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A SIMPLE MODEL FOR THE PROBABILITY DENSITY FUNCTION OF CONCENTRATION FLUCTUATIONS IN ATMOSPHERIC PLUMES

EUGENE YEE

Defence Research Establishment Suffield, Box 4000, Medicine Hat, Alberta, T1A 8K6 Canada

and

R. CHAN

Kosteniuk Consulting Ltd, 429 6th Street East, Saskatoon, Saskatchewan, S7H 1B7 Canada

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Abstract—Simple closure hypotheses for the conditional pseudo-diffusion and pseudo-dissipation terms given a fixed concentration level in a dispersing plume are formulated, and used to obtain a simple functional form for the one-point probability density function (PDF) of concentration. These closure assumptions lead to a left-shifted and clipped-gamma PDF. A simple parametric expression for the observed relationship between the intermittency factor (i.e. the probability of non-zero concentrations) and the normalized second concentration moment is incorporated in the specification of the clipped-gamma PDF. This expression allows the form of the clipped-gamma PDF to be completely specified in terms of only two parameters; namely, the mean concentration and concentration variance. Comparisons of the clipped-gamma PDF to some atmospheric and large-eddy simulation concentration data show good agreement over a wide range of plume locations and atmospheric conditions. Copyright © 1997 Elsevier Science Ltd

Key word index: Plume dispersion, plume concentration statistics, hazard assessment.

1. INTRODUCTION

The probability density function (PDF) of concentration in an atmospheric plume is an important quantity used to describe and discuss environmental diffusion. The plume concentration PDF forms the basis for the definition and computation of a number of relevant parameters intervening in various practical problems. For example, assessment of reactant material flammability and of the level of harm arising from toxic materials requires a knowledge of the frequency of exceedance of critical threshold levels by peak concentrations. That is the reason why a considerable effort has been made to measure, model, and understand the concentration PDF.

Measurements of the frequency distribution of fluctuating plume concentrations (e.g. Fackrell and Robins, 1982; Sawford *et al.*, 1985; Sawford, 1987; Dinar *et al.*, 1988; Mylne and Mason, 1991; Yee *et al.*, 1993a, 1994; among many others) have revealed that the PDF is generally strongly skewed to the right with an upper tail that is heavier than that of the Gaussian form. A large number of mathematical forms have been used to model the concentration PDF shape [e.g. the lognormal distribution (Csanady, 1973), the exponential distribution (Barry, 1975; Hanna, 1984), the clipped-normal distribution (Lewellen and Sykes, 1986), the beta-Jacobi distribution (Derksen and Sullivan, 1990), and a linear combination of exponential and generalized Pareto PDFs (Lewis and Chatwin, 1995)]. However, despite this enormous effort, there is still no general agreement on the parametric PDF model that provides the best fit to plume concentration data over a wide range of experimental conditions.

Normally, turbulent diffusion models predict concentration moments rather than the concentration PDF itself. Currently, the most advanced atmospheric dispersion models can predict only the two lowest-order concentration moments (i.e. the mean concentration and concentration variance) of the instantaneous plume concentration field using either a second-order closure model for concentration fluctuation variance (e.g. Sykes *et al.*, 1984, 1986), or a two-particle Lagrangian stochastic model (e.g. Durbin, 1980; Kaplan and Dinar, 1988; Thomson, 1990). Hence, for the practical purpose of model parametrization, we need to formulate an appropriate

two-parameter function for the concentration PDF that is valid over a wide range of conditions, since the parameters of any such model can then be uniquely determined from the first two moments (e.g. the concentration mean and variance which can be predicted using an atmospheric dispersion model).

Of the mathematical PDF forms considered by various researchers, only the lognormal, exponential, and clipped-normal distributions are fully specified by two parameters, so that they may be estimated by predicting the concentration mean and variance. Although a physical justification has been proposed for the lognormal distribution (Csanady, 1973), it has been found that this distribution does not fit the concentration data very well (Sawford, 1987; Yee *et al.*, 1993a). Furthermore, the lognormal distribution cannot be used to predict the intermittency effect as the probability that the concentration is zero. Sawford (1987) tested the exponential and clipped-normal distributions against some concentration data, and noted that these model distributions give better agreement with the field observations than does the lognormal distribution. Mylne and Mason (1991) found that the clipped-normal PDF provided a reasonable fit to the concentration PDF over a wide range of experimental conditions, and that the exponential PDF was less flexible in fitting the full range of observed PDF behavior. Even so, the clipped-normal distribution appears to consistently underestimate the intermittency factor γ (i.e. the probability of non-zero concentrations) (Sykes and Henn, 1992). Furthermore, in plumes dominated by meandering, the upper tail of the clipped-normal distribution appears to underestimate the probability of occurrence of large concentrations.

Hence, all the two-parameter functions for the concentration PDF considered to date do not appear to be completely adequate in providing a statistical description of the measured concentration fluctuations. The objective of this paper is to construct a simple mathematical form of a PDF that is, nevertheless, flexible enough to fit measured concentration data over a wide range of experimental conditions. The concentration PDF form is obtained from closure hypotheses concerning the effects of turbulent stirring and molecular diffusion on the plume concentration structure.

2. CONCENTRATION PDF FORM

The equation for the instantaneous concentration $\Psi(\mathbf{x},t)$ of a passive scalar dispersing in a random incompressible velocity field $\mathbf{v}(\mathbf{x},t)$ is given by

$$\frac{\partial \Psi(\mathbf{x},t)}{\partial t} + \mathbf{v}\cdot\nabla\Psi(\mathbf{x},t) = D\nabla^2\Psi(\mathbf{x},t), \quad \nabla\cdot\mathbf{v}(\mathbf{x},t) = 0 \tag{1}$$

where $\mathbf{x}$ is the position vector, t is time, and D is the molecular diffusion coefficient. The solution of equation (1) reduces in Lagrangian terms to the analysis of the motion of contaminant tracer elements along the Wiener path $\mathbf{r}_t$:

$$\mathbf{r}_t = \mathbf{x} - \int_0^t \mathbf{v}(\mathbf{r}_\tau, t-\tau)\,d\tau + (2D)^{1/2}\,\mathbf{w}(t) \tag{2}$$

where $\mathbf{w}(t)$ is a Wiener process (Durbin, 1980). Equation (2) describes the displacement, during the interval $(0,t)$, of all fluid elements that arrive at the point $\mathbf{x}$ at time t.

We are interested in the concentration PDF at a given point, i.e.

$$f_\Psi(\psi;\mathbf{x},t) \equiv \langle \delta(\psi - \Psi(\mathbf{x},t))\rangle \tag{3}$$

where $\langle\cdots\rangle$ denotes the ensemble average and $\delta(\cdot)$ denotes the Dirac distribution [i.e. $\delta(\psi - \Psi(\mathbf{x},t))$ is the instantaneous or fine-grained PDF]. Since attention will be restricted to statistically stationary situations (e.g. steady plume emissions into a stationary atmosphere), the PDF of equation (3) will be independent of t, and the ensemble average can be replaced by the time average (ergodicity). Without molecular diffusion ($D = 0$), the Wiener paths of equation (2) coincide with the Lagrangian trajectory (viz., the concentration in a fluid element is unchanged as it is carried about). Assuming that the source concentration ψ_s is uniform, the random trajectories of fluid parcels that pass point $\mathbf{x}$ can carry either the scalar concentration $\Psi = 0$ (i.e. the fluid element did not pass through the source on its way to $\mathbf{x}$ and, hence, is devoid of scalar) or $\Psi = \psi_s$ (i.e. the fluid element emanated from the source and, hence, is marked by the source concentration). Thus, in the absence of molecular diffusion, the concentration PDF reduces to the following simple form:

$$f_\Psi(\psi;\mathbf{x}) = \gamma(\mathbf{x})\,\delta(\psi - \psi_s) + (1 - \gamma(\mathbf{x}))\,\delta(\psi) \tag{4}$$

where $\gamma(\mathbf{x})$ is the intermittency factor (viz., the probability of $\mathbf{x}$ being in contaminant fluid).

More realistically, the concentration in a marked fluid element moving along the random Wiener trajectory $\mathbf{r}_t$ and passing through the point $\mathbf{x}$ at time t [cf. equation (2)] is the result of the simultaneous effects of turbulent stirring (i.e. strain-induced stretching of the intermaterial surface of the fluid element) and molecular mixing (i.e. molecular diffusion of the contaminant across the intermaterial surface). The latter process results in the "spreading" of the ideal two-state concentration PDF of equation (4) into a range of concentration values that varies continuously between 0 and ψ_s. Hence, the development of the long upper tail observed in the concentration PDF at $\mathbf{x}$ simply reflects the various strain histories and strain-induced mixing events along all the Wiener trajectories that bring fluid elements through the point $\mathbf{x}$. We wish to determine a functional form for the concentration PDF which captures the physics of turbulent transport and molecular diffusion (i.e. $D > 0$) relevant to the actual situation.

To that end, consider an arbitrary stationary random function $Y(\mathbf{x},t)$ that is statistically dependent on the random concentration $\Psi(\mathbf{x},t)$. Recall the following important formula from probability theory (Parzen, 1960):

$$\langle g(\Psi)Y\rangle = \int_0^\infty g(\psi')\langle Y|\Psi=\psi'\rangle f_\Psi(\psi')\,\mathrm{d}\psi' \quad (5)$$

where $\langle Y|\Psi=\psi\rangle$ denotes the expected value of Y conditioned upon $\Psi(\mathbf{x},t)=\psi$, and $g(\cdot)$ is an arbitrary function. The choice $g(\Psi)=\delta(\psi-\Psi(\mathbf{x},t))$ reduces equation (5) to

$$\langle Y(\mathbf{x},t)\,\delta(\psi-\Psi(\mathbf{x},t))\rangle = \langle Y(\mathbf{x},t)|\Psi(\mathbf{x},t)=\psi\rangle f_\Psi(\psi). \quad (6)$$

By stationarity, the ensemble average on the left-hand side of equation (6) should not depend on time, and if we differentiate this quantity with respect to t we get

$$0 = \langle Y_{,t}|\Psi=\psi\rangle f_\Psi(\psi) - \frac{\partial}{\partial\psi}[\langle Y\Psi_{,t}|\Psi=\psi\rangle f_\Psi(\psi)]. \quad (7)$$

In equation (7), we use the convenient notation which represents partial derivatives by a comma; i.e. $Y_{,t}\equiv\partial Y/\partial t$, etc. Equation (7) defines a transport equation for the concentration PDF. The right-hand side of equation (7) can be interpreted as a probability flux in the positive direction of the ψ-axis (scalar concentration space) and, as such, is determined by the specification of two conditional expectations; namely, $\langle Y_{,t}|\Psi=\psi\rangle$ and $\langle Y\Psi_{,t}|\Psi=\psi\rangle$.

We are free to choose the random function Y in equation (7). Because we are interested in determining how turbulent stirring and molecular diffusion at the smallest length scales of the flow determine the concentration PDF form, it is convenient to choose $Y\equiv\Psi_{,t}$. This choice is sensible because the concentration derivative emphasizes the information on the distribution of the small concentration eddies in the plume and, as a result, should embody information on the small-scale mixing processes in the concentration field (viz., the concentration derivative encodes information on the stochastically distributed strainings of the scalar-marked fluid elements). Hence, letting $Y\equiv\Psi_{,t}$ in equation (7), the concentration PDF is defined by the following differential equation:

$$\frac{\partial}{\partial\psi}[\langle(\Psi_{,t})^2|\Psi=\psi\rangle f_\Psi(\psi)] - \langle\Psi_{,tt}|\Psi=\psi\rangle f_\Psi(\psi) = 0. \quad (8)$$

Equation (8) determines the PDF for any stationary random process (e.g. the fluctuating concentration in a steady plume dispersing in a stationary atmosphere) in terms of two conditional time derivatives of the process. For fluctuating plume concentrations, these two conditional expectations can be interpreted as embodying the effects of turbulent stirring and molecular diffusion on the concentration PDF form. Recall that without molecular diffusion, the concentration PDF would simply be the two-state function summarized in equation (4).

Can we intuit reasonable forms for $\langle(\Psi_{,t})^2|\Psi=\psi\rangle$ and $\langle\Psi_{,tt}|\Psi=\psi\rangle$ on purely *a priori* grounds? Indeed, closure of equation (8) requires one to specify appropriate forms for these two conditional expectations. At this point, there seems to be no *a priori* way of knowing what forms these conditional expectations should take. The only reasonable way to proceed is to make some informed guesses for the two conditional expectations and see whether they lead to a successful prediction for the concentration PDF. From this viewpoint, it should be emphasized that the specification of these two conditional expectations, however plausible, should be interpreted simply as closure hypotheses for the determination of an appropriate concentration PDF form and, as such, will need to be carefully tested and validated against experimental data. Nevertheless, we propose to make the required guesses as plausible as possible by drawing analogies to a simple phenomenological picture of a dispersing plume; namely, the simultaneous effects of turbulent stirring and molecular mixing produce a plume that consists of thin strands or sheets of high concentration embedded in a low concentration background (e.g. Batchelor, 1959; Chatwin and Sullivan, 1979) [viz., the turbulent advection draws out contaminant blobs into thin sheets and strands which are subsequently molecularly diffused into the background fluid].

We first consider the specification of a plausible form for $\langle(\Psi_{,t})^2|\Psi=\psi\rangle$, which we will refer henceforth to as the conditional pseudo-dissipation term. This nomenclature arises from the observation that if some form of Taylor's frozen flow hypothesis is valid, then this term may provide a plausible one-dimensional surrogate for the conditional scalar dissipation defined to be the expected value of the squared gradient of Ψ conditioned on the scalar concentration (viz., $\langle(\Psi_{,t})^2|\Psi=\psi\rangle \sim U^2\langle(\nabla\Psi)^2|\Psi=\psi\rangle$, where U is a typical advection velocity). However, in what follows, we do not require any assumptions either on the validity of Taylor's frozen turbulence hypothesis or on the nature of the conditional scalar dissipation in order to specify an appropriate form for $\langle(\Psi_{,t})^2|\Psi=\psi\rangle$ [i.e. it is the latter quantity, and not the conditional scalar dissipation, which is required in equation (8)]. To that end, we first note that the term $\langle(\Psi_{,t})^2|\Psi=\psi\rangle$ controls the rate of decay of the concentration increments at a fixed point. To model this decay, we make the assumption that the average of the square of the concentration derivative at a fixed concentration level is linearly related to the concentration level, so

$$\langle(\Psi_{,t})^2|\Psi=\psi\rangle = a(\psi+\lambda) \quad (9)$$

where a and λ are parameters with $a>0$ and $\lambda\geqslant 0$. Equation (9) implies that there exists a statistical dependence between the concentration and the square of

the concentration derivative (e.g. pseudo-dissipation). Indeed, some measurements of the conditional pseudo-dissipation in steady plumes (Yee *et al.*, 1993b) indicate that $\langle(\Psi_{,t})^2|\Psi=\psi\rangle \sim \psi^{17/20}$. However, for simplicity, we will assume that the conditional pseudo-dissipation depends linearly on the concentration level as expressed in equation (9). As will be shown below, the hypothesis that $\langle(\Psi_{,t})^2|\Psi=\psi\rangle$ grows with ψ results in a concentration PDF that decreases with ψ more slowly than the Gaussian distribution. In particular, our conjecture of the linear dependence of the conditional pseudo-dissipation on the concentration level [cf. equation (9)] implicitly recognizes the action of turbulent straining upon the concentration gradients and the concomitant enhancement of dissipative molecular effects by that action.

Next, we consider the modeling of $\langle\Psi_{,tt}|\Psi=\psi\rangle$ which we will refer to as the conditional pseudo-diffusion term. This term is so named because invoking Taylor's frozen flow hypothesis allows a connection to be made to the expected value of the Laplacian of Ψ conditioned on the scalar concentration (viz., $\langle\Psi_{,tt}|\Psi=\psi\rangle \sim U^2\langle\nabla^2\Psi|\Psi=\psi\rangle$), although the physical validity of this connection is not required in order to specify an appropriate form for $\langle\Psi_{,tt}|\Psi=\psi\rangle$. To proceed further, we note that turbulent stirring is effective at redistributing the concentration field at all scales above the Batchelor scale. In consequence, we expect the average instantaneous rate of change of the concentration increments ($\Psi_{,t}$) to grow (increase) or decay (decrease) when the effects of turbulent stirring are, respectively, greater than or smaller than those of molecular diffusion. This simple physical picture for the growth and decay of instantaneous concentration increments (derivatives) can be modeled with the following simple form:

$$\langle\Psi_{,tt}|\Psi=\psi\rangle = b(d-h(\psi)) \tag{10a}$$

where $h(\psi)$ is a non-decreasing function defined on $(0,\infty)$, and b and d are two parameters with $b>0$.

Equation (10a) implies that $\langle\Psi_{,tt}|\Psi=\psi\rangle>0$ when the concentration is less than the threshold concentration $\psi_T \equiv h^{-1}(d)$. In other words, turbulent stirring is effective at increasing the concentration increments (or, gradients) for concentration levels less than ψ_T. On the other hand, when the concentration is greater than the threshold level ψ_T, concentration increments are decaying since $\langle\Psi_{,tt}|\Psi=\psi\rangle<0$. This effect has the physical justification that regions in the scalar field that have been highly strained typically experience stronger molecular diffusion than other regions. This causes the concentration fluctuations in the highly strained regions to decay rapidly. A simple choice for the non-decreasing function $h(\psi)$ is $h(\psi)=(\psi+\lambda)$ [cf. equation (9)]. With this choice, equation (10a) reduces to

$$\langle\Psi_{,tt}|\Psi=\psi\rangle = k' - \frac{(\psi+\lambda)}{s'} \tag{10b}$$

where $k' \equiv bd$ and $s' \equiv b^{-1}$ are parameters with $k'>0$ and $s'>0$. For the choice $h(\psi)=(\psi+\lambda)$, a positive threshold concentration $\psi_T \equiv h^{-1}(d) = (d-\lambda)$ requires $d>\lambda\geqslant 0$, so $k'>0$.

We have made some simple, pragmatic, and arguably plausible guesses for the form of the conditional pseudo-dissipation and pseudo-diffusion terms in equations (9) and (10b), respectively. Future efforts should perhaps focus on providing more physically based forms for these conditional expectations (i.e. forms that have a stronger connection to the fundamental processes underlying turbulence and turbulent diffusion). Nevertheless, with the specification of the closure hypotheses for the conditional pseudo-dissipation and pseudo-diffusion terms, the concentration PDF can be uniquely determined from the solution of the differential equation of equation (8), once an appropriate boundary condition for $f_\Psi(\psi)$ is imposed at the lower endpoint $\psi=0$ of the domain of definition for the PDF.

To that end, we remark that equation (8) has the integrating factor

$$u(\psi) = \exp\left[-\int^{\psi}\frac{\langle\Psi_{,tt}|\Psi=\psi'\rangle}{\langle(\Psi_{,t})^2|\Psi=\psi'\rangle}\,\mathrm{d}\psi'\right]$$

which simplifies to $u(\psi)=(\psi+\lambda)^{-k}\exp(\psi/s)$ on insertion of equations (9) and (10b). Here, $k\equiv k'/a>0$ and $s\equiv s'a>0$. For $\lambda=0$, note that $\lim_{\psi\to 0^+}u(\psi)\to\infty$ implying in this case that the lower boundary $\psi=0$ is inaccessible (viz., $\psi=0$ acts as a natural boundary here). In this case then, no boundary condition needs to be imposed at $\psi=0$. The solution of equation (8) that satisfies equations (9) and (10b) with $\lambda=0$ is given by

$$f_\Psi(\psi) = M\psi^{k-1}\exp(\psi/s) \tag{11}$$

where $M^{-1}\equiv M^{-1}(k,s)=s^k\Gamma(k)$ [$\Gamma(k)$ denotes the Gamma function] is chosen to ensure proper normalization of the PDF in $(0,\infty)$.

On the other hand, for $\lambda>0$ we note that $\lim_{\psi\to 0^+}u(\psi)\to A$ where A is a *finite* positive constant, implying that the lower boundary $\psi=0$ is accessible. In this case, we need to specify an explicit boundary condition at $\psi=0$ in order to uniquely determine the probability law. To that end, we note that when the plume is absent at a receptor point due to meandering, the concentration can admit the value zero with *positive* probability, say, $(1-\gamma)$ ($\gamma\in[0,1]$). In view of this, it is appropriate to adjoin the following boundary condition at $\psi=0$ (intermittent boundary condition):

$$F_\Psi(0^+)-F_\Psi(0^-)\equiv\int_{0^-}^{0^+}f_\Psi(\psi)\,\mathrm{d}\psi=(1-\gamma)$$

where $F_\Psi(\psi)$ is the cumulative distribution function (CDF) for the concentration. Inserting equations (9) and (10b) in equation (8), and solving the resulting differential equation for $f_\Psi(\psi)$, subject to the intermittent boundary condition, leads to the following

concentration PDF form:

$$f_\Psi(\psi) = N(\psi + \lambda)^{k-1} \exp\left(-\frac{\psi + \lambda}{s}\right) + (1 - \gamma)\,\delta(\psi), \quad \lambda > 0 \tag{12a}$$

where $N = N(\gamma, k, s, \lambda)$ is the normalization constant chosen to make the integral of equation (12a) over the sample space $[0, \infty)$ equal to unity, so

$$N^{-1} = \gamma^{-1} \int_0^\infty (\psi + \lambda)^{k-1} \exp\left(-\frac{\psi + \lambda}{s}\right) d\psi = \gamma^{-1} s^k \Gamma(k; \lambda/s). \tag{12b}$$

Here, $\Gamma(v; x)$ denotes the incomplete Gamma function. The concentration PDF embodied in equation (12) is completely determined by four parameters; namely, γ, k, s, and λ.

There is no conceptual difficulty in considering γ to be an independent parameter in equation (12). However, in order for the concentration PDF to be useful for practical applications, it is desirable to choose PDF forms with as few parameters as possible. To that end, we will reduce the number of independent parameters required to specify the concentration PDF in equation (12) by assuming that γ can be uniquely determined by k, s, and λ [viz., $\gamma = \gamma(k, s, \lambda)$]. To proceed further, we first note that the in-plume concentration PDF [i.e. the continuous part of the PDF defined over $(0, \infty)$ that is determined exclusively by the conditional pseudo-dissipation and pseudo-diffusion terms] of equation (12) reduces to a gamma distribution when $\gamma \to 1^-$ and $\lambda \to 0^+$ (e.g. in this case, s and k are the scale and shape parameters, respectively, of the gamma distribution). This special case reduces to the concentration PDF form of equation (11). The introduction of the parameter λ to this PDF corresponds then to a shift (translation) of the gamma distribution to the left by the amount λ [i.e. $\psi \to \psi' \equiv (\psi + \lambda)$ in the gamma distribution]; and, hence it seems reasonable to relate the magnitude of the left-shift to γ in order to reduce the number of independent parameters required to specify the concentration PDF of equation (12). To that end, it is assumed that γ can be uniquely determined as the area remaining under the gamma PDF curve for $\psi > 0$ after a left-shift of ψ by the amount λ, viz.

$$\gamma \equiv \gamma(k, s, \lambda) = \int_\lambda^\infty \left(\frac{\psi}{s}\right)^{k-1} \frac{\exp(-\psi/s)}{s\Gamma(k)}\, d\psi = \frac{\Gamma(k; \lambda/s)}{\Gamma(k)}. \tag{13}$$

Obviously, with this assumption we have that $\gamma \to 1^-$ (i.e. non-intermittent plume concentration) when $\lambda \to 0^+$ (i.e. no left-shift), and this is consistent with equation (11) and the discussion preceding it (viz., $\gamma = 1$ for this case because zero concentrations are inaccessible). The assumption that γ in equation (12) can be uniquely determined as a function of k, s, and λ in accordance to equation (13) was introduced here as a practical constraint aimed at minimizing the number of parameters required to specify the concentration PDF. As such, we will need to appeal to experimental data for the real justification of this assumption.

In summary, the concentration PDF can be formally expressed as

$$f_\Psi(\psi) = \left(\frac{\psi + \lambda}{s}\right)^{k-1} \frac{\exp(-(\psi + \lambda)/s)}{s\Gamma(k)} + (1 - \gamma)\,\delta(\psi) \tag{14}$$

with $\gamma = \gamma(k, s, \lambda)$ determined in accordance to equation (13). Hence, the concentration PDF is composed of a mixed fluid part (e.g. continuous part of the distribution of Ψ for $\Psi > 0$) that results from in-plume mixing of eddies that contain the scalar contaminant, and an unmixed ambient fluid part (e.g. discrete part of the distribution of Ψ at $\Psi = 0$) that is caused by plume meandering producing intermittent periods of zero concentration for a fraction of time $(1 - \gamma)$. Note that equation (14) includes equation (11) as a special case; namely, for the case where $\lambda \to 0^+$ and $\gamma \to 1^-$. The closure hypotheses of equations (9) and (10b) have resulted in a conditional PDF of Ψ given that $\Psi > 0$ which is a left-shifted and clipped-gamma distribution. Finally, the discrete component represented by γ [cf. equation (13)] is obtained by replacing any unrealizable negative tail in the left-shifted gamma distribution by a delta function at zero concentration with an integrated probability of the negative concentrations. Consequently, we will henceforth refer to the concentration PDF of equation (14) as the clipped-gamma PDF.

Finally, we note that the clipped-normal PDF would have resulted if we specified the following forms for the conditional pseudo-dissipation and pseudo-diffusion terms:

$$\langle (\Psi_{,t})^2 | \Psi = \psi \rangle = a \tag{15a}$$

and

$$\langle \Psi_{,tt} | \Psi = \psi \rangle = b - c\psi \tag{15b}$$

where a, b, and c are parameters with $a > 0$ and $c > 0$. In contrast to equation (10b), which postulates that $\langle (\Psi_{,t})^2 | \Psi = \psi \rangle$ depends linearly on the concentration level, equation (15b) assumes that $\langle (\Psi_{,t})^2 | \Psi = \psi \rangle$ maintains a constant value that is independent of ψ. Subject to the closure hypotheses of equations (15a) and (15b), the solution of equation (8) that satisfies the intermittent boundary condition yields the following Gaussian form for the concentration PDF:

$$f_\Psi(\psi) = N \exp\left(-\frac{1}{2}\left(\frac{\psi - \mu_c}{\sigma_c}\right)^2\right) + (1 - \gamma)\,\delta(\psi) \tag{15c}$$

where the normalization constant $N \equiv N(\gamma, \mu_c, \sigma_c)$ is determined from

$$N^{-1} = \gamma^{-1} \int_0^\infty \exp\left(-\frac{1}{2}\left(\frac{\psi - \mu_c}{\sigma_c}\right)^2\right) d\psi$$

$$= \gamma^{-1} (\pi/2)^{1/2} \sigma_c \left(1 + \operatorname{erf}\left(\frac{\mu_c}{\sqrt{2}\sigma_c}\right)\right). \quad (15d)$$

Here, $\operatorname{erf}(x)$ denotes the error function, and $\mu_c \equiv b/c$ and $\sigma_c^2 \equiv a/c$ are the location and scale parameters, respectively. Again, it is useful to assume that $\gamma \equiv \gamma(\mu_c, \sigma_c)$ in order not to add any additional independent parameters. To that end, it is assumed that γ is the unique function of μ_c and σ_c that is determined from the integrated probability for the usual Gaussian form in the positive concentration range [e.g. compare with the similar condition embodied in equation (13) for the clipped-gamma distribution]. With this assumption, we obtain the clipped-normal concentration PDF (Lewellen and Sykes, 1986):

$$f_\Psi(\psi) = \frac{1}{\sqrt{2\pi}\sigma_c} \exp\left(-\frac{1}{2}\left(\frac{\psi - \mu_c}{\sigma_c}\right)^2\right) + (1 - \gamma)\,\delta(\psi) \quad (16)$$

where

$$\gamma = \gamma(\mu_c, \sigma_c) \equiv \frac{1}{2}\left(1 + \operatorname{erf}\left(\frac{\mu_c}{\sqrt{2}\sigma_c}\right)\right). \quad (17)$$

3. DETERMINATION OF MOMENTS AND PARAMETERS OF THE PDF

We will use the clipped-normal PDF as the standard for comparison because this two-parameter model appears to provide the best agreement with plume concentration data (e.g. Sawford, 1987; Dinar *et al.*, 1988; Mylne and Mason, 1991). The integral moments of the clipped-normal PDF [cf. equations (16) and (17)] can be determined as follows:

$$\left\langle \left(\frac{\Psi}{C}\right)^n \right\rangle = \frac{\Lambda^n}{\sqrt{\pi}} \sum_{j=0}^{n} \binom{n}{j} \phi^j I_{n-j}, \quad 2 \leqslant n, \quad n \in \mathbf{N}, \quad (18a)$$

where $\phi \equiv \mu_c/(\sqrt{2}\sigma_c)$ and $C \equiv \langle \Psi \rangle$ is the mean concentration. Furthermore,

$$\Lambda \equiv \left(\gamma\phi + \frac{1}{2\sqrt{\pi}} \exp(-\phi^2)\right)^{-1} \quad (18b)$$

and, I_n is defined in terms of the following two-term recurrence relation:

$$I_n = \frac{1}{2}(-1)^{n-1} \phi^{n-1} \exp(-\phi^2) + \frac{n-1}{2} I_{n-2}, \quad 2 \leqslant n, \; n \in \mathbf{N}, \quad (18c)$$

with $I_0 = \sqrt{\pi}\gamma$ and $I_1 = \exp(-\phi^2)/2$.

The integral moments of the clipped-gamma PDF [cf. equation (14)] can be determined as follows:

$$\langle \Psi^n \rangle = \sum_{j=0}^{n} \binom{n}{j} (-\lambda)^j s^{n-j} \frac{\Gamma(n - j + k; \lambda/s)}{\Gamma(k)}, \quad n \in \mathbf{N}. \quad (19)$$

The associated CDF assumes the form

$$F_\Psi(\psi) \equiv \int_0^\psi f_\Psi(\psi')\, d\psi' = 1 - \frac{\Gamma(k; (\psi + \lambda)/s)}{\Gamma(k)}. \quad (20)$$

The model concentration PDF parameters are k, s, and λ with $k > 0$, $s > 0$, and $\lambda \geqslant 0$. To estimate these parameters, we need first to derive relationships between the first and second concentration moments and k, s, and λ. In light of equations (13) and (19), the first and second concentration moments impose the following two constraints on the parameters:

$$C = (-\lambda + sk)\gamma + \frac{s}{\Gamma(k)} \left(\frac{\lambda}{s}\right)^k \exp(-\lambda/s) \quad (21)$$

and

$$\langle \Psi^2 \rangle = \lambda s \gamma + (-\lambda + s(k + 1)). \quad (22)$$

Equations (21) and (22) together with equation (13) enable k, s, and λ to be determined from a knowledge of the mean concentration, C, the mean-square concentration, $\langle \Psi^2 \rangle$, and the intermittency factor, γ.

The preceding methodology for determination of k, s, and λ requires the prediction of three parameters by an atmospheric dispersion model, namely C, $\langle \Psi^2 \rangle$, and γ. Given that most current models can predict the mean concentration only, the requirement to predict two further parameters imposes a major restriction on the use of the clipped-gamma PDF. To further reduce the number of independent parameters in the clipped-gamma PDF, we will assume that there exists a direct functional relationship between the fraction γ of non-zero concentrations and the normalized second concentration moment, $\langle (\Psi/C)^2 \rangle$. In order to determine the relationship between γ and $\langle (\Psi/C)^2 \rangle$, we will use some plume concentration fluctuation data.

To that end, we selected 1038 individual concentration time series from a set of plume concentration fluctuation measurements that formed part of a cooperative Concentration Fluctuation Experiments (CONFLUX) project. The data were collected in September 1991, November 1992, May 1993, and May 1994 at a site near Tower Grid on U.S. Army Dugway Proving Ground, Utah (40°06′N, 112°59′W). In each experiment, a plume was formed in the atmosphere by releasing propylene (C_3H_6) from a continuously emitting point source. The measurements were made with very fast-response concentration detectors having a frequency response in excess of 100 Hz. Further details of the experimental system and concentration measurements have been given by Yee *et al.* (1993c, 1994, 1995).

The data used for the present study were selected to approximately bracket the range of conditions measured in the CONFLUX project. The measurements covered downwind fetches x of between 12.5 and 1000 m from the source. The concentration data were measured under moderately convective to extremely stable atmospheric conditions with wind speeds in the range from about 0.5 to 6.0 m s^{-1} at 3 m height. The plume concentration data were sampled over a wide range of receptor positions in both lateral and vertical cross-sections through the plume. Lateral plume positions sampled varied from the mean-plume centerline at $y/\sigma_y = 0$ (where y is crosswind distance from the mean-plume centerline, and σ_y is the mean-plume width) to the plume fringes at $y/\sigma_y \approx 3.5$. Vertical plume positions sampled ranged from 0.5 to 16 m height above ground. For all the experiments, the source was located at heights above ground of from 1 to 5 m.

In Fig. 1, observations of $\langle(\Psi/C)^2\rangle$ are plotted as a function of the measured intermittency factor, γ, for the selected field data. Only concentration data with $\langle(\Psi/C)^2\rangle < 200$ have been included in Fig. 1. The dashed line in Fig. 1 shows the normalized mean-square concentration vs intermittency relationship predicted by the clipped-normal distribution [cf. equation (17)]. It is noteworthy that the clipped-normal PDF systematically underpredicts the intermittency factor γ (viz., overpredicts the fraction of zero concentrations) for a given value of the normalized mean-square concentration. This fact is consistent with the results of Sykes and Henn (1992) who showed that the probability of zero concentration from their large-eddy simulation plume concentration data generally falls below that predicted by the clipped-normal distribution [cf. Fig. 14 in Sykes and Henn (1992)]. A close examination of Fig. 1 suggests that γ is inversely proportional to $\langle(\Psi/C)^2\rangle$. Hence, it appears that a simple relation between the normalized mean-square concentration and plume intermittency can be represented as follows:

$$\langle(\Psi/C)^2\rangle = 3/\gamma. \tag{23}$$

The constant 3 in equation (23) was obtained by a least-squares fit to the data. Equation (23) is shown in Fig. 1 by the solid curve, and it appears that this relation represents the data well.

In view of equation (23), it is now possible to determine the parameters k, s, and λ of the clipped-gamma PDF from a knowledge of only the mean and mean-square concentrations. The parameters of the clipped-gamma PDF determined below are appropriate for an instantaneous concentration that has been normalized by C (i.e. for Ψ/C). In view of equations (13), (21)–(23), the parameters k, s, and λ are determined by an equation for the mean concentration,

$$1 = (-\lambda + sk)\gamma + \frac{s}{\Gamma(k)}\left(\frac{\lambda}{s}\right)^k \exp(-\lambda/s) \tag{24}$$

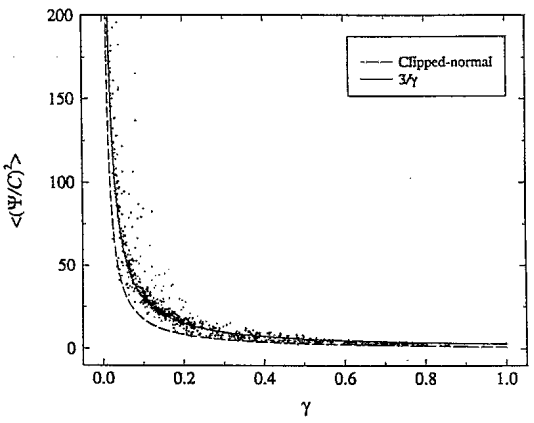

Fig. 1. Normalized mean-square concentration, $\langle(\Psi/C)^2\rangle$, plotted against the intermittency factor, γ.

an equation for the normalized mean-square concentration,

$$\langle(\Psi/C)^2\rangle = \lambda s\gamma + (-\lambda + s(k+1)) \tag{25}$$

and, an equation relating the intermittency factor to the normalized mean-square concentration,

$$\gamma = 3\langle(\Psi/C)^2\rangle^{-1} = \frac{\Gamma(k;\lambda/s)}{\Gamma(k)}. \tag{26}$$

The non-dimensional relations of equations (24)–(26) require only the normalized mean-square concentration, $\langle(\Psi/C)^2\rangle$, to determine the clipped-gamma PDF parameters k, s, and λ.

4. COMPARISON WITH MEASUREMENTS

We first compare higher-order moments predicted by the clipped-gamma PDF with some measured moments. To that end, the concentration time series were processed to determine the normalized concentration moments up to order eight (i.e. we calculated $\langle(\Psi/C)^n\rangle$ for $n = 2, 3, 4, \ldots, 8$ from the concentration data). These results are presented in Fig. 2 in the form of a number of normalized moment diagrams (viz., scattergrams of $\langle(\Psi/C)^n\rangle$ for $n = 3$, 4, 5, 6, 7, and 8 plotted against $\langle(\Psi/C)^2\rangle$ on a double-logarithmic scale). A noteworthy feature in all these moment diagrams is that there appears to exist strong correlations between the various higher-order and second-order non-dimensional concentration moments. In particular, note the collapse of the concentration data in each moment diagram onto a single "universal" curve. Thus, it appears that the concentration PDF over a wide range of conditions in a dispersing plume is determinable effectively from the first two concentration moments.

The normalized second-order concentration moment was used to estimate the parameters required to specify the clipped-gamma and clipped-normal PDFs;

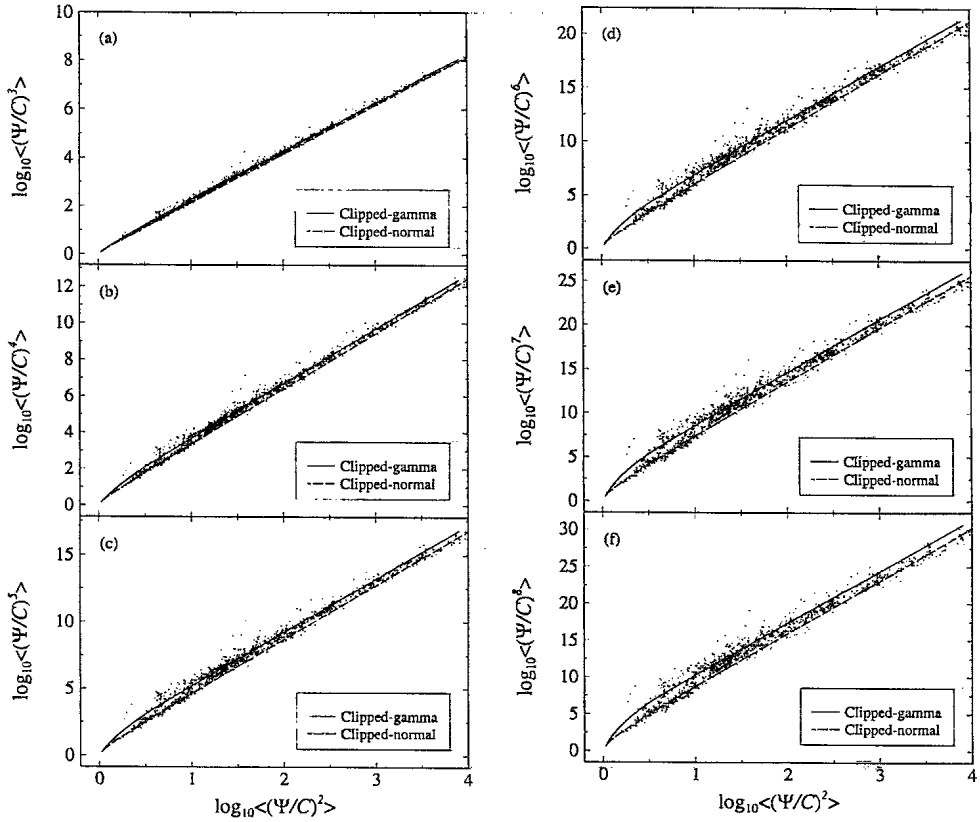

Fig. 2. Moment diagrams of (a) $\langle(\Psi/C)^3\rangle$; (b) $\langle(\Psi/C)^4\rangle$; (c) $\langle(\Psi/C)^5\rangle$; (d) $\langle(\Psi/C)^6\rangle$; (e) $\langle(\Psi/C)^7\rangle$; and, (f) $\langle(\Psi/C)^8\rangle$ plotted against $\langle(\Psi/C)^2\rangle$ on a double-logarithmic scale. The plot also includes the theoretical one-dimensional curves generated by the clipped-gamma and clipped-normal PDFs.

and, the resulting estimated PDFs were then used to calculate the higher-order concentration moments [cf. equations (18) and (19)]. The predicted values of $\langle(\Psi/C)^n\rangle$ ($n = 3$, 4, 5, 6, 7, and 8) obtained from the two model PDFs have been superimposed on the moment diagrams in Fig. 2 for comparison. Note that the clipped-normal PDF appears to provide a slightly better fit to the data for small values of $\langle(\Psi/C)^2\rangle$ (i.e. for $\langle(\Psi/C)^2\rangle \to 1^+$) than that provided by the clipped-gamma PDF. However, in general, the concentration moments appear to be slightly underpredicted by the clipped-normal PDF (i.e. the majority of the data points in the moment diagrams lie slightly above the theoretical curve generated by the clipped-normal PDF). Because the characteristics of a probability distribution in the upper tail are emphasized in the higher-order moments, this observation suggests that the clipped-normal PDF possesses a shorter upper tail than the concentration data. On the other hand, the concentration moments are in good agreement with the predictions provided by the clipped-gamma PDF. In particular, there appears to be no discernible bias in the prediction of the higher-order moments, although the maximum deviation between the observed and predicted moments appears to increase with increasing moment order. This behavior is most likely attributed to the increased sampling errors arising from the measurement of the higher-order moments (viz., decreased statistical convergence as moment order increases).

Since the moments only embody information on the behavior of the PDF integrated over the entire concentration range, the moments themselves are relatively insensitive to the precise PDF shape. The most difficult test for a model concentration PDF is to predict accurately the quantiles over the entire concentration range (e.g. from the extreme lower to upper tails). A particular concentration quantile of interest which can be derived from the PDF is the peak-to-mean concentration ratio, Ψ_{99}/C, where Ψ_{99} is the quantile of order 0.99 (i.e. Ψ_{99} is the concentration which is exceeded for 1% of the time). Figure 3 shows a scatterplot of the peak-to-mean concentration ratio plotted against $\langle(\Psi/C)^2\rangle$. Predictions of the relationship between Ψ_{99}/C and $\langle(\Psi/C)^2\rangle$ provided by the clipped-gamma and clipped-normal PDFs are also shown in Fig. 3. With a judgement based on a visual comparison only, and an emphasis on overall shape and magnitude, the predicted Ψ_{99}/C values from the clipped-gamma PDF appear to be in better agreement with the measured values than those provided by the clipped-normal PDF. In particular, the

location and height of the maximum in the relationship between Ψ_{99}/C and $\langle(\Psi/C)^2\rangle$ appear to be well represented by the clipped-gamma PDF. Again, note that the data in Fig. 3 for $\langle(\Psi/C)^2\rangle \gtrsim 100$ are rather scattered. This increased scatter reflects the decreased statistical convergence as $\langle(\Psi/C)^2\rangle$ increases (e.g. near the plume fringes).

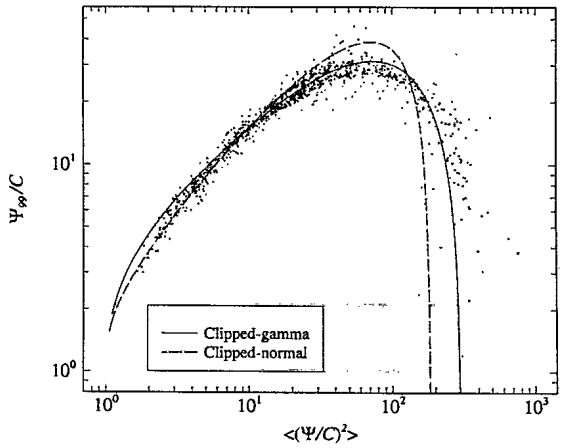

Fig. 3. Double-logarithmic scatterplot of the peak-to-mean concentration ratio, Ψ_{99}/C, plotted against the normalized second concentration moment, $\langle(\Psi/C)^2\rangle$. The plot also includes the predictions of the relationship generated by the clipped-gamma and clipped-normal PDFs.

Next, we compare the shape of the model probability distributions to the observed concentration data at a number of plume locations. Figure 4 presents measured CDFs of the normalized concentration obtained from various plume locations. The clipped-gamma and clipped-normal distributions, with the same fluctuation intensity $i[\langle(\Psi/C)^2\rangle = i^2 + 1]$ as the measured result, are also shown in Fig. 4 for comparison. Generally, the fits of the clipped-normal distribution to the present concentration data over a wide range of conditions were found to be not as good as for the clipped-gamma distribution. As noted earlier (see Fig. 1), γ is underpredicted by the clipped-normal distribution [i.e. the probability of zero concentration, $(1-\gamma)$, which for all practical purposes can be taken to be the probability that $\Psi < 0.01C$, appears in many cases in Fig. 4 to be significantly smaller than that predicted by the clipped-normal distribution]. In consequence, the lower tail of the clipped-normal PDF is generally heavier than that for the observed concentration probability distribution.

Generally, for a number of practical applications, it is the largest peak concentrations that are of greatest

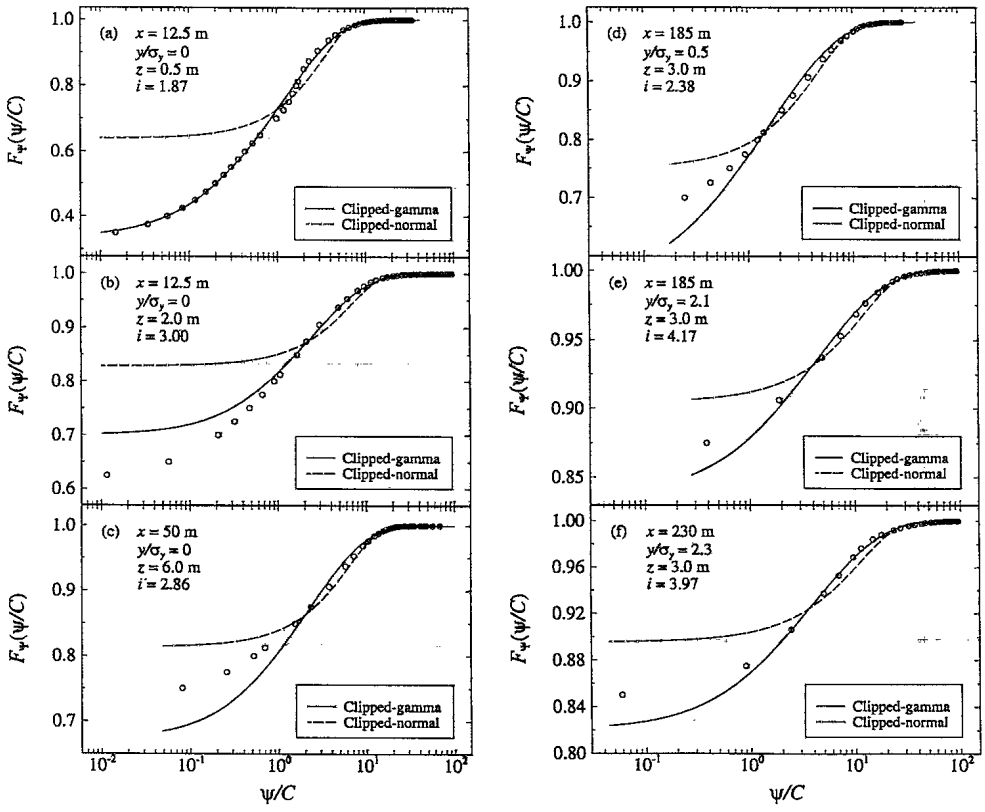

Fig. 4. Cumulative distribution function (CDF), $F_\Psi(\psi)$, of the concentration measured at various receptor positions in a plume dispersing in the atmosphere. The downwind distance, x; normalized crosswind distance, y/σ_y; vertical height, z; and, fluctuation intensity, i, which identify the receptor location are given in each panel. The plot also includes the curves provided by the clipped-gamma and clipped-normal CDFs which were generated using the observed fluctuation intensity at each plume location.

interest. Hence, since it is the prediction of the likelihood of extreme events that is important in the hazard assessment of flammable or toxic gas releases, it is important to examine also the upper tail of the concentration distribution. To that end, we present in Fig. 5 the exceedance probability distributions [viz., the complement of the CDF, $1 - F_\Psi(\psi)$] for the same plume locations depicted in Fig. 4. The exceedance probability distributions have been plotted on a logarithmic scale in order to emphasize the upper tail. Figure 5 indicates that the clipped-gamma distribution predicts the upper tail remarkably well. In contrast, the clipped-normal distribution tends to underpredict the probability of occurrence of large concentrations (see, also Fig. 2 where the clipped-normal PDF is seen to underpredict the concentration moments). This underprediction of the upper tail of the concentration PDF is quite significant in some cases, particularly at those plume locations where the distribution of contaminant material is developing increasing structure and becoming increasingly patchy, but before the much slower process of molecular mixing is complete (e.g. in the turbulent-convective regime of plume development). Here, the concentration PDFs tend to exhibit exponential-like tails rather than more nearly Gaussian ones.

A well-documented set of concentration CDFs, that illustrates the evolution in the CDF form as the plume develops downwind, has been provided by Sykes and Henn (1992). These concentration CDFs have been obtained from a large-eddy simulation (LES) of turbulent plume dispersion from a localized source of passive tracer. The published CDF data [cf. Fig. 14 in Sykes and Henn (1992)] have been reproduced in Fig. 6 along with a comparison with the clipped-gamma and clipped-normal distributions. The model distributions were chosen to match the observed fluctuation intensity. Near the source (e.g. at $x/H = 0.5$, where x is downwind distance and H is boundary-layer height), the plume begins as an instantaneously smooth distribution of contaminant material, and the clipped-normal distribution is seen to agree well with the measured concentration distribution. Here, the only source of fluctuations is the meandering motion of the plume [viz., at this early location, there has not been enough time for the turbulent convective motions to stretch and fold the marked fluid parcels into a complex and highly intertwined set of contorted sheet-like and (or) strand-like structures to produce a significant fine-scale texture in the plume], and if this is true, it seems unlikely for the closure assumptions of

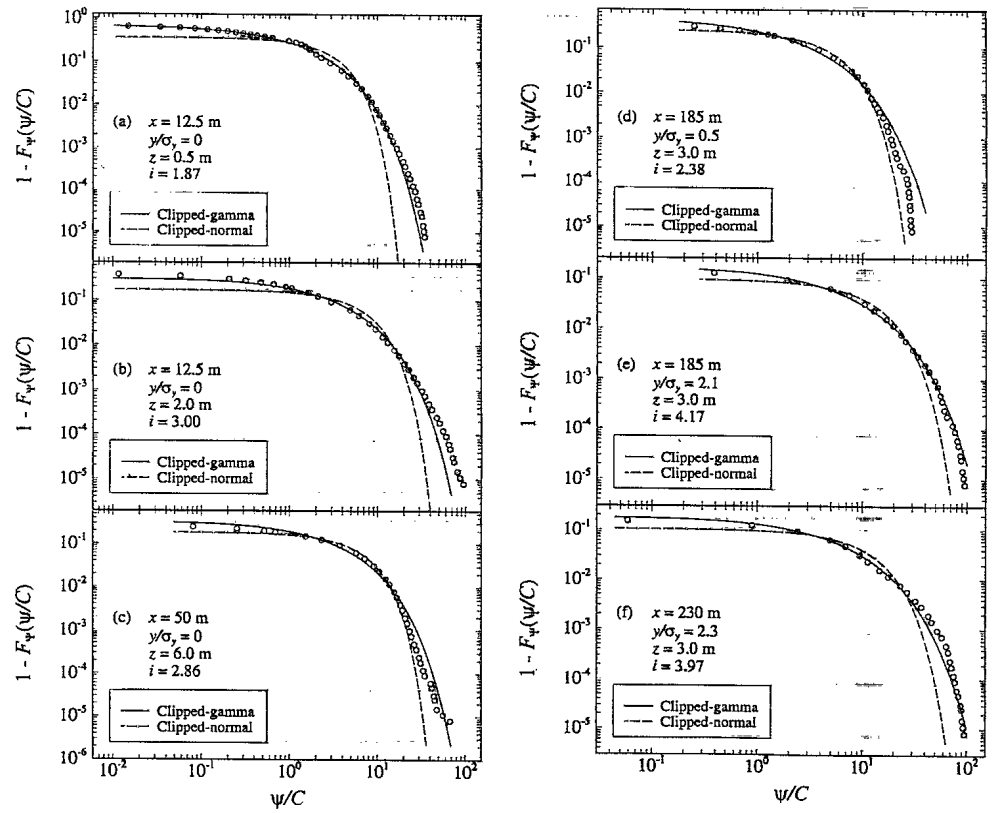

Fig. 5. Exceedance distribution function (EDF), $(1 - F_\Psi(\psi))$, of the concentration measured at the same receptor positions in the plume as documented in Fig. 4. The downwind distance, x; normalized crosswind distance, y/σ_y; vertical height, z; and fluctuation intensity, i, which identify the receptor location are given in each panel. The plot also includes the curves provided by the clipped-gamma and clipped-normal EDFs which were generated using the observed fluctuation intensity at each plume location.

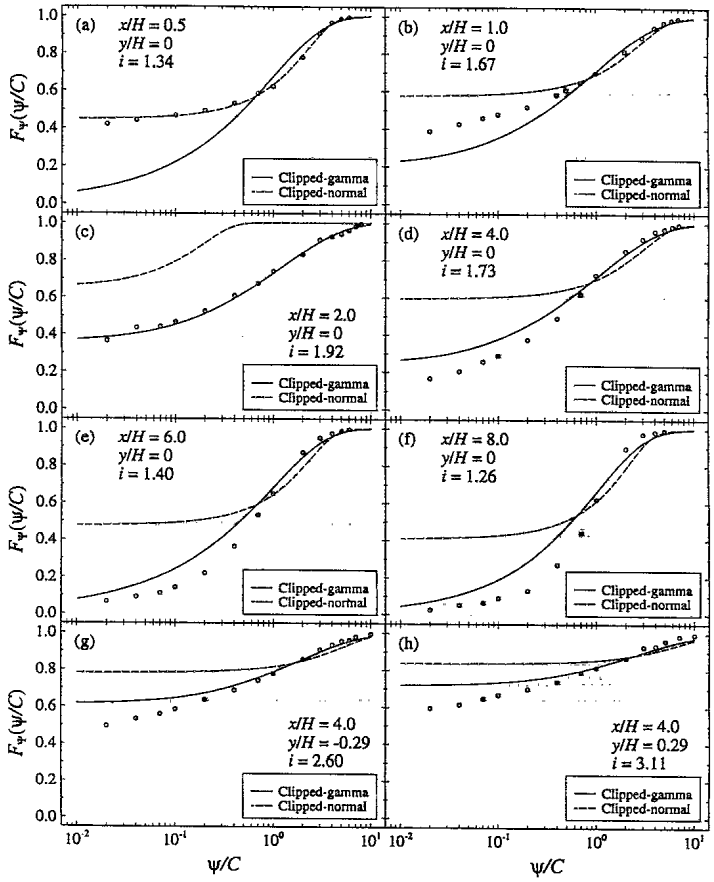

Fig. 6. Cumulative distribution function (CDF), $F_\psi(\psi)$, of the LES plume concentration data at various locations. All locations are taken at source height. The normalized downwind distance, x/H, where H is the boundary-layer height; normalized crosswind distance, y/H; and, fluctuation intensity, i, which identify the receptor location are given in each panel. The plot also includes the curves provided by the clipped-gamma and clipped-normal CDFs which were generated using the observed fluctuation intensity at each plume location.

equations (9) and (10b) to be valid at this early stage of plume development. However, further downwind, when turbulent mixing processes have had more time to act and the internal fluctuations are progressively becoming more important as a source of fluctuations, the clipped-gamma PDF generally fits the LES concentration data better than the clipped-normal PDF.

Sykes and Henn (1992) suggested that the concentration PDF transitions toward a lognormal distribution as the plume progressively develops downwind toward an approximately well-mixed state. In this regime of plume development, the intermittency effect and the presence of large concentrations have been reduced by continued mixing; also, the instantaneous concentration gradients throughout the internal plume have been reduced so that molecular mixing becomes increasingly slower. The transition to the lognormal distribution in the well-mixed state of plume development was interpreted by Sykes and Henn as the result of the cumulative effect of a large number of turbulent mixing events [proposed originally by Csanady (1973)] that become established once the plume has had the time to travel far enough downwind (i.e. several large-eddy time scales). Alternatively, the clipped-gamma PDF model [cf. equation (14)], based as such on the closure assumptions embodied in equations (9) and (10b), predicts that with continued mixing so that $\gamma \to 1$ (i.e. $\lambda \to 0$), the equilibrium PDF in the well-mixed state transitions toward the familiar gamma PDF. Indeed, the results of Fig. 6 do appear to provide some support for the latter prediction. In particular, the concentration CDF obtained from the LES data appears to be better represented by the gamma distribution than by the lognormal distribution [e.g. cf. Fig. 6 with Fig. 14 in Sykes and Henn (1992), the latter of which contains the lognormal distribution for comparison]. Finally, experimental measurements by Deardorff and Willis (1988) appear to suggest that the gamma PDF for plume concentrations emerges in the turbulent-diffusive regime of plume development when the instantaneous plume has grown to fill the mean-plume width, and a self-similar state is reached in which the contaminant parcels are "well-mixed" in the plume.

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5. CON

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Simple closure assum
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and molecular diffusion) on plume concentrations. These closure assumptions have been used to derive the functional form of the one-point concentration PDF, and results in a left-shifted and clipped-gamma PDF. This scheme provides a basis for the physical interpretation of the concentration PDF form.

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Utilization of a simple parametric expression for the observed relationship between γ and $\langle(\Psi/C)^2\rangle$ allows the 3 parameters of the clipped-gamma PDF to be completely specified from a knowledge of the mean and mean-square concentrations, only. This feature is attractive because use of the present model concentration PDF allows estimates of various concentration fluctuation statistics to be obtained simply from parameters that are normally predicted by a modern atmospheric dispersion model (e.g. mean concentration and concentration variance).

Comparison of the clipped-gamma PDF with some atmospheric and LES concentration data shows that it provides a good representation of the first eight concentration moments, the peak-to-mean concentration ratio, the general distribution shape, and the form of both the lower and upper tails of the concentration distribution over a wide range of locations in a dispersing plume. Furthermore, the clipped-gamma distribution appears to fit the concentration data rather better than the clipped-normal distribution and, in particular, appears to be more flexible in fitting the full range of observed PDF behavior in a dispersing plume. Finally, the clipped-gamma distribution asymptotes to the familiar gamma distribution in the limit $\gamma \to 1$, so the model predicts that the gamma probability law is obtained for plume concentrations in the approximately well-mixed state of development (e.g. when the plume has traveled far enough so that the continued molecular mixing process has produced a plume with little or no intermittency).

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