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CONCENTRATION FLUCTUATIONS

IN

ATMOSPHERIC DISPERSION

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by

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Final technical report on Agreement No.2066/62 (CDE)

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CONTENTS

Summary

This report summarizes work done at Brunel University under Agreement No.2066/62 from 15 July 1986 to 14 July 1989. The title of the project was Concentration Fluctuations in Atmospheric Dispersion. The report has three principal components. These are:

(i) theoretical work on the electrostatic effects associated with dispersing charged tracers.

(ii) extensive analysis of several datasets taken with the CDE sensor system, particularly one obtained at RAF Cardington on 10 May 1988;

(iii) interpretation of the results of the analysis.

The conclusions of the report include recommendations for further work to exploit the advantages that the system has over many others.

Chapter 1.

INTRODUCTION

1.1 Description of work

In the Agreement, dated 12 February 1986, the Description of Work given was:

"(i) Studies (largely theoretical) of:

(a) The behaviour of the probability density function (pdf) of concentration in relation to position downwind of a continuous point source.

(b) The effect of sampler dimensions etc. on the perceived pdfs.

(c) Extension of the analysis of electrostatic effects of charged tracers.

(d) Liaison with other studies of atmospheric dispersion being conducted by CDE.

(ii) To submit reports in accordance with the requirements of Clause 8."

This programme has been followed for the duration of the Agreement (15 July 1986 to 14 July 1989). The work was carried out by Dr. N.T. Hajian at Brunei University, under the day-to-day supervision of Professor P.C. Chatwin, and the CDE monitor was Dr. CD. Jones. Dr. D.J. Ride of CDE was also in close contact with progress. Frequent meetings between the persons named took place to review the research, to discuss the progress reports, and to set priorities.

1.2 Brief summary of scientific background

Atmospheric dispersion is (arguably) the most important practical application of the branch of science known as *turbulent diffusion* (or *dispersion*). *Turbulence* is the term used to denote the random (or "gusty" or "chaotic") type of fluid flow that is- overwhelmingly - the most commonly occurring in the atmosphere, as elsewhere. Despite its very complicated structure, turbulence is governed by the most basic laws of classical physics, namely the *principle of mass conservation*, and *Newton's laws of motion*. (Applied to fluid flow, these laws express themselves as the equation of continuity, and the Navier-Stokes equations, respectively.) It is clear that any foreign material present in a turbulent flow will spread in a way which depends significantly on the properties of this flow. The term turbulent diffusion is that given to (the study of) this spread.

The distribution of foreign material within the flow can be measured by its *Concentration* $\mathbf{\Gamma}(x,t)$, where x, and t, denote position, and time, respectively. If the volume of a small region of space surrounding the point x at time t is δV, then $\mathbf{\Gamma}(x,t)\delta V$ is the quantity of foreign material within this region. It is normally convenient to measure this quantity as a mass, in which case the units of $\mathbf{\Gamma}$ are those of density (kg m^{-3}). However there is no reason why other units should not be used if appropriate; for example $\mathbf{\Gamma}$ could be expressed as a volume ratio for the case of a tracer gas dispersing in the atmosphere. In fact a substantial part of this report deals with the analysis of data in which the dispersing material is *ions* and the units used for $\mathbf{\Gamma}$ are quite naturally, those of *electric charge density* (Cm^{-3} or - more practically - nCm^{-3}).

The variation of Γ(x,t) with x and t is determined by the two processes of *advection* (transport by the ambient fluid) and *molecular diffusion.* As already noted above, the velocity field of a fluid in turbulent motion is *random;* consequently so is Γ(x,t). It therefore follows that it is appropriate to apply the techniques of *statistics* to Γ(x,t). In particular there exists a *probability density function* p(θ;x,t) defined by the equation:

$$p(\theta; x, t) = \frac{d}{d\theta}\{\text{prob}[\Gamma(x, t) \leq \theta]\}. \tag{1.1}$$

More immediate understanding of p(θ;x,t) is, perhaps, provided by an alternative definition equivalent to (1.1). If δθ is small then

$$p(\theta; x, t)\delta\theta \approx \text{prob}[\theta \leq \Gamma(x, t) < \theta + \delta\theta]. \tag{1.2}$$

As will be seen later, it is the definition (1.2) that was directly applied to data during this project to provide the estimates of p(θ;x,t) and other statistical properties of Γ(x,t), whose consideration forms an important part of this final technical report.

For later reference it is convenient to summarise here some important properties of p(θ;x,t), and some associated key definitions. Since concentrations cannot be negative:

$$p(\theta; x, t) - 0 \text{ for all } \theta < 0 \tag{1.3}$$

Also, since p(θ ;x,t) is a probability density function (abbreviated to *pdf* from now on),

$$\int_0^\infty p(\theta; x, t)d\theta = 1 \tag{1.4}$$

Note that in any particular ensemble of experiments, there will always be a finite maximum concentration, say $\theta_M(x,t)$, with p(θ;x,t) zero for all θ > θ_M clearly (1.4) holds with θ_M (or - indeed - any value of θ > θ_M) replacing ∞, However the dependence of θ_M on x and t is unknown; therefore it is simplest to use (1.4) when listing general properties.

The (ensemble) mean *concentration* C(x,t) is defined by

$$C(x, t) = \int_0^\infty \theta p(\theta; x, t)d\theta \tag{1.5}$$

and higher moments $\overline{c^n}(x, t)$ by

$$\overline{c^n}(x, t) = \int_0^\infty (\theta - c)^n p(\theta; x, t)d\theta \tag{1.6}$$

for n = 2,3,... It is sometimes convenient to express Γ as the sum of its mean C and *fluctuation* c, i.e.

$$\Gamma(x,t) - c(x,t) + c\ (x,t)\ , \qquad (1.7)$$

with $\bar{c}(x,t) = 0,$ where the overbar denotes the operation in (1.6). The quantity $\overline{c^2}(x,t)$ is the *variance* of $\Gamma(x,t)$ or the (*mean square*) fluctuation intensity. (Standard statistical notation for C and $\overline{c^2}$ would be μ *and* σ^2 respectively.) The *skewness* $s(x,t)$ and the *kurtosis* $k(x,t)$ are defined by

$$s(x,t) - \overline{c^3}(x,t)/(\overline{c^2}(x,t))^{3/2},\ k(x,t) - \overline{c^4}(x,t)/\left\{\overline{c^2}(x,t)\right\}^2, \qquad (1.8)$$

and are among the simplest non-dimensional measures of the *shape* of the graph of p(θ;x, t) against θ. Thus S = 0 if this graph is symmetric about its mean, i.e. about θ = C; hence non-zero values of S indicate a lack of such symmetry. Similarly K = 3 if Γ is *Normally distributed*, i.e. if

$$p(\theta;x,t) - (2_{\varpi}\overline{c^2})^{-\frac{1}{2}} \exp\left\{-(\theta - c)^2/(2\overline{c^2})\right\}; \qquad (1.9)$$

thus values of K different from 3 indicate that Γ is not Normally distributed. (In fact (1.3) shows that the positive random variable Γ can never be *exactly* Normally distributed but there is no reason why this should not be a good *practical* approximation under certain circumstances.) Examination of the data records considered in this project shows that the time series of Γ at any particular position x is always *intermittent*, and usually highly so. In this report the definition used for the *intermittency factor* $\gamma(x,t)$ will be

$$\gamma(x,t) \quad = \text{prob}\left[\Gamma(x,t) = 0\right]; \qquad (1.10)$$

high values of $\gamma(x,t)$ therefore indicate a high probability ofencountering uncontaminated fluid - always atmospheric air in the experiments considered here.

It is necessary to record here that p(θ;x, t) satisfies a differential equation which can be obtained from the equation governing Γ (x, t) - see (2.1), and is believed to be a well-defined and non-random quantity. Unfortunately, like all statistical properties associated with turbulence and turbulent diffusion, the equation for p(θ;x,t) exhibits the closure problem which has always bedevilled theoretical research on turbulence and for which no solution is known or, perhaps, attainable. Hence this equation will not be given here. However further details on this problem, and on the whole of this section can be found in Chatwin (1989). Other references that may be interesting are Chatwin (1982), Cam and Chatwin (1985), Ride (1987,1988), Chatwin and Sullivan (1989a,b).

1.3 Structure of report

Most of the original work undertaken falls into (a) and (c) of the Description of Work given in section 1.1. It was in fact decided to carry out (c) first, and Chapt2 summarises the results and the conclusions of those theoretical studies. In terms of time and effort by far the most substantial work was the analysis of many data records obtained by Dr. C.D. Jones and his collaborators, notably Dr. R.F. Griffiths of UMIST and his colleagues. The methodology and results of these analyses are described in Chapter 3. Finally Chapter 4 presents the conclusions obtained, and discusses research that remains to be done.

Chapter 2.

ELECTROSTATIC EFFECTS

2.1 Background

Experimental studies of atmospheric dispersion have taken place in laboratories and in the field. Many different sensor systems have been deployed in both environments and it is now generally recognised that the characteristics of the sensor system (e.g. time response, sensor size, ...), and the mode of deployment, can both affect the data, often significantly. It is particularly difficult to obtain accurate and reproducible measurements of properties such as the intermittency which depend in an important way on the fine spatial structure present in a dispersing cloud or plume (typically on length scales of order $10^{-3}m)$ and the associated rapid changes (typically over times of order $10^{-2}-10^{-1}$s). For various reasons it is believed that an understanding of such phenomena is needed for the assessment of hazards associated with toxic agents released into the atmosphere.

Dr. CD. Jones of CDE has pioneered the development and use of a novel sensor system which has many advantages from this point of view, and which has the necessary robustness for reliable use near the ground in the atmospheric boundary layer (Jones 1979, 1983). Briefly the system consists of a constant source of unipolarly ionised air produced by a stabilised corona discharge, and a series of detectors located at appropriate positions downwind of the source. In the experiments considered in this project detectors were in some cases up to 100m from the source, but the bulk of the data was taken with them no more than 25m away. The detectors operate on a biased coaxial cylinder electrode principle in which the ionised air tracer is drawn into the detector by means of a low-power suction fan whereupon the ions experience a strong electric field and are repelled onto a collecting electrode. The collected current, typically $10^{-12}-10^{-8}$A, is sensed by a high-speed feedback amplifier that produces a voltage output proportional to that current, and thus to the local instantaneous ion concentration. The special feature of this system which, as noted above, makes it potentially so useful and attractive is the extremely fast response time $(\sim 10^{-2}s)$, which allows high resolution measurements up to 100Hz bandwidth.

However the interpretation of data taken with this system obviously requires consideration of the inevitable electrostatic effects that influence the dispersion in addition to the basic processes of advection and molecular diffusion noted in Chapter 1. During the first year of research under the Agreement, earlier theoretical work on this problem (Chatwin 1985) was continued and extended. Since a full account of all these theoretical studies has recently been published (Chatwin, Hajian, Mole and Jones 1989), it is appropriate in this final technical report to give an extended summary only.

Furthermore it is also necessary to emphasize that the dispersion of *passive* tracers in the atmosphere is not yet understood, especially in regard to quantitative predictions of $p(\theta;x,t)$ and associated properties involving the concentration fluctuations. Indeed gaining such understanding is the purpose of all the experiments using *charged* tracers! Realistically therefore, it is impossible at present to provide a conclusive account of the quantitative differences between the dispersion of passive and charged tracers; therefore the work referred to above (and summarized below) uses simple physics and aims at producing order of magnitude estimates for the most important phenomena.

2.2 Some global results

The velocity of charged particles differs from that of the ambient air, denoted here By u(x,t), by an amount $\mu E(x,t)$, where μ is the mobility and E is the electric field. Let ρ (x,t) be the charge density; the equation of charge conservation that is thought to be adequate (see Chatwin, Hajian, Mole and Jones 1989 and references in that paper, especially to earlier papers by Dr. C.D. Jones) is

$$\frac{\partial \rho}{\partial_t} + \nabla.\left[\rho(u + \mu E)\right] - K\nabla^2 \rho, \qquad (2.1)$$

where *k* is the molecular diffusivity. In addition the velocity field is incompressible so that

$$\nabla.u - 0, \qquad (2.2)$$

and the electric field can be regarded as conservative so that there is a potential $\varphi(x,t)$ with

$$E = -\nabla\varphi, \nabla^2\varphi = -\frac{\rho}{\in_o}, \qquad (2.3)$$

Where $\in_o$ is the *permittivity* whose value can be taken to be the vacuum permittivity with value $\in_o \approx 8.85 \times 10^{-12} cv^{-1} m^{-1}$.

The equation governing the concentration $\Gamma(x,t)$ of an uncharged tracer is (2.1) with $\mu = 0$ and Γ replacing ρ.

In simple physical terms, the main differences between the dispersion of charged tracers and the dispersion of uncharged passive tracers are:

(1) The cloud or plume of charged tracer will be larger than the corresponding cloud or plume of passive tracer because of electrostatic repulsion. (Here the word "corresponding" implies the same source geometry and release conditions, and the same atmospheric conditions.)

(2) The possibility of charge leaking to earth which implies, for assessing the comparable dispersion of passive tracers, that the total mass of material is not conserved. (Although, even for a passive tracer, conditions will often be such - e.g. absorption by humans, animals or vegetation - that material is lost, it is prudent from the point of view of assessing danger when the passive tracer is toxic to assume that total mass is conserved.)

The sensor system can be operated in several modes. Usually unipolar ions are injected at a constant rate, with a discharge current i_o of the order of $10^{-8} - 10^{-9}\,A$. This leads to dispersing *plumes,* which can be regarded as statistically steady once the effects of starting up have become small. In another mode a fixed quantity of charge is injected almost instantaneously; this leads to dispersing clouds in which, by contrast, statistical conditions change with time. The second mode is easier to analyse; moreover it can (in certain circumstances) act as a canonical case from which the behaviour in other modes can be inferred. In particular, results for steady plumes can be deduced from those for clouds.

In Chatwin (1985) an exact solution for dispersing clouds was obtained, generalizing to arbitrary shape an earlier result for spherical clouds by Jones (1977). The solution is realistic for those global properties of clouds that do not depend on molecular diffusion to any significant extent. For the large *Péclet numbers* $(\sim 10^6)$ occurring in practice, such properties include the cloud size and the orders of magnitude of the mean, and the variance, of the charge density. In this final technical report, to avoid unnecessarily profuse notation, the symbols C and $\overline{c^2}$ will be used for these latter quantities, i.e. C(x,t) will be the ensemble mean of $\rho(x,t)$ and $\overline{c^2}\,(x,t)$ will be the ensemble mean of $\{\rho(x,t) - c(x,t)\}^2$ (This use is entirely consistent with the definitions (1.5) and (1.6); in Chatwin (1985) and Chatwin, Hajian, Mole and Jones (1989) the symbols used for C and $\overline{c^2}$ were ρ and ρ'^2 respectively.)

Suppose a cloud of initial volume V_o and initial charge density ρ_o is instantaneously released. The solution of (2.1) referred to showed that, at time t later, the volume V(t) of the cloud and the charge density p(t) within it satisfied

$$V(t) - V_o\left[1 + \frac{t}{T_e}\right], \quad \rho(t) - \rho_o\left[1 + \frac{t}{T_e}\right]^{-1}, \tag{2.4}$$

Where T_e is a constant time scale defined by

$$T_e = \frac{\in_o}{o}. \tag{2.5}$$

(Without electrostatic repulsion V(t) and ρ(t) would be equal to V_o and ρ_0 for all t.) The time scale T_e is characteristic of that in which electrostatic effects become important and its values in actual use of the sensor system have ranged from about $5 \times 10^{-4}\,S$ to about 20s; further numerical estimates are given by Chatwin (1985, Table 1on p.18). Provided the total duration of the experiments is much less than T_e, it is clear from (2.4) that the data taken with the sensor system will apply directly to clouds of passive tracer. Unfortunately other practical considerations, including the need to take measurements at relatively substantial distances downwind of the source, mean that this condition has not normally been a realistic one to require. Nevertheless it is clear that T_e should be as *large* as is practically possible. A first recommendation of this work is therefore:

RECOMMENDATION 1 : In designing experiments using the CDE *sensor system the value of* T_e *should be estimated in advance, and made as large (by choosing* ρ_0 *small) as is possible given the other practical requirements.*

From (2.4), a mean cloud radius of the cloud of charged tracer at time t is of order $3V(t)/4\pi)1/3$. To minimise interaction with the earth this should be much less than the height H_0 of the centre of the cloud above the earth. Provided the initial cloud radius R_0 is also much less than H_0, as has always occurred in practice, this condition is equivalent, using (2.4), to $(3V_0 t/4\pi T_e)^{1/3} << H_0$. If measurements are taken at a distance d downwind of the source and the wind speed is u, this requirement is equivalent to:

$$\frac{\epsilon_o u}{\rho_o}\left[\frac{H_o}{R_o}\right]^3 >> 1 . \qquad (2.6)$$

Alternative ways of writing this condition are given in Chatwin (1985) and Chatwin, Hajian, Mole and Jones (1989). It was shown in the first of these references that satisfaction of (2.6) does, in practice, ensure that image charge effects are also negligible.

RECOMMENDATION 2: When the sensor system is used with a cloud of charged tracer, the initial radius should be much less than the source height, and (2.6) should be satisfied.

Application of the above ideas to steady plumes of charged tracer is given in the references. In preparing the present report a new solution has been found for plumes; according to this the plume spreads somewhat less rapidly than is given by the first equation in (5.1) of Chatwin, Hajian, Mole and Jones (1989). As a consequence (2.6) is replaced for plumes by

$$\frac{\epsilon_0 u}{d\rho_0}\left[\frac{H_0}{R_0}\right]^4 >> 1. \qquad (2.7)$$

where R_0 is now the cross-sectional radius of the plume immediately after leaving the source. Then *Recommendation 2* becomes:

RECOMMENDATION 3: When the sensor system is used with a plume of charged racer, the initial cross-sectional radius of the plume should be much less than the source height, and (2.7) should be satisfied.

It is important to note that (2.6) and (2.7) have always been satisfied for the datasets collected by the CDE sensor system that have been examined at Brunel.

It is well-known (e.g. Chatwin and Sullivan 1979) that, although its total volume V_0 stays constant, the *shape* of a cloud of passive tracer dispersing in the atmosphere rapidly becomes very complicated and unpredictable; this is due to random advection. Evidently therefore, even though its volume increases according to (2.4), the same statement applies to a dispersing cloud of charged tracer. Detailed mathematical analysis by Chatwin (1985) showed that for very small clouds the only important difference between the shape of a cloud of passive tracer and that of a corresponding cloud of charged tracer was that electrostatic repulsion slowed down the rate at which the *thinnest* dimensions decreased with time. This was entirely responsible for the growth of the total volume V(t) since the largest dimensions of the small cloud were not affected by repulsion in any significant way. Real clouds are not very small. However they can, for present purposes, be regarded as composed of many very small clouds to each of which the above conclusions apply.

It was therefore argued in Chatwin (1985) and Chatwin, Hajian, Mole and Jones (1989) that electrostatic repulsion would not change the overall size and shape of the *mean* cloud since these depend entirely (Chatwin and Sullivan 1979) on the behaviour of the largest dimensions in each realisation of the dispersion. This is an important conclusion.

The different behaviour of the thinnest dimensions mainly affects the *dissipation* of $\overline{c^2}$, an important and complicated phenomenon in which molecular diffusion has a crucial role. The further discussion of dissipation in Chatwin (1985), which was extended during the work on the Agreement now being reported and discussed in full in Chatwin, Hajian, Mole and Jones (1989), is not summarised here since it does not affect the data analysis and subsequent interpretation.

Clearly the arguments and conclusions above for clouds apply equally to plumes.

2.3 The orders of magnitude of C and $\overline{c^2}$

Provided (2.6) is satisfied the total charge Q_0 in the cloud is conserved throughout each realisation of the dispersion, where $Q_0 = P_0 V_0$. Therefore, for all time t, and in each realisation of the dispersion,

$$\int \rho(x, t > dv = Q_0 \qquad (2.8)$$

irrespective of whether the charge density ρ is uniform within the cloud, or not. (The assumption of uniformity is a good approximation for all the experiments and is, in any case, justified mathematically when deriving (2.6), which deals with orders of magnitude.) In each realisation the volume V(t) of the cloud is given by (2.4), at least to an order of magnitude. However the cloud is positioned, and oriented, *randomly* in space. Consequently the volume of space occupied by the (ensemble) mean cloud is greater, generally much greater, than V(t); let its order of magnitude be $L^3(t)$. According to the discussion in section 2.2, $L^3(t)$ is unaffected by electrostatic repulsion and its dependence on t and atmospheric conditions is a classical problem summarised, for example, in Pasquill and Smith (1983). On taking the ensemble mean of (2.8) it then follows that the order of magnitude of the mean charge density C is given by

$$C \sim \frac{Q_0}{L^3(t)}, \tag{2.9}$$

and this estimate is independent of the fact that the dispersing material is charged tracer, i.e. it is independent of what Q_0 is the total quantity of.

By contrast, electrostatic repulsion does affect the order of magnitude of $\overline{c^2}$; so also, because of dissipation, does molecular diffusion. For the moment however suppose there is no molecular diffusion, i.e. k = 0 in (2.1). Then, for a spatially uniform distribution of charge, it follows from (2.4) that

$$\int \rho^2(x,t)dv = \rho_0 Q_0 \left[1+\frac{t}{T_e}\right]^{-1} \tag{2.10}$$

and this will be correct to much better than an order of magnitude even when ρ is not uniform. Now, from (1.6) with n = 2, the ensemble mean of ρ^2 is $C^2 + \overline{c^2}$, so that the ensemble mean of (2.10) is

$$\int c^2 dv + \int \overline{c^2} dv = \rho_0 Q_0 \left[1+\frac{t}{T_e}\right]^{-1} \tag{2.11}$$

Since, by (2.9), the order of magnitude of C^2 is $Q_0{}^2/L^6(t)$, it follows that (2.11) can be satisfied only if the integral involving $\overline{c^2}$ dominates the left-hand side of (2.11) - since $L^3(t) >> V_0(1 + t/Te)$. Therefore the order of magnitude of $\overline{c^2}$ must be given by (Chatwin 1985; Chatwin, Hajian, Mole and Jones 1989):

$$\overline{c^2} \sim \frac{\rho_0 Q_0}{L^3(t)\left[1+\frac{t}{T_e}\right]} = \frac{Q_0{}^2}{L^3 V}, \tag{2.12}$$

where V = V(t) is defined in (2.4). It follows by combining (2.9) and (2.12) that, in the absence of molecular diffusion,

$$\frac{\overline{c^2}}{C^2} \sim \frac{L^3}{v} >> 1. \tag{2.13}$$

The corresponding estimate for clouds of passive tracer (Chatwin and Sullivan 1979) is L^3/V_0 ; hence in clouds of any tracer (passive or charged) the mean square fluctuation is much greater than the square of the mean until molecular diffusion becomes significant but in the charged case $\overline{c^2}$ is less than its value in the corresponding uncharged case by a factor of order $V_0/V = (1+t/T_e)^{-1}$.

Corresponding estimates to those above for a steady plume were given by Chatwin

(1985) and presented in section 5 (pp.107-108) of Chatwin, Hjian, Mole and Jones (1989). As noted above, immediately prior to (2.7), a new solution has been found for steady plumes that somewhat changes those estimates. The new estimates at a distance x downwind for a plume of initial cross-sectional area A_0 are

$$C \sim \frac{\rho_0 A_0}{L^2}; \overline{c^2} \sim \frac{\rho_0{}^2 A_0}{L^2}\left[1+\frac{2x}{uTe}\right]^{\frac{1}{2}}; \ \frac{\overline{c^2}}{C^2} \sim \frac{L^2}{A_0}\left[1+\frac{2x}{uTe}\right]^{-\frac{1}{2}}, \tag{2.14}$$

where L = L(x) is now a mean plume radius. According]'to the new solution the area A = A(x) of the instantaneous plume is equal to A0(l + 2x/uTe)$^{\frac{1}{2}}$ so the results for $\overline{c^2}$ and $\overline{c^2}/C^2$ in (2.14) can be rewritten

$$\overline{c^2} \sim \frac{i_0^2}{u^2 L^2 A}, \frac{\overline{c^2}}{C^2} \sim \frac{L^2}{A}, \tag{2.15}$$

where $i_0 = u\rho_0 A_0$ is the discharge current. In this form the estimates are directly comparable with those in (2.12) and (2.13); since $L^2/A >> 1$, it remains true that $\overline{c^2} >> C^2$.

These estimates do not include the effects of molecular diffusion; as noted in section 2.2 the primary role of molecular diffusion is to dissipate $\overline{c^2}$. Eventually both C and $\overline{c^2}$ tend to zero everywhere, and C does so in accordance with the estimates already given since the mean concentration is never significantly affected by molecular diffusion. Unfortunately it is possible to include the dissipation of $\overline{c^2}$ in quantitative models only in the case of statistically steady and quasi-homogeneous situations involving passive tracers, and then only tentatively via empirical approximations. For clouds and plumes of charged tracer therefore no progress is possible at present, except as pure speculation. However it is reasonable to make three statements about $\overline{c^2}$ in real clouds and plumes of charged tracer:

(a) the above estimates of $\overline{c^2}$ will be useful for *large* clouds and plumes (including essentially all those occurring in experiments) in a period immediately after release;

(b) molecular diffusion will eventually invalidate these estimates as it does for passive tracers (Chatwin and Sullivan 1979);

(c) all experimental data give values of $\overline{c^2}/C^2$ that are everywhere at least of order unity, and much greater than unity at the edges of the cloud or plume.

Some data taken by Jones (1983) that illustrate (c) were given in Table 1 of Chatwin, Hajian, Mole and Jones (1989). The theoretical estimates there need changing because of the new solution, and the amended results are given below.

x (m)	L (m)	$(L^2/A)^{\frac{1}{2}}$	$(\overline{c^2}/C^2)^{\frac{1}{2}}$ Measured	$(\overline{c^2})^{\frac{1}{2}}$ Estimate (2.15)	$(10^{-9}\,cm^{-3})$ Measured
2	0.39	7.9	3.0	33	13
5	1.10	17.9	4.2	9.4	2.2
10	1.63	22.3	3.4	05.3	0.8
15	2.00	24.8	2.1	3.9	0.35
		(1)	(2)	(3)	(4)

Table 1: Comparison between estimates in (2.15) and data from Jones (1983).

It will be noted that (2.15) overestimates both $\overline{c^2}/C^2$ and $\overline{c^2}$; this can be attributed to molecular diffusion but also to smoothing effects caused by the finite size of the ion collector (diameter $\approx$ 0.036m).

Chapter 3

DATA ANALYSIS AND RESULTS

3.1 Preliminary remarks

All data examined were taken in the field at RAF Cardington using the CDE sensor system described in section 2.1 of Chapter 2 of this report. During the work covered by this Agreement, essentially all the analysis was of three datasets which divided naturally into two groups. The first group consists of the results from two experiments conducted by the UMIST team, led by Dr. R.F. Griffiths, in July 1986. The second group is data from experiments on 10 May 1988. These were designed after the results from the first group had been analysed at Brunei by Dr. N.T. Hajian (with invaluable assistance from Mr. M. Wellham, a Brunei undergraduate), and, naturally, took account of what had been Learned from that analysis. Given, also, that the May 1988 experiments, unlike those in July 1986, were specifically designed to meet the aims of the Brunei project, this third dataset proved more valuable and will receive most emphasis in this report.

The results of all the analyses have been included in progress reports submitted to CDE in accordance with the terms of the Agreement. Therefore the stress here will be on giving a consolidated overview.

3.2 The July 1986 experiments

One set of experiments took place on 9 July 1986 and the second set on 15 July 1986. In all cases, the ion generator and collectors were 4.65m above level unobstructed terrain, and the 5 collectors were located, in each run, at the same downwind distance. They were however separated in the crosswind direction at intervals of 1.6m so the overall lateral coverage was 6.4m.

In the three runs on 9 July 1986, each of 1-2 hours, the collectors were placed successively at downwind distances of 32m, 75m and 100m from the generator.

In the one run on 15 July 1986 the 5 ion collectors were at 50m downwind. On this occasion data were also taken with a T.I.P. (Total Ionisables Present) transducer system operated by a team from the Meteorological Office led by Mr. K.R. Mylne. The aim of this experiment was to compare readings, taken under *identical* atmospheric conditions, from two very different systems; to this end the T.I.P. instruments were placed as close as possible to the CDE instruments. In practice the T.I.P. source (a propylene jet) was 0.50m crosswind from the ion generator and at the same height, and each T.I.P. collector was 0.35m vertically above an ion collector. In view of the known dependence of concentration fluctuation data on the instruments used to obtain them, the aim of the 15 July 1986 experiment was very worthwhile. It fits in with (b) in the Description of Work (see section 1.1 above), and also with the objectives of another Brunei project (almost simultaneous with the CDE funded project) involving Dr. N. Mole and Professor P.C. Chatwin and funded by SERC. Unfortunately the aim has not yet been achieved for this particular experiment. This is for two reasons. The first is the quality of the data obtained with the CDE system (see below), and the second is that the Meteorological Office data was not made available to Brunei in an appropriate format.

The July 1986 data from the CDE sensor system were recorded in analogue form, digitised at UMIST, and recorded at a sampling rate of 1kHz on $\frac{1}{2}$" magnetic tape. The Data Acquisition System (DAS) at UMIST has a different machine code from that routinely available at Brunei and it was eventually necessary to write new software to read the data. This was done with the help of Dr. Mole and Mr. Wellham but took several months. Before detailed statistical analysis occurred, the data had to be, and were, inspected visually. The production of appropriate graphical print-outs was also done at UMIST since a multi-channel digital chart recorder of the appropriate type was not available at Brunel.

Once the graphical print-outs had been obtained they were inspected visually and discussed with Dr. Jones before more detailed statistical analysis took place. It was agreed that, unfortunately, data on each of the two dates from 2 of the 5 ion collectors were of too poor a quality for such analysis to be worthwhile. The reasons for rejection are the same as those discussed in more detail later (see section 3.3) in the same connection for the 10 May 1988 data; in brief the visual print-outs from the faulty channels were different in type from those that long experience has shown were provided by correctly functioning collectors. Even in the remaining data, there were occasional suspicious regions such as very short lasting (~ 4-5ms) and abnormally high peaks. There was also some base line instability, assumed to be natural, whose root mean square magnitude (of the order of l00mV) was about 2-3 bin widths of the bins used in digitisation (8V divided into 204 bins with 1 bin corresponding to a ρ of 0.1 nCm^{-3}), and was compared with expected peak values of ρ of the order of 30-35 bin widths.

The analysis of the remaining data from 12 channels (3 from each of 32m, 75m, 100m downwind on 9 July 1986, and 3 from 50m downwind on 15 July 1986) produced full histograms - see section 3.5 for explanation of this term - for up to 60 minutes in each case. In addition the means, variances and intermittencies were estimated. The results were fully discussed by Dr. Hajian at a symposium entitled *Concentration Fluctuations in the Atmosphere* held at CDE Porton on 5 February 1988 and were also formally presented to CDE. The results showed that the peak signal for most channels was only about 10-12 bin widths, about one-third of that expected. They also strongly suggested that a period of 30-40 min was adequate in statistically stationary atmospheric conditions for stable estimates to be obtained. Here *stable* implies that the unavoidable effects of *statistical noise* due to finite record length are acceptably low. However the results were not of direct benefit to the Brunel project for the following reasons:

(a) The signal to instrument noise ratio of the data was far too small.

(b) The amplitude resolution was too coarse because too few digits and, consequently, too few bins were used in the digitisation process.

(c) As noted above the base line drift was a substantial fraction, of the order of 20%, of the peak signal and in view of (a) and (b) there was a strong risk of losing real data if this was simply removed by automatically setting to zero.

Despite these drawbacks, which became apparent in the early summer of 1987 before the bulk of the analysis had been performed, the lengthy exercise involving the July 1986 data proved worthwhile since it provided (i) experience, and (ii) precise guidance. Both of these enable future experiments to be designed much more helpfully; in particular they allowed Dr. Hajian to suggest precise experiments to meet aim (a), i.e. investigation of the evolution of the pdf of concentration with downwind distance.

A meeting involving Professor Chatwin, Dr. Griffiths, Dr. Jones and Dr. Ride to discuss the analysis of data from ion collector experiments took place at Brunei on 7 October 1987, i.e. after the problems summarised above had been identified but before the detailed analysis. Notes of the meeting were made by Dr. Ride on 8 October 1987 (paper headed LM to IE1244 and IE1275). The principal points agreed, and relevant to the Brunei project, can be stated as follows.

(1) Experiments should be designed with an analysis strategy in mind.

(2) It was more important than had, perhaps, been realised before to make realistic and - as far as possible - complete estimates of the resources required for analysis (man-power, hardware, software) prior to conducting experiments.

(3) Given that (consistent with the Brunel experience with the July 1986 data) the bulk of these resources were needed for data access, and not subsequent processing and calculation, it was extremely important to make data available from future experiments on a medium and in a format which could be easily accessed by the most common systems now used. For example, block lengths should be of a standard size and use of the ASCII code was suggested. It was agreed that wide accessibility was far more important than compactness.

(4) While it is important to *validate* data, experience (*not* mainly from experiments using the CDE sensor system) has shown that many experimenters make unjustified assumptions or subjective choices when preparing "validated" data from raw data. A common example is the removal of raw data by the application of a *threshold*, and then the precise choice of the threshold value. This results in good data being lost, and being lost arbitrarily.

(5) The following procedure was agreed to be desirable for ensuring the most useful and efficient data analysis:

- The people who will conduct the analysis should examine the field notes and talk to the experimenter. (If possible and convenient they might attend the actual experiments.)
- Inspect the raw data visually, looking for features such as drift, and unusual noise (the latter could signal equipment failure).
- Select the portion of the data for analysis.
- Pre-treat the data *if necessary* (e.g. thresholding, averaging, "removing" noise and drift).
- Compute the pdf, and other statistics.

3.3 The May 1988 experiments. I Experimental details

At a meeting on 17 March 1988 involving Dr. Jones, Professor Chatwin and Dr. Hajian, Dr. Hajian was invited to make proposals for new experiments to be carried out at Cardington. Dr. Griffiths had arranged to carry out field trials there in the second week of May 1988 with Dr. Jones. He had 4 ion collectors available and it was agreed to make 1 day available for experiments relevant to the Brunei project.

The proposals were delivered to Dr. Jones on 30 March 1988. In these, stress was put on meeting condition (2.7) - see *Recommendation 3* in Chapter 2 - and on performing preliminary spot checks at the beginning of each run to ensure acceptably high signal to noise ratios. The proposals contrasted the low signal to noise ratio (no greater than 10) achieved during the 15 July 1986 runs using the CDE sensor system with that achieved by the T.I.P. transducer system (between 200 and 2000).

In the event 6 runs, essentially fulfilling these proposals, took place on the afternoon of 10 May. Dr. Hajian was present as well as Dr. Griffiths and Dr. Jones. The weather throughout was dry with steady winds from the NW; the measured wind speed was $3.5 ms^{-1}$. The ion generator and collectors were at a height of 1.9m. Table 2 gives further details. Visual monitoring of the data outputs from the four ion collectors took place two by two in a random manner since there was only one 2 channel oscilloscope available.

The data were recorded on $\frac{1}{2}$" magnetic tape using analogue techniques; each record was a continuous time series of voltages, i.e. frequency modulated signals. Subsequently the data were digitised at Brunel using an ISC-16 system consisting of an A-D converter and an IBM PC. Obviously this process required a prior choice of the sampling rate; given the extremely fast response to changes of the CDE sensor system (already noted in section 2.1) and the importance and desirability of exploiting this, it was decided to sample at 1kHz. A run lasting 40 minutes therefore yielded 2.4×10^6 readings of which only a very small proportion could not be validly analysed because they were taken right at the beginning of the run during the plume start-up period. The digitised data were stored on floppy discs, each of which contained 5 minutes of data for 2 collectors. It is noted here that *no* pre-processing of the data (e.g. to remove noise) took place before digitisation.

RUN NO.	ION OUTPUT CURRENT (nA)	DURATION (mins)	ION COLLECTORS NO. 1 d(m)	AG	NO. 2 d(m)	AG	NO. 3 d(m)	AG	NO. 4 d(m)	AG
1	10	45	3*	33	6	33	9	100	12*	100
2	3	40	3*	33	6	33	9	100	12*	100
3	10	35	9*	100	12	100	15	100	18*	100
4	10	38	15*	100	18	100	21	333	24*	333
5	3	40	15*	100	18	100	21	333	24*	333
7	10	37	21*	333	24	333	27	333	30	333

Table 2: Details of the 10 May 1988 experiments at Cardington. d is downwind distance from the ion generator and AG is amplifier gain. Asterisks denote the datasets analysed.(Run No.6 was a very short run of only 5 minutes designed to assess the collector signal at a collector close to the generator; its results were not analysed.)

However - see point (5) in section 3.2 - the analogue data had been inspected at Brunel as soon as possible after the experiments (and before digitisation). Unfortunately it quickly became clear, in consultation with Dr. Jones, that the data from collectors 2 and 3 (i.e. channels 2 and 3) were unfit for analysis. Figures 1-6 show the time series generated by the remaining collectors (channels 1 and 4). The time series for channel 4 in run 7 (see Figure 6) is clearly different in quality from all the others. In particular there is substantial drift, and the signal does not display the substantial intermittency (i.e. for much of the time the signal is of pure noise without any contribution from the plume of ions) so evident elsewhere. Such features were observed throughout the visual inspection of channels 2 and 3 (though not during the visual monitoring that occurred at the time of the experiments), and were the cause of the rejection of the data from these collectors. Moreover the data from channel 4 in run 7 were also rejected. There remained apparently valid data from 11 channels, and these were analysed in detail. Even in these channels there were occasional suspicious readings such as extraneous troughs (or peaks) lasting for a very short period. One example occurs about 7 minutes into the record from channel 1 in run 1 (see Figure 1).

3.4 The May 1988 experiments. II Analysis methodology

The raw data were recorded as voltages V, and these were converted into concentrations, i.e. charge densities ρ, by means of the calibration equation:

The following Figures 1-6 show the time series obtained from, channels 1 and 4 in each run. For logistical and display reasons, each time series is broken down into 6 graphs each showing the record from both channels for a five minute period. The graphs are consecutive and one labelled, for example, 15 min. gives the record for the five minute period terminating 15 minutes after the start of the run. Thus, for each run, a total of 30 minutes is shown for each of the two channels. Experimental details are summarized in Table 2, and reference should be made to the text for further explanation.

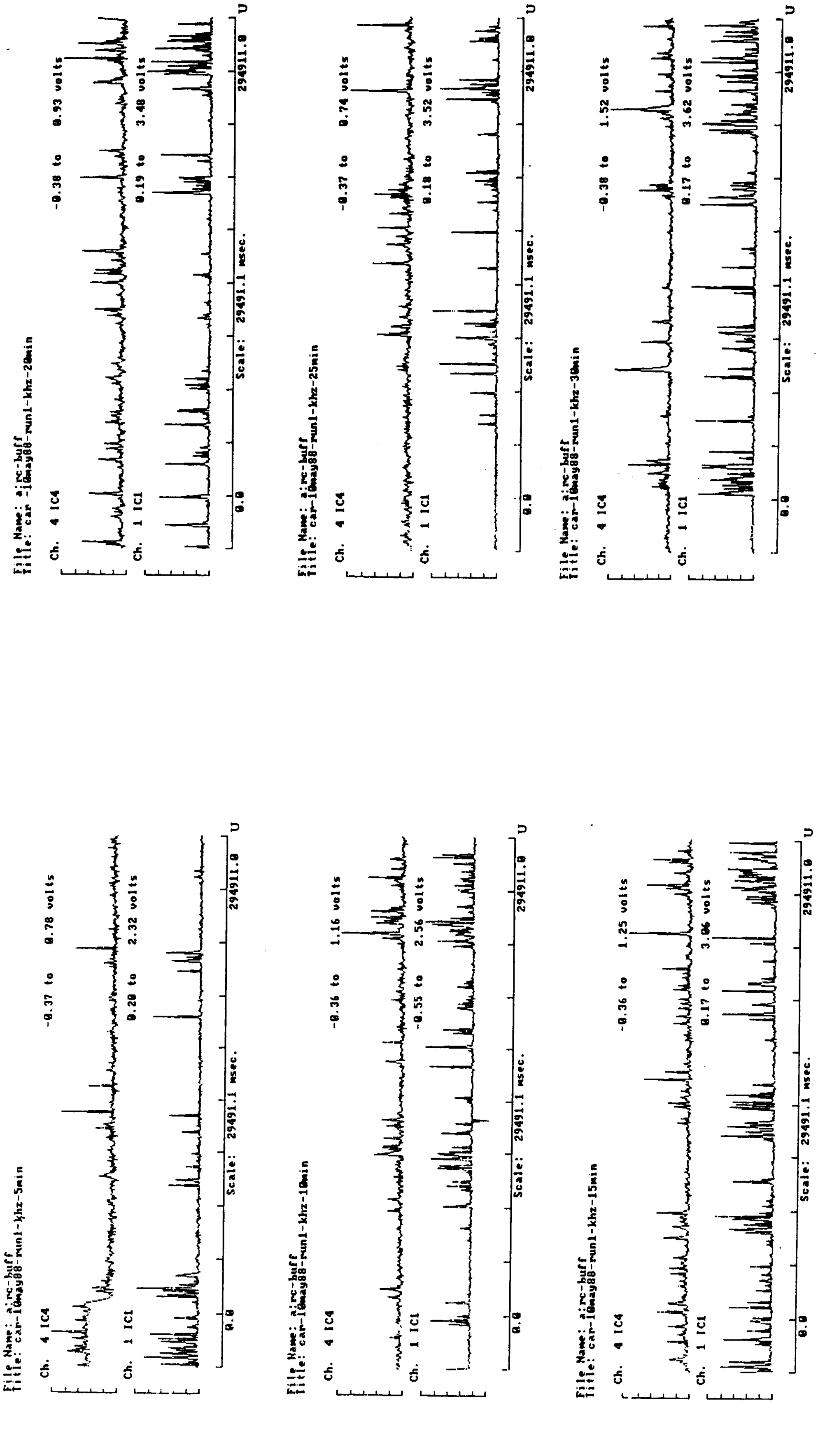


FIGURE 1. RUN 1.

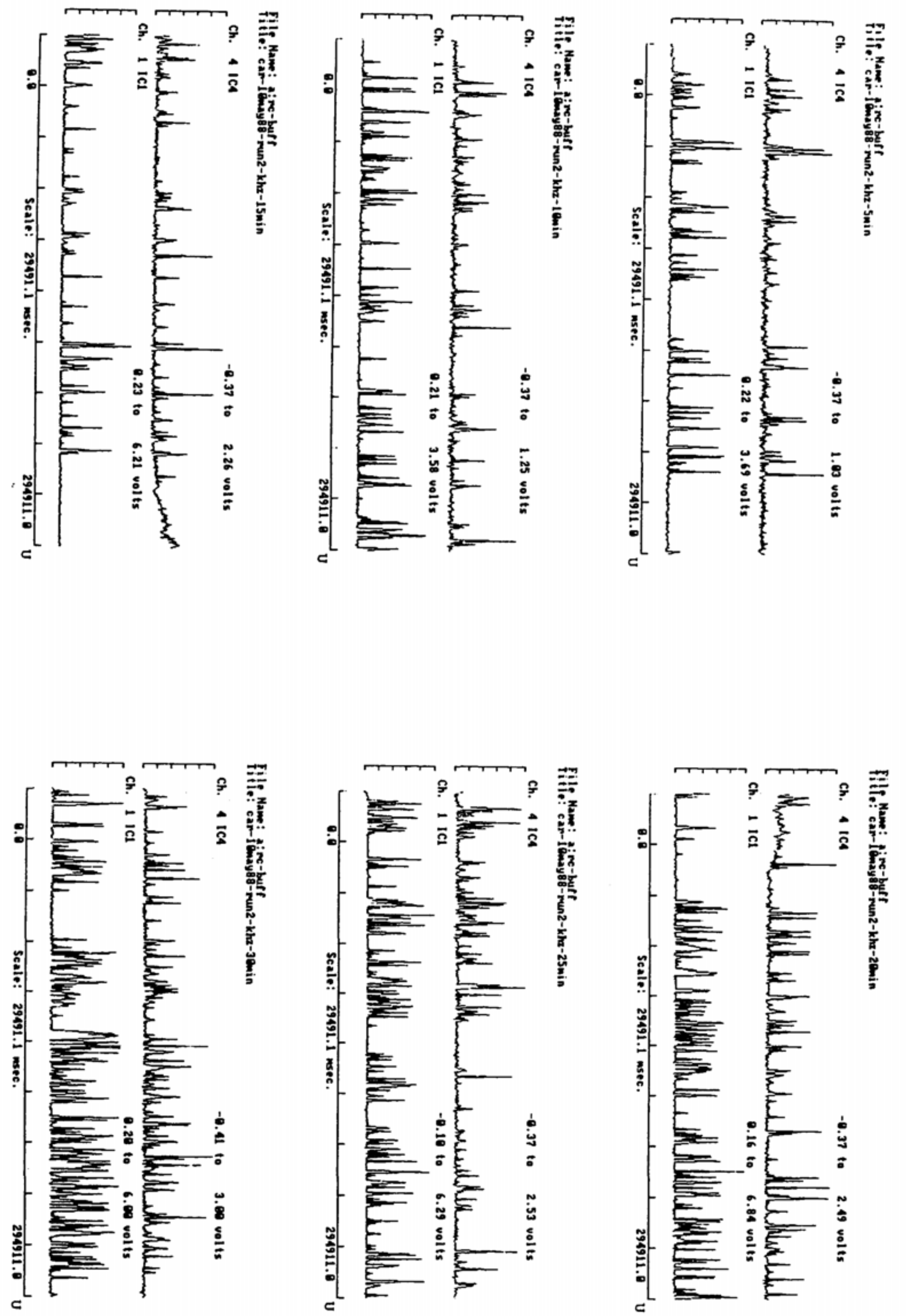


FIGURE 2. RUN 2.

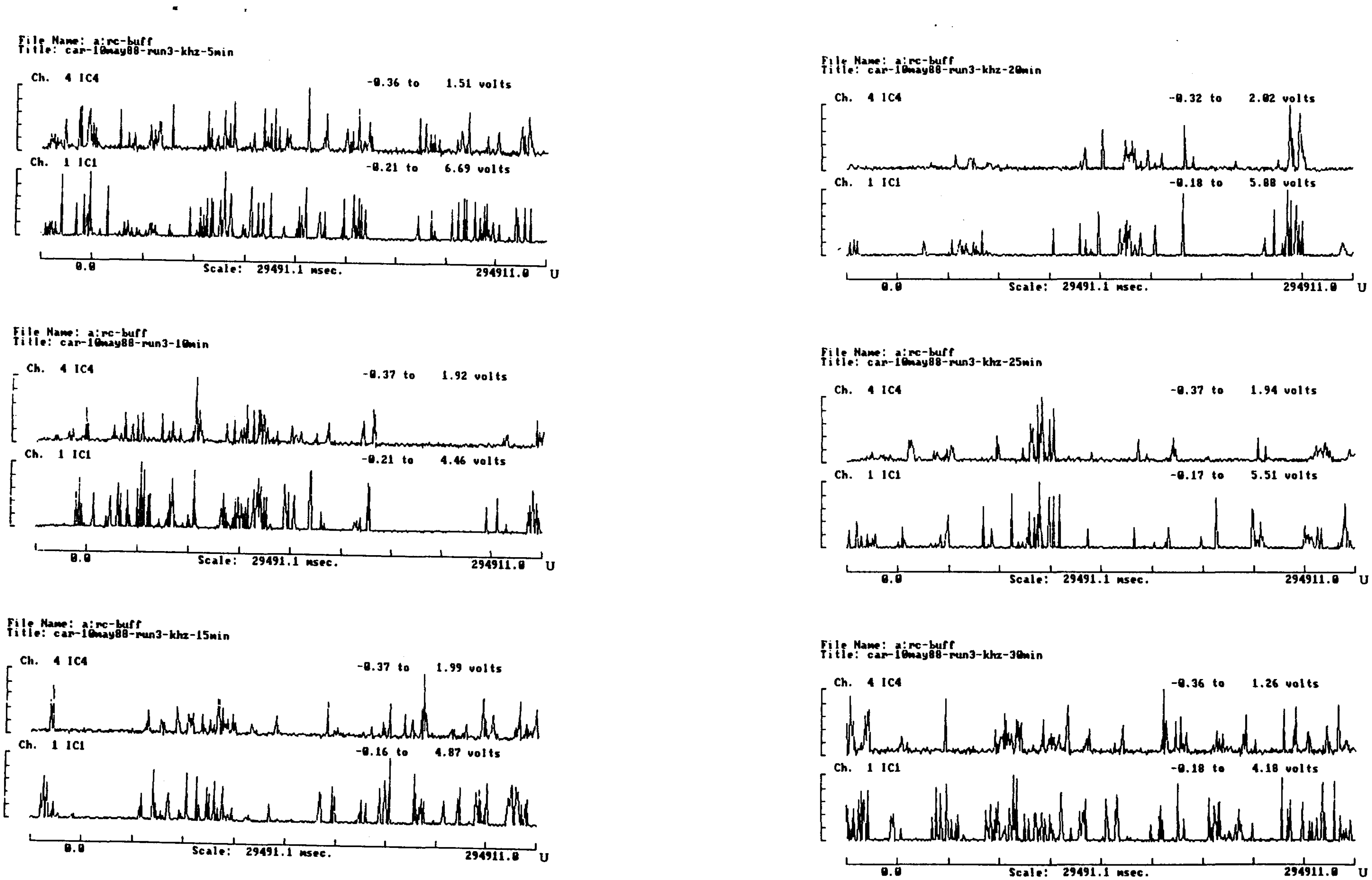


FIGURE 3. RUN 3

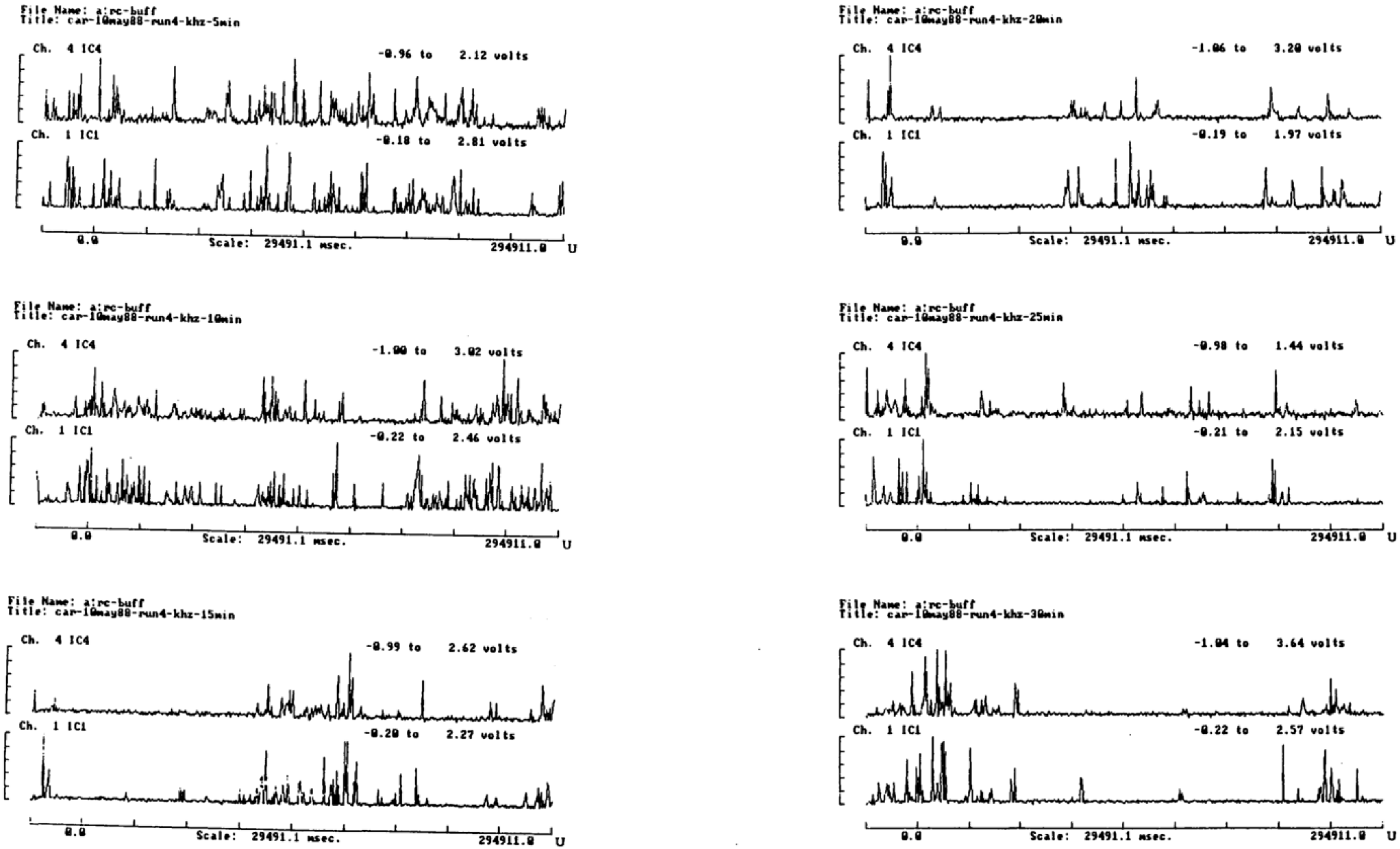


FIGURE 4. RUN 4.

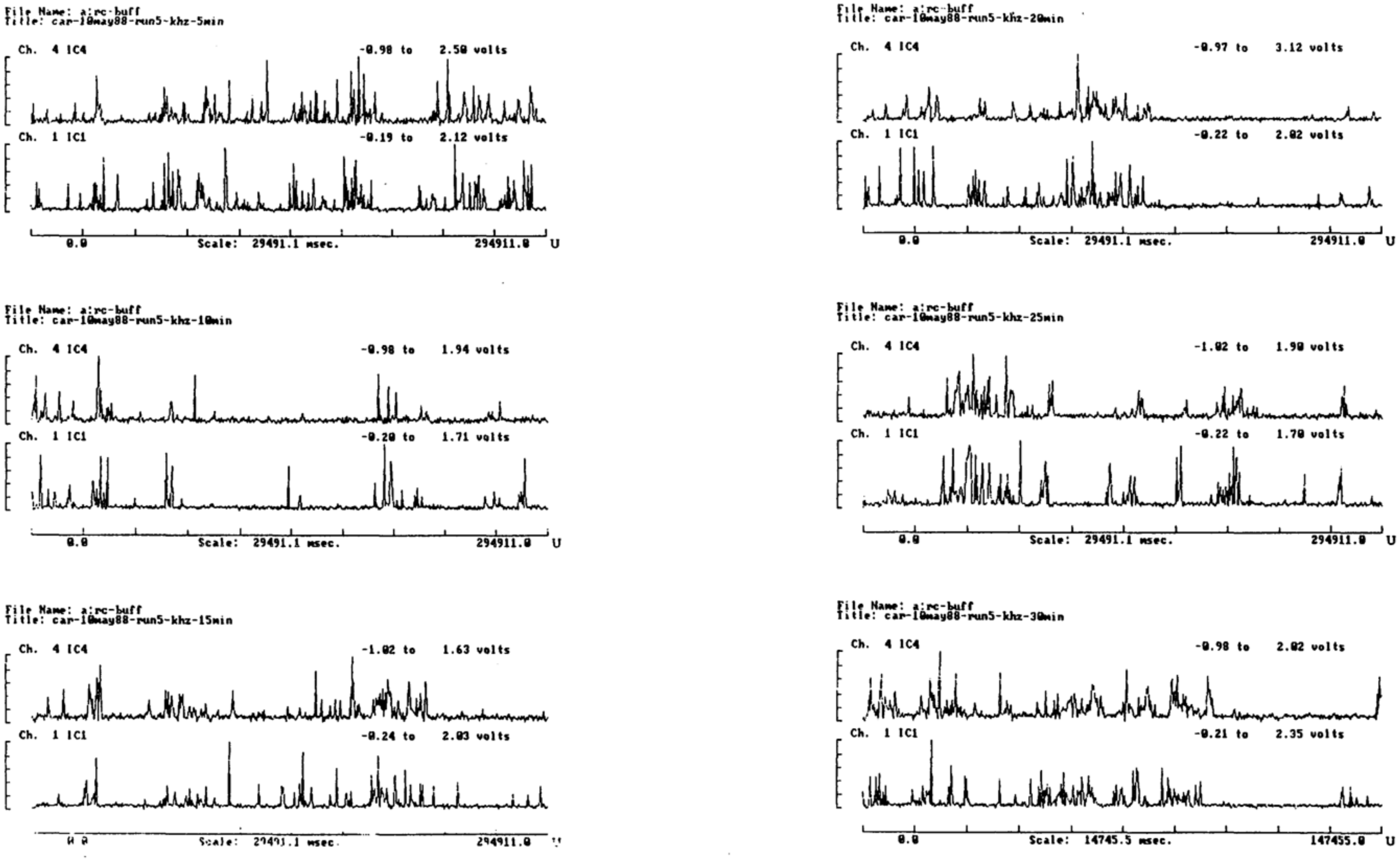


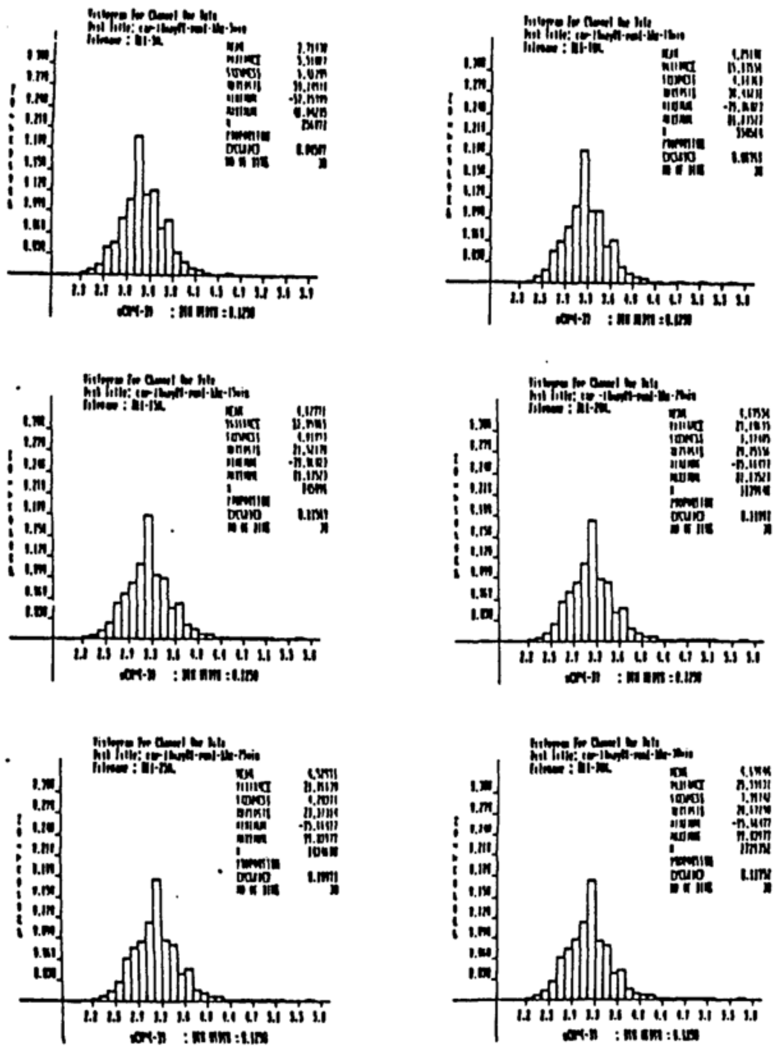
FIGURE 5. RUN 5.

Thus histograms are constructed for a dataset using the above ideas as follows. First, the complete range of values that the observed random variable – here ρ - can take is divided up (in an arbitrary but convenient way). In the case of the 10 May 1988 trials the division adopted was naturally that into the classes defined above. Once the division has been chosen the number of observations within each element of the division is obtained by counting. The abscissa ("x-axis") of the histogram is the whole range of θ, i.e. the set of possible observed values of the random variable, and the ordinate ("y-axis") is *frequency density*, i.e. number of observations per unit of θ. For each element of the division a rectangle is drawn with that element as base and with area proportional to the number of observations in the division. When each element of the division has *equal* width (which is common but by no means universal), the area of each rectangle is obviously proportional to its height with the constant of proportionality the same for each element. Therefore the ordinate of the histogram can then be taken as frequency or proportion. The latter choice was made in the present analysis in which, except for the extreme classes, each element of the division has width $0.125nCm^{-3}$. The advantage of choosing proportion as the ordinate is that, for the reasons given in the previous paragraph, the height of each rectangle is then an estimate of the average value of p(θ) over the appropriate range of θ; thus the shape of the histogram is an estimate of the graph of p against θ, and the accuracy of this estimate would be expected to increase with (a) the total number of observations, and (b) decrease in width of the elements of the division. (The reason for (b) is that p(θ) will vary less over the division so that it is more legitimate to approximate it by its average value.)

As is now clear from section 3.4, the method of analysis of the data from the 10 May 1988 trials was chosen so that histograms could be routinely drawn. When this was done, it was immediately apparent that in all cases the large majority of the data points fell in only about 30 of the 202 classes; this was confirmed by printing out proportions. An important practical point was that the graphics facilities available at Brunel allowed an ordinate resolution of 200 pixels so that a proportion would be *visually* recorded as non-zero only if it exceeded 1/200 = 0.005. The following steps were therefore taken in order to give the most useful visual representation. The class with the highest proportion was identified; this highest proportion was almost always less than 0.4. The ordinate scale on the histogram was expanded so that the maximum value displayed was no longer 1 (the theoretical maximum proportion) but this maximum proportion rounded up to the nearest 0.1; this meant that the observed proportions that would be visually recorded as non-zero fell from 0.005 to e.g. 0.4/200 = 0.002 (for a case when the observed maximum proportion fell between 0.3 and 0.4). Having done this the classes with *visible* proportions were identified, and the scale on the abscissa chosen so that it began with the class with the lowest θ (and a visible proportion) and ended 30 consecutive classes later.

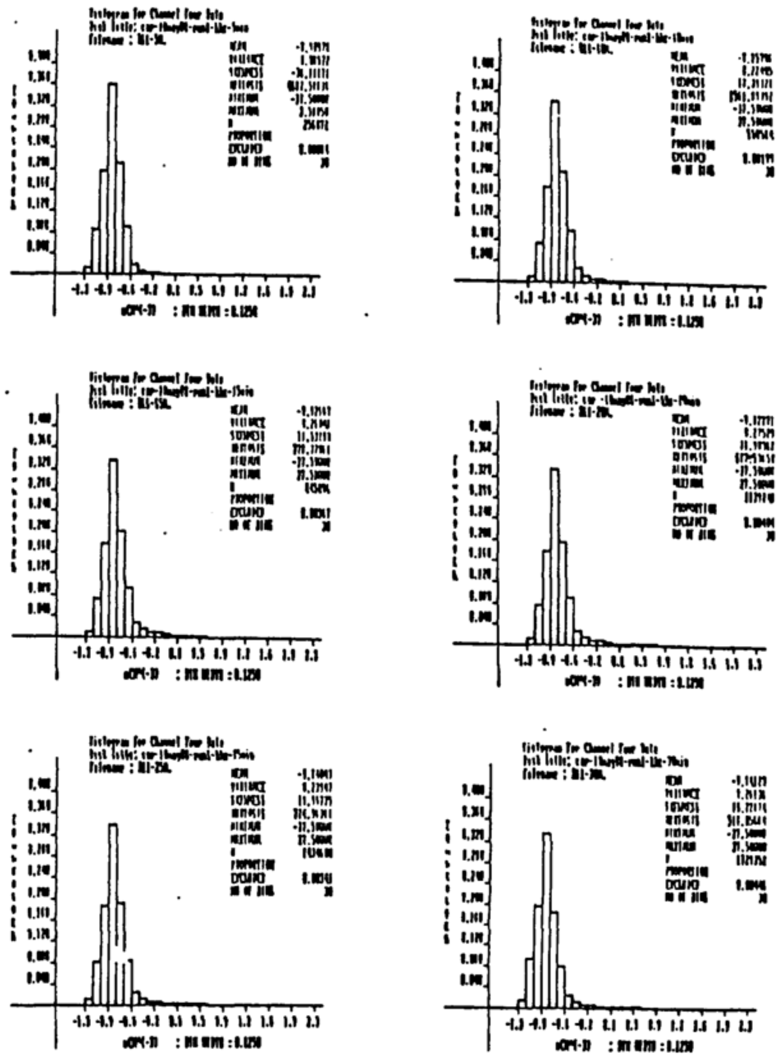
The resulting histograms are shown in Figures 7-12. Interpretation of them will be given in section 3.7 but, for clarity, it is necessary to make two points here. For each

The following Figures 7-12 show the histograms recorded for the 11 channels for which valid data were obtained. Reference should be made to the text for details of their construction.

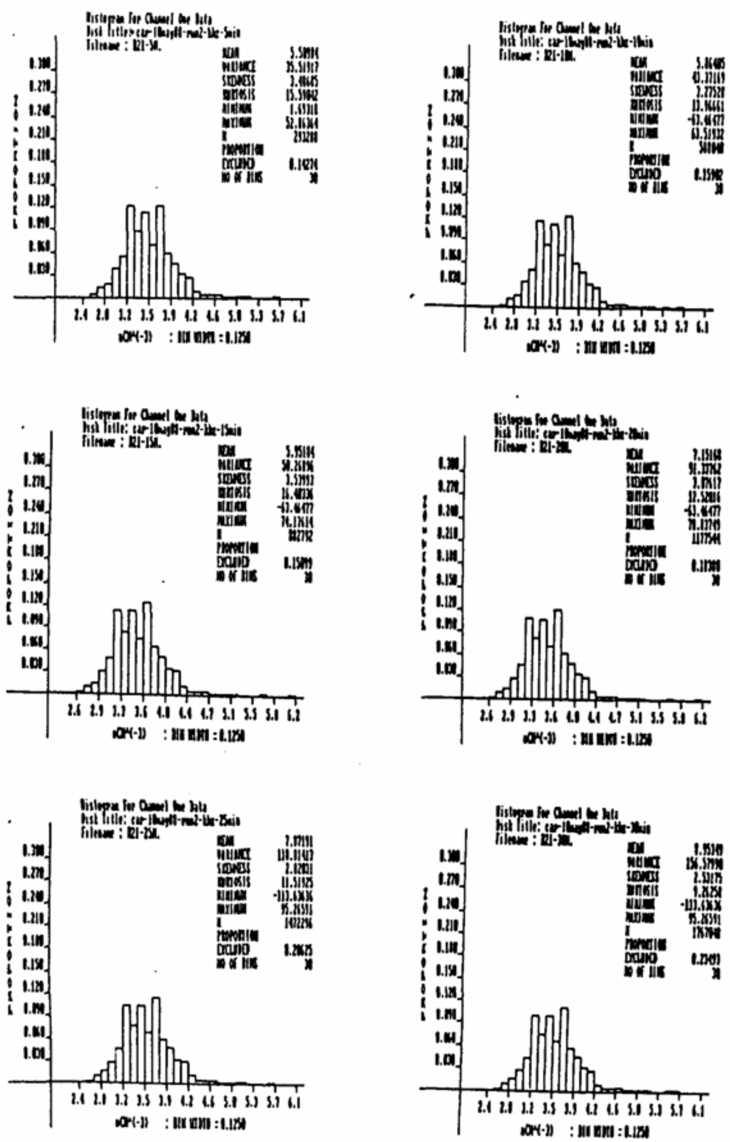


7(a) Run 1 Channel 1

FIGURE 7

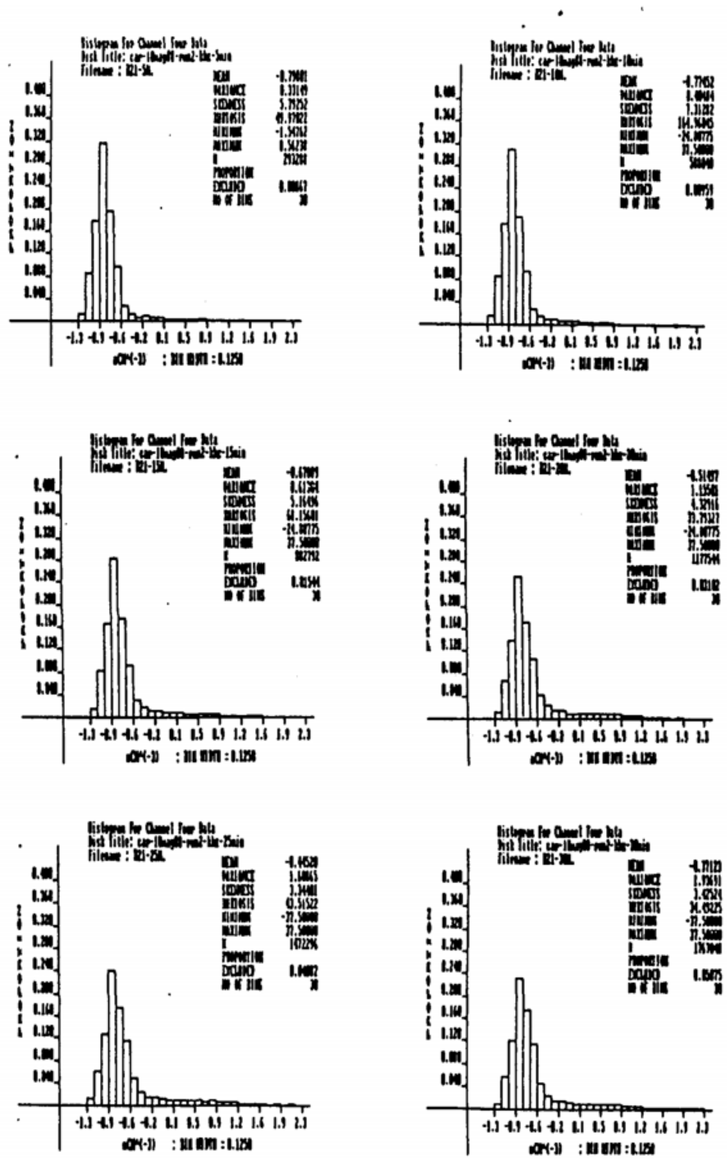


7(b) Run 1 Channel 4

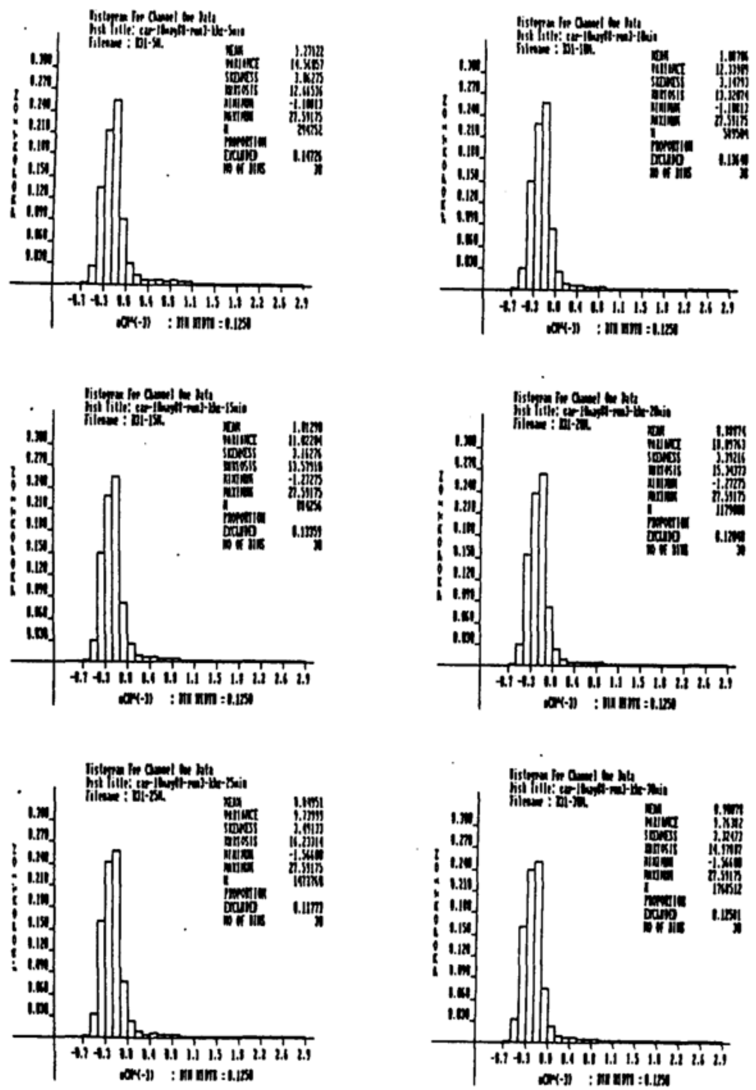


8(a) Run 2 Channel 1

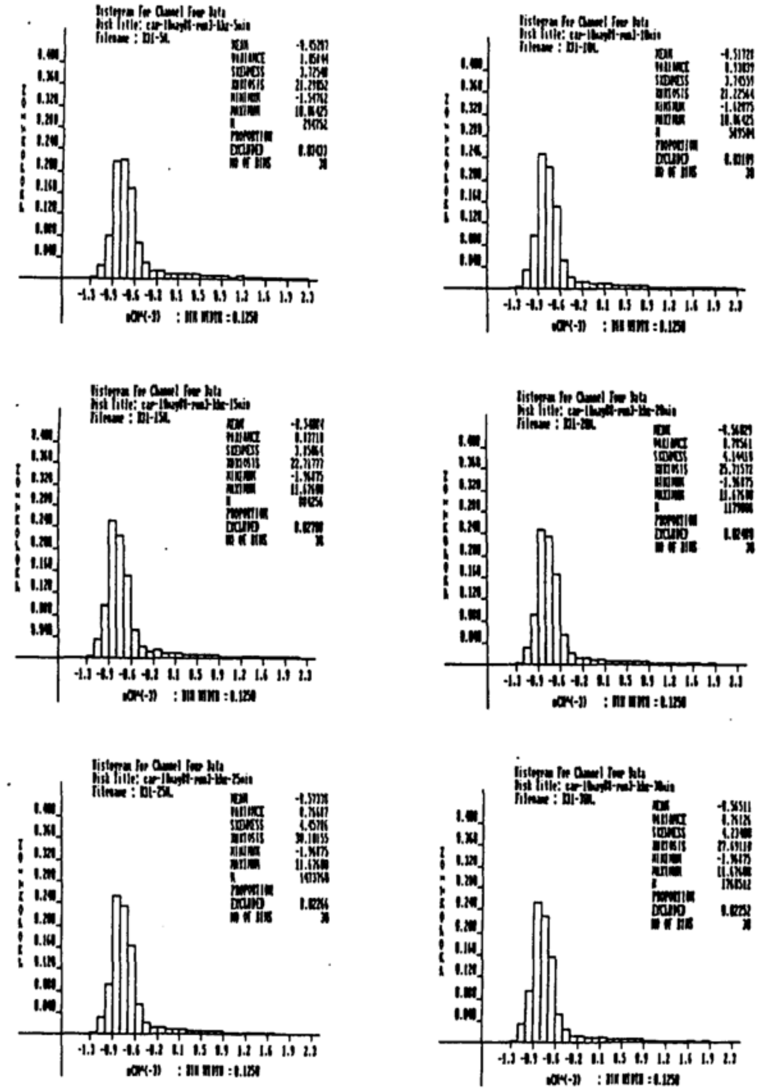
FIGURE 8



8(b) Run 2 Channel 4

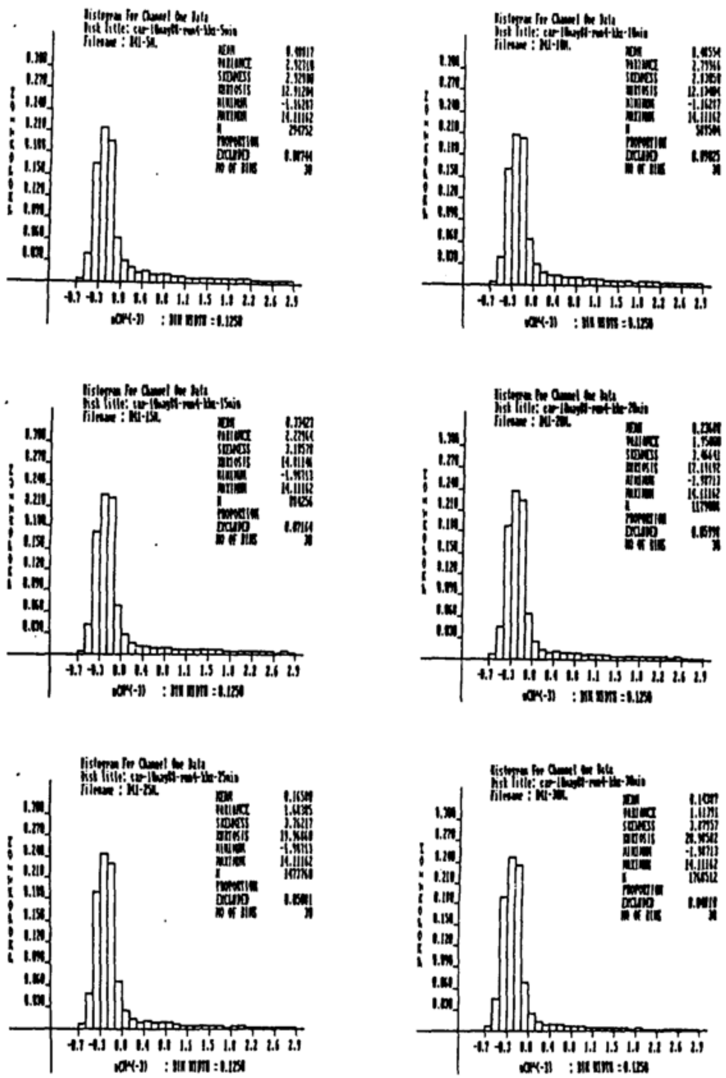


9(a) Run 3 Channel 1



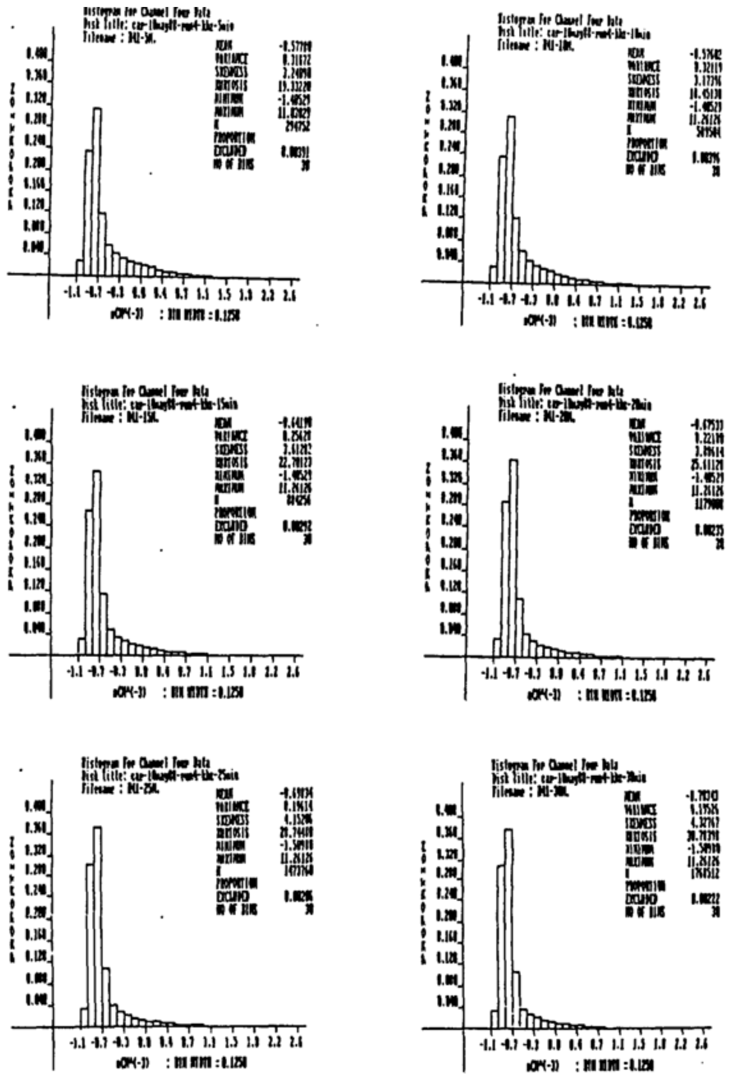
9(b) Run 3 Channel 4

FIGURE 9

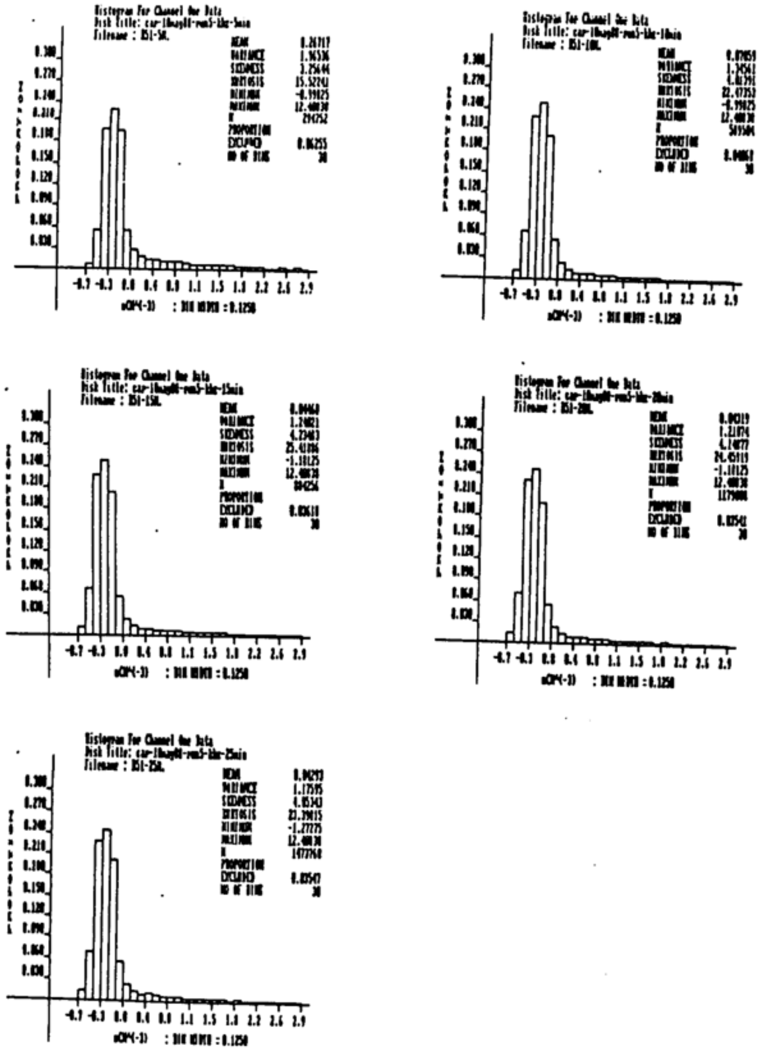


10(a) Run 4 Channel 1

FIGURE 10

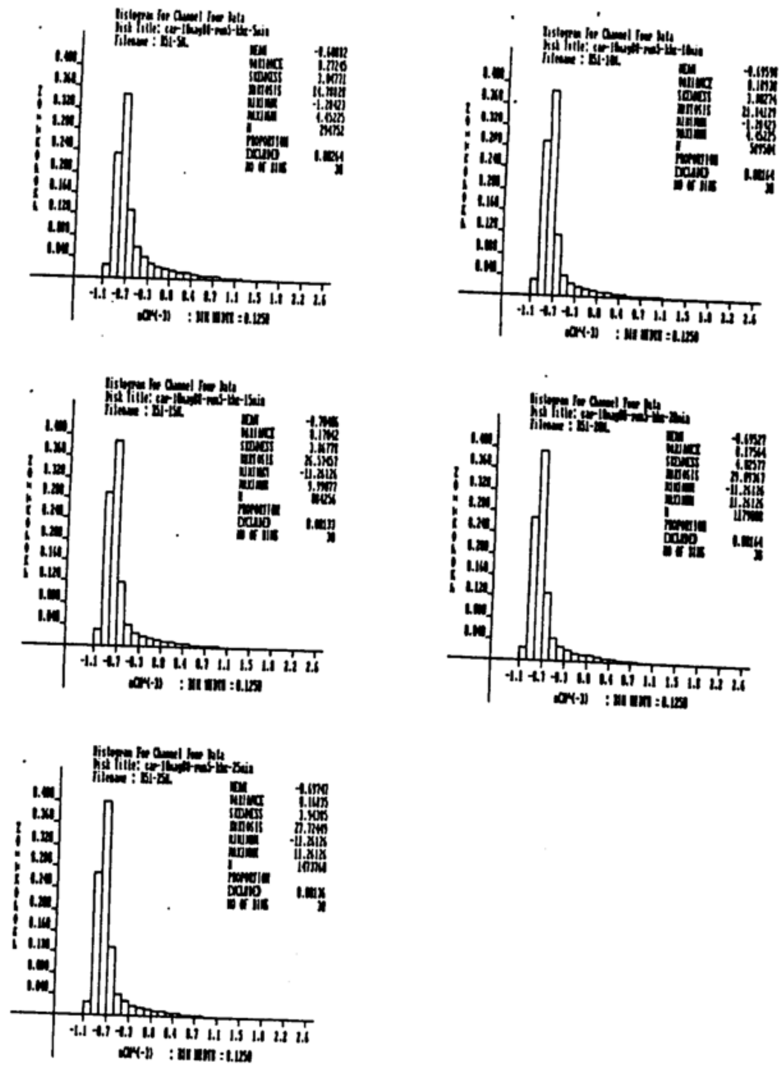


10(b) Run 4 Channel 4



11(a) Run 5 Channel 1

FIGURE 11



11 (b) Run 5 Channel 4

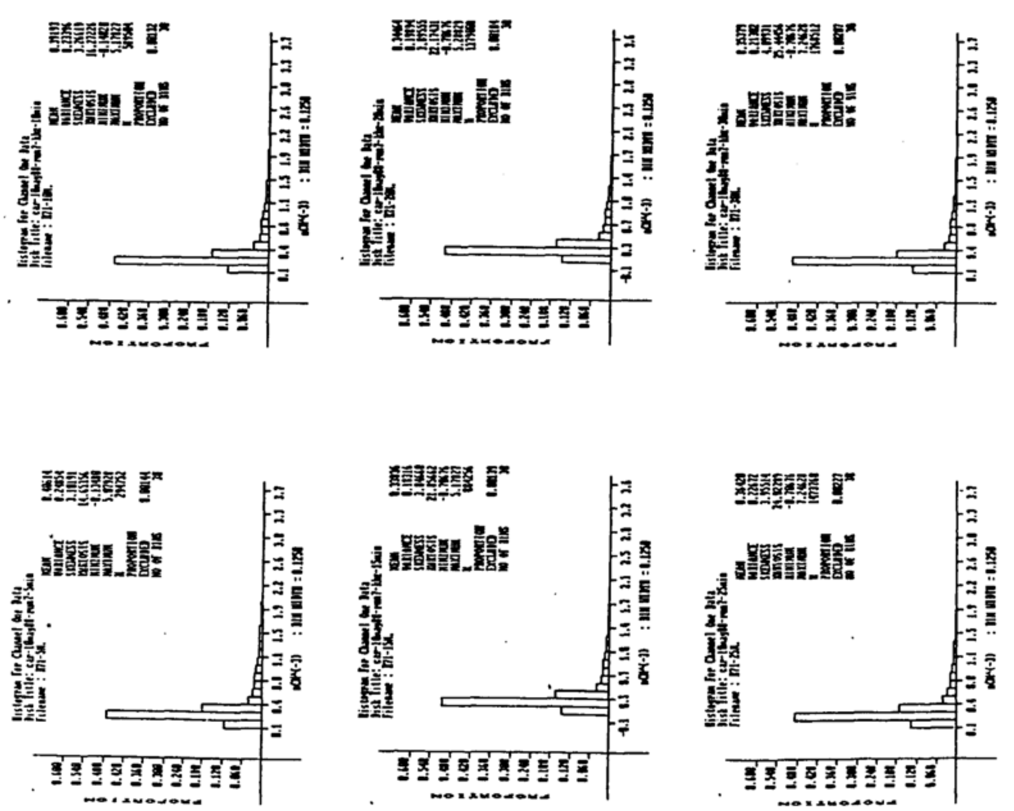


FIGURE 12. Run 7 Channel 1

collector (channel) 5 or 6 histograms are shown and labelled with a time (5min, l0min,...). These histograms are *cumulative,* that is they record all the data collected from the start of the experiment to the time indicated. This is evident from the number N of observations included and shown near the bottom of the list in the top right-hand part of each histogram. The main purpose of this is to judge the length of record needed to obtain acceptable estimates of the pdf; conversely this method of display will show up trends due, for example, to meteorological changes during an experiment. The second point is that some data are not visible on the histograms for the practical reasons discussed in the previous paragraph. For each histogram the proportion of points not shown is recorded; normally this is low but for collectors near the source (as in channel 1 data for run 2) it can reach 10-20%.

3.6 The May 1988 experiments. IV Statistical properties

The data were also used to estimate several basic statistical properties for each dataset using standard statistical formulae. In all cases the calculations were performed using the 'exact' (i.e. subject only to the tolerances imposed by the digitisation) value of each individual observation of ρ. However the results were randomly spot-checked with a different program applied to the *grouped* data used to construct the histograms. This program assumed that *all* observations in a class represented readings at the mid-point of that class. In all cases the results of the checks were satisfactory.

The parameters estimated were the *mean, variance, skewness* and *kurtosis*. For a set of N observations of ρ, the mean was estimated by $\hat{C}'$ where

$$\hat{C}' - \frac{1}{N} \sum_{i=1}^{N} \rho_i \ , \tag{3.1}$$

and ρ_1, ρ_2...., ρN are the N separate observations of ρ. The *variance* (mean square fluctuation intensity) was estimated by $\hat{\overline{c^2}}$, where

$$\hat{\overline{c^2}} - \frac{1}{N} \sum_{i=1}^{N} (\rho_i - \hat{C}')^2 - \frac{1}{N} \left[\sum_{i=1}^{N} \rho_i^2 \right] - (\hat{C}')^2 \ , \tag{3.2}$$

with the equality of the last two expressions following immediately from (3.1). (Although it is customary to use the denominator (N-l), rather than N, in (3.2) to ensure that the estimator is unbiased the criterion of unbiasedness is no more fundamental than several other sensible possibilities. The use of N is more convenient for the algebra needed to combine data from separate 5 minute intervals. Furthermore the *minimum* value of N occurring in calculating any of the estimates was over 250000 so the numerical difference was always negligible.) From equation (1.8), the estimates of skewness and kurtosis were taken as $\hat{S}$ and $\hat{K}$ respectively, where

The following Tables 3 - 8 summarise the statistical properties of the data from the 11 channels. The units for mean, displacement and revised mean at nCm^{-3}, for variance they are $\left(\mathrm{nCm}^{-3}\right)^2$, and the other quantities (skewness, kurtosis, intensity) are dimensionless. Here intensity I - $\frac{\wedge}{\int (c^2)} / \hat{C}$.

	Period covered from start of trial (min)					
	0 - 5	0 - 10	0 - 15	0 - 20	0 - 25	0 - 30
mean: $\hat{C}'$	3.71	4.25	4.63	4.68	4.53	4.70
variance: $\overline{\hat{c}^2}$	5.51	15.34	22.09	23.81	21.87	25.60
skewness: $\hat{S}$	6.41	4.68	4.01	3.97	4.20	3.99
kurtosis: $\hat{k}$	59.15	30.43	21.52	20.86	23.37	20.67
displacement	3.125	3.125	3.125	3.125	3.125	3.125
revised mean: $\hat{c}$	0.59	1.13	1.50	1.55	1.40	1.57
Intensity: I	3.98	3.48	3.13	3.15	3.33	3.22

Table 3(a) : Run 1 collector 1 (d = 3m, i_0 = 10nA)

	Period covered from start of trial (min)					
	0 - 5	0 - 10	0 - 15	0 - 20	0 - 25	0 - 30
mean: $\hat{c}^1$	-0.91	-0.86	-0.83	-0.82	-0.84	-0.84
variance: $\overline{\hat{c}^2}$	0.09	0.22	0.26	0.28	0.24	0.26
skewness: $\hat{S}$	-36.11	17.39	11.53	11.90	12.00	10.22
kurtosis: $\hat{k}$	4687.59	1561.89	778.37	687.54	724.96	518.06
displacement	-1.000	-1.000	-1.000	-1.000	-1.000	-1.000
revised mean: $\hat{c}$	0.09	0.14	0.17	0.18	0.16	0.16
Intensity: I	3.25	3.31	2.94	2.96	3.07	3.22

Table 3(b) : Run 1 collector 4 (d = 12m, i_0 = 10nA)

		Period covered from start of trial (min)					
		0 - 5	0 - 10	0 - 15	0 - 20	0 - 2	0 - 30
mean:	$\hat{C}^1$	5.51	5.86	5.95	7.15	7.87	8.95
variance:	$\overline{\hat{c}^2}$	35.52	43.37	50.27	91.34	118.81	156.58
skewness:	$\hat{S}$	3.49	3.28	3.54	3.08	2.82	2.53
kurtosis:	$\hat{K}$	15.59	13.97	16.40	12.52	11.52	9.26
displacement		3.625	3.625	3.625	3.625	3.625	3.625
revised mean:	$\hat{c}$	1.88	2.24	2.33	3.53	4.25	5.33
intensity:	I	3.16	2.94	3.05	2.71	2.57	2.35

Table 4(a): Run 2 Collector 1 (d = 3m, i_0 = 3nA)

		Period covered from start of trial (min)					
		0 - 5	0 - 10	0 - 15	0 - 20	0 – 25	0 - 30
mean:	$\hat{C}^1$	-0.80	-0.77	-0.68	-0.52	-0.45	-0.37
variance:	$\overline{\hat{c}^2}$	0.33	0.40	0.61	1.16	1.61	1.94
skewness:	$\hat{S}$	5.79	7.31	5.16	4.33	3.34	3.43
kurtosis:	$\hat{K}$	49.97	164.96	68.16	33.79	43.52	34.49
displacement		-1.000	-1.000	-1.000	-1.000	-1.000	-1.000
revised man:	$\hat{C}$	0.20	0.23	0.32	0.49	0.55	0.63
intensity:	I	2.85	2.82	2.44	2.22	2.29	2.21

Table 4(b): Run 2 Collector 4 (d = 12m, i_0 = 3nA)

		Period covered from start of trial (min)					
		0 - 5	0 - 10	0 -15	0 - 20	0 - 25	0 – 30
mean:	$\hat{C}^1$	1.27	1.09	1.10	0.89	0.85	0.90
variance:	$\overline{\hat{c}^2}$	14.56	12.34	11.02	10.10	9.74	9.76
sewness:	$\hat{S}$	3.06	3.15	3.16	3.39	3.49	3.32
kurtosis:	$\hat{K}$	12.67	13.32	13.58	15.34	16.23	14.98
displacement		-0.250	-0.250	-0.250	-0.250	-0.250	-0.250
Revised mean:	$\hat{c}$	1.52	1.34	1.26	1.14	1.10	1.15
intensity:	I	2.51	2.63	2.63	2.79	2.84	2.72

Table 5(a): Run 3 Collector 1 (d = 9m, i_0 = l0nA)

		Period covered from start of trial (min)					
		0 - 5	0 - 10	0 - 15	0 - 20	0 - 25	0 - 30
mean:	$\hat{C}^1$	-0.45	-0.52	-0.54	-0.56	-0.57	-0.57
variance:	$\overline{\hat{c}^2}$	1.06	0.93	0.84	0.79	0.77	0.76
skewness:	$\hat{S}$	3.73	3.75	3.85	4.14	4.46	4.23
kurtosis:	$\hat{K}$	21.29	21.23	22.72	25.72	30.10	27.69
displacement		-1.000	-1.000	-1.000	-1.000	-1.000	-1.000
evised mean:	$\hat{c}$	0.55	0.48	0.46	0.44	0.43	0.43
Intensity:	I	1.88	2.00	1.99	2.02	2.05	2.01

Table 5(b): Run 3 Collector 4 (d = 9m, i_0 = l0nA)

		Period covered from start of trial (min)					
		0 - 5	0 - 10	0 - 15	0 - 20	0 - 25	0 - 30
mean:	$\hat{C}^1$	0.49	0.49	0.33	0.24	0.17	0.14
variance:	$\overline{\hat{c}^2}$	2.93	2.80	2.28	1.95	1.68	1.61
skewness:	$\hat{S}$	2.93	2.83	3.19	3.47	3.76	3.88
kurtosis:	$\hat{K}$	12.91	12.13	14.81	17.19	19.97	20.99
displacement		-0.375	-0.375	-0.375	-0.375	-0.375	-0.375
evised mean:	$\hat{c}$	0.86	0.86	0.71	0.61	0.54	0.52
intensity:	I	1.98	1.94	2.13	2.28	2.40	2.45

Table 6(a): Run 4 Collector 1 (d = 15m, i_0 = l0nA)

		Period covered from start of trial (min)					
		0 - 5	0 - 10	0 - 15	0 - 20	0 - 25	0 – 30
mean:	$\hat{C}^1$	-0.58	-0.58	-0.64	-0.68	-0.70	-0.70
variance:	$\overline{\hat{c}^2}$	0.32	0.32	0.26	0.22	0.20	0.20
skewness:	$\hat{S}$	3.24	3.17	3.61	3.89	4.15	4.33
kurtosis:	$\hat{K}$	19.33	18.45	22.70	25.61	28.74	30.78
displacement		-0.875	-0.875	-0.875	-0.875	-0.875	-0.875
evised mean:	$\hat{c}$	0.30	0.30	0.23	0.20	0.18	0.17
intensity:	I	1.90	1.90	2.17	2.36	2.51	2.58

Table 6(b): Run 4 Collector 4 (d = 9m, i_0 = 10nA)

		Period covered from start of trial (min)				
		0 - 5	0 - 10	0 - 15	0 - 20	0 - 25
mean:	$\hat{C}^1$	0.27	0.07	0.04	0.04	0.04
variance:	$\overline{\hat{c}^2}$	1.97	1.35	1.24	1.21	1.18
skewness:	$\hat{S}$	3.26	4.01	4.23	4.15	4.05
kurtosis:	$\hat{K}$	15.52	22.47	25.42	24.46	23.40
displacement		-0.375	-0.375	-0.375	-0.375	-0.375
revised mean:	$\hat{c}$	0.64	0.45	0.42	0.42	0.42
intensity:	I	2.18	2.60	2.65	2.63	2.59

Table 7(a): Run 5 Collector 1 (d = 15m, i_0 = 3nA)

		Period covered from start of trial (min)				
		0 - 5	0 - 10	0 - 15	0 - 20	0 - 25
mean:	$\hat{C}^1$	-0.61	-0.70	-0.70	-0.70	-0.70
variance:	$\overline{\hat{c}^2}$	0.27	0.19	0.17	0.18	0.17
skewness:	$\hat{S}$	3.05	3.80	3.87	4.03	3.94
kurtosis:	$\hat{K}$	14.79	21.84	26.57	29.09	27.72
displacement		-0.875	-0.875	-0.875	-0.875	-0.875
revised mean:	$\hat{c}$	0.27	0.18	0.17	0.18	0.18
intensity:	I	1.96	2.43	2.42	2.33	2.31

Table 7(b): Run 5 Collector 4 (d = 24m, i_0 = 3nA)

		Period covered from start of trial (min)					
		0 - 5	0 - 10	0 - 15	0 - 20	0 - 25	0 - 30
mean:	$\hat{C}^1$	0.41	0.39	0.34	0.34	0.36	0.35
variance:	$\overline{\hat{c}^2}$	0.25	0.23	0.18	0.20	0.23	0.21
skewness:	$\hat{S}$	3.10	3.27	3.85	3.90	3.96	4.10
kurtosis:	$\hat{K}$	14.65	16.27	21.86	22.17	24.02	25.44
displacement		0.125	0.125	0.125	0.125	0.125	0.125
revised mean:	$\hat{c}$	0.28	0.27	0.21	0.22	0.24	0.23
intensity:	I	1.77	1.81	2.01	2.03	1.99	2.02

Table 8: Run 7 Collector 1 (d = 21m, i_0 = 10nA)

$$\hat{s} = \frac{1}{N}\sum_{i=1}^{N} (\rho_i - \hat{c}^1)^3 / \widehat{(c^2)^{3/2}}, \qquad \hat{K} = \frac{1}{N}\sum_{i=1}^{N} (\rho_i - \hat{C}^1)^4 / (\overline{\hat{c}^2})^2. \qquad (3.3)$$

These estimates were calculated for each 5 minute interval for each of the 11 channels and - for each channel - the results for the separate 5 minute intervals were combined (using simple standard algebra) to give results for 0-5min, 0-l0min..... The results are recorded in each of the histograms in Figures 7-12. For ease of reference and comparison the results are also collected, correct to 2 decimal places, in Tables 3-8.

Superficially it appears that the values of many of the estimated means are inconsistent with the histograms since, by (3.1) and ignoring any corrections for grouping, $\hat{C}'$ is the x-coordinate of the centroid of the histogram. But there is no inconsistency. For the reasons explained in section 3.5, the histograms in Figures 7-12 are *not* the true histograms since only 30 classes are shown and proportions less than about 0.002 are not visible. In fact nearly all the datapoints not included have higher values of ρ than those shown. The true histogram therefore has a low-height positive 'tail', confirmed by the positive skewnesses in all except one case. The estimated mean must then be to the right of the centroid of the approximate histograms. In cases like Figures 7(a) (Run 1 Channel 1) and, especially, 8(a) (Run 2 Channel 2) where the apparent discrepancies are most marked, it is easy to check the sense of the above explanation by noting that the proportions of observations not included are relatively large.

For all the channels analysed, it is apparent from Figures 1-6 that the base line did not significantly drift during the run. Any small drift there may have been is less than, and masked by, *noise*. Unfortunately however this steady base line corresponded to different values of ρ from run to run, and even from collector to collector during one run, apparently because instrument settings made prior to each run were not calibrated. This important point will be discussed later from the general perspective. Here it is necessary to note that there is no *certain* method of adjusting the raw data so that the observed voltages, and hence the inferred values of ρ, are *absolutely* correct and, in particular, there is no certain method of establishing the true zero level of ρ. To illustrate the effect of this point on the estimated mean, the data were assumed to be such that the *true zero* of ρ occurred for the maximum observed proportion with all lesser readings influenced by noise. While there is, on the surface, plenty of evidence from other sources that this often occurs for atmospheric dispersion pdfs, the scientific quality of such evidence is somewhat dubious since other assumptions involving arbitrary choices, such as thresholding, have usually been made for the data. The assumption gives a displacement value, constant for each channel, and this is recorded in Tables 3-8. The resulting revised mean $\hat{C}$, obtained by simple subtraction, is also recorded, as is the *intensity* $(\overline{\hat{c}^z})\frac{1}{2}/\hat{C}$. Given the arbitrary nature of the assumptions,the revised histograms have not been drawn but they would be identical in shape to those in Figures 7-12 with a revised scale on the abscissa. The other calculated parameters are not affected in any way by a constant displacement.

3.7 The May 1988 experiments. V Discussion of results

It is unfortunate of course that more than half of the recorded data proved unsuitable for analysis. The cause was apparently the malfunctioning of two of the four collectors; hence valid observations were taken at only two downwind distances in 5 of the 6 runs and at only one downwind distance in run 7. Given also the uncertainty about the true zero level referred to at the end of section 3.6, which translates into uncertainty about comparing results from different runs, it is clear that the results of the analysis will be much less useful than was hoped for (and anticipated) in meeting one principal aim of the Agreement, namely the study of the evolution of the pdf with downwind distance. These points will be referred to again in Chapter 4.

All the histograms in Figures 7-12 contain the effect of inevitable, but unwanted, *instrument noise* whose own statistical properties were not separately measured. It seems almost certain that this noise and the concentration will be statistically independent, and a substantial body of previous research suggests that the pdf of the noise alone will be very nearly, if not exactly, Normal with mean zero - see equation (1.9). (Here "zero" of course means an absolute value; as noted at the end of the previous section this does not necessarily mean a *measured* value of zero.) Given that concentrations are positive the parts of the histograms to the *left* of the peaks must contain substantial contributions from the noise. While it is by no means certain that the assumption made at the end of the previous section that the peaks correspond to an absolute zero of ρ is correct, it must be true that noise makes a relatively increasing contribution to the histograms as the measured value of ρ decreases from the value at which the peak occurs. To judge by eye from the histograms, it then follows that the *standard deviation* (i.e. root mean square value) of the noise in the May 1988 experiments was of order 0.2nCm^{-3} giving a *variance* of order $0.05\,(\text{nCm}^{-3})^2$. Since the variance of the sum of two independent random variables is the sum of their variances, the variance of the concentration signal alone will be obtained by subtracting the variance of the noise from the values given in Tables 3-8. It can be seen that in some cases (e.g. Run 1, Channel 4, 12m; Run 4, Channel 4, 24m; Run 5, Channel 4, 24m; Run 7, Channel 1, 21m) this causes a significant reduction. Since the pdf of the noise is likely to be almost, if not exactly, symmetric it makes "equal" contributions to the observed histograms on both sides of (absolute) $\rho = 0$. That for positive ρ is, of course, masked by the larger concentration signal.

The above discussion and that immediately below takes no separate account of *instrument smoothing*, by which is meant changes, other than those attributable to pure noise, caused by characteristics of the sensors and - more generally - of the whole instrumentation system. Such changes include time and space averaging, and filtering of the input signal. They have been examined in depth by Mole in various papers (e.g.

Mole 1989); following detailed analysis of a dataset collected by the CDE sensor system (Griffiths and Jones 1986), he concluded that instrument smoothing made an insignificant contribution in that case to estimates of $\overline{c^Z}$. In this report this conclusion is extrapolated and the assumption has been made that instrument smoothing can be ignored throughout the data analysis and interpretation.

Models of pdfs in atmospheric dispersion, while still relatively rare, have almost all (e.g. Hanna 1984; Ride 1987; Dinar, Kaplan and Kleiman 1988; Chatwin 1989) given an important role to the intermittency factor γ defined in (1.10). A standard model form for the pdf p(θ) is

$$P(\theta) = \gamma^{\delta} +(\theta) + (1-\gamma)P_c(\theta) \ , \tag{3.4}$$

where δ+(θ) is a one-sided *delta function* (δ+(θ) is zero for $\theta \neq 0$, and infinite for $\theta = 0$ in such a way that it has integral 1 over every interval $0 \leq \theta \leq$ Є, where Є is *any* positive number) and $p_c(\theta)$ is the so-called *conditional* pdf of ρ, i.e. the pdf of ρ restricted to those cases where ρ is greater than 0. In the experiment the measurements are not of ρ but of ρ+N, where N = N(t) is the noise. It follows that the pdf of the measured (*output*) signal PM(θ) say, is the *convolution* of p(θ) in (3.4) with the pdf of the noise, PN(x) say. Therefore

$$P_M(\theta) = \gamma P_N(\theta) + (1-\gamma)\int_0^{\infty} P_c(x) P_N(\theta - x)dx \tag{3.5}$$

This formula shows that negative readings will occur from both contributions to p(θ) in (3.4). The probability of a negative reading is P, where

$$P = \int_{-\infty}^{0} P_M(\theta)d\theta = \gamma \int_{-\infty}^{0} P_N(\theta)d\theta + (1-\gamma) \int_0^{\infty} P_c(x)\left\{\int_{-\infty}^{0} P_N(\theta - x)d\theta\right\}dx \ , \tag{3.6}$$

When, as normally anticipated, PN(θ) is symmetric about θ=0, this becomes

$$P = \frac{1}{2}\gamma + (1-\gamma)\int_0^{\infty} P_C(x)\left\{\int_x^{\infty} P_N(y)dy\right\}dx \ . \tag{3.7}$$

The second term in (3.7) is the contribution from cases where the true concentration ρ is positive but of lesser magnitude than the negative noise. Its precise value depends on p_c and PN; in two idealised model calculations that have been made it is of order $(1-\gamma)(\sigma_N/\sigma_c)$, where σ_N and σ_c are the standard deviations associated with p_N and pc respectively. The inferred values of σ_N and σ_c for the May 1988 experiments (see above and Tables 3-8) strongly suggest that in some cases the second term in (3.6) made a significant contribution to P which *cannot* therefore be satisfactorily approximated by $\frac{1}{2}\gamma$.

Leaving aside the difficulties considered above about the true zero, it follows, conversely, that γ cannot be estimated by 2P nor, *a priori,* in any other way since it is not known what $p_N(x)$ is. This repor therefore contains no estimates of the intermittency factor γ.

It should also be noted that Chatwin and Sullivan (1989a,b) have criticised the definition (1.10) of γ on the grounds that it is meaningless because *molecular diffusion* definition ensures that zero concentrations do not exist, i.e. a strict application of (1.10) gives $\gamma = 0$ everywhere.

Visual inspection of the 11 series of histograms shows little difference between the last two or three in each series. This confirms that 25-30 min, with sampling at 1kHz, is a sensible run length, i.e. that *statistical* noise is then sufficiently small for stable estimates to be achieved provided the dispersion remains *statistically stationary* during the run. (Above all this requires atmospheric conditions to remain stationary - see below.) The two series of histograms taken at 3m from the source (Figures 7(a) and 8(a) - Channel 1 for Runs 1 and 2) appear to be almost symmetric (even though the estimates $\hat{S}$ of skewness are not small). All the other series of histograms have a familiar and characteristic general shape, namely a single peak {*mode*) at a low value of ρ with the histogram falling off to zero much more slowly to the right of the peak than to the left. This gives positive skewness, and Tables 3-8 confirm that $\hat{s}$ is greater than zero in all cases except one. (This negative value is derived from 5 min of data only - Table 3(b): Run 1, Channel 4 - so that it is very likely due to the shortness of the length of record since all remaining values for this series are positive.)

Before discussing the evolution of the histograms with downwind distance, it is useful to consider Figure 13. This shows the dependence of the revised means $\hat{C}$ on length of data record as given in Tables 3-8. Except for Run 2 (both collectors), the values of $\hat{C}$ appear to be stabilising within the bounds expected because of statistical noise. It can be seen from Tables 3-8 that the values of the estimated variance $\widehat{\overline{c^2}}$ have the same features. Again Run 2 is anomalous.

It is superficially curious that the histograms in Figure 8 do not immediately show up the non-stationary behaviour in Run 2 that is obvious from Table 4. However a casual glance at the time series in Figure 2 shows activity in the last 15 minutes that is markedly enhanced relative to the first 15 minutes. The means and variances for this first part of the record are certainly of the same order that they are somewhat higher could be due entirely to statistical noise) as those recorded at the same distances downwind (but with different values of i_0) in Run 1. An obvious explanation for the rapid growth of $\hat{C}$ and $\widehat{\overline{c^2}}$ during the latter half of Run 2 is that this was a period when the wind speed, and hence the turbulence, was increasing significantly so that the collectors recorded the ion plume at earlier and earlier stages of its evolution than in Run 1. There is no apparent reason to reject this explanation. Other possible causes that have been considered, but rejected, are (i) malfunctioning of the measurement system (no visual evidence and the results would require the highly improbable event that both collectors malfunction *simultaneously*), and (ii) effects due to plume *meandering* (trends in Figure 13 are the "wrong way", assuming that the record in the first part of Run 2 is from the same region of the (mean) plume as the other records; see below for further

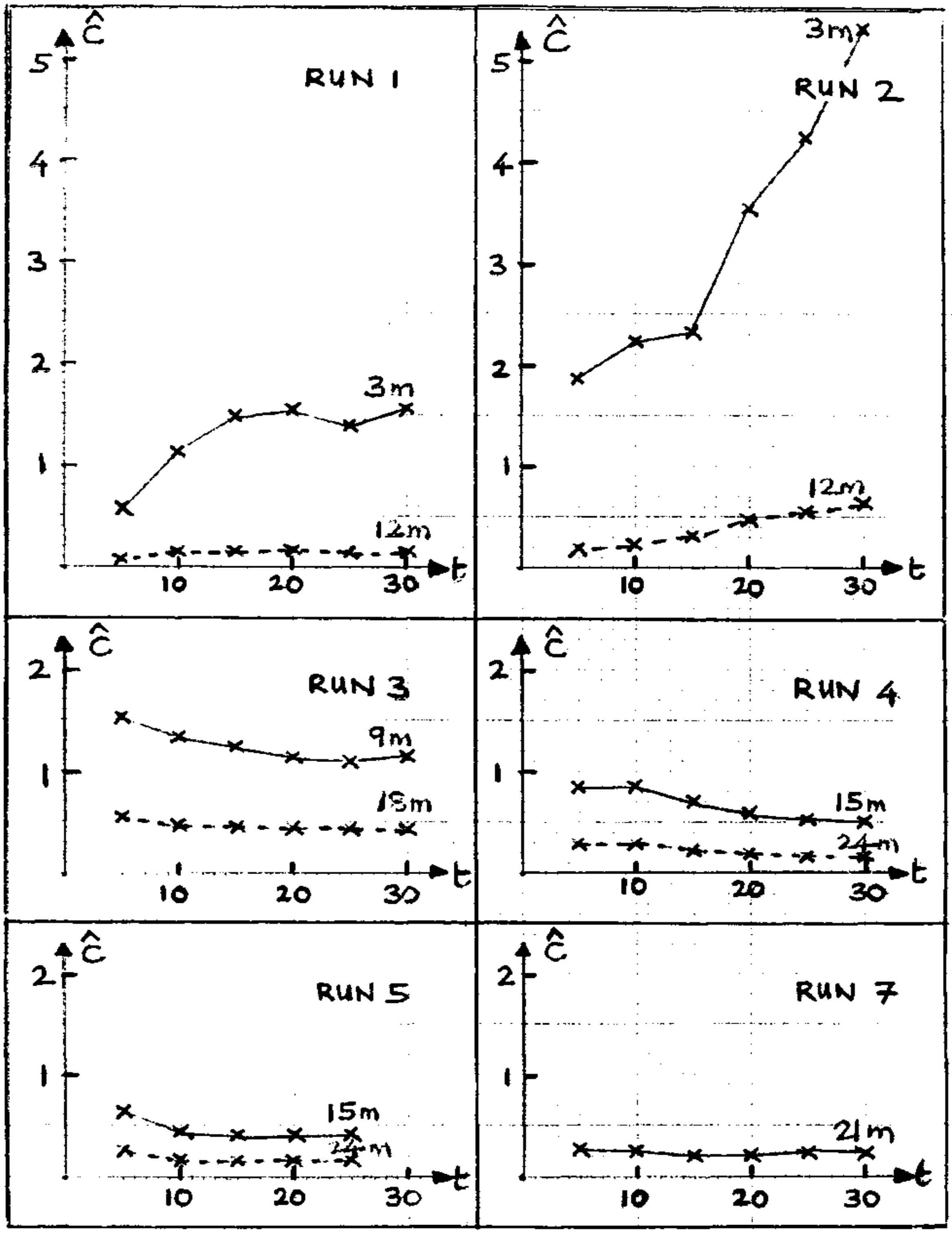


FIGURE 13. Revised means $\hat{C}$ from Tables 3-8 as functions of length of data record. In each case the units of $\hat{C}$ are nCm $^{-3}$, those of t are min. and the distances above each plot are the distances downwind from the source

discussion).

In Figure 14 and 15 are plotted the asymptotic (i.e. those obtained at the end of the whole data record that was analysed) values of the statistical properties as functions of downwind distance from the source; Run 2 values are excluded. There are several interesting features of these graphs.

At 12m downwind (Run 1, Channel 4) the values of and $\hat{C}$ and $\hat{\overline{c^2}}$ are much lower, and those of ŝ and $\hat{K}$ much higher, than the trends obvious from all the other points. These observations are all consistent with plume meandering. This is the term for the phenomenon in which the plume axis - strictly the mean plume axis - wanders due to changes in wind direction. It is known (see Chatwin and Sullivan (1990) for more detail) that, for crosswind traverses, C has a maximum on the axis, and $\overline{c^2}$ has an off-axis maximum (at about one plume width crosswind) but an axial value of the same order of magnitude as its maximum; both C and $\overline{c^2}$ tend to zero as the crosswind distance from the axis increases. The behaviour of S and K is somewhat more complex but for present purposes it is relevant only that both tend to plus infinity as the crosswind distance increases, with K doing so much faster than S. Therefore all the observations at 12m downwind are consistent with the plume having meandered (veered) between 3m and 12m (since the properties at 3m downwind from the same run - Run 1 - appear, admittedly on relatively little evidence, to fit the trends in Figures 14 and 15) and with the data at 12m having been taken at several plume widths crosswind from the axis. These comments make the implicit inference that all the points in Figures 14 and 15 come from data taken on or near the mean plume axis - hence the rejection above of plume meandering as an explanation of the anomalous appearance of the observations from Run 2. The large magnitude of the changes in the statistical properties due to meandering is important (but perhaps not welcome!) since it indicates the experimental, and subsequent analytical, effort needed to provide a complete map of the pdf associated with a plume dispersing in the atmosphere, even in stationary conditions. (In addition there are practical, even philosophical, considerations connected with the question of what is meant by "stationary" conditions in theatmosphere, giventhe presence of changes on time-scales of the order of months, years and - even, climatically speaking - hundreds of years and more.)

The values of the intensity $(\hat{\overline{c^z}} / \hat{C}^2)^{\frac{1}{2}}$ in Figure 14 are roughly constant with downwin d distance, consistent with *self-similarity* (Chatwin and Sullivan 1990) and of order 2-3. This should be compared with values of order 1 obtained by Dinar, Kaplan and Kleiman (1988), also in the atmosphere, but using fog-oil smoke as the dispersing material and an optically based detection system with data collected at a rate of 30Hz. There has been some controversy about the limiting value (if any) of the axial intensity as downwind distance increases. It is difficult to understand why some researchers apparently believe that this issue is one of fundamental importance. Suffice it to report here that, like many other datasets, usually taken in a laboratory, the present results are inconclusive; they could be consistent with a limit of zero, or a positive constant.

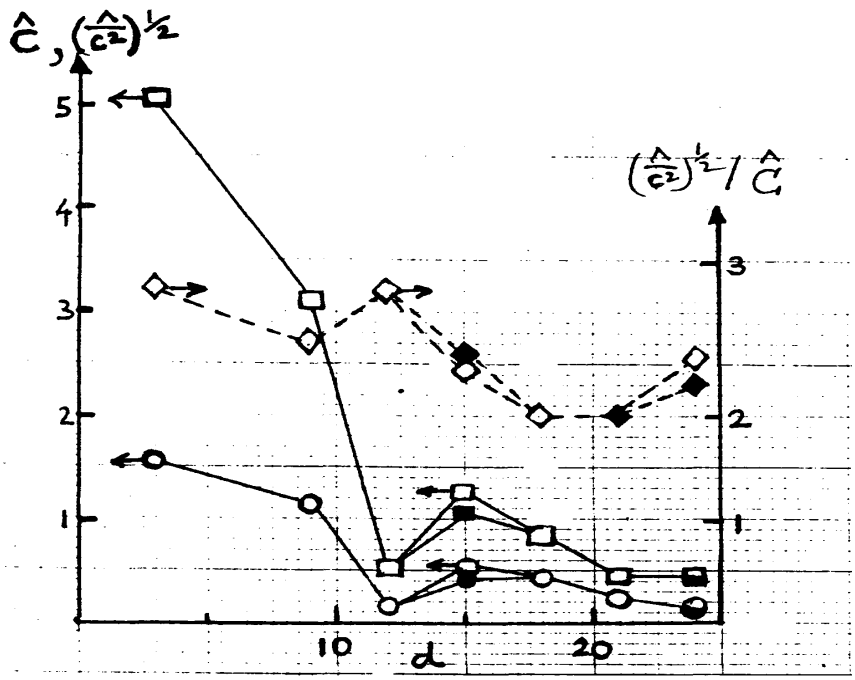


FIGURE 14. Asymptotic values of ĉ(circles), $(\hat{c^2})^{\frac{1}{2}}$ (squares) and $(\hat{c^2})^{\frac{1}{2}}/\hat{c}$ (diamonds) versus downwind distances d(metres). Open symbols are for i_0 = l0nA, closed symbols are for i = 3nA and half-open for 2 (almost) identical readings - one for each value of i_{fl}.

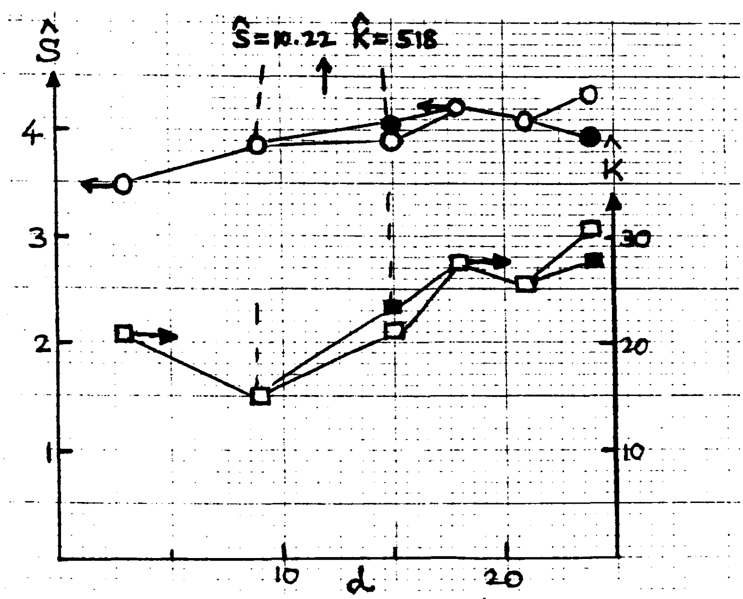


FIGURE 15. Asymptotic values of ŝ (circles) and (squares). $\hat{k}$ Open and closed have the same meanings as in Figure 14.

Likewise the values of $\hat{S}$ in Figure 15 are roughly constant and so also are those of $\hat{K}$ given that the higher the moment of the pdf p(θ), the slower is the approach to self-similarity.

It is also reassuring to note from Figures 14 and 15 that there is no discernible dependence on i_0 of the *non-dimensional* parameters characterising the pdfs; this suggests that (2.7) ensures that electrostatic effects are indeed negligible (as intended).

In summary the histograms and the calculated statistical properties appear to be consistent with a self-similar structure (or an approach to such structure) of the type analysed for laboratory data by Chatwin and Sullivan (1990). It is certainly possible to fit the dependence of $\hat{C}$ and $\hat{\overline{c^z}}$ on downwind distance shown in Figure 14 by lines (or curves) using standard techniques such as *least squares*. But the statistical significance of the results would be low given the (regrettably) small number of points. Similar remarks apply to possible comparisons of this dependence with *theoretical* predictions such as those in Pasquill and Smith (1983). In both cases, there would be several good reasons, discussed above, for deviations of individual readings from the fitted line or theoretical prediction and no foundation, on the basis of the data taken on 10 May 1988, for choosing between these reasons. A *fortiori,* it is not sensible to attempt to discriminate at the moment between the various models that have been proposed for the form of the pdf.

Chapter 4

CONCLUSIONS AND RECOMMENDATIONS

4.1 Introduction

The main purposes of this final Chapter are to summarize the principal *general* conclusions and to recommend further *research*. These two purposes will be considered together in the next section since it is rather unnatural to separate them.

4.2 What has been learned and what remains to be done

The work has closely followed the Description of Work (see section 1.1) *except* that there were no useful new data to enable further studies of point (b) to be undertaken. (This is the important question of the effect of sampler dimensions, source size etc. on the perceived pdfs.) The experiments that were conducted and analysed (see Chapter 3) led to many interesting discoveries. First of all, they seemed to confirm, where appropriate, the correctness of the theoretical studies (Chatwin 1985; Chatwin, Hajian, Mole and Jones 1989) performed both before, and as part of, this work and summarised in Chapter 2 of this report. Secondly the analysis was far more detailed than of any other datasets collected by the CDE sensor system, and showed, once more, its peculiar advantages as a tool for studying atmospheric dispersion.

In slightly more detail, the analysis of the data showed that under stationary weather conditions, run lengths of about 30 minutes with sampling at 1kHz are sufficient to enable statistically stable estimates of $p(\theta)$ to be made, and therefore of its most important parameters. The results were consistent with the achievement of self-similar structure at distances of the order of a few metres downstream of the source (at least at the height of about 2m at which most of the analysed data were taken). However, for various reasons, the results did not allow deductions that were statistically significant to be made about the evolution of the pdf and its associated statistical properties with downwind distance.

The principal one of these reasons was the loss of more than half of the expected data on 10 May 1988 due to the malfunction of 2 of the 4 collectors. Moreover it was the two middle collectors that were faulty (see Table 2); given that two collectors out of four were to fail, these would clearly have been the worst ones to choose from the point of view of analysing dependence on downwind distance! These remarks lead to:

> **RECOMMENDATION 4**: *The causes of the malfunction of ion collectors during prolonged runs should be further investigated and, where possible, rectified. Attention should be given to the tighter monitoring of collector performance during runs and to the possibility of providing replacements for faulty collectors.*

In making this recommendation, it is recognized that some of the commonest causes of malfunction are beyond the control of the experimenters. These include, for example, collision with insects. It is also understood that the design and construction of the ion source and collectors is already receiving substantial attention.

Two other reasons why the data analysis could not be taken in this work as far as had been hoped have been discussed in section 3.7. These are: substantial uncertainty about the measured signal that corresponds to a true zero of ρ (or - perhaps more exactly - the ambient value of ρ in the absence of injected ions) *and* ignorance of the precise form of the pdf $p_N(x)$ of the noise. Both problems would be improved by implementing:

RECOMMENDATION 5: *In a prolonged run in the field the system should be operated for two relatively short additional periods (up to 5 minutes in each case), one immediately prior to switching on the ion source and one immediately after it has been switched off.*

The purpose of the second short period is, of course, to check on changes in system performance during the run, particularly to examine whether there has been baseline drift.

It has been noted in section 3.7 that Mole (1989) has analysed the instrument smoothing for one dataset obtained with the CDE sensor system and found it to be insignificant as far as estimates of c^2 were concerned. For this reason no formal recommendation about assessing instrument smoothing is made here; nevertheless, if resources permit, the integrity of a dataset would be enhanced if a method could be devised whereby the response of the system to a known input could be measured during a run or very close in time to it.

Despite the meetings prior to the 10 May 1988 experiments, it was clear subsequently that the Brunei team underestimated the magnitude of the data analysis task. Although it had recognized in advance almost all of the steps that would be necessary, insufficient attention had been given to resourcing, particularly time. This could be made the subject of a formal recommendation. However, given the close connection that there ought to be between experimenters and analysts (and which is certainly possible in even greater degree between CDE, UMIST and Brunei) it is more helpful to make a recommendation of wider scope.

RECOMMENDATION 6: *Further research programmes using the CDE sensor system and involving more than one institution should be planned as a whole in more detail than hitherto. All collaborating teams have to be involved from the start. In particular more attention should be given to: (i) ensuring that the data (perhaps in conjunction with other data such as that analysed in the present Agreement) are adequate for achieving the stated objectives; (ii) realistic assessment of the resources needed for successful completion of all stages of the programme; (iii) guaranteeing that the resources are, or will be, available when required.*

This recommendation reinforces, and extends, the conclusions of the 7 October 1987 meeting that were discussed in section 3.2.

There is no doubt that it was correct to emphasize further studies of the pdf in the Agreement. Understanding the structure of p(θ;x,t) is the major scientific problem of atmospheric dispersion, indeed of turbulent diffusion. It has already been noted that Dr. N. Mole and Professor P.C. Chatwin were involved in an SERC-funded project from 1 February 1986 to 31 January 1989, i.e. over a three year period beginning about six months before the Agreement that is described in the present report. The SERC project was a theoretical one concerned with the effects of instrument smoothing on measured concentrations of dispersing contaminants. There was close liaison between the two projects; one outcome is:

> **RECOMMENDATION 7**: *In view of the pioneering status (worldwide) of research on the structure of p(θ;x,t), it is important to be aware of, and where necessary support, theoretical developments.*

It should be noted, however, that CDE has already made clear that it accepts this recommendation since it is now supporting a further Agreement (No.2066/71) with Brunel. This began on 1 February 1989 and is entitled *Fluctuations in Atmospheric Contaminants.* The Agreement provides the salary for three years of Dr. Mole who is working with Professor Chatwin; the CDE monitor is Dr. D.J. Ride. The overall aim is to develop and validate robust models for p(θ;x,t), and reference should be made to Mole and Chatwin (1990) where the encouraging progress during the first year is summarised.

4.3 Acknowledgements

Above all we wish to thank our CDE monitor Dr. Chris Jones who provided such advice and encouragement that it was sometimes difficult to believe that he was not working full-time at Brunei! Sterling help beyond the call of duty was provided throughout by Dr. Nils Mole and Dr. David Ride. We also gratefully acknowledge the invaluable help of Dr. Richard Griffiths and his team at UMIST, and of Robert Hamilton and Mark Wellham. Finally it is a pleasure to thank Mrs. Molly Demmar for achieving her usual high standard of work in typing this report.

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