RECURRENCE STATISTICS OF CONCENTRATION FLUCTUATIONS IN PLUMES WITHIN A NEAR-NEUTRAL ATMOSPHERIC SURFACE LAYER

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Abstract. This study examines the statistical properties of the concentration derivative, χ', for a dispersing plume in a near-neutrally stratified atmospheric surface layer. Towards this goal, the probability density function (pdf) of χ', and the conditional pdf of χ' given a fixed concentration level, χ, have been measured. These pdfs are found to be modeled well by a generalized q-Gaussian (gqG) distribution with intermittency exponent, q, equal to 0.3 and 3/4, respectively. These results highlight the strong intermittency effect (patchiness) of the small-scale concentration eddy structures in the plume. The distribution of time intervals between successive high peaks in the squared derivative process, χ'^2, is found to be well approximated by a power-law distribution, implying that occurrences of these high peaks are much more clustered than would be predicted by a Poisson or shot-noise process. The results are used to improve models for the joint pdf of χ and χ', and for the expected number of upcrossings per unit time interval of a fixed concentration level that have been proposed by Kristensen *et al.* (1989). The predictions of the improved models are in accord with observations, and suggest that the intercorrelation between χ and χ' must be explicitly incorporated if good estimates of the upcrossing intensity are to be obtained.

1. Introduction

The statistical properties of scalars dispersing in a turbulent boundary layer have been the subject of numerous experimental investigations in recent years (e.g., Fackrell and Robins, 1982; Deardorff and Willis, 1984; Hanna, 1984; Sawford *et al.*, 1985; Lewellen and Sykes, 1986; Sawford, 1987; Dinar *et al.*, 1988; Mylne and Mason, 1991; Wilson *et al.*, 1991; Bara *et al.*, 1992; Yee *et al.*, 1993). The results of these studies have provided conclusive evidence that the concentration fluctuations in a dispersing plume are of the same order of magnitude as the mean concentration itself. Consequently, for risk assessment of the consequences of the release of a toxic and/or flammable material into the atmosphere, there has been an increased emphasis in recent years to predict higher-order moments of the instantaneous concentration field (e.g., relative fluctuation intensity, skewness, and kurtosis), and even the single-point probability density function (pdf) of concentration (Wilson *et al.*, 1985; Kaplan and Dinar, 1988).

A statistical framework for the prediction of the effects and consequences of accidental releases of hazardous materials, based on the single-point pdf of concentration, has been described by Chatwin (1982). Whilst a knowledge of the concentration pdf enables a prediction of the probability that a certain critical threshold concentration is exceeded at any given instant of time, it does not provide information that can be used to predict the likelihood of the fluctuating concentration crossing a particular concentration level at least once within a specified time frame, or to predict the expected number of such level-crossings within this time frame (i.e., recurrence statistics). An understanding of recurrence statistics of concentration fluctuations is very beneficial to the assessment of the potential hazards from the release of toxic materials (both gases and aerosols) into the atmosphere. This is particularly so in the chemical and biological defence arena, where this information is required for the evaluation of the effectiveness of agent detectors and of protective equipment (e.g., respiratory protective devices, protective clothing, etc.).

Despite its importance to hazard assessment, no measurements of recurrence statistics of concentration fluctuations have been reported. Nevertheless, a model of the functional dependence of recurrence statistics on the fluctuation intensity of the dispersing scalar, on the instrument averaging time, and on various atmospheric surface-layer variables has been developed recently by Kristensen *et al.* (1989). However, this model depends on certain assumptions that have not been verified because of the lack, *a fortiori*, of measurements of recurrence statistics of concentration fluctuations. This study presents some observations of the pdf of concentration time derivative, and of the joint pdf of concentration and concentration derivative for a plume dispersing under near-neutral conditions in the atmospheric surface layer. This work represents an extension of previous experimental investigations which have focussed exclusively on the concentration pdf (e.g., Hanna, 1984; Lewellen and Sykes, 1986; Sawford, 1987; Dinar *et al.*, 1988; Mylne and Mason, 1991; Yee *et al.*, 1993). The objective is to use the information on the observed statistical properties of the concentration and concentration derivative to provide some guidance on the modeling of recurrence statistics of concentration fluctuations.

2. Rice's Theory and Recurrence Statistics

Let $\chi(t)$ denote the fluctuating concentration measured at a fixed point in a dispersing plume. We say that $\chi(t)$ exhibits an upcrossing of the constant level,

χ_L, at time t_0 if $\chi(t_0) = \chi_L$ and $\chi'(t_0) > 0$. Here, $\chi'(t)$ denotes the concentration time derivative. Similarly, we say that a χ_L-downcrossing occurs at time t_0 if $\chi(t_0) = \chi_L$ and $\chi'(t_0) < 0$. An excursion above the constant level, χ_L, is defined as an upcrossing followed by the next downcrossing of the given concentration level. For simplicity of notation, we use the same symbol to denote the random variable and a value assumed by the random variable.

The joint probability density function, $f(\chi, \chi')$, of the scalar concentration, χ, and its time derivative, χ', contains a great deal of information on the recurrence statistics of concentration fluctuations. Let $N(\chi_L, T)$ denote the number of χ_L-upcrossings of $\chi(t)$ in the interval $[0, T]$. To simplify notation, we write $N(\chi_L) \equiv N(\chi_L, 1)$ for the number of upcrossings of level χ_L in the unit interval $[0, 1]$ (viz., $T = 1$). Rice (1945) demonstrated that the expected (mean) number of upcrossings of the level χ_L by $\chi(t)$ per unit time, $\langle N(\chi_L)\rangle$, is given by

$$\langle N(\chi_L)\rangle = \int_0^\infty \chi' f(\chi_L, \chi')\, \mathrm{d}\chi' . \tag{1}$$

Applying the law of conditional probabilities, the joint pdf $f(\chi, \chi')$ can be expressed as the product $f(\chi, \chi') = f_1(\chi) f_{2|1}(\chi' \mid \chi)$, where $f_1(\chi)$ is the (marginal) pdf of concentration and $f_{2|1}(\chi' | \chi)$ is the conditional pdf of χ' given χ (viz., conditional on the concentration being fixed at some value χ). In consequence, Equation (1) can be written as

$$\begin{aligned}\langle N(\chi_L)\rangle &= f_1(\chi_L) \int_0^\infty \chi' f_{2|1}(\chi' \mid \chi_L)\, \mathrm{d}\chi' \\ &\equiv f_1(\chi_L) \langle \chi'^{+} \mid \chi_L \rangle ,\end{aligned} \tag{2}$$

where $\chi'^{+} \equiv \max(0, \chi')$ and $\langle \chi'^{+} \mid \chi_L \rangle$ is the expected (mean) positive slope of χ evaluated at the crossing level χ_L. Equation (2) implies that the mean upcrossing rate (or, equivalently, the mean excursion rate) of level χ_L can be calculated as the product of the concentration pdf evaluated at χ_L and the mean positive slope of $\chi(t)$ at χ_L (viz., the mean positive slope of $\chi(t)$ at the χ_L-upcrossings).

The determination of the mean number of upcrossings per unit time of an arbitrary constant level χ_L permits the computation of a number of other exceedance statistics of interest. For example, the return period, $R(\chi_L)$, between consecutive occurrences of upcrossings of a fixed level χ_L is given by

$$R(\chi_L) = 1/\langle N(\chi_L)\rangle . \tag{3}$$

Hence, $R(\chi_L)$ is simply the mean value of the random time between successive χ_L-upcrossings of the fluctuating concentration. The characteristic largest concentration value, χ_{T_s}, for a sampling period T_s, is defined as the (fixed) concentration level for which the mean number of exceedances of that level in the sampling period is unity. In consequence, the characteristic largest concentration value satisfies the equation

$$\langle N(\chi_{T_s})\rangle T_s = 1\,. \tag{4}$$

A knowledge of the characteristic largest concentration value, χ_{T_s}, as a function of the sampling (exposure) period, T_s, allows one to answer the question posed by Deardorff and Willis (1988): "How long must one wait in exposure to a pollutant before there is an even chance that a fluctuation in concentration exceeding a particular value will actually occur?" The mean duration of excursions, $\tau_{\chi L}$, above the fixed level χ_L is given by

$$\begin{aligned}\tau_{\chi L} &= \frac{1}{\langle N(\chi_L)\rangle}\int_{\chi L}^{\infty} f_1(\chi)\,\mathrm{d}\chi \\ &\equiv \frac{1 - F_1(\chi_L)}{\langle N(\chi_L)\rangle}\,,\end{aligned} \tag{5}$$

where $F_1(\chi)$ is the cumulative distribution function (cdf) of concentration.

Equation (2) can be simplified if the process $\chi(t)$ is such that

$$\langle \chi'^{+} \mid \chi_L\rangle = \langle \chi'^{+}\rangle\,. \tag{6}$$

The latter relationship implies that the mean positive slope of $\chi(t)$ is independent of the given fixed level χ_L. Assuming that $\chi(t)$ is a stationary process, it is straightforward to demonstrate that χ and χ' are uncorrelated random variables. However, this condition is not sufficient in general to ensure that the simplification provided by Equation (6) will hold. A stronger condition is required, namely that the conditional pdf of χ' given $\chi = \chi_L$ is identical to the pdf of χ', $f_2(\chi')$, viz.

$$f_{2|1}(\chi' \mid \chi_L) = f_2(\chi')\,. \tag{7}$$

However, Equation (7) is equivalent to the statement that χ and χ' are statistically independent random variables, so that the joint pdf of χ and χ' factors according to

$$f(\chi, \chi') = f_1(\chi) f_2(\chi')\,. \tag{8}$$

In consequence, the condition embodied in Equation (6) must be verified for at least one type of stationary process, namely a process in which χ and χ' are jointly Gaussian because, in this case, the fact that χ and χ' are uncorrelated is sufficient to guarantee the statistical independence of these two random variables.

Kristensen *et al.* (1989) assumed that χ and χ' are statistically independent in their formulation of a simple model for the recurrence statistics of concentration fluctuations. In addition, they assumed that the pdf of χ', $f_2(\chi')$, is Gaussian. At present, there are no observations which can be used either to support or to reject these assumptions. However, there is almost conclusive evidence to support the fact that the concentration pdf, $f_1(\chi)$, is positively skewed (i.e., it is non-Gaussian with a mode less than the mean), although there is no consensus at present whether the exact form of $f_1(\chi)$ for dispersing plumes is exponential, lognormal, gamma, clipped normal, etc. (e.g., Hanna, 1984; Lewellen and Sykes, 1986; Sawford, 1987; Mylne and Mason, 1991; Yee *et al.*, 1993). However, the strongly non-Gaussian form of the concentration pdf provides some circumstantial evidence that the conditional pdf of χ' given χ, is not independent of χ (viz., the conditional pdf of χ' given χ does not reduce to the (unconditional) pdf of χ'). With this motivation, we shall use some measurements of concentration fluctuations in a plume dispersing in a near-neutral atmospheric surface layer to try to address the following questions:

1. Is the pdf of the scalar derivative, χ', Gaussian? If not, what is the form of the pdf of χ'?
2. Is the pdf of χ' given the value of concentration χ, independent of χ? If not, what is the form of the conditional pdf of χ' given χ? What is the form of the joint pdf of χ and χ'?

3. Experimental Results

To test the hypotheses that χ and χ' are statistically independent and that the pdf of χ' is Gaussian, we use some data of concentration fluctuations in a dispersing plume obtained from a concentration fluctuation field test of chemical hazards (Biltoft, 1991; Yee *et al.*, 1993). This field test is part of an extensive program of field trials planned under the Tripartite Concentration Fluctuation Field Trials (TCFFT) project that involves the participation of a number of defence and industrial research organizations from the United States, Great Britain, and Canada. The test was conducted near Tower Grid on U.S. Army Dugway Proving Ground in September 1991, and involved the continuous release of a tracer gas (e.g., propylene (C_3H_6)) into the atmosphere from an elevated point source at a height, $h = 2.5$ m.

In all our experiments, propylene was released at a steady rate from a specially designed gas dissemination system, which consisted of 2 propylene cylinders connected in parallel and immersed in a hot water bath. A Matheson 1L-510 regulator was connected to the outlet of the cylinders to ensure a constant downstream pressure, and a flexible hose was used to connect the regulator to the inlet fitting of the mass flow controller (Teledyne Hastings-Raydist). This controller, which consisted of a sensor, electronic circuitry, a shunt, and a valve, was used to set, control, and measure the flow rate of gas through the dissemination system. The controller allowed a steady gas flow rate to be maintained at a user-selected reference level (between 1.67×10^{-4} and $1.67 \times 10^{-3}\ \mathrm{m^3\,s^{-1}}$) to within about 2%. Finally, a quick-release connector mated the outlet fitting of the mass flow controller to the dissemination hose, the latter of which was connected to the base of the disseminator, a section of schedule 40 PVC pipe 1 m in length and 0.05 m in diameter. These dimensions were chosen to ensure an approximate parabolic velocity profile at the exit of the dissemination system, with a mean velocity less than about 1 m s^{-1} at the maximum flow rate of 1.67×10^{-3} m^3 s^{-1}. This allowed the tracer gas to be released isotropically from the source without forming a jet.

In this study, we use time series of fluctuating concentrations measured in dispersing plumes with a crosswind array of modified fast-response photoionization detectors (designed and constructed by S & J Engineering, Inc.; see, Chandler (1991)). The frequency response (-6 dB point) of the detectors is approximately 100 Hz. It can be shown that this response is sufficient to resolve the fluctuation variance contributed by the energetic subrange and most of the inertial-convective subrange, with a reduction in the fluctuation variance due to instrument smoothing of the finest scales (e.g., scales in the dissipation subrange at which molecular diffusion becomes important) of at most 4% (Yee *et al.*, 1993). However, our detectors are not sufficiently responsive to resolve the fine-scale structure in the dissipation subrange (i.e., Kolmogorov microscale-size concentration eddies). Consequently, some of the small-scale plume statistics to be presented may be specific to our instrument which possesses the following low-pass frequency response function:

$$H(n) = \frac{1}{(1 + 2\pi j n \beta)^{\alpha}}, \tag{9}$$

where n is frequency (Hz), $j \equiv \sqrt{-1}$, β is a scale parameter whose value is approximately 0.00109 s, and α is a shape parameter whose value is approximately 3.85. The analog signals from the detectors were fed into a computer interface containing a fast sample-and-hold A/D converter with 16-bit resolution (Keithley Metrabyte, DAS-HRES) operating at a sampling rate of 1000 Hz.

The data for the present study were carefully selected according to two criteria. Firstly, we selected data from field experiments that were conducted under near-neutral stratification only. Secondly, we searched for long uninterrupted high signal-to-noise ratio (SNR) time series with a sampling time of 16 minutes or greater that were statistically stationary over the sampling period. We found a number of time series that satisfied these two criteria. These time series corresponded to measurements at various positions in a crosswind cross-section through the dispersing plume at three downwind distances (e.g., at $x = 25$, 50, and 100 m, or, equivalently, at $x/h = 10.0$, 20.0, and 40.0) at a height of 3 m above the ground. Each of these time series was corrected for any discernible baseline drift, the DC offset was removed, and the resulting sequence was clipped to a concentration threshold. The latter was determined from a visual inspection of all the background noise segments in the time series.

Concentration time derivatives were obtained from the samples of $\chi(t)$ by a two-step procedure: (1) least-squares fitting a polynomial of degree M, using n_L and n_R points in the data sequence to the left and right, respectively, of the given time instant at which the derivative is required; and, (2) differentiating the resulting fitted polynomial to give the estimated time derivative of concentration at the

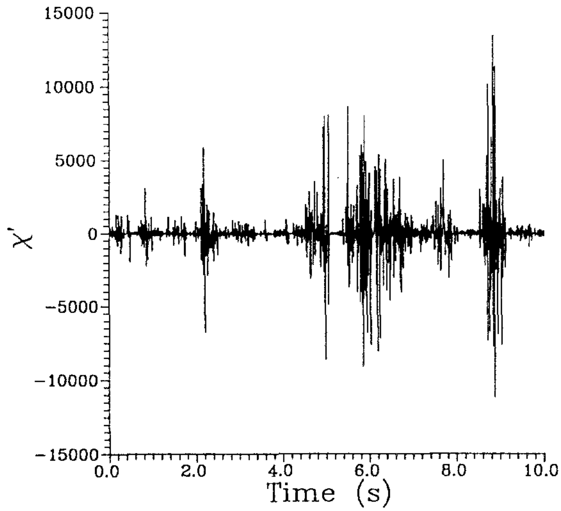

Fig. 1. An example of 10 s of a concentration derivative time series of in-plume fluctuations illustrating the strong intermittency of the small-scale concentration eddies in the plume.

given time instant. We note that the first step of this procedure serves to smooth the time series in order to reduce the effects of measurement noise. The two-step procedure for obtaining the concentration time derivative was implemented in the form of a Savitzky-Golay least-squares filter (Press and Teukolsky, 1990). This filter provides a local least-squares polynomial fit within a moving window that is sequentially centered around each data point at which a derivative is required.

Figure 1 shows a 10 s segment of the time derivative of a conditionally sampled (in-plume) concentration time series (i.e., zeros ignored) computed with $n_L = n_R = 2$ and $M = 2$. The intervals of zero concentration arise primarily from meandering of the plume and, as such, contribute only zero to the concentration derivative time series. In consequence, the intervals of zero concentration represent superfluous information once the intermittency factor, γ, defined as the total time that non-zero concentrations are observed, is known. Unless otherwise indicated, all the statistics in the present study are determined from the conditionally sampled concentration time series. One interesting feature of the concentration derivative time series (cf. Figure 1) is the conspicuous intermittent nature of the process that occurs on a time scale of fractions of a second. Note the occurrence of bursts of high-frequency fluctuations in the concentration derivative. These bursts provide strong visual evidence of the small-scale intermittency of the in-plume fluctuations. We note that peak values of $\chi(t)$ are about 10 times the conditional mean concentration, C_p, whereas peak values of $|\chi'(t)|$ are about 1000 times the conditional mean value of $|\chi'(t)|$. This is not surprising because the concentration derivative emphasizes the information associated with the smallest scales of the dispersing plume.

The concentration derivative is a two-point statistic of the dispersing plume that reflects the information on the dynamics of the small-scale mixing processes in the plume. This can be seen by noting that

$$\frac{\partial \chi}{\partial x} = -\frac{1}{U}\frac{\partial \chi}{\partial t} = \lim_{\delta x \to 0} \frac{\chi(x + \delta x) - \chi(x)}{\delta x}, \tag{10}$$

where we have used Taylor's frozen flow hypothesis to interpret time intervals δt as space separations, $\delta x = -U\delta t$, in the alongwind direction x (U is the mean windspeed). Hence, the concentration derivative emphasizes the information on the nature of the small-scale structures in the dispersing plume. An examination of the concentration-difference (increment) time series, $\chi(x + \delta x) - \chi(x)$, shows that the intermittency becomes increasingly stronger at the smaller scales (viz., as the separation distance, δx, which is indicative of the scale of concentration eddies considered, is reduced). This is reflected in the tendency of the concentration-difference statistics to become increasingly non-Gaussian with decreasing scale, δx.

The derivative, χ', of concentration fluctuations exhibited in Equation (10) is easy enough to write down, but this formulation raises a basic problem – namely, what do we mean by the derivative of $\chi(t)$? The derivative makes sense only if $\chi(t)$ is assumed to vary continuously in time (or, equivalently, in space if we apply Taylor's frozen field hypothesis). The latter requires the dispersing scalar field to be regarded as a continuum (viz., the limit $\delta x \approx \delta V^{1/3} \to 0$, where δV represents an incremental volume of the scalar field, is interpreted as a macroscopic limiting value, and the derivative associated with this limiting operation involves taking differences of the instantaneous concentration as δx (or, δt) becomes infinitesimally small compared to the field dimensions, but not so small as to be influenced by the individual gas molecules themselves). In other words, the limiting value in Equation (10) is confined to scales that are macroscopically small (i.e., much less than distances characteristic of the fine-scale and high-frequency structure of the scalar field in the continuum, the latter of which is determined by the Batchelor length, λ_B ($\approx 10^{-3}$ m)), but microscopically large (i.e., much greater than distances characteristic of the mean free path, λ_P, between gas molecules ($\approx 10^{-9}$ m)). Hence, the limit as $\delta x \to 0$ is interpreted as an outer region (continuum) limit meaning $\delta x/\lambda_B \to 0$. However, the spatial and/or temporal resolution achieved by our concentration sensors with frequency response characteristics described by Equation (9), is not sufficiently responsive to resolve the smallest scales (e.g., those responsible for smearing by molecular action) present in the plume. Since it is these small scales that contribute most to the concentration derivative, it is expected that the derivative statistics are strongly dependent on instrument resolution. In this regard, all concentration fluctuation statistics (e.g., pdf of concentration derivative, recurrence statistics, etc.) should strictly be described in terms of the temporal (or frequency) resolution of the instrument with which $\chi(t)$ is mea-

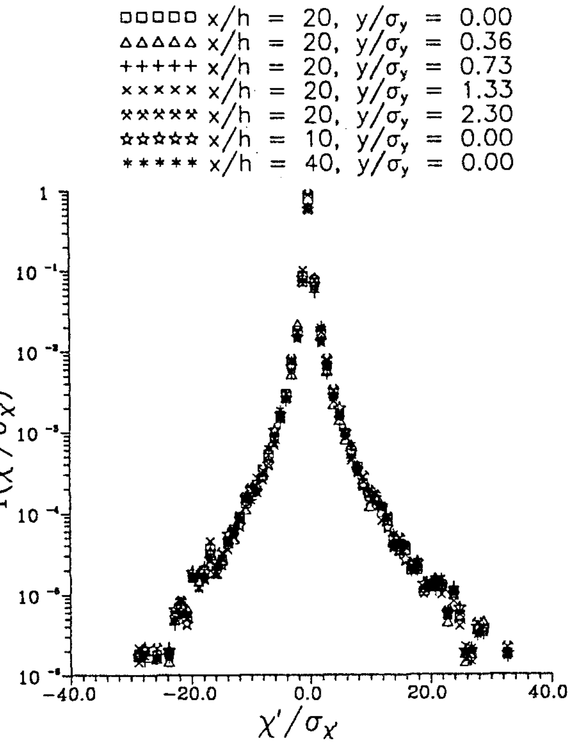

Fig. 2. Probability density function of normalized concentration derivative, $\chi'/\sigma_{\chi'}$, measured at a number of crosswind positions, y/σ_y, and downwind distances, x/h, in the dispersing plume under near-neutral stability conditions.

sured. In what follows, the concentration fluctuation statistics presented are appropriate for the frequency resolution of our concentration sensors. The effect of instrument smoothing on recurrence statistics is explored further in Appendix A.

Figure 2 shows the pdfs of the concentration time derivative, χ', normalized by the corresponding standard deviation of the derivative, $\sigma_{\chi'}$. The pdfs have been measured at various positions in the lateral cross-section of a dispersing plume at three downwind distances. These pdfs exhibit a remarkable degree of universality in their form over a wide range of positions in the dispersing plume. We note that the pdfs of concentration derivative appear to exhibit a higher degree of self-similarity (universality) in their form than do the corresponding pdfs of concentration (e.g., cf. with concentration pdfs in Yee *et al.*, 1993). Because the concentration derivative pdf emphasizes the information on the distribution of the small concentration eddies in the plume, the latter observation suggests that the dynamics important in determining the distribution (i.e., statistics) of the small-scale structures in the plume approach self-similarity much sooner than the plume as a whole. Indeed, the plume evolves toward a self-similar state only when the

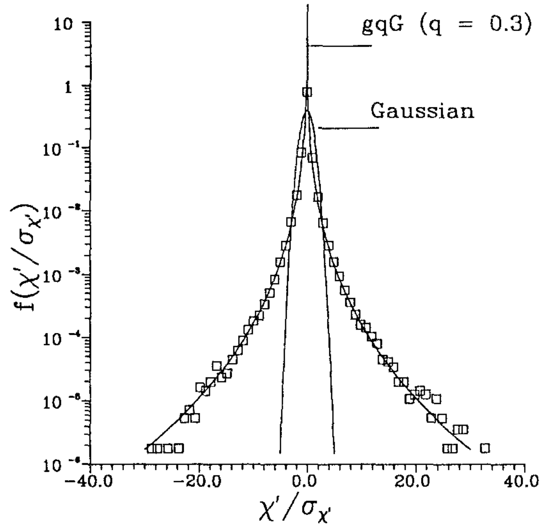

Fig. 3. Probability density function of normalized concentration derivative, $\chi'/\sigma_{\chi'}$, compared with the standard Gaussian distribution and the generalized q-Gaussian (gqG) distribution ($q = 0.3$).

instantaneous plume has grown to fill the mean-plume width (turbulent-diffusive regime), but the self-similarity of the small scales in the plume appear to be established well before this stage of plume development.

In contrast to the concentration pdfs which are strongly positively skewed, the pdfs of concentration derivative are remarkably symmetric. Figure 2 demonstrates clearly the remarkable symmetry in the concentration derivative probability distribution, a result that is consistent with a visual inspection of the time trace of χ' shown in Figure 1. Furthermore, the concentration derivative probability distribution possesses heavy lower and upper tails. In this regard, we see that values of the concentration derivative greater than 10 times the standard deviation are observed with a probability of about 7×10^{-4}, which is about 7 and 10^{20} times larger than what would have been predicted by the Laplacian (i.e., two-sided exponential) and Gaussian distributions, respectively.

The strong departure of the concentration derivative from Gaussian behavior is clearly illustrated in Figure 3 which compares the sample pdf, $f(\chi'/\sigma_{\chi'})$, of the normalized derivative with the standard Gaussian distribution. The probability of very small values of $|\chi'|$ or of values of $|\chi'|$ greater than about 3.5 standard deviations is larger than what would be expected from a Gaussian distribution, whereas the probability of occurrence of the intermediate values is smaller. The mode of the concentration derivative pdf occurs at zero, implying that the derivative series is dominated by small values of the derivative near zero. This fact is consistent with a visual inspection of the concentration derivative time series exhibited in Figure 1. The concentration derivative pdf exhibits wider and more flared out "skirts" than a Gaussian pdf, emphasizing the dominant effect of the intermittency of small-scale structures in the plume.

We have fitted a "stretched" Gaussian, $f(\chi'/\sigma_{\chi'}) \sim \exp(-\lambda|\chi'/\sigma_{\chi'}|^q)$, to the sample pdf of the normalized concentration derivative. The normalization for the "stretched" Gaussian was chosen so that the functional form reduces to that of the standard Gaussian when the "stretching" exponent, q, is equal to 2. In consequence, the following *ansatz* was chosen as a model for the "stretched" Gaussian pdf:

$$f(\chi'/\sigma_{\chi'}) = \frac{\tilde{\alpha}}{\sigma_{\chi'}} \exp\left\{ -\lambda \left| \frac{\chi'}{\sigma_{\chi'}} \right|^q \right\}, \tag{11}$$

where

$$\alpha \equiv \frac{\tilde{\alpha}}{\sigma_{\chi'}} = \frac{q}{2\Gamma(1/q)[\Gamma(1/q)/\Gamma(3/q)]^{1/2}\sigma_{\chi'}}, \tag{12}$$

$$\lambda \equiv \left[\frac{\Gamma(3/q)}{\Gamma(1/q)}\right]^{q/2}, \tag{13}$$

and $\Gamma(x)$ is the gamma function. We refer to the "stretched" Gaussian pdf exhibited in Equations (11) to (13) as the generalized q-Gaussian (gqG) pdf. Clearly, the gqG pdf is symmetric, and reduces to the standard Gaussian and Laplacian (i.e., two-sided exponential) pdfs when $q = 2$ and 1, respectively.

Figure 3 compares the sample concentration derivative pdf and the fitted gqG pdf with q determined to be 0.3. Although this gqG pdf ($q = 0.3$) overestimates the sample pdf for concentration derivatives near zero, it, nevertheless, provides a very good fit to an extensive region of the pdf corresponding to the lower and upper tails (viz., for regions of the pdf extending up to about 30 times the standard deviation of the concentration derivative). The "stretching" exponent, q, can be interpreted as a measure of the degree of intermittency of the fluctuations (viz., the smaller the value of q, the more intermittent are the underlying fluctuations). Clearly, Figure 3 provides quantitative evidence in support of a substantial intermittency effect of the small-scale structures in the dispersing plume. This result must be a consequence of the intermittency of the multiplicative process associated with the eddy cascade of both the turbulent energy and the concentration fluctuation variance. Hence, the concentration derivative pdf, which depends strongly on the characteristics of the smaller scales (i.e., smaller concentration eddies), is found to deviate considerably from both a Gaussian and an exponential law.

Next, we investigate the conditional statistics of concentration derivative, χ', given the value of the concentration, χ. Towards this objective, we determine the rms of the normalized concentration derivative, $\xi \equiv \chi'/\sigma_{\chi'}$, conditioned on a given normalized concentration value, χ/C_p (viz., we determine the functional form of $\sigma_\xi(\chi/C_P) \equiv (\langle\chi'^2/\sigma_{\chi'}^2 \mid \chi/C_p\rangle)^{1/2}$, which provides an important indicator of the

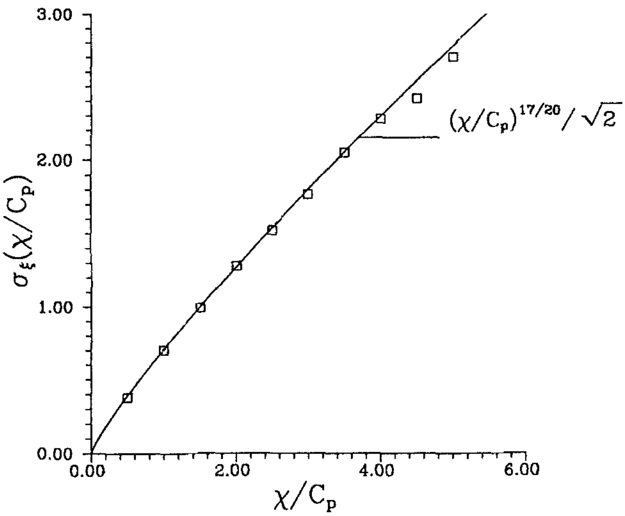

Fig. 4. Conditional standard deviation (root-mean-square (rms)), σ_ξ, of normalized concentration derivative, $\xi \equiv \chi'/\sigma_{\chi'}$, given a fixed normalized concentration level, χ/C_p. The solid curve is a power-law fit to the conditional rms fluctuation in the normalized concentration derivative.

interdependence between χ' and χ). Figure 4 displays the conditional standard deviation, $\sigma_\xi(\chi/C_p)$, as a function of the normalized concentration, χ/C_p. The data in Figure 4 were measured near the mean-plume centerline at $x/h = 20.0$. Measurements at other lateral and/or downwind positions in the plume, not displayed here, show similar behavior (viz., there were no systematic differences between the various measurements, only a dissimilarity in the degree of scatter, which appears to be larger for points measured near the plume fringes).

Figure 4 demonstrates that the statistical independence assumption between χ and χ' is too strong – this assumption would imply that $\sigma_\xi(\chi/C_p) \equiv 1$ for all values of the normalized concentration, χ/C_p. However, we see that $\sigma_\xi(\chi/C_p)$ is not a constant function of χ/C_p, implying that χ' is statistically dependent on the concentration, χ. To account for the correlation between these two variables, we use a power-law relationship of the form $\sigma_\xi(\chi/C_p) = a(\chi/C_p)^b$, where a and b are undetermined parameters. Accordingly, the solid line in Figure 4 represents the power-law form $a(\chi/C_p)^b$, which has been least-squares fitted to the measured conditional standard deviation, $\sigma_\xi(\chi/C_p)$. We find that $\sigma_\xi(\chi/C_p)$ exhibits a power-law dependence that can be described well with $a = 1/\sqrt{2}$ and $b = 17/20$ (at least over the range of the values of normalized concentration observed). The number of χ/C_p-upcrossing and downcrossing events decreases roughly exponentially with the normalized concentration level. In consequence, it is difficult to estimate accurately the conditional standard deviation, $\sigma_\xi(\chi/C_p)$, for large values of χ/C_p, where the upcrossing and downcrossing events of such levels become increasingly rare. We expect $\sigma_\xi^2(\chi/C_p) \equiv \langle \chi'^2/\sigma_{\chi'}^2 \mid \chi/C_p \rangle$ to vanish at the extremal points (i.e.,

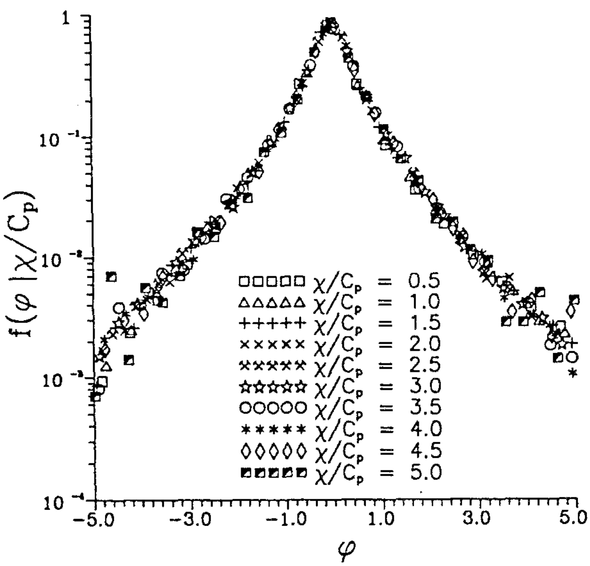

Fig. 5. Conditional probability density function of normalized concentration derivative, $\phi \equiv \xi/\sigma_\xi(\chi/C_p)$, where $\xi \equiv \chi'/\sigma_{\chi'}$, determined for a number of fixed normalized concentration levels, χ/C_p.

minimum and maximum values) of the concentration because the concentration derivative must always be zero there. The power-law form for $\sigma_\xi(\chi/C_p)$ verifies this constraint at the lower bound for concentration (viz., at zero concentration). However, the power-law form increases monotonically and, in consequence, we expect the power-law form for $\sigma_\xi(\chi/C_p)$ to break down eventually as the maximum in the normalized concentration is approached. Physically, χ/C_p has an upper bound, and we expect $\sigma_\xi(\chi/C_p)$ to decrease to zero again as this upper bound is approached because the derivative of χ/C_p evaluated at its maximum value is always identically zero.

Figure 5 displays the pdf of the normalized derivative, $\phi \equiv \xi/\sigma_\xi(\chi/C_p)$, conditioned on a fixed normalized concentration value, χ/C_p. Note that ϕ is the nondimensional derivative, $\xi \equiv \chi'/\sigma_{\chi'}$, evaluated at the χ/C_p-upcrossings and scaled by the conditional standard deviation of ξ, $\sigma_\xi(\chi/C_p)$. We see that the conditional pdfs, $f(\phi \mid \chi/C_p)$, at each of the given values of χ/C_p are symmetric. Furthermore, these pdfs are seen to collapse remarkably well onto one universal form, indicating that the normalized derivative, ξ, scaled by the conditional standard deviation of ξ, $\sigma_\xi(\chi/C_p)$, is a proper similarity variable. The functional dependence of the conditional standard deviation on the normalized concentration value is displayed in Figure 4. Although the data used to construct Figure 5 were measured at the mean-plume centerline at $x/h = 20.0$, we found that data measured at other locations in the plume (viz., at different lateral positions and/or different downwind distances) show similar behavior. The high degree of universality exhibited by the

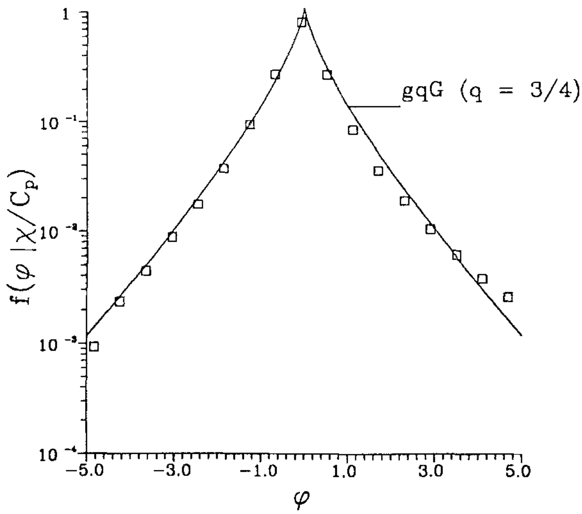

Fig. 6. Conditional probability density function of normalized concentration derivative, $\phi \equiv \xi/\sigma_\xi(\chi/C_p)$, where $\xi \equiv \chi'/\sigma_{\chi'}$, compared with the generalized q-Gaussian (gqG) distribution ($q = 3/4$).

conditional pdf, $f(\phi \mid \chi/C_p)$, is rather remarkable. As in the case for the pdf of ξ (cf. Figure 3), the conditional pdf of ϕ is seen to exhibit considerably longer tails than the bell-shaped curve of a Gaussian pdf. We found that the conditional pdf of ϕ can be approximated well by a gqG distribution (cf. Equations (11) to (13)) with intermittency exponent, $q = 3/4$, as is shown in Figure 6, implying that the tails of the pdf fall off more slowly than even the exponential form.

As noted earlier, the concentration derivative emphasizes the relevant smaller eddy sizes (scales) in the dispersing plume. The pdf (both unconditional and conditional) of the concentration derivative provides information on the non-uniformity of the spatial structure of the concentration field on the smaller scales and, in consequence, reflects the distribution and structure of these smaller concentration eddies. This small-scale eddy structure is seen to exhibit a strong tendency towards universality, as reflected in the self-similarity of the unconditional concentration derivative pdfs across the mean-plume lateral cross-section, and in the collapse of the conditional concentration derivative pdfs onto a universal curve. Both the unconditional and conditional concentration derivative pdfs correspond to rather peaked intermittent distributions with intermittency exponents q less than unity (viz., the tails of these distributions are heavier than those of the exponential distribution). This is indicative of the strongly intermittent nature of the small concentration eddies (viz., the eddies at smaller and smaller scales become less and less space-filling), and this physical effect is manifested in the strong small-scale intermittency exhibited by the concentration derivative. To investigate this small-scale concentration eddy structure in greater detail, we deter-

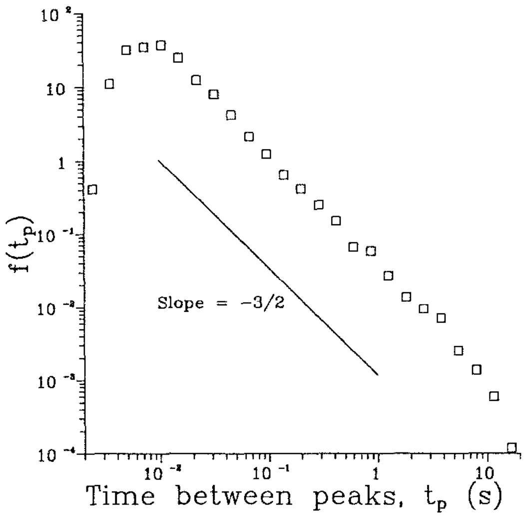

Fig. 7. Probability density function of the time interval, t_p, between high peak values in the normalized squared concentration derivative time series, $\chi'^2/\langle\chi'^2\rangle$. For time intervals between peaks greater than about 0.01 s, $f(t_p)$ can be well approximated with a power-law distribution with $f(t_p) \sim t_p^{-3/2}$.

mine the probability distributions of the time interval, t_p, between high peak values in the normalized squared concentration derivative, $\chi'^2/\langle\chi'^2\rangle$, and the temporal width, t_w, of these high peaks.

The pdf, $f(t_p)$, of the time interval between the occurrence of high peak values in the normalized squared concentration derivative signal, is displayed in Figure 7. Here, we define high peak values to have an amplitude, χ'^2, that is 25 times the mean value, $\langle\chi'^2\rangle$. In consequence, a stretch of data corresponding to the high peak values is a set of ordered numbers $\tau_1 < \tau_2 < \cdots < \tau_K$ associated with the times of upcrossing events of the fixed level 25 in the $\chi'^2/\langle\chi'^2\rangle$ process measured over the sampling time, T_s. The pdf, $f(t_p)$, was constructed by histogramming the elapsed time between these upcrossing events and normalizing the resulting bin counts by bin interval and sample size. The expected time between high peak values in χ'^2 is about 0.266 s, which corresponds to a mean occurrence rate of about 3.75 s^{-1}. The mode of the pdf of the time interval between the occurrences of high peak values is seen to be at about 0.01 s. For time intervals between high peaks greater than about 0.01 s, $f(t_p)$ appears to follow a power-law distribution with $f(t_p) \sim t_p^{-r}$, where the exponent r is 3/2. In the turbulent-convective regime of plume development, the exponent r maintains an almost constant value across the mean-plume lateral cross-section.

Figure 7 illustrates clearly that the time intervals between the high peaks in the squared derivative process, χ'^2, are not Poisson distributed in time (viz., the stochastic point process corresponding to the times of high peak events in χ'^2 is not a shot-noise process). To see this, we note that if the times of occurrence of the high peak events are distributed uniformly in time such that the probability of occurrence of a peak is independent of the preceding one, then the probability of occurrence of peaks is described by the well-known Poisson distribution,

$$P_{T_s}(n) = \frac{(\Lambda T_s)^n \exp(-\Lambda T_s)}{n!}, \quad n \in N, \tag{14}$$

where $P_{T_s}(n)$ is the probability of observing n high peaks in sampling time T_s and $\Lambda > 0$ is the mean number of occurrences of peaks per unit time (viz., the mean rate of peak occurrence). The mean and variance of the distribution, $P_{T_s}(n)$, are equal to ΛT_s. For Poisson-distributed events, the time intervals between events are distributed exponentially:

$$f(t_p) = \frac{1}{\Lambda} \exp\left(-\frac{t_p}{\Lambda}\right), \quad t_p \in \mathbf{R}^+ . \tag{15}$$

However, the distribution of the time intervals between high peaks is poorly approximated by an exponential (cf. Equation (15)) with parameter Λ set to the observed mean occurrence rate for high peaks. The exponential distribution greatly underestimates the pdf of the interval, t_p, between successive high peaks for t_p less than about 0.5 s, and overestimates the pdf for t_p greater than about 0.5 s. This implies that the probability of occurrence of high peaks in χ'^2 cannot be considered to be statistically independent – rather, the high peaks tend to occur in localized clusters (cf. Figure 1). This implies that small-scale eddy structures in the concentration field are not space-filling.

The sample pdf, $f(t_w)$, of peak width, t_w, is displayed in Figure 8. Here, the peak width, t_w, is defined as the time interval between an upcrossing and the next downcrossing of the threshold value used to define high peaks in χ'^2 (e.g., $\chi'^2/\langle\chi'^2\rangle = 25$). We found an expected peak width of about 0.006 s. It is interesting to note that the expected peak width is comparable to the Taylor microscale, t_χ, of concentration fluctuations. Here, t_χ was found to be about 0.015 s, which we determined from the definition $t_\chi^2 \equiv \sigma_\chi^2/\sigma_{\chi'}^2$ with the zero periods included (viz., all points in the total time series were used to determine t_χ^2). Interestingly, the definition of t_χ coincides with the inverse of the mean frequency, ν, of the concentration fluctuations: $\nu \equiv \{\int_0^\infty \omega^2 S(\omega)\, d\omega / \int_0^\infty S(\omega)\, d\omega\}^{1/2} = (\sigma_{\chi'}^2/\sigma_\chi^2)^{1/2}$, where ω is angular frequency and $S(\omega)$ is the power spectral density function for concentration fluctuations. This suggests that the width of the high peaks in the squared concentration derivative field determines the Taylor concentration microscale (or, equivalently, the mean frequency of the process). These high peaks are probably the result of the turbulent stretching effect of the velocity field (which operates on the time scale $\tau_S \sim \sigma_i/U$, where σ_i is the instantaneous plume width and U is the wind speed), which progressively draws out the plume material into contorted sheets and strands (Chatwin and Sullivan, 1979). This serves to break the concentration eddies into smaller and smaller sizes resulting in the development of large

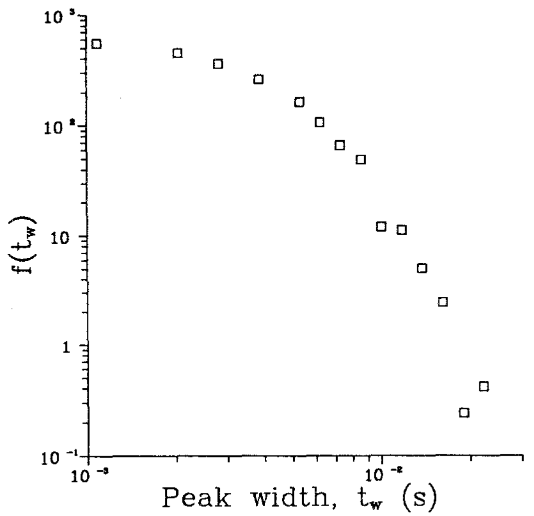

Fig. 8. Probability density function of the width, t_w, of high peak values in the normalized squared concentration derivative time series, $\chi'^2/\langle\chi'^2\rangle$.

concentration gradients in the plume that enhance the action of molecular mixing. The distribution of peak widths, $f(t_w)$, is indicative of the range of different speeds and orientations of small-scale concentration eddy structures in the dispersing plume that are advected past the detector. The intervals corresponding to the peak widths are associated with those time periods where large scalar dissipations are occurring in the dispersing plume – these periods probably coincide with the passage of the high-gradient shear zone regions between the small-scale concentration eddy structures. The intervals between successive high peaks correspond to periods of inactivity in χ'^2 and, as such, coincide with the passage of concentration eddy structures of varying sizes.

4. Applications

Our measurements of the statistics of the concentration derivative can be used to refine the model for recurrence statistics of concentration fluctuations developed by Kristensen *et al.* (1989). The important result is that the conditional pdf of the concentration derivative, χ', conditioned on a fixed value of the concentration, χ, exhibits a strong dependence on χ. It is unclear how the dependence of χ' on χ arises, but it seems plausible to identify two causes: (1) the correlation introduced by the turbulent (random) velocity field that is responsible for the turbulent stretching and diffusion of the scalar; and, (2) the correlation between the scalar and its gradient that is imposed by the source (release) conditions. The intercorrelation between χ and χ' implies that the (marginal) pdf of concentration derivative cannot be used in Rice's formula for the expected number of upcrossings of a fixed level.

Rice's formula for the expected number of upcrossings in a fixed time interval requires a knowledge of the joint probability density function of the concentration, χ, and its derivative, χ' (cf. Equation (1)). The joint pdf of χ and χ', $f(\chi, \chi')$, provides information on the probability that χ and χ' jointly lie in a given interval, and can be determined in terms of the marginal pdf of χ, $f_1(\chi)$, and the conditional pdf of χ' given χ, $f_{2|1}(\chi' \mid \chi)$, as follows:

$$f(\chi, \chi') = f_1(\chi) f_{2|1}(\chi' \mid \chi) . \tag{16}$$

Equation (16) requires that we model both $f_1(\chi)$ and $f_{2|1}(\chi' \mid \chi)$ if we are to determine the upcrossing intensity in accordance with Equation (1).

Although there is no agreement on the "correct" form of the pdf of χ, it was found in Yee *et al.* (1993) that under near-neutral conditions, the lognormal and gamma distributions provide good approximations to the in-plume concentration pdfs in the turbulent-convective and turbulent-diffusive regimes of plume development, respectively. To provide a concrete example, we consider points in the plume measured in the turbulent-convective regime of plume development. In consequence, we use the lognormal distribution as the model pdf for the in-plume concentration fluctuations. Hence, we choose the marginal pdf of χ to have the following form:

$$f_1(\chi) = \frac{1}{\sqrt{2\pi}\sigma\chi} \exp\left(- \frac{\ln^2(\chi/m)}{2\sigma^2} \right), \tag{17}$$

where

$$m = \frac{C_p}{\sqrt{i_p^2 + 1}}, \tag{18}$$

and

$$\sigma^2 = \ln(i_p^2 - 1) . \tag{19}$$

Here, C_p and i_p are the conditional (in-plume) mean and fluctuation intensity of concentration fluctuations, respectively. As a model for the conditional pdf of $\chi'/\sigma_{\chi'}$ given χ/C_p, the measurements presented in the previous section suggest that we could use a gqG distribution with the following form:

$$f_{2|1}(\xi \mid \chi/C_p) = \frac{\tilde{\alpha}}{\sigma_\xi(\chi/C_p)} \exp\left\{ -\lambda \left| \frac{\xi}{\sigma_\xi(\chi/C_P)} \right|^q \right\}, \tag{20}$$

where $\xi \equiv \chi'/\sigma_{\chi'}$, $\tilde{\alpha}$ and λ are normalization constants defined in Equations (12) and (13), respectively, and the intermittency exponent, $q = 3/4$. Furthermore,

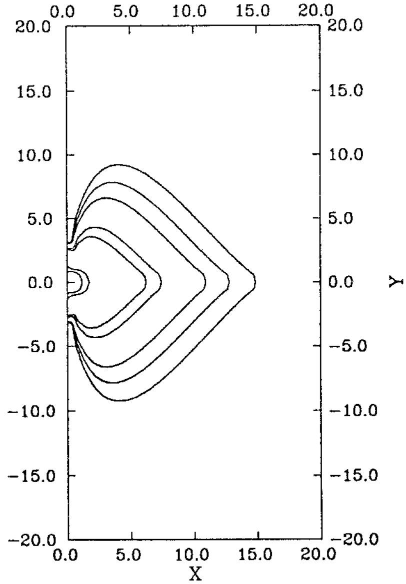

Fig. 9. Model joint probability density function between normalized concentration, $X \equiv \chi/C_p$, and normalized concentration derivative, $Y \equiv \chi'/\sigma_{\chi'}$. Contour levels (inner to outer) are 0.1, 0.05, 0.001, 0.005, 0.0001, 0.00005, and 0.000025, respectively.

$$\sigma_\xi(\chi/C_p) = \frac{1}{\sqrt{2}}\left(\frac{\chi}{C_p}\right)^{17/20}, \tag{21}$$

is the conditional standard deviation of the normalized derivative, $\chi'/\sigma_{\chi'}$, for a given value of the normalized concentration, χ/C_p.

The model joint pdf, $f(\chi, \chi') = f_1(\chi) f_{2|1}(\chi' \mid \chi)$, with $f_1(\chi)$ and $f_{2|1}(\chi' \mid \chi)$ determined according to Equations (17) to (21) is exhibited in Figure 9. The joint pdf shown here has been nondimensionalized with the mean and standard deviation of χ and χ', C_p and $\sigma_{\chi'}$, respectively (i.e., $X \equiv \chi/C_p$ and $Y \equiv \chi'/\sigma_{\chi'}$). The contour levels shown (inner to outer) correspond to 0.1, 0.05, 0.001, 0.005, 0.0001, 0.00005, and 0.000025, respectively. The parameters for $f_1(\chi)$ (viz., m and σ defined in Equations (18) and (19)) have been selected to match the measured values of conditional mean, C_p, and fluctuation intensity, i_p, of the concentration time series whose (in-plume) derivative is displayed in Figure 1 (measured at lateral position $y/\sigma_y \approx 0.36$ at downwind distance $x/h = 20.0$). In consequence, the model joint pdf of Figure 9 can be directly compared with the observed joint pdf of χ and χ' for the concentration time series associated with Figure 1. Isojoint pdf contours of $X \equiv \chi/C_p$ and $Y \equiv \chi'/\sigma_{\chi'}$ for this time series are shown in Figure 10. The contour levels shown in Figure 10 (inner to outer) correspond to 0.5, 0.1, 0.05, 0.001, 0.005, 0.0001, 0.00005, and 0.000025, respectively, and with the exception of the innermost contour level (i.e., 0.5) can be compared directly with those in

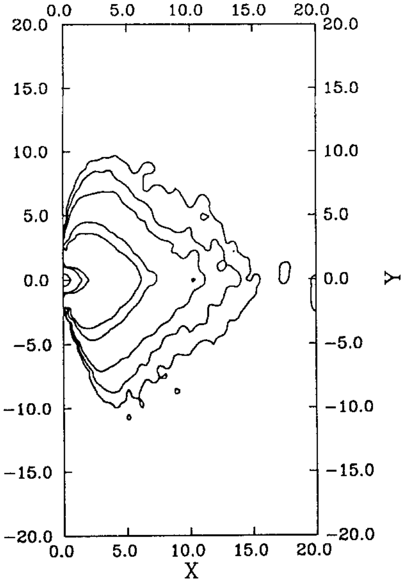

Fig. 10. Measured joint probability density function between normalized concentration, $X \equiv \chi/C_p$, and normalized concentration derivative, $Y \equiv \chi'/\sigma_{\chi'}$. Contour levels (inner to outer) are 0.5, 0.1, 0.05, 0.001, 0.005, 0.0001, 0.00005, and 0.000025, respectively.

Figure 9. Such a comparison shows that the model joint pdf (cf. Equations (16) to (21)) reproduces the dominant features in the observed joint pdf. Both show contour shapes that approximately resemble parallelograms. There is a high probability that small values of concentration, χ, are associated with small values of absolute concentration derivative, $|\chi'|$. Furthermore, there is greater probability for large (small) concentrations to occur together with small (large) concentration derivatives than for large concentrations and concentration derivatives to occur simultaneously. In consequence, although χ and χ' are uncorrelated random variables, their joint pdf suggests a dependence between two variables where the activity of one appears to suppress that of the other.

The model joint pdf of χ and χ', described by Equations (16) to (21), can be used in Rice's formula to determine the expected number of upcrossings (or, equivalently, excursions) of a fixed concentration level in a particular sampling period, T_s. Hence, the mean number of χ/C_p-upcrossings per unit time of the normalized concentration is given by (cf. Equations (2), (17), (20), and (21))

$$\langle N(\chi/C_p)\rangle = \frac{1}{2}\frac{\Gamma(2/q)}{[\Gamma(1/q)\Gamma(3/q)]^{1/2}}\,\sigma_{\chi'/C_P}(\chi/C_p)f_1(\chi/C_p)\,, \tag{22}$$

where

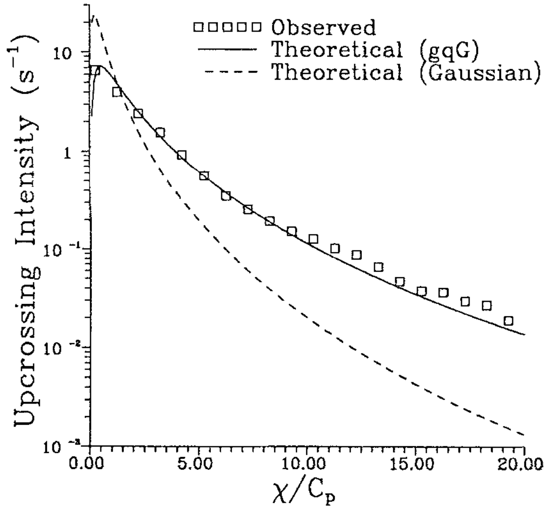

Fig. 11. Comparison between the experimental data for the upcrossing intensity over normalized concentration level, χ/C_p, and the theoretical result given by Rice's formula using the generalized q-Gaussian (gqG) distribution as a model for the conditional probability density function of concentration derivative. Also shown is the theoretical result given by Rice's formula using the Gaussian distribution as a model for probability density function of concentration derivative.

$$\sigma_{\chi'/C_p}(\chi/C_p) = \frac{1}{\sqrt{2}}\frac{\sigma_{\chi'}}{C_p}\left(\frac{\chi}{C_p}\right)^{17/20} . \tag{23}$$

Here, $\sigma_{\chi'/C_p}(\chi/C_p)$ is the conditional standard deviation of the normalized derivative, χ'/C_p, and $q = 3/4$. Note that χ' is normalized by C_p in this case (and not by $\sigma_{\chi'}$).

This model for the mean upcrossing rate can be tested against some of the available data. Accordingly, Figure 11 exhibits the observed number of upcrossings per unit time as a function of the normalized concentration, χ/C_p, in comparison with the predicted number of upcrossings per unit time as determined using Equations (22) and (23) (solid curve). The lognormal distribution was used to model $f_1(\chi/C_p)$. In general, the agreement between the predicted and observed upcrossing rate is very good. The model slightly underpredicts the upcrossing intensity for normalized concentration levels, χ/C_p, greater than about 10. This may be due to the fact that the conditional standard deviation, $\sigma_{\chi'/C_p}(\chi/C_p)$, is not accurately characterized by the relationship embodied by Equation (23) for values of χ/C_p greater than 10. In this regard, it should be recalled that this functional form was derived from $\sigma_\xi(\chi/C_p)$, which was determined based on values of χ/C_p less than about 6 (cf. Figure 4), and there is no guarantee that extrapolations of this relationship beyond this range are valid. Furthermore, some of the discrepancy may be the result of statistical scatter due to the paucity of data at large values of the concentration level. Data measured at other plume positions, not displayed here, show a similar behavior to that exhibited in Figure 11.

It would be interesting to compare the model of the mean upcrossing rate (cf. Equations (22) and (23)) with that which would have been obtained by assuming that the pdf of the concentration derivative, χ', given a concentration, χ, is statistically independent of χ, and that the concentration derivative pdf is Gaussian. Adopting these assumptions leads to the following model for the upcrossing intensity of the normalized concentration:

$$\langle N(\chi/C_p)\rangle = \frac{\sigma_{\chi'/C_p} f_1(\chi/C_p)}{\sqrt{2\pi}} . \tag{24}$$

Equation (24) is the model proposed by Kristensen *et al.* (1989) for the mean upcrossing rate. Figure 11 compares the upcrossing intensity with that predicted by the model embodied in Equation (24) (dashed curve). The model of Equation (24) results in a drastic underestimation of the upcrossing intensity for normalized concentration levels, χ/C_p, greater than about 4. On the other hand, the mean upcrossing rate is overestimated for normalized concentration levels less than about 3. In general, we found that failure to account for the statistical dependence of χ' on χ leads to poor estimates of the upcrossing rate.

The model for the upcrossing intensity embodied in Equations (22) and (23) is applicable only to in-plume concentrations (or, equivalently, to situations where intermittency due to plume meander is negligible). The upcrossing rate can be predicted from a knowledge of C_p and i_p for the concentration time series, and $\sigma_{\chi'}$ for the concentration derivative time series. Models for the prediction of the spatial distribution of C_p and i_p exist for plumes dispersing in a neutrally stratified turbulent boundary layer (Wilson *et al.*, 1985; Sawford and Stapountzis, 1987). A model for the spatial distribution of the unconditional standard deviation, $\sigma_{\chi'}$, of the concentration derivative has been developed by Kristensen *et al.* (1989) for plume dispersion from an elevated point source in a neutral boundary layer. In consequence, all the components exist for the prediction of recurrence statistics of concentration fluctuations (e.g., upcrossing intensity, return periods between successive upcrossings, mean excursion durations, etc.). To incorporate the effects of intermittency due to plume meandering, it is straightforward to show that the upcrossing intensity, $\langle N(\chi/C)\rangle$, of the normalized concentration level, χ/C, for the total concentration time series (zero concentrations included) is related to the upcrossing intensity, $\langle N(\chi/C_p)\rangle$, of the conditional concentration time series (cf. Equations (22) and (23)) as follows (C is the (total) mean concentration):

$$\langle N(\chi/C)\rangle = \gamma\langle N(\chi/(\gamma C_p))\rangle , \tag{25}$$

where γ is the intermittency factor. To derive Equation (25), we have used the relationship $C = \gamma C_p$. Equation (25) allows one to determine the upcrossing rate for the total concentration time series, once the upcrossing rate for the conditional time series has been obtained.

5. Summary and Conclusions

To verify certain assumptions underlying the simple model developed recently by Kristensen *et al.* (1989) for estimating the number of upcrossings per unit time of a given fixed concentration level, this study has analyzed some concentration fluctuation data from dispersing plumes in a near-neutral atmospheric surface layer, with the objective of extracting information on the statistical properties of the concentration derivative. Our results have shown that the pdf of concentration derivative, although symmetric, is highly non-Gaussian with wide flaring tails that are indicative of a significant intermittency effect of the small-scale concentration eddy structures in the dispersing plume. We show that the pdf of χ' can be well approximated by a gqG distribution with an intermittency exponent, $q = 0.3$. This result was obtained from concentration fluctuations measured at various positions in the crosswind cross-section of a dispersing plume in a near-neutral atmospheric surface layer over a limited downwind range from 25 to 100 m. Within these constraints, we found that the pdf of normalized concentration derivative, $\chi'/\sigma_{\chi'}$, displayed a remarkable universality in form. Because the concentration derivative emphasizes the information in the small eddy scales, the universality of the concentration derivative pdf is indicative of the local isotropy and homogeneity of the statistical characteristics of the small-scale structures in the plume.

Although χ and χ' are necessarily uncorrelated for a stationary process, our data indicate that they are not statistically independent as is assumed by Kristensen *et al.* (1989). The dependency between χ and χ' is such that they tend to inhibit each other in the sense that large (small) values of χ are preferentially associated with small (large) values of χ'. We found that the conditional pdf of the concentration derivative, χ', given a fixed value of concentration, χ, is well described by a gqG distribution with intermittency exponent $q = 3/4$. Because the conditional concentration derivative pdf exhibits a constant intermittency exponent in the dispersing plume (at least over the range of positions covered by the available data), its form is completely specified by the conditional standard deviation, $\sigma_{\chi'}(\chi/C_p)$. Under near-neutral stratification, we found that the conditional standard deviation of the normalized derivative, $\chi'/\sigma_{\chi'}$, reduces to a universal form which can be well approximated by $\sigma_{\chi'}(\chi/C_p)/\sigma_{\chi'} = (\chi/C_p)^{17/20}/\sqrt{2}$. It would be interesting to investigate (when more data become available) whether this universal form for the conditional pdf and the corresponding standard deviation of the normalized concentration derivative are maintained at greater downwind distances, or under diabatic conditions (e.g., very unstable thermal stratification, strong stable vertical stratification, etc.).

The information on the conditional statistics of the concentration derivative was used to revise a model for recurrence statistics of concentration fluctuations developed by Kristensen *et al.* (1989). This revised model agrees very well with measurements of the mean upcrossing rate obtained in a dispersing plume under near-neutral stratification. In consequence, development of a model to predict the mean concentration, C, the intermittency factor, γ, the fluctuation intensity, i, and the concentration derivative standard deviation, $\sigma_{\chi'}$, would allow determination of not only the single-point concentration pdf, but also multi-point statistics as embodied in the joint pdf of concentration and its derivative. The latter statistics reflect the small-scale properties of the dispersing plume and, in consequence, embody the information on the dynamics of the turbulent mixing processes. In turn, the joint pdf of concentration and its derivative allows the prediction of a number of recurrence statistics of concentration fluctuations such as upcrossing intensity, mean excursion duration, return period (inverse time factor), etc.

Appendix A. Effect of Instrument Smoothing on Recurrence Statistics

It is clear that the measured upcrossing intensity will be influenced significantly by the high-frequency content of the signal. In accordance with Equations (22) and (23), the effect of instrument smoothing on the upcrossing intensity will depend on the relative contribution made by the scales which are not resolved to fluctuation statistics such as the concentration pdf, $f_1(\chi/C_p)$, the conditional standard deviation, $\sigma_\xi(\chi/C_p)$, of the normalized derivative, $\xi \equiv \chi'/\sigma_{\chi'}$, the standard deviation, $\sigma_{\chi'}$, of the concentration derivative, and the intermittency exponent, q. The effect of low-pass filtering imposed by an instrument on the concentration pdf, f_1, has been studied elsewhere (e.g., Dinar *et al.*, 1988; Mylne and Mason, 1991) and, consequently, will not be considered here. The effect of time averaging on the standard deviation, $\sigma_{\chi'}$, of the concentration derivative can be determined analytically as follows:

$$\sigma^2_{\chi'}(f_R) = \int_0^\infty \omega^2 |H(\omega)|^2 S(\omega)\, \mathrm{d}\omega\,, \tag{A1}$$

where f_R is some measure of the frequency resolution of the instrument (e.g., -6 dB point of the frequency response function), ω is angular frequency, $S(\omega)$ is the power spectral density of concentration fluctuations, and $H(\omega)$ is the frequency response function of the instrument.

The influence of frequency resolution on the determination of the conditional standard deviation, $\sigma_\xi(\chi/C_p)$, and the intermittency exponent, q, cannot be determined analytically. In this case, we evaluated the influence by passing the original signal (measured by our concentration sensors) through a sequence of zero-phase Butterworth filters of order 8 with -6 dB cut-off frequency, f_R, selected between 1 and 100 Hz. We recall that the magnitude-squared of the frequency response function of a Butterworth filter of order r is

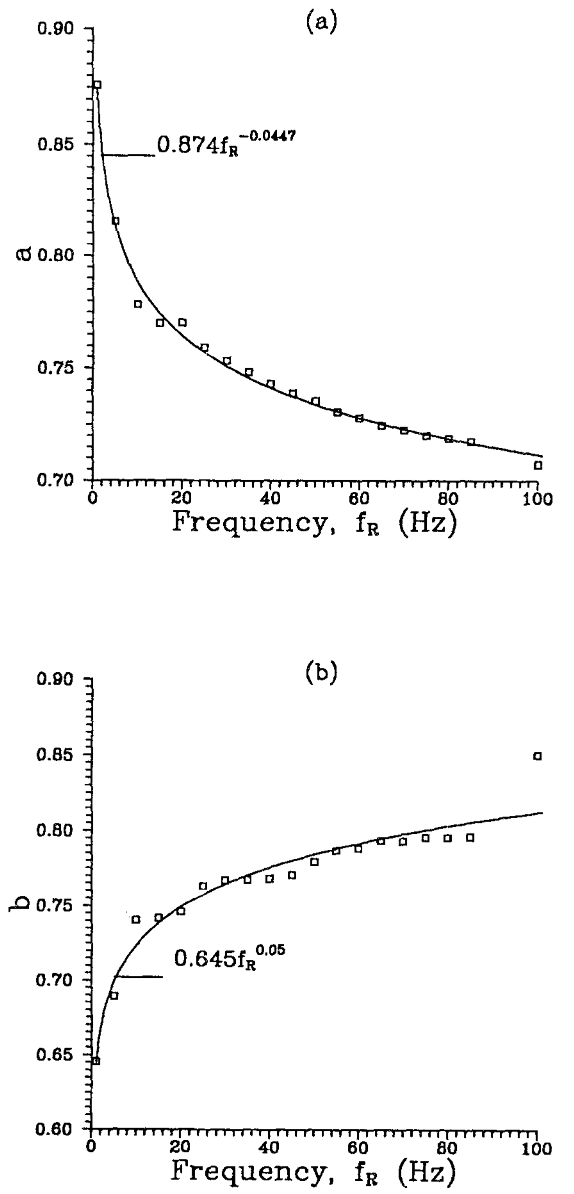

Fig. 12. Variation of a and b with the cut-off frequency, f_R. The solid curves are power-law fits of a and b to f_R.

$$|H(n)|^2 = \frac{1}{1 + (3^{1/2r} n/f_R)^{2r}}, \tag{A2}$$

where f_R is the frequency for which $|H(f_R)| = 1/2$ (-6 dB cut-off frequency). At a frequency resolution measured by f_R, we found that $\sigma_\xi(\chi/C_p; f_R)$ can be well

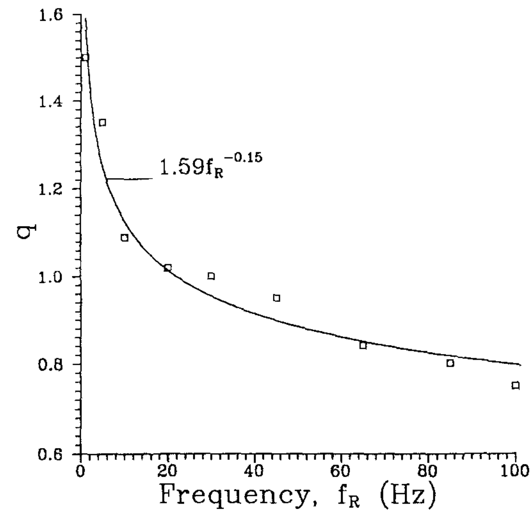

Fig. 13. Variation of intermittency exponent, q, with the cut-off frequency, f_R. The solid curve is a power-law fit of q to f_R.

approximated by a power-law form $a(f_R)(\chi/C_p)^{-b(f_R)}$, where a and b are constants at any frequency resolution that do not depend on χ/C_p (cf. Figure 4 which was constructed for the original signal measured with an instrument with -6 dB point, $f_R = (4^{1/\alpha} - 1)^{1/2}/(2\pi\beta) \approx 100$ Hz (cf. Equation (9))). The variation of a and b with the cut-off frequency is exhibited in Figure 12. As expected, the conditional standard deviation, $\sigma_\xi(\chi/C_p)$, is seen to decrease monotonically with the progressive loss of high-frequency content in the signal. Figure 12 indicates that the dependence of a and b on f_R can be approximated with the power-law forms 0.874 $f_R^{-0.0447}$ and $0.645 f_R^{0.05}$, respectively.

The variation of the intermittency exponent, q, for the conditional pdf of normalized concentration derivative, $\phi \equiv \xi/\sigma_\xi(\chi/C_p)$, (cf. Figure 6) with the cut-off frequency is shown in Figure 13. We found that $q(f_R)$ can be well approximated by the power-law form 1.59 $f_R^{-0.15}$. It is seen that the observed intermittency exponent, q, increases with the loss of high-frequency content in the signal. At high frequency resolutions, the conditional pdf of normalized derivative becomes more peaked around the center and its tails "skirt out" more, and the more so the greater the frequency resolution. Hence, as expected, the intermittency becomes increasingly stronger at the smaller scales, and this physical effect is observed, provided that the instrument is sufficiently responsive. For instruments with large averaging times, the conditional pdf of normalized derivative becomes increasingly parabolic (i.e., Gaussian) in shape and the value of the intermittency exponent approaches 2.

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