbf{x},t)$ does not depend explicitly on t, or where this is a reasonable practical approximation. This has to be regarded as an exceptional circumstance, but it is sometimes appropriate 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, on average, not change with t during the dispersion period of interest; the technical term for such an ensemble is statistically stationary. Although statistical properties like p, C and $\overline{c^2}$ can still be estimated by averages over repeat experiments, it is much more convenient (and much cheaper) to obtain such estimates by exploiting a mathematical property that holds for such ensembles, namely that ensemble means can be estimated from the results of one experiment by appropriate time averaging. For statistically stationary ensembles, the statistical properties do not depend on t; thus, for example, $C(\mathbf{x},t) = C(\mathbf{x})$, and it can be estimated by $\hat{C}(\mathbf{x};T)$, where

$$\hat{C}(x,T) = \frac{1}{T}\int_{t-\frac{1}{2}T}^{t+\frac{1}{2}T}\Gamma(x,s)\,ds\ , \qquad (9)$$

where the integrand $\Gamma(\mathbf{x},s)$ is obtained from the record of a single experiment.

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 very often assumed that all averages (or means) considered in studies of phenomena like atmospheric dispersion that involve turbulence are time averages. Such an assumption is wrong and potentially dangerous.

I want to make one further comment on this important point. Not only is the simplest (and most natural) theory of atmospheric dispersion not concerned with time-averaged quantities, but these quantities are themselves random variables with their own statistical theory (which is not simply related to that given above). It is still, unfortunately, common to see the word "concentration" used without qualification in papers dealing with safety standards or, inexcusably, papers reporting original research, and only careful perusal makes it clear that what is meant is a "time-averaged concentration". Moreover the period over which the time average is taken is often not stated, or difficult to discover. (I have seen periods ranging from 10s to 1hr!) However, as noted, the time-averaged concentration is a random variable, whose statistical properties depend explicitly on the period of integration, i.e. on T in equation (9). This is illustrated by Figure 3. Hanna (1984) [*Atmos. Envir.*, 18, 1091-1106] quotes evidence that, even when T is as large as 1 hr, the time-averaged concentrations can exhibit a factor of two variability even for a fixed hourly average wind velocity. If time-averaged concentrations are to be considered, it should be because they are what is relevant to the assessment of a particular environmental hazard and then the choice of T should be determined by the same considerations. The subject of the last few paragraphs is discussed further by Chatwin and Allen (1985) [*Tellus*, 37B, 46-49].

3. *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 $I(\mathbf{x},t)$, where

$$I(x,t) = \sqrt{\overline{c^2}(x,t)}/C(x,t) \ . \qquad (10)$$

Typically values of I are of order unity, as evidenced for example by the 0.07s data in Figure 3. The variation of I with $\mathbf{x}$ in statistically stationary ensembles is interesting. Consider, as a specific example, the plume downwind of a factory chimney. At a fixed distance x downwind, experiments show that I has a minimum value on the centre-line whose value depends on factors like the chimney diameter and is usually of order 1. As distance from the centre-line increases, so do the values of I; some typical profiles are shown in Figures 4 and 5. The variation of I with downwind distance x along the centre-line is a problem that has generated some controversy; that controversy will not be discussed here since it is difficult to understand why some researchers regard it as so important.

There is therefore no doubt nowadays that the magnitude of the statistical variability in any measurement of $\Gamma(\mathbf{x},t)$ is (at least) comparable with the value of the measurement itself. There is therefore very much purely scientific interest in gaining a deeper understanding of the quantitative behaviour of properties like $\overline{c^2}(\mathbf{x},t)$ and $p(\theta;\mathbf{x},t)$.

But is it necessary to take account of variability in assessing the environmental impact of air pollution? Or is it adequate for practical purposes to continue to use, and to refine, models - like Gaussian plume models - that consider only the mean concentration (or time-averaged concentrations) and take no account of fluctuations? To some extent the answers to these important questions depend on the particular pollutant and on the particular hazards that are of concern. But I am in no doubt that, in general, fluctuations ought to be an integral part of air quality models, and I shall devote the remainder of this lecture to discussing two particular examples.

4. *FLAMMABLE GASES*

A mixture of a flammable gas like methane (CH_4) and air will support a flame in the presence of an ignition source only if the concentration by volume of the flammable gas lies between the appropriate flammable limits (or stoichiometric limits) for that gas, i.e. only if

$$\theta_1 < \Gamma(x,t) < \theta_2 \ , \qquad (11)$$

where θ_1 and θ_2 are properties of the gas. For CH_4, $\theta_1 = 0.05$ and $\theta_2 = 0.15$. With the statistical viewpoint considered in these lectures, it follows that the probability $P(\mathbf{x},t)$ that flammable conditions (i.e. potential danger) exist at point $\mathbf{x}$ at time t in a gas-air mixture is given by

$$P(x,t) = \int_{\theta_1}^{\theta_2} p(\theta;x,t)d\theta \ . \qquad (12)$$

Thus $P(\mathbf{x},t)$ is equal to the area between that part of the curve of $p(\theta;\mathbf{x},t)$ agaionst θ, and the θ-axis, that is bounded by the lines $\theta = \theta_1$ and $\theta = \theta_2$.

This definition is illustrated in Figure 6, taken from Birch, Brown and Dodson 1980 [*18th International Symposium of the Combustion Institute*, Waterloo, Canada; also *Report No. MRS E 374* (June 1980), Midlands Research Station, British Gas]. This section of the lecture is based on that work. The shaded area in two of the diagrams in Figure 6 is equal to $P(\mathbf{x},t)$ defined in equation (12). By means of a series of experimental measurements of $p(\theta;\mathbf{x},t)$, Birch, Brown and Dodson were able to evaluate $P(\mathbf{x},t)$ by integration, and the solid curves in Figures 7(a) and 7(b) show some of their results. (The methane jets used in these experiments were statistically stationary so, in fact, p and

P are independent of t. Also the curves are smooth because numerical interpolation and smoothing techniques were applied to the values of P determined through use of (12).)

Birch, Brown and Dodson also made direct measurements of $P(\mathbf{x})$ by counting the proportion of 400 repetitions in which a spark of fixed duration and energy (100mJ) led to flame formation or ignition. The results are shown by the solid points (and associated error bars) in Figures 7(a) and 7(b). It can be seen that there is very good agreement between the two independent sets of measurements of $P(\mathbf{x})$. This is strong experimental confirmation for the validity of the statistical description of the dispersion process that led to equation (12).

By contrast, Birch, Brown and Dodson asserted that the mean concentration $C(\mathbf{x})$ is "of little value in assessing flammability", and this remark, that invalidates many conventional methods, is based on comparisons like that shown in Figure 7(b) where the dashed curve, which is the profile of $C(\mathbf{x})$, bears no relation to the data.

5. *TOXIC GASES*

This section summarizes some results in three papers [Griffiths and Megson 1984 *Atmos. Envir.* 18, 1195–1206; Ride 1984 *J. Haz. Mat.* 9, 235–240; Griffiths and Harper 1985 *J. Haz. Mat.* 11, 369–372].

Early models of the degree of harm produced in a population by exposure to a toxic gas were based on the dosage, defined as the product of the "concentration" and the time of exposure. Use of this definition presumed – wrongly – that the concentration was a constant, but it was for other reasons that such models were replaced. In particular limited experimental data indicated that it was necessary to account for the different physiological response to **high** concentrations by **weighting** the concentration in the definition of dosage. This led to the consideration of a new dosage (which Ride calls "dosement" to distinguish it from the earlier usage) D where, for a time of exposure T to a concentration Γ,

$$D = \int_0^T \Gamma^n dt \ , \tag{13}$$

and the index n is greater than 1 and depends on the gas. Understandably (and fortunately) there is little experimental evidence to fix values of n, but Griffiths and Megson suggest 2.00–2.75 for NH_3 and 2.75 for Cl_2, while Ride quotes a value of 1.8 for HCN.

In practice, use of D continued to assume that Γ in equation (13) was constant in time so that D was replaced by $\Gamma^n T$. As is obvious from Figures 1(a) to 1(f) this assumption is manifestly incorrect. The papers quoted above showed by very simple (and still unrealistic) modelling that inclusion of the fluctuations in Γ would give greatly enhanced mortality rates. Figure 8, taken from Griffiths and Harper, shows the type of argument that was used. It was based on comparing the effects of the two different exposure patterns shown in the top diagram, and the remarkable change in mortality rates is shown in the bottom diagram. In technical terms, the differences in the exposure patterns illustrate the phenomenon of **intermittency**, to be considered in some detail in my second lecture.

Lecture 2. Air Pollution Probability Density Functions

1. *SOME THEORETICAL CONSIDERATIONS*

Given that it is important for practical, as well as scientific, reasons to know more about the statistical properties of $\Gamma(\mathbf{x},t)$, this lecture will consider some of the problems involved in this, and some recent theoretical and experimental research. Although I have not, and shall not, put primary emphasis on complicated mathematics, I do want at the beginning of this lecture to discuss briefly some theory because it provides a clear context for the remainder of the lecture.

Manipulation of equation (2), using equation (1) and (in some situations) the Navier-Stokes equations, establishes equations for the statistical properties that are of concern. In the same way that C and c denote – see equation (7) – the mean and fluctuating components of Γ, I use U and u to denote, respectively, the mean and fluctuating components of the velocity field T, so that T = U + u. Both U and u are functions of **x** and t, and both can be shown to have zero divergence, i.e. they satisfy the equation of continuity – equation (1). Then equation (2) is the same as

$$\frac{\partial C}{\partial t} + \frac{\partial c}{\partial t} + \nabla.(UC+Uc+uC+uc) = \kappa\nabla^2 C + \kappa\nabla^2 c \;. \qquad (14)$$

The mean of equation (14) – and I emphasize again that the term "mean" signifies a probability average like those in equations (6) and (8) and not a time-average (or a space-average) – is

$$\frac{\partial C}{\partial t} + \nabla.(UC) + \nabla.(\overline{uc}) = \kappa\nabla^2 C \;, \qquad (15)$$

where the overbar denotes an ensemble mean (or probability average) as is still (unfortunately) the conventional notation – see the remarks after equation (8).

Equation (15) is perhaps the simplest equation in the whole of turbulence that exhibits the closure problem which bedevils all theoretical research into the subject. Even if it can be assumed that the mean velocity field $U(\mathbf{x},t)$ is known, or if it can be modelled with sufficient accuracy, the term involving uc has introduced a new unknown field that is important (for otherwise there would be no effect of turbulence) and is not closely related to $C(\mathbf{x},t)$. It is easy to obtain an equation for uc but that introduces new unknowns that are important. There is no known way of terminating the process that is theoretically satisfactory in the sense, particularly, that the termination process (or closure hypothesis) that may be chosen for equation (15) does not lead to any scientifically well-based termination process for equations – see equations (16) and (17) below – for other statistical properties of $\Gamma(\mathbf{x},t)$, equations that also exhibit – but more severely – the closure problem. This serious objection applies to the concept of eddy diffusivity that is frequently used – and with some success – to close equation (15). I will therefore make no further reference to eddy diffusivities in these lectures, although I expect other Workshop lecturers to do so!

It is straightforward to derive equations for other statistical properties. Those for $\overline{c^2}(\mathbf{x},t)$ and $p(\theta;\mathbf{x},t)$ are:

$$\frac{\partial \overline{c^2}}{\partial t} + \nabla.(U\overline{c^2}) + \nabla.\left\{\overline{uc^2}\right\} + 2\overline{uc}.\nabla C = \kappa\nabla^2(\overline{c^2}) - 2\kappa\overline{(\nabla c)^2} \;, \qquad (16)$$

and

$$\frac{\partial p}{\partial t} + \nabla.(Up) + \nabla.(\overline{u\delta[\Gamma(x,t)-\theta]}) = \kappa\nabla^2 p - \kappa\frac{\partial^2}{\partial\theta^2}(\overline{(\nabla\Gamma)^2\delta[\Gamma(x,t)-\theta]}) \quad . \qquad (17)$$

In equation (17), the symbol δ denots the **Dirac delta function**.

The left-hand sides of both equations (16) and (17) exhibit the closure problem in the same way that equation (15) does, namely by the appearance of new unknown terms involving combinations of the velocity and concentration fields. But in those equations there are also new closure problems with the last term on the right-hand side of each equation. The term in equation (16) represents the **dissipation** of $\overline{c^2}$ through the action of molecular diffusion; this is an essential process in understanding the behaviour of $\overline{c^2}(\mathbf{x},t)$. Except for the mean concentration $C(\mathbf{x},t)$ – the term involving κ in equation (15) can probably be neglected for all practical purposes – molecular processes are important to the proper understanding of all statistical properties of $\Gamma(\mathbf{x},t)$. Unfortunately such understanding is not yet available (except for special circumstances not relevant to atmospheric dispersion).

Further discussion of the points summarized above, and more mathematical details, are given in Pope (1985) [*Prog. Energy Combust. Sci.* 11, 119–192] and Chatwin (1989) [*Lecture Series 1989 –03, Turbulent Shear Flows*, von Karman Institute for Fluid Dynamics, Rhode-St-Genese, Belgium; also *Brunel University Department of Mathematics and Statistics Technical Report TR/02/89*].

2. *THE EXPERIMENTAL DETERMINATION OF* $p(\theta;\mathbf{x},t)$

Given that the theoretical difficulties associated with the full equations governing $p(\theta;\mathbf{x},t)$ and the other statistical properties of $\Gamma(\mathbf{x},t)$ have so far proved insuperable, progress must be made in other ways. This section deals with experimental methods.

Laboratory determinations of $p(\theta;\mathbf{x},t)$ for statistically stationary ensembles have been commonly made for about 15 years. Some examples have already been seen in Figure 6, and others are shown in Figures 9 and 10. These graphs show many interesting features. In the first place, it is obvious from Figure 6 that the **shape** of the graph of p against θ varies substantially from place to place in the flow. On the centre-line, the graph has a single maximum (is **unimodal**) at a non-zero value of θ, and has approximately the shape associated with the familiar **Normal distribution**. (Since concentrations cannot be negative the distribution cannot be exactly Normal, and the graph has indeed a slight negative **skewness** with a tail towards low values of θ.) As one moves away from the centre-line, the value of p that is measured at $\theta = 0$ first becomes non-zero and then increases. Also the position of the maximum moves towards $\theta = 0$, the value of p at this maximum decreases, and two of the curves have as a consequence two maxima (are **bimodal**), one at $\theta = 0$. The explanation of these facts is easy to understand, at least qualitatively, and applies to all pdfs of concentration including those in Figures 9 and 10, and all those that apply to atmospheric dispersion. As dispersion progresses following release, the turbulent motion causes more and more **clean fluid** (i.e. ambient air in atmospheric dispersion) to mix with the pollutant. This mixing is more "advanced" at the edges of a cloud or plume (because the ambient fluid is "nearer"); hence the probability of encountering clean fluid (corresponding to $\theta = 0$) is greater at the edges than in the centre. This explanation does not predict that bimodal distributions are inevitable in some parts of the cloud or plume and, indeed, there are many investigations in which the experimentally determined curves of p against θ are **everywhere** unimodal. The maximum is at $\theta = 0$ near the edges and, in some cases, at all points. In other cases the position of the maximum switches to a non-zero value of θ at points near enough to the centre, and this value of θ increases as the centre is approached.

Most experimental determinations of p are made by smoothing **histograms** like those shown in Figure 10, but other methods have been used. The curves in Figure 6, for example, were obtained by determining the first eight **central moments** of $\Gamma(\mathbf{x},t)$ as functions of $\mathbf{x}$, and then applying a maximum likelihood procedure. Given the enormous number of readings of Γ that can now be handled with modern computing techniques, there is a need for more research into the most efficient way of estimating p, and into the statistical errors associated with the chosen method.

Another factor in such experimental work is the types of instrument response that can contaminate experimental readings of Γ. One type is instrument smoothing, occurring because the input concentration signal is averaged, sometimes over time, sometimes over volume (and sometimes both). Such averaging is also inevitable because there is very fine-scale spatial (and temporal) structure within dispersing distributions of pollutant, down to scales of order 10^{-4}m, and no existing instrument can yet resolve such scales with guaranteed accuracy. The records in Figures 1(c) to 1(f) are, of course, output signals of concentration that have resulted from such smoothing; even if, as is likely, these records do not reproduce the input signals with total integrity, they do indicate the presence of the fine-scale structure. A second type of instrument response is noise arising from a variety of sources including physical vibrations and electronics. Such noise is itself random, and has its own pdf. Even if there were no instrument smoothing the output pdf would be the convolution of the input pdf (which is what is required) and the noise pdf. It is therefore necessary to consider methods of deconvolution; any such method obviously requires independent knowledge of the noise pdf which can be obtained by operating the measurement system in the absence of pollutant. The histograms in Figure 10 are believed to be significantly affected by noise. Unfortunately many experimenters account for noise in an unsatisfactory way by choosing a threshold value θ_T of concentration and assuming that all measured values of concentration below θ_T are really zero. Leaving aside doubts about the precise choice of θ_T, which sometimes appears to be very arbitrary, the whole procedure cannot be acceptable since it does not distinguish between true zero values of concentration (if any - see discussion of intermittency later) and small non-zero values below θ_T. This objection has added weight in those frequent situations where the maximum of the graph of p against θ is observed to be at $\theta = 0$ (but see the discussion below). Moreover it allows the inclusion of measured values of Γ above θ_T that, because of contamination by noise, are really below θ_T. All the questions in this paragraph have been investigated for several years by a team at Brunel that includes myself and Dr. Nils Mole. A paper by Mole is to appear soon in *Atmospheric Environment* and earlier papers can be obtained by writing to me.

For reasons of cost and convenience, much experimental work whose results are intended to be relevant to atmospheric dispersion is carried out in wind tunnels. Clearly the value of the results for atmospheric dispersion depends on the accuracy with which salient features of the real situation are modelled. Nowadays there is little fundamental difficulty with reproducing terrain or topography or source characteristics, although each requires great care and effort by the experimental team. But it is difficult, perhaps impossible, to model certain features of the air flow in the atmosphere especially those that involve large scales of the order of tens of metres, or even kilometres. Such features include the deep convective mixing associated with unstable atmospheres and, above all, the meandering of a plume or cloud caused by relatively large-scale horizontal eddies. In a wind tunnel such motions are inevitably inhibited by the presence of the roof and the side walls. (By meandering is meant the motion of the plume (or cloud) as a whole, and this phenomenon is normally responsible for a substantial fraction of the intensity of the concentration fluctuations, the remainder being due to within-plume structure. See, for example, Ride (1988) [*J. Haz. Mat.* 19, 131-137].)

3. *INTERMITTENCY*

All the concentration records in Figures 1(a) to 1(f) have a characteristic feature, namely that there are periods of high "activity" separated by periods of quiescence. This feature is invariably described as intermittency and measured by the intermittency factor $\gamma(x,t)$, defined by

$$\gamma(x,t) = \text{prob}[\Gamma(x,t) > 0] \ . \qquad (18)$$

(This definition is consistent with the use of the symbol γ in Figure 8.) As will be seen briefly in the next section, the concepts of intermittency and intermittency factor are widely used in models of $p(\theta;\mathbf{x},t)$ for the assessment of air quality.

However Chatwin and Sullivan (1989a) [*Phys. Fluids* **A1**, 761–763) and (1989b) [*7th Symposium on Turbulent Shear Flows*, Stanford Univ., 29.4.1 – 29.4.6] have recently cast doubt on the validity of the definition in (18). I believe the points we made there are conceptually so important that I would like to indulge myself by summarising them here. First of all, for reasons discussed above, the presence of noise and fine-scale spatial structure has led many experimenters to use a **concentration threshold** θ_T, so that they replace (18) in practice by

$$\gamma(x,,t) = \text{prob}[\Gamma(x,t) > \theta_T] \ . \tag{19}$$

For reasons discussed in the second of our papers cited above, use of (19) gives measured values of γ that must depend significantly on the choice of θ_T. This is not satisfactory since θ_T is not connected with the real concentrations of the pollutant.

However there is an even more fundamental objection to either (18) or (19). It is well-known that the presence of **molecular diffusion** (the term in equation (2) involving κ) ensures that $\Gamma(\mathbf{x},t)$ is everywhere **positive**, i.e. equation (18) strictly applied gives

$$\gamma(x,t) = 1 \tag{20}$$

for all $\mathbf{x}$ and all $t > t_1$ (where t_1 is the time when dispersion began with a steady continuous source represented by $t_1 = -\infty$). Thus equation (18) is a meaningless definition.

These objections are not to the concept of intermittency itself but to the use of the definition (18) of intermittency factor to quantify the concept. In fact intermittency is intended to measure a property of the dispersion of a pollutant that is entirely **independent** of the existence of molecular diffusion, but depends only on the statistical properties of the turbulent velocity field $T(\mathbf{x},t)$ and factors such as the source geometry and the pollutant release details. In the **hypothetical** situation in which there is no molecular diffusion and in which the pollutant has concentration θ_1 at release, there is no mechanism for transferring pollutant from one fluid (air) volume to another. This hypothetical situation is illustrated schematically in Figure 11 and it is clear that then $p(\theta;\mathbf{x},t)$ has the simple form:

$$p(\theta;x,t) = \gamma_0(x,t)\delta(\theta-\theta_1) + [1-\gamma_0(x,t)]\delta(\theta) \ . \tag{21}$$

Here γ_0 is the intermittency factor defined by equation (18) – which is sensible when there is no molecular diffusion – and depends only on the velocity field and on source properties. A consequence of (21) is that the mean concentration $C(\mathbf{x},t)$ is related to γ_0 by $\gamma_0\theta_1 = C$. But, as already noted, $C(\mathbf{x},t)$ is the one statistical property of the concentration field unaffected by molecular diffusion. Sullivan and I then took the natural step of proposing a new definition of intermittency factor in real situations with molecular diffusion that was consistent with this result for the hypothetical situation. This new definition to replace (18) is:

$$\gamma(x,t) = C(x,t)/\theta_1 \ . \tag{22}$$

The practical worth of the ideas in this section awaits further testing.

4. *THE STRUCTURE OF* $p(\theta;\mathbf{x},t)$ *IN ATMOSPHERIC DISPERSION*

It has been conventional to use the intermittency factor in models for $p(\theta;\mathbf{x},t)$ for use in atmospheric dispersion. The model used has been

$$p(\theta;x,t) = \gamma(x,t)f(\theta;x,t) + [1-\gamma(x,t)]g(\theta;x,t) \ , \qquad (23)$$

where f and g are themselves pdfs with the properties given in equations (4) and (5). Equation (23) is an obvious generalisation of equation (21) and f is the conditional pdf of Γ when attention is restricted to cases where, using the conventional (but misguided) ideas that motivate equation (18), $\Gamma(\mathbf{x},t)$ is positive; in the same way $g(\theta;\mathbf{x},t)$ must then be $\delta(\theta)$.

Fortunately, when equation (22) is adopted as a definition of $\gamma(\mathbf{x},t)$, equation (23) is an exact representation of $p(\theta;\mathbf{x},t)$. However, while f and g still have the properties in equations (4) and (5), their interpretations are somewhat different. Thus $f(\theta;\mathbf{x},t)$ is the pdf of $\Gamma(\mathbf{x},t)$ conditional on the concentration in the hypothetical situation, identical in all respects except for the absence of molecular diffusion, being equal to the source concentration θ_1; it follows, for example, that

$$C_f(x,t) = \int_0^\infty \theta f(\theta;x,t)d\theta \ , \qquad (24)$$

is the ensemble mean concentration over the fluid (air) particles that emanate from the source. The function $g(\theta;\mathbf{x},t)$ is the conditional pdf when the hypothetical concentration is known to be zero; the presence of molecular diffusion in the real situation means that it can no longer be exactly $\delta(\theta)$.

Irrespective of the invalidity of the definition of γ in equation (18), there can be no doubt of the potential practical utility of equation (23). Much attention has been focussed on the ability of a simple structural form for $f(\theta;\mathbf{x},t)$ to model adequately hazards arising in practice. By the term "simple" is meant a pdf in which only a few parameters need to be specified, e.g. two or three. Such parameters include, for example, the (conditional) mean and variance. Among the forms for f that have been, and are being, investigated are the lognormal, the truncated (or clipped) Normal and the beta. Details of some recent investigations can be found in many of the references cited in these lectures but there is no sign yet of a consensus. I would hope to be able to discuss some of the issues in this research in conjunction with my lectures.

I would like to thank Dr. Tirabassi for inviting me to give these lectures, and ICTP for arranging financial support. The research of myself and my colleagues in the areas of turbulent diffusion and atmospheric dispersion has been sponsored by many organisations and I would particularly like to acknowledge the UK Ministry of Defence, the Common Market, the Natural Science and Engineering Research Council of Canada and NATO.

Figure 1 (a)

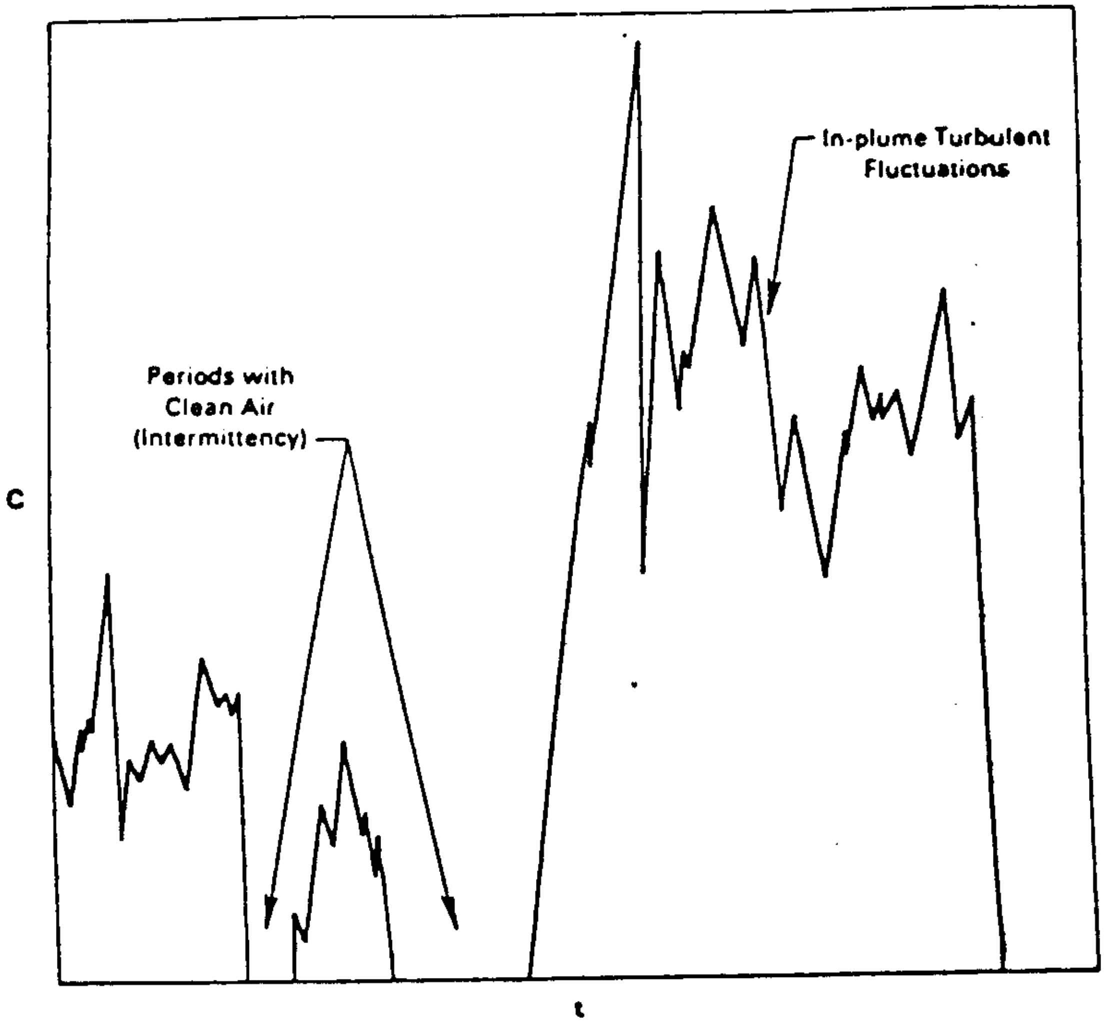


Fig. 1. Example of typical stripchart of concentration as a function of time.

From: Hanna, S.R. 1984 Concentration fluctuations
in a smoke plume. Atmos. Envir. 18, 1091-1106.

Figure 1 (b)

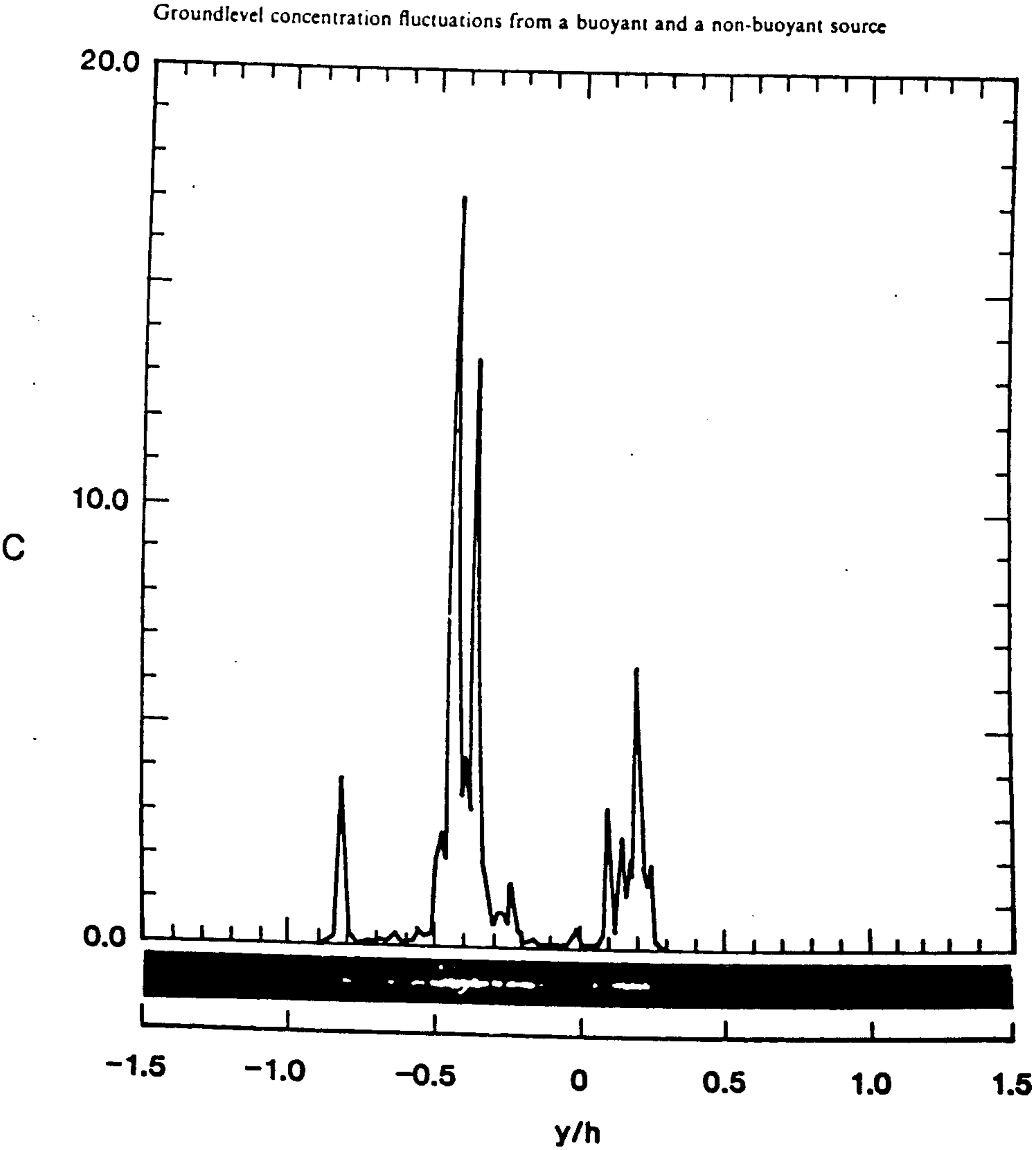


Fig. 7. Upper: Profile of C in an experiment with non-buoyant effluent, at $X = 1.33$. Lower: photograph of fluorescing dye within the laser beam at the same time.

From: Deardorff, J.W. and Willis, G.E. 1984 Groundlevel concentration fluctuations from a buoyant and a non-buoyant source within a laboratory convectively mixed layer. *Atmos. Envir.* **18**, 1297–1309

Figure 1(c)

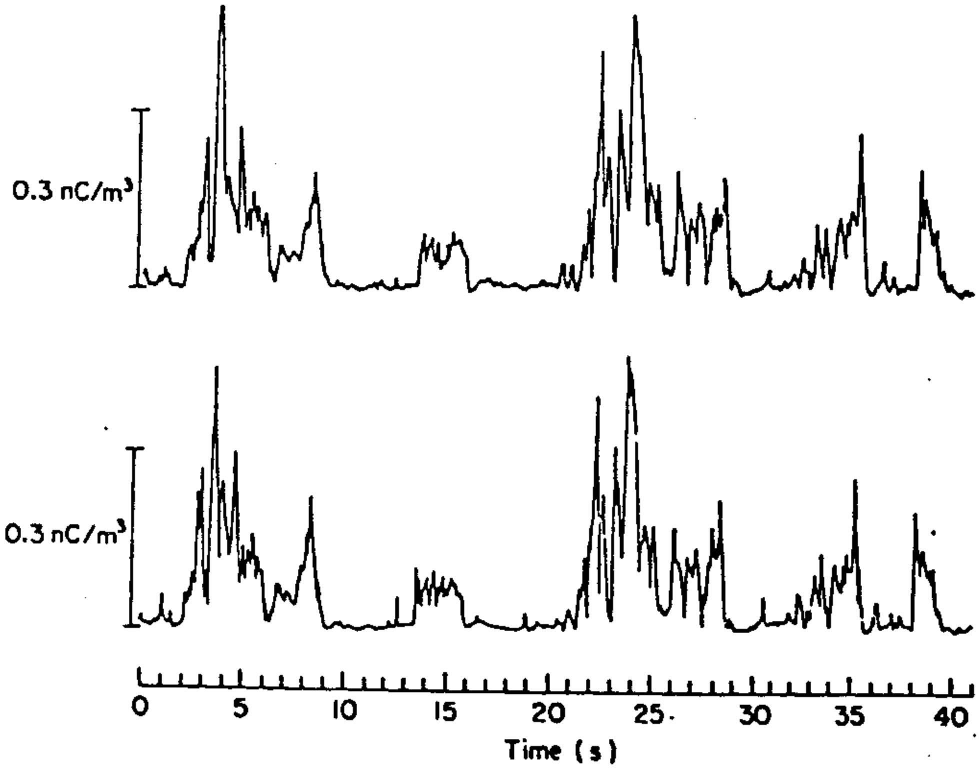
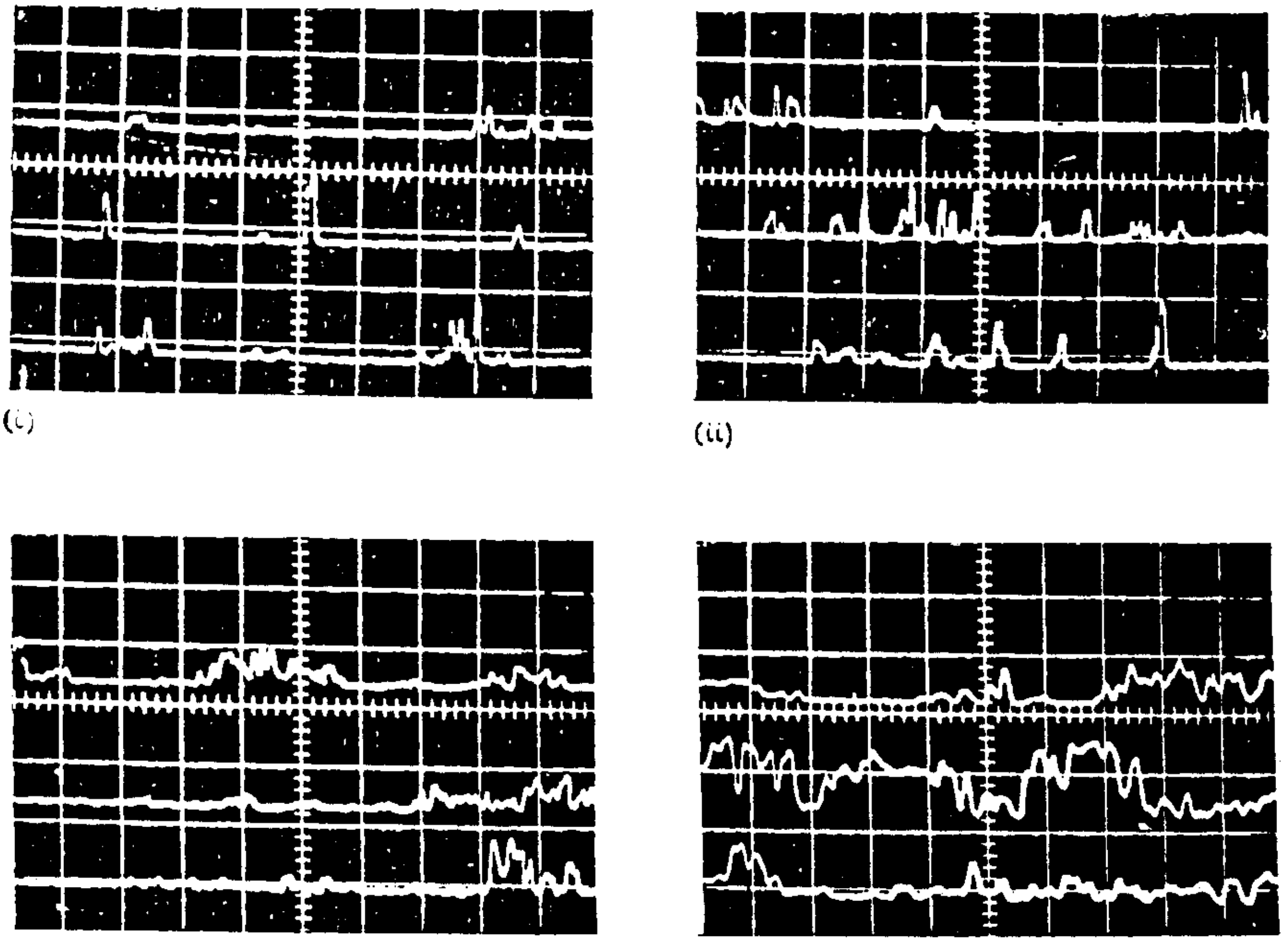


Figure 7. Simultaneous ion concentration measurements at vertically separated points. Upper trace, 1.5 m; lo 1.45 m; ion collectors, 30 m downwind of a continuous point source of ~0.15 μA at 2.2 m height; wind sp 2–5 m/s; slightly unstable conditions, 22.8.75.

From: Jones, C.D. 1977 Ion concentration variations at short distances downwind of continuous and quasi-instantaneous point sources. Pestic. Sci. 8, 84–95

Figure 1(d)



(i) (ii) (iii) (iv)

Fig. 1. Concentration signals on plume centre line, showing effects of fetch and source size. Elevated source, $z_s = 0.19\,H$. (i) $d = 3$ mm, $x/z_s = 2$, (ii) $d = 8.5$ mm, $x/z_s = 2$, (iii) $x/z_s = 16$ (iv) $x/z_s = 32$. 50 m s per division.

From: Fackrell, J. E. and Robins, A.G. 1982 The effect of source size on concentration fluctuations in plumes. *Bound.-Lay. Meteor.* **22**, 335-350.

Figure 1(e)

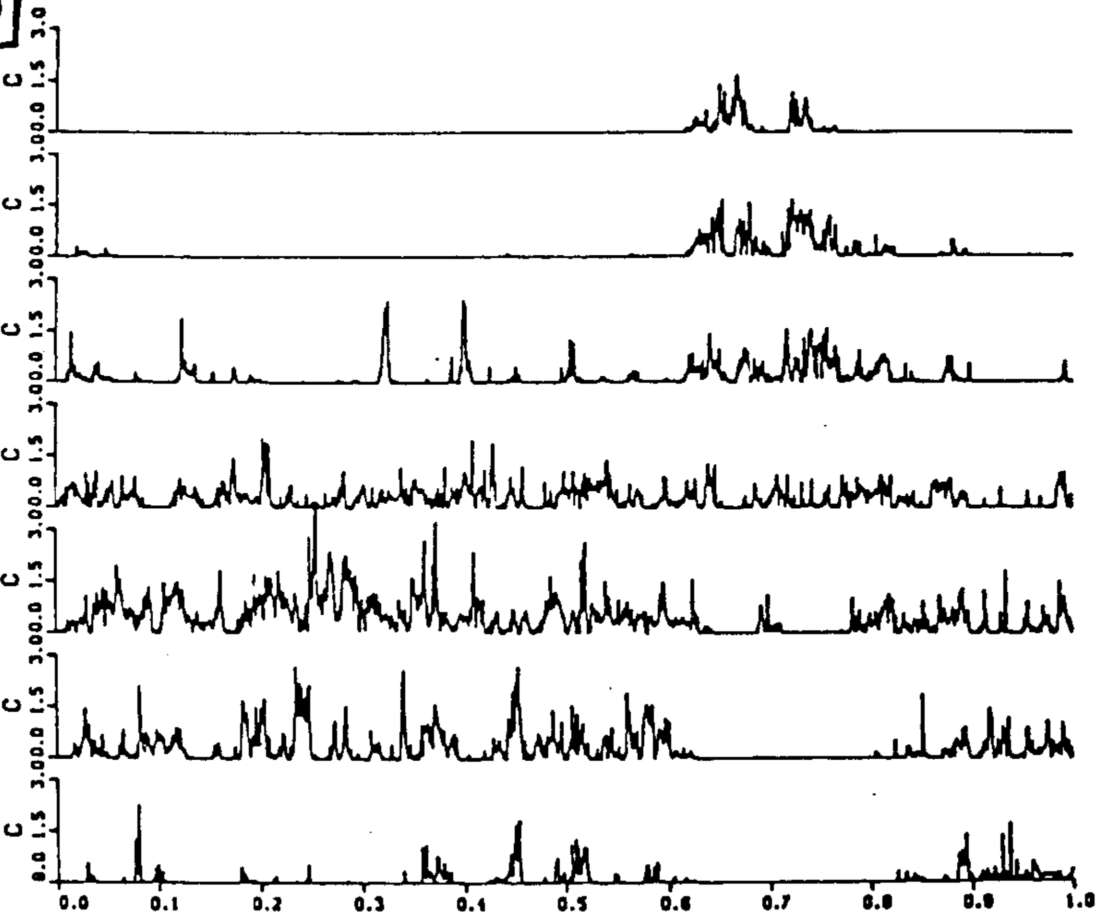


Fig. 2. An example of the signal which appears in the seven detectors in the cross-wind direction during one minute

From: Dinar, N., Kaplan, H. and Kleinan, M. 1988 Characterization of concentration fluctuations of a surface plume in a neutral boundary layer. *Bound.-Lay. Meteor.* **45**, 157-175.

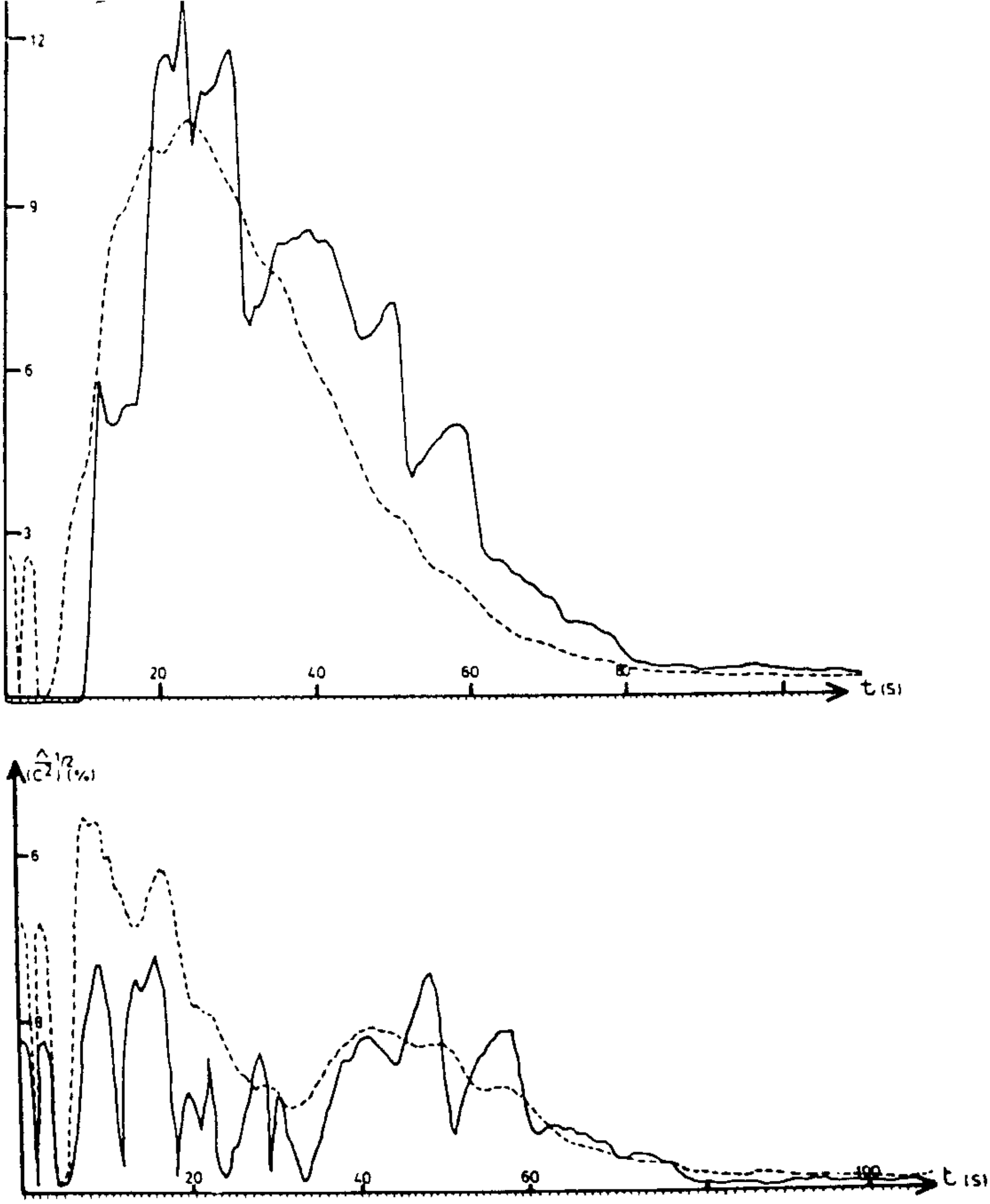
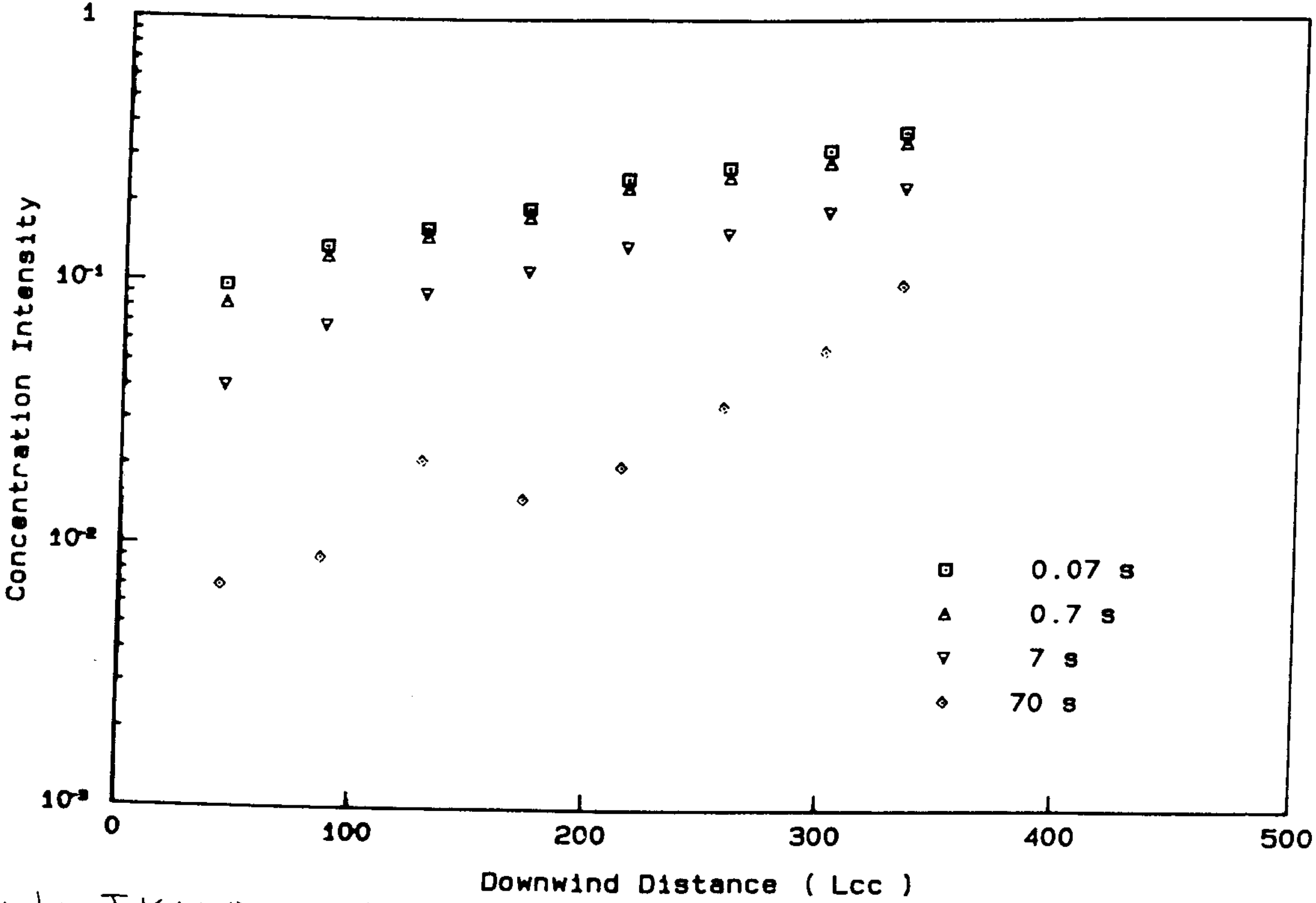


Fig. 3 Estimates of C and $(\overline{c\ })^{\frac{1}{2}}$ for the mast at (400m,250m) in trial 019. Data taken at 0.4m height.
3(a): ——— data; ----- $\hat{C}$
3(b): ——— data; ----- $(\overline{c^2})^{\frac{1}{2}}$
Recall that the dashed curves in these two figures are estimates for the 5 trials that were combined, and therefore do not vary from trial to trial for a given sensor location. Of course the solid curves do vary from trial to trial since they are the observed values of concentration.

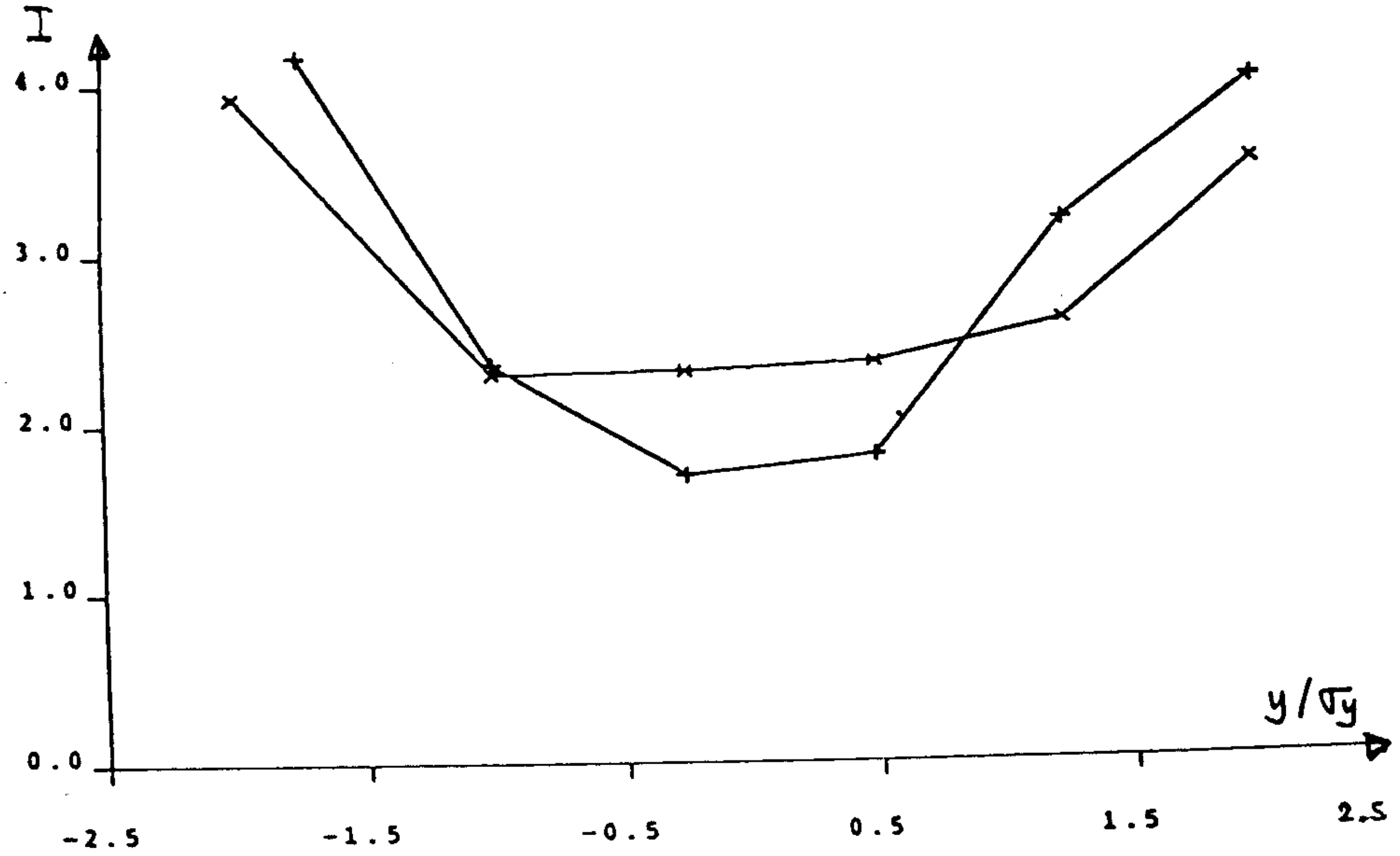
From: Carn, K.K., Sherrell, S.J. and Chatwin, P.C. 1988 Analysis of Thorney Island data: variability and box models. In Stably Stratified Flow and Dense Gas Dispersion (OUP, edited by J.S. Puttock), 205-231.

Figure 5



Analysis by J.K.W. Davies (HSE, Sheffield) of data taken by K. Marotzke and M. Schatzmann (Univ. of Hamburg). Concentration intensity = $\sqrt{\{\overline{c^2}\}}/C$.

Figure 4



From: Mylne, K.R. 1989 See Figure 1(f).

The two sets of points (+ and ×) are from different plumes.

NOTE

In both Figure 4 and Figure 5, y is measured horizontally in a direction perpendicular to the plume centre-line, and σ_y (which depends on downwind distance x) is the mean plume width (in the y direction).

Figure 5

From: Sawford, B. L., Frost, C.C. and Allan, T.C. 1985
Atmospheric boundary-layer measurements of concentration statistics from isolated and multiple sources. Bound.-Lay. Meteor. 31, 249-268.

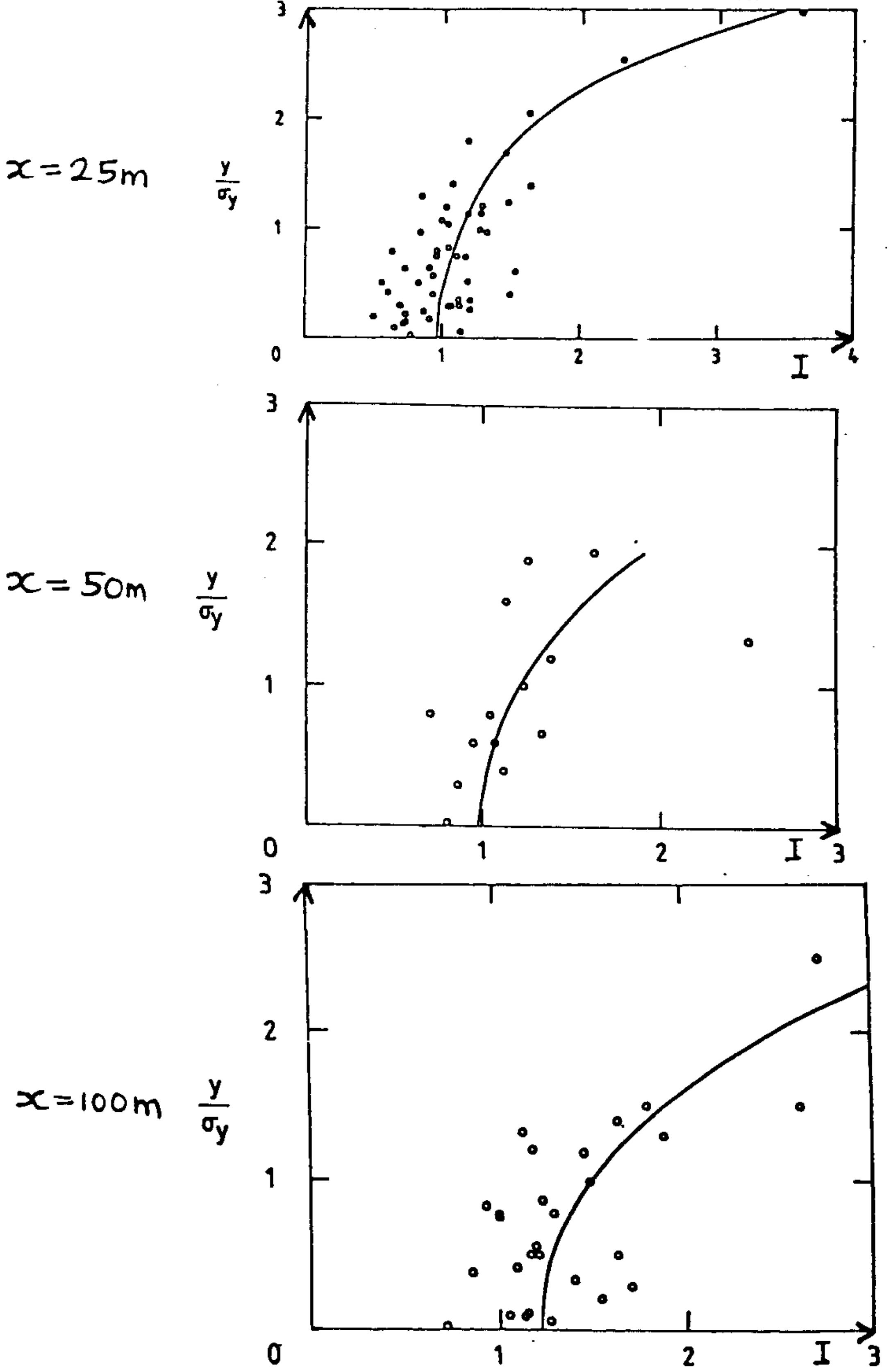
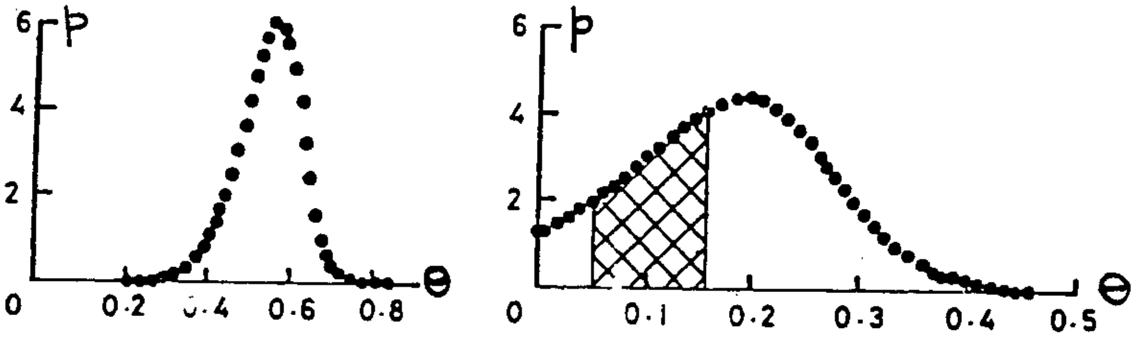
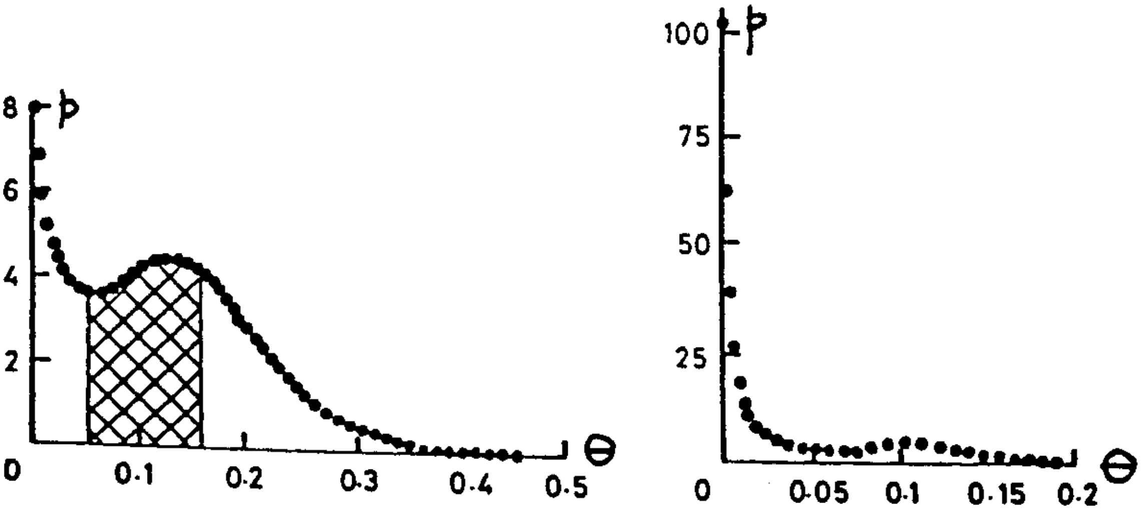


Figure 6



r/d = 0

r/d = 1·3



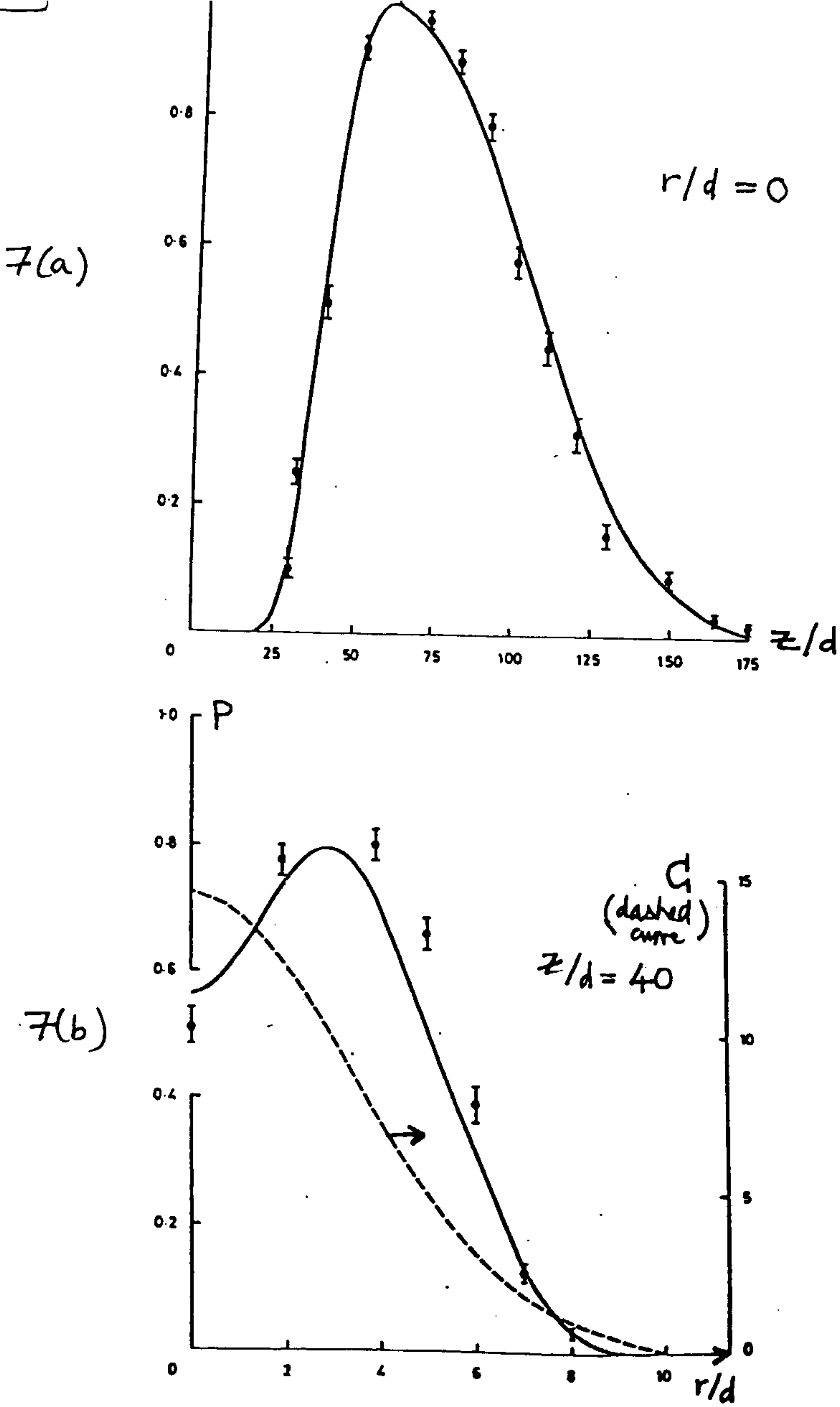
r/d = 1·49

r/d = 1·8

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.

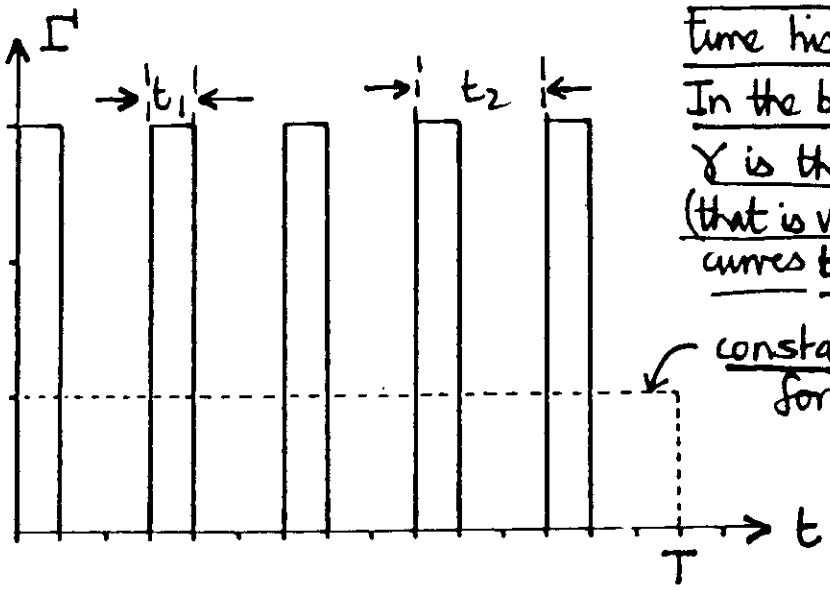
NOTE

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.

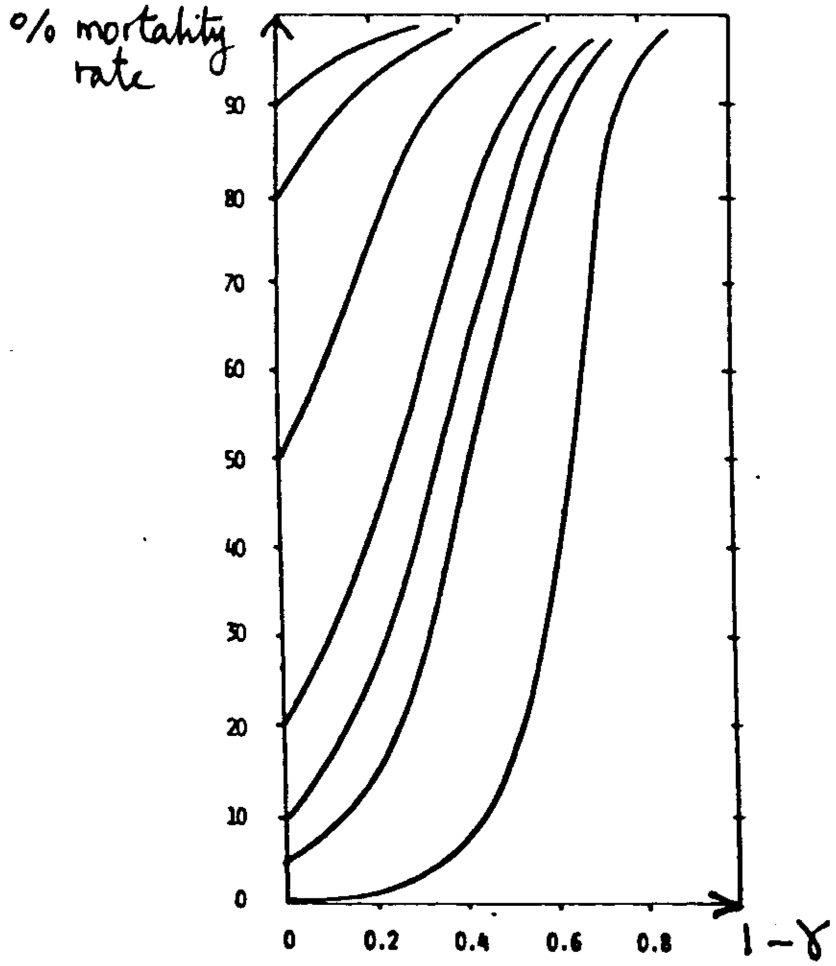


Sources as in Figure 6. z denotes downwind distance. Other notation is given in the text.

The two different concentration-time histories considered. In the bottom diagram, γ is the value of t_1/t_2 (that is varied to obtain the curves there).



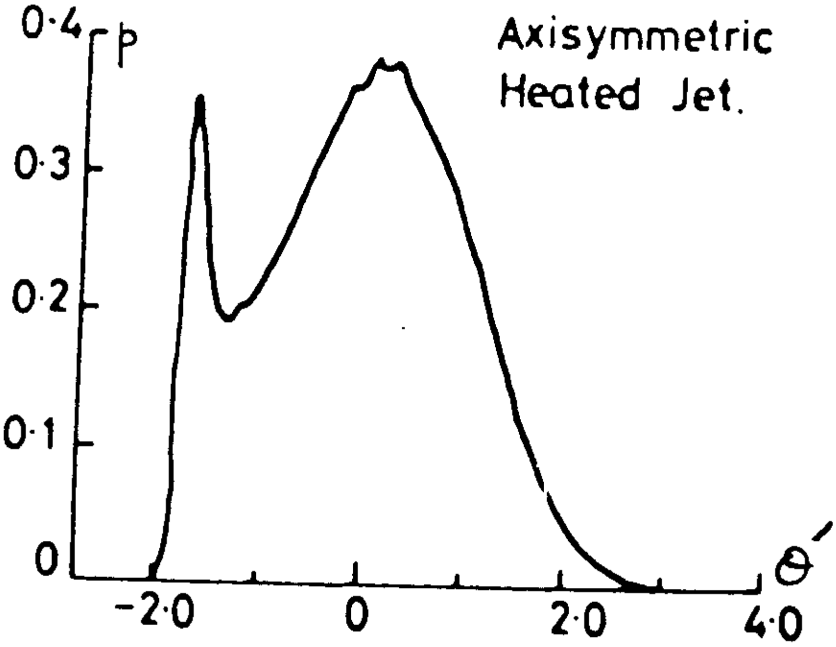
constant concentration for time T



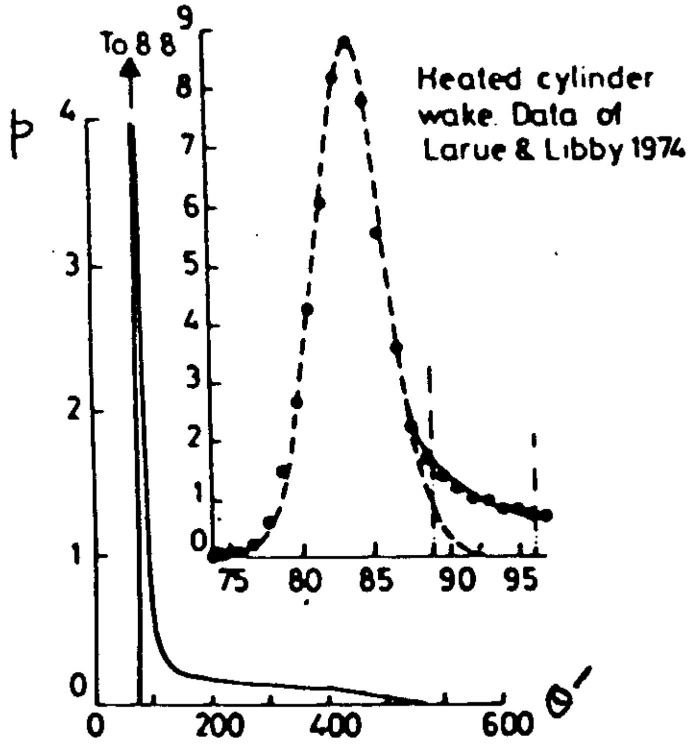
Definition of intermittency γ is given above and in the text.

From: 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. Haz. Mat. 11, 369–372.

Figure 9. From Bilger, R.W., Antonia, R.A. and Sreenivasan, K.R. 1976 Determination of intermittency from the probability density function of a passive scalar. *Phys. Fluids* 19, 1471-1474.

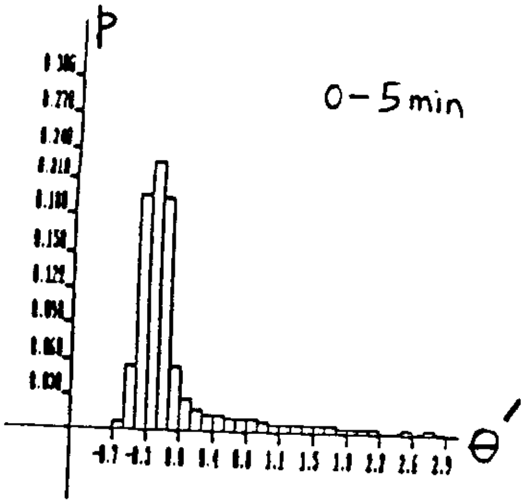


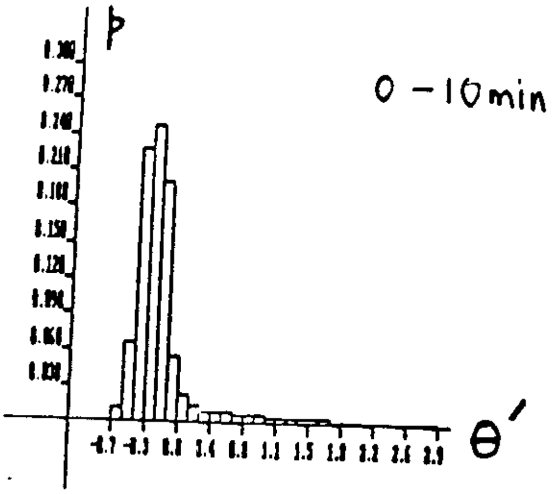
From: Bilger, Antonia & Sreenivasan 1976

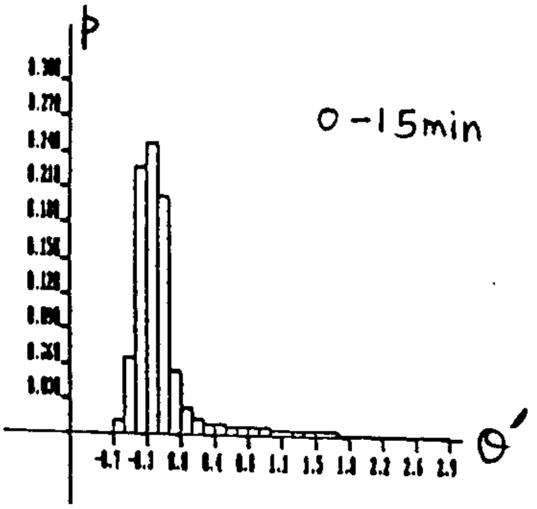


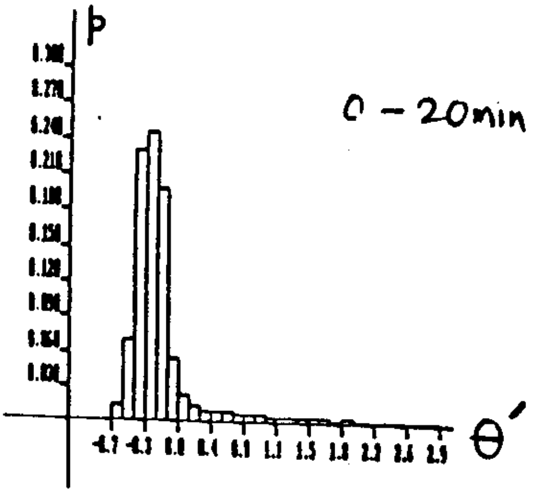
From Bilger, Antonia & Sreenivasan 1976.

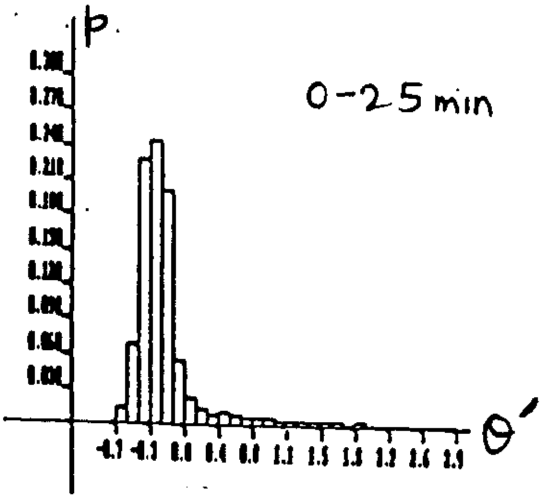
Figure 10.











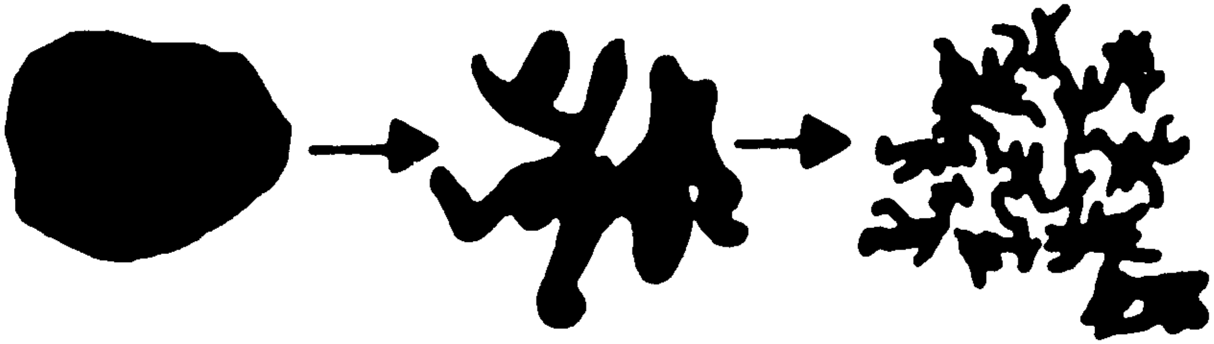
Analysis by N.T. Hajian (Brunel Univ.) of data taken in the field at RAF Cardington by R.F. Griffiths (UMIST) and C.D. Jones (CDE Porton).

In Figures 9 and 10, θ' is $\theta + \theta_0$, where θ is defined in the text and θ_0 is a constant.

Figure 11

$$p(\theta;\underset{\sim}{x},t) = \gamma_0\,\delta(\theta-\theta_1) + [1-\gamma_0]\,\delta(\theta)$$

NO MOLECULAR DIFFUSION



Adapted from: Corrsin 1959

(*J. Geophys. Research* **64**, 2134 - 2150)