# COMMENTS ON "RELATIONSHIPS BETWEEN HIGHER MOMENTS OF CONCENTRATION AND OF DOSE IN TURBULENT DISPERSION"

*Letter to the Editor*

EUGENE YEE and R. CHAN

*Defence Research Establishment Suffield, P.O. Box 4000, Medicine Hat, Alberta, T1A 8K6 Canada*

(Received in final form 2 July, 1996)

Mole and Clarke (1995, hereafter MC) demonstrated that there was a collapse in plume concentration data when certain normalized higher moments of the fluctuating concentration were plotted against each other. In particular, MC demonstrated that the concentration kurtosis and skewness exhibit a characteristic collapse onto a single curve when plotted against each other, as did the normalized 5th moment of concentration (super-skewness) against either skewness or kurtosis. This observation has an important implication with regard to the parametrization of the concentration probability density function (PDF); namely, the concentration PDF requires the specification of at most one shape parameter (viz., three parameters in total including those for location and scale) for its proper representation over a wide range of plume positions. Although MC investigated the relationship between skewness and kurtosis (viz., between the normalized third and fourth moments), they did not consider the next lower-order relationship between fluctuation intensity (i.e., standard deviation of concentration divided by mean concentration) and skewness (viz., between the normalized second and third moments). Nevertheless, MC remarked that when the normalized higher moments (e.g., kurtosis $K$, etc.) were plotted against the normalized second moment (i.e., fluctuation intensity, $i$), **no collapse** onto a single curve was observed [cf. Figure 2(a) in MC where the plot of $K$ versus $i$ exhibits considerable scatter]. The latter observation, if true, must also necessarily imply that the concentration PDF requires the specification of a minimum of three parameters (viz., location, scale, and shape; or, equivalently, mean, variance, skewness) for its proper characterization. Indeed, if the concentration PDF can be represented by only two parameters (e.g., location and scale), then these parameters (degrees of freedom) can be uniquely prescribed using only the first two integral moments of concentration. Once so prescribed, the normalized higher concentration moments (e.g., $S$, $K$, etc.) can be written as a function of $i$ only (because one degree of freedom has been eliminated in the normalization), hence implying that plotting any higher moment (e.g., $K$, etc.) against $i$ should collapse the data.

Most current models of atmospheric dispersion can predict only the mean concentration. However, in recent years, some advanced dispersion models have been developed that predict both the mean concentration and mean-square concentra-

*Boundary-Layer Meteorology* **82:** 341–351, 1997.

tion (or, equivalently, concentration variance) using either a second-order closure model for the concentration variance (Sykes et al., 1984; Sykes et al., 1986; Sykes et al., 1993), or a two-particle Lagrangian stochastic model based on the adoption of various models of fluid particle motion (Durbin, 1980; Thomson, 1990; Borgas and Sawford, 1994). Because the mean concentration and concentration variance do not usually provide an adequate statistical description of the concentration fluctuations, it is frequently required to specify the form of a concentration PDF that combines all the statistical moments into a single function. Obviously, for the practical purpose of model parametrization, the concentration PDF must be specified by two independent parameters only, since this then allows the PDF form to be completely defined using only the predicted concentration mean and variance from an atmospheric dispersion model. However, as noted above, the results of MC appear to suggest that at least one additional parameter, namely, the third moment of concentration would need to be predicted in order to adequately represent the concentration PDF form in a dispersing plume. In view of the difficulty in predicting even two parameters, it is important to verify or refute experimentally the far-reaching implication of the MC result that, in general, the concentration PDF would require three parameters for its proper representation.

The experimental data, used by MC to test the relationships between the concentration moments, consist of only 28 individual concentration time series obtained from 7 experiments involving four detectors placed at downwind distances of between 5 and 15 m from a steady continuous tracer source (Mole and Jones, 1994). These experiments encompass a rather restrictive range of experimental configurations and atmospheric conditions. In each of these experiments, the source was placed at about 3-m height and the detectors were placed between 3 and 4 m above the ground. The sampling time for the experiments varied from 25 to 80 min. Five of the experiments were conducted under convective conditions, and two were conducted under stably stratified conditions, although turbulence measurements were not available for the quantitative assessment of surface-layer stability.

This correspondence is concerned with using a more comprehensive concentration fluctuation data set covering a much wider range of experimental conditions to test the relationships between the higher moments of concentration. The data were collected in September 1991, November 1992, May 1993, and May 1994 during the cooperative Concentration Fluctuation Experiments (CONFLUX) project whose scientific objective was the detailed study of the fine-scale structure and dynamics in dispersing plumes in relationship with the turbulence structure of the atmosphere (see Yee et al., 1993; Yee et al., 1994; Yee et al., 1995a; Yee et al., 1995b). We extracted 1038 individual concentration time series from a large number of different experiments covering a very wide range of conditions. All the experiments considered here involved the continuous and controlled release of a tracer gas (propylene) into the atmosphere. The measurements were made at distances $x$ of 12.5 to 1000 m from the source, under moderately convective to extremely stable atmospheric conditions in which mechanical turbulence was suppressed by the stable stratification. The concentration time series were measured over a wide range of plume positions in both lateral and vertical cross-sections through the plume—lateral plume positions varied from the mean-plume centerline at $y/\sigma_y = 0$ to the extreme plume fringes at $y/\sigma_y \approx \pm 3.5$, where $y$ is the crosswind distance from the mean-plume centerline and $\sigma_y$ is the mean-plume dispersion; and, vertical plume positions ranged from 0.5 to 16 m. The source was placed at heights above the ground ranging from 1 to 5 m. The sampling time for the concentration data ranged from 16 to 64 min for experiments conducted in September 1991, and 30 or 35 min for experiments conducted in November 1992, May 1993, and May 1994.

We will consider moments of the normalized concentration, $\chi/C$, where $\chi$ is the instantaneous concentration and $C$ is the mean concentration. Let $M_n$ denote the $n$-th moment of the normalized concentration, viz.

$$M_n \equiv \left\langle \left( \frac{\chi}{C} \right)^n \right\rangle, \qquad n \in \mathbf{N}.$$

Here, $\langle \cdots \rangle$ denotes the ensemble average and $\mathbf{N}$ denotes the set of natural numbers. For the case of a steady plume dispersing in a stationary atmosphere considered here, the ensemble average can be replaced by the time average (ergodicity). We present $M_n$ ($n \geq 2$) rather than intensity, $i$, skewness, $S$, and kurtosis, $K$, etc. in order to maintain a consistency in the non-dimensionalization for all higher moments (e.g., whereas $i$ uses the mean concentration to non-dimensionalize the second central moment, $S$, on the other hand, uses the concentration standard deviation to non-dimensionalize the third central moment). In any case, there is a straightforward algebraic relationship between the two sets of normalized moments (Kendall and Stuart, 1977). Figure 1 presents scatterplots of various higher-order concentration moments $M_n$ ($n$ = 3, 4, 5, 6, 7, and 8) plotted against the second-order concentration moment $M_2$. There appears to be more scatter in the plots of $M_n$ versus $M_2$ for the higher values of $n$. This scatter appears to be random, and is most likely attributed to the difficulty of measuring higher moments arising from enhanced sampling errors (viz., the standard errors of the measured moments rapidly increase with order).

Each of the scatterplots in Figure 1 appears to exhibit a collapse of the data onto a single curve. The remarkably systematic dependencies of $M_n$ ($n > 2$) on $M_2$ are all the more interesting because they encompass a very large range in plume positions and atmospheric stratification. In particular, the collapse that is observed in $(M_2, M_3)$ and $(M_2, M_4)$ appears to contradict the results presented by MC. It is noteworthy that the collapse here occurs over four decades in $M_2$; and, the latter range in $M_2$ is considerably more extensive than that covered by the data used in MC. If the lower-order concentration moments [e.g., $(M_2, M_3)$, $(M_2, M_4)$, etc.] do not exhibit any collapse as suggested by the experimental observations used by MC, then we would expect the latter effect to be accentuated with the use of a larger concentration fluctuation data set that encompasses a considerably wider range of experimental configurations and atmospheric conditions. The fact that this is not the case supports the view that the collapse of the lower-order moment relationships is genuine [and, particularly, the collapse observed in $(M_2, M_3)$].

It is of interest to compare the relationships between $M_n$ $(n > 2)$ and $M_2$ shown in Figure 1 with three simple models for these relationships that have been proposed in the literature. To this purpose, we will consider the following three models: the exponential PDF proposed by Barry (1977); the clipped-normal PDF suggested by Lewellen and Sykes (1986); and the $\alpha$-$\beta$ model proposed by Chatwin and Sullivan (1990).

The exponential PDF for intermittent concentrations is a two-parameter PDF that has the following form:

$$f(\chi) = \frac{\gamma^2}{C} \exp\left(-\frac{\gamma\chi}{C}\right) + (1-\gamma)\delta(\chi), \tag{1}$$

where $\delta(\chi)$ is the Dirac delta function, and $\gamma$ is the intermittency factor [viz., the discrete part of the distribution in Equation (1) arises from the fact that $\chi$ admits the value 0 with probability $(1-\gamma)$]. The concentration moments for the exponential distribution satisfy the following relationship:

$$\left\langle\left(\frac{\chi}{C}\right)^n\right\rangle = \frac{n}{2}\left\langle\left(\frac{\chi}{C}\right)^{n-1}\right\rangle\left\langle\left(\frac{\chi}{C}\right)^2\right\rangle, \qquad n = 3, 4, 5, \ldots. \tag{2}$$

The exponential PDF imposes the constraint that $i^2 \equiv \langle(\chi/C)^2\rangle - 1 \geq 1$ (e.g., $i \to 1$ as $\gamma \to 1$), so that Equation (2) is strictly valid only for $\langle(\chi/C)^2\rangle \geq 2$.

The clipped-normal PDF is a two-parameter PDF that is defined as

$$f(\chi) = \frac{1}{\sqrt{2\pi}\sigma_c} \exp\left(-\frac{1}{2}\left(\frac{\chi-\mu_c}{\sigma_c}\right)^2\right) + (1-\gamma)\delta(\chi), \tag{3a}$$

where

$$\gamma \equiv \frac{1}{2}\left(1 + \operatorname{erf}\left(\frac{\mu_c}{\sqrt{2}\sigma_c}\right)\right), \tag{3b}$$

and $\mu_c$ and $\sigma_c$ are location and scale parameters, respectively. In the clipped-normal PDF, a Gaussian PDF is used to model the continuous part of the distribution of $\chi$ for $\chi > 0$, with the probability of negative concentrations being transferred into a delta function at $\chi = 0$ to represent the intermittency effect. In the clipped-normal PDF, the intermittency factor $\gamma$ is uniquely determined as a function of $\mu_c/\sigma_c$. Explicit expressions for the higher-order concentration moments are rather complicated for the clipped-normal PDF, but can be obtained from the following recurrence relationship:

$$\left\langle\left(\frac{\chi}{C}\right)^n\right\rangle = \frac{\Lambda^n}{\sqrt{\pi}} \sum_{k=0}^{n} \binom{n}{k} \phi^k I_{n-k}, \qquad n = 2, 3, 4 \ldots, \tag{4a}$$

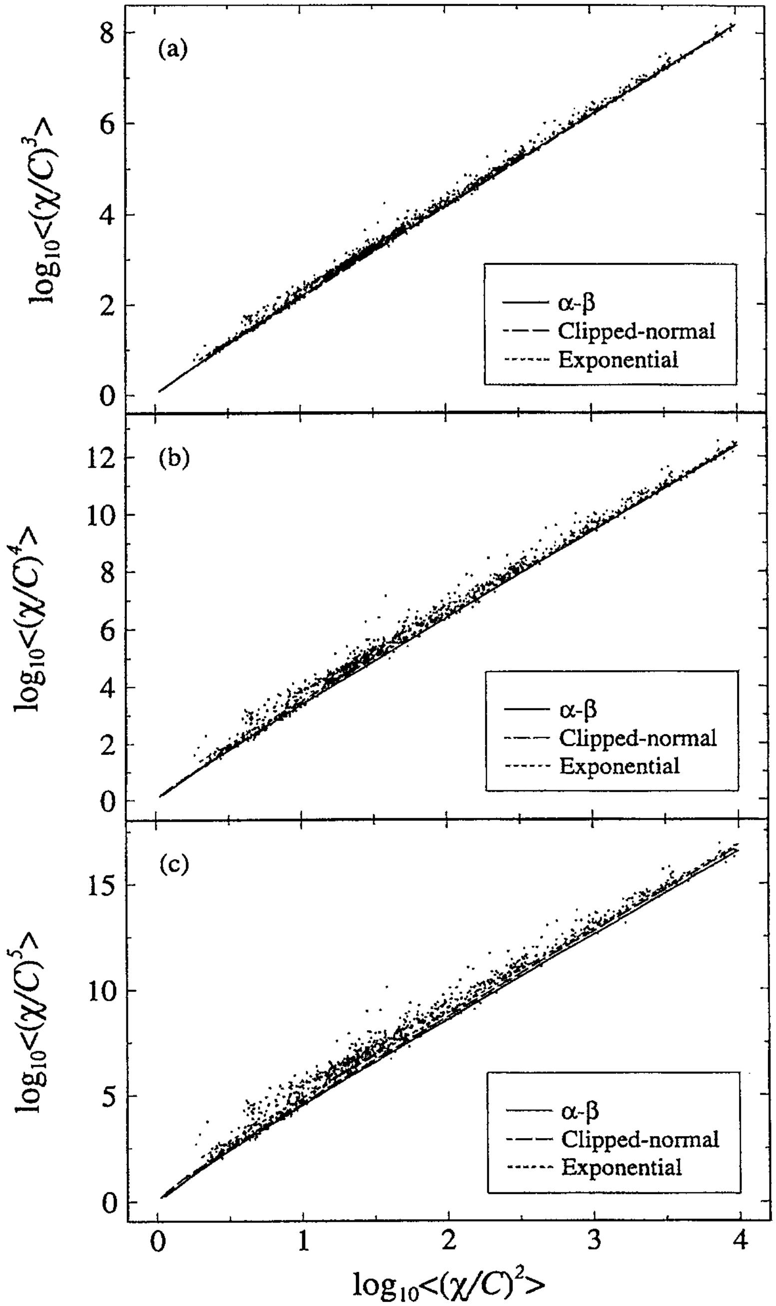


*Figure 1.* Double-logarithmic scatterplots of (a) $M_3$ versus $M_2$; (b) $M_4$ versus $M_2$; (c) $M_5$ versus $M_2$; (d) $M_6$ versus $M_2$; (e) $M_7$ versus $M_2$; and, (f) $M_8$ versus $M_2$, where $M_n \equiv \langle (\chi/C)^n \rangle$. Additional curves show the analytically predicted relationships between these normalized concentration moments provided by the exponential PDF, the clipped-normal PDF, and the $\alpha$-$\beta$ model (with $\beta$ set to 0.64 to optimize the fit with the observed relationship between $M_2$ and $M_3$).

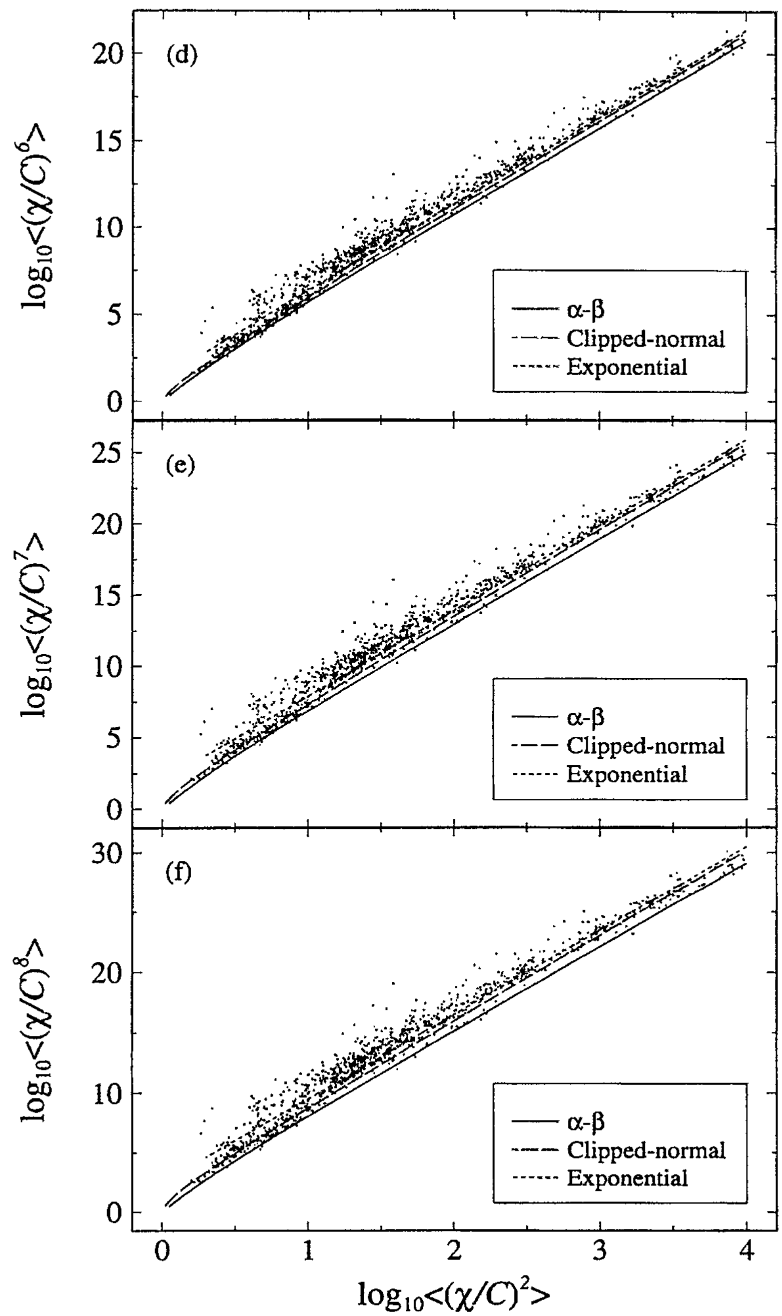


*Figure 1.* Continued.

where $\phi \equiv \mu_c/(\sqrt{2}\sigma_c)$ and

$$\Lambda \equiv 1 \Big/ \left(\gamma\phi + \frac{1}{2\sqrt{\pi}}\exp(-\phi^2)\right). \tag{4b}$$

Furthermore, $I_n$ is determined from the two-term recurrence relation

$$I_n = \frac{1}{2}(-1)^{n-1}\phi^{n-1}\exp(-\phi^2) + \frac{n-1}{2}I_{n-2}, \tag{4c}$$

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

The $\alpha$-$\beta$ model in its simplest form is based on the following three-parameter concentration PDF (Ye, 1995):

$$f(\chi) = \frac{C}{\alpha C_0}\delta(\chi - \chi_1) + \left(1 - \frac{C}{\alpha C_0}\right)\delta(\chi - \chi_2), \tag{5a}$$

where $C_0$ is the maximum value of $C$ at each cross-section (e.g., a local scale for $C$), $\alpha$ and $\beta$ are positive parameters, and

$$\chi_1 \equiv \alpha\beta C_0 + (1-\beta)C, \tag{5b}$$

and

$$\chi_2 \equiv (1-\beta)C. \tag{5c}$$

The concentration PDF model of Equation (5) yields the following relationships for the concentration moments:

$$\left\langle\left(\frac{\chi}{C}\right)^n\right\rangle = (1-\beta)^n\left[\frac{\vartheta}{\alpha}\left(1 + \frac{\beta}{(1-\beta)}\frac{\alpha}{\vartheta}\right)^n - \frac{\vartheta}{\alpha} + 1\right], \tag{6a}$$

where

$$\vartheta \equiv \frac{C}{C_0}, \tag{6b}$$

and

$$\frac{\alpha}{\vartheta} = \frac{1}{\beta^2}\left(\left\langle\left(\frac{\chi}{C}\right)^2\right\rangle - 1\right) + 1. \tag{6c}$$

In the simplest form of the $\alpha$-$\beta$ model, the higher moments are related to the mean concentration $C$ (location parameter) through two parameters, $\alpha$ and $\beta$. In the most general form of the $\alpha$-$\beta$ model, an extra parameter $A_n$ is introduced for each moment higher than the second (i.e., for $n > 2$), although Chatwin and Sullivan (1990) argue $A_n^{1/n}$ is of order unity.

Equations (6a) and (6c) imply that the relationship between $M_2$ and $M_3$ depends explicitly on $\beta$. Because $\beta$ in general depends on plume position and atmospheric conditions, the $\alpha$-$\beta$ model implies that the $(M_2, M_3)$ relationship would not be expected to exhibit a collapse of data onto a single curve in general (e.g., this lack in collapse would simply reflect the spatial and temporal variations of $\beta$ in general). In particular, the parameter $\beta$ in the $\alpha$-$\beta$ model was introduced to account for the dissipation of the concentration variance and, as such, would be expected to decrease with travel time or downwind distance from the source. However, Figure 1(a) suggests that there is a collapse when $M_2$ is plotted against $M_3$, implying a definite functional relationship between these two quantities. Hence, the collapse of the $(M_2, M_3)$ relationship does not appear to support the applicability of the $\alpha$-$\beta$ model. However, the $\alpha$-$\beta$ scheme can be made consistent with the observed relationship between $M_2$ and $M_3$ [cf. Figures 1(a)] if it is assumed that $\beta$ is a constant (e.g., $\beta$ does not vary with plume position or atmospheric conditions). In this case, $\beta$ must be considered simply to be a constant parameter whose value can be fixed to optimize quantitative agreement with the data in Figure 1(a). From this perspective, $\beta$ loses its interpretation as a measure of the amount of concentration variance dissipation arising from molecular diffusion. To that end, a good fit to the measurements in Figure 1(a) can be obtained with $\beta = 0.64$ (which was determined from a least-squares fit of the measured relationship between $M_2$ and $M_3$). Using this value of $\beta$, we plotted the curves given by Equations (6a) and (6c) in Figure 1 for comparison. Even though the $\alpha$-$\beta$ model fits the $(M_2, M_3)$ relationship well owing to the fact that $\beta$ was chosen in this case to give the best fit curve for this model, nevertheless, it appears that the $\alpha$-$\beta$ model curves consistently underestimate the observed relationship between $M_n$ ($n \geq 4$) and $M_2$, the more so with increasing moment order $n$.

We have also plotted in Figure 1, the moment relationships predicted by the exponential and clipped-normal PDFs. It is noteworthy that both the exponential and clipped-normal PDFs appear to provide rather good fits to the measurements over the full range of conditions covered by the data. Furthermore, the simple PDF forms appear to generally characterize the concentration moment relationships better than the $\alpha$-$\beta$ model over almost the entire range of $M_2$, despite the fact that the $\alpha$-$\beta$ model required the least-squares fitting (tuning) of a parameter (i.e., $\beta$) in order to optimize quantitative agreement with all the data. Finally, the $\alpha$-$\beta$ model implies a concentration PDF that is not realistic (viz., the model gives a PDF consisting of two delta functions [cf. Equation (5)]). In consequence, while the exponential and clipped-normal PDF models can be used to provide useful estimates of various concentration quantiles required for a number of practical applications, the $\alpha$-$\beta$ model cannot be used to provide such estimates.

Figure 2 shows a plot of kurtosis, $K$, against skewness, $S$, showing an expected collapse onto a universal curve [viz., because plotting any $M_n$ ($n \geq 3$) against $M_2$ collapses the data, it follows that relationships between any pair of normalized moments higher than second order should also exhibit a similar collapse]. Also

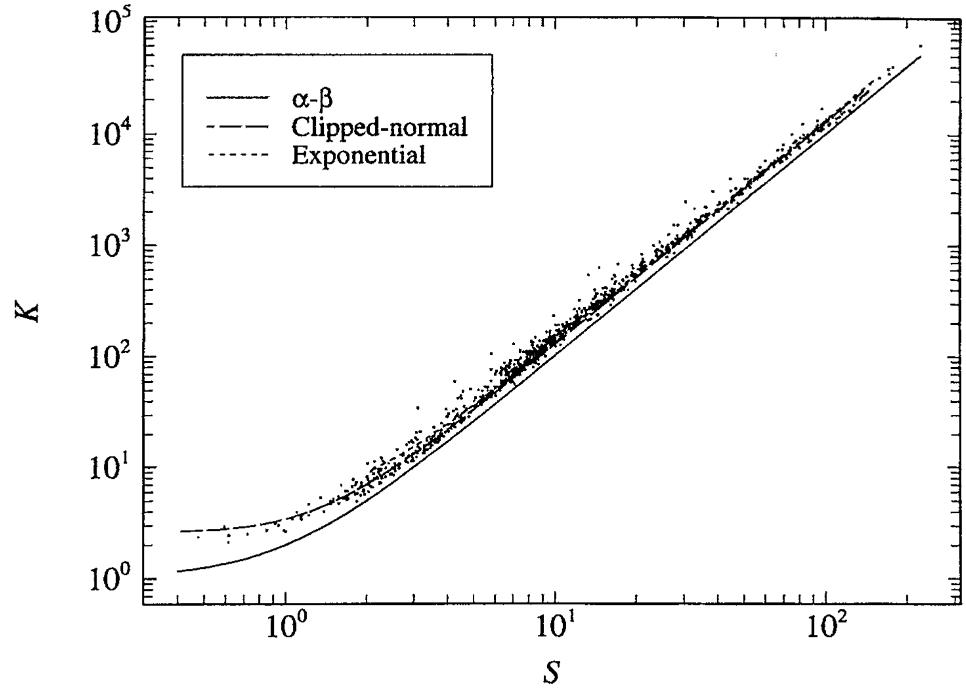


*Figure 2.* Kurtosis, $K$, plotted against skewness, $S$. Additional curves show the analytically predicted relationships between $S$ and $K$ provided by the exponential PDF, the clipped-normal PDF, and the $\alpha$-$\beta$ model.

displayed in Figure 2 are the curves predicted by the exponential and clipped-normal PDFs, as well as the quadratic relationship

$$K = S^2 + 1, \tag{7}$$

predicted by the $\alpha$-$\beta$ model (MC). A key observation in MC is that the relationship in Equation (7) is parameter free and can be used as a check of the consistency of the $\alpha$-$\beta$ model with concentration data, without any reference to the values for $\alpha$ and $\beta$. It was argued in MC that the collapse of concentration data in a plot of $K$ versus $S$ onto a universal curve near the quadratic curve of Equation (7) provides support for the applicability of the $\alpha$-$\beta$ model. However, Figure 3 shows that this observation could, in addition, be explained using some other simple models (paradigms). In particular, note that the exponential and clipped-normal PDF models appear to describe the observed $(S, K)$ relationship better than that given by the $\alpha$-$\beta$ model. In fact, the quadratic curve given by Equation (7) consistently underestimates the observed relationship between $S$ and $K$ (and, in fact, the same effect was noted in MC). Indeed, it is known that for all probability distributions, $K \geq S^2 + 1$, with equality attained only when the PDF reduces to the weighted sum of two delta functions as for the $\alpha$-$\beta$ model [cf. Appendix in MC]. In consequence, the $\alpha$-$\beta$

model, which corresponds to a physically unacceptable two-state PDF given by Equation (5a), generally provides a poorer fit to the $(S, K)$ relationship than that provided by some very simple PDF models such as the exponential or clipped-normal distributions that predict a continuous range of values for $\chi$ (arising, as such, from the molecular diffusive action).

In summary, the present experimental data clearly show that there are strong correlations between the various normalized higher-order concentration moments and the normalized second-order concentration moment. The latter observation suggests that the concentration PDF over a wide range of receptor positions in a dispersing plume and under a variety of meteorological conditions can be represented (approximately or better) by a functional form that can be specified by two parameters, one for location (e.g., mean concentration) and one for scale (e.g., concentration standard deviation).

Presently, the exact concentration PDF form cannot be derived rigorously from the advection-diffusion equation without invoking some form of closure hypothesis. Nevertheless, the exponential and clipped-normal PDF forms (both of which are fully specified, including the intermittency effect, by two parameters) appear to provide good and useful approximations to the exact (but unknown) concentration PDF form. Furthermore, the concentration moment relationships appear to be better represented by the exponential and clipped-normal PDFs than by the $\alpha$-$\beta$ model. There exists also independent evidence supporting the observation that the exponential and clipped-normal distributions generally provide good fits to concentration data (e.g., Hanna, 1984; Sawford, 1987; Dinar et al., 1988; Mylne and Mason, 1991). In particular, it has been found that the exponential PDF appears to fit better in the regime of plume development dominated by meandering, whereas the clipped-normal PDF appears to provide the best agreement in the regime of plume development where the internal plume structure is becoming dominant (Mylne and Mason, 1991). Other PDF models involving three or more parameters have been suggested by a number of researchers (e.g., Yee, 1990; Derksen and Sullivan, 1990; Lewis and Chatwin, 1995; Sullivan and Ye, 1995; among others) and may be capable of fitting concentration data better because of their greater mathematical flexibility. These models are probably of little use for practical applications since they require the prediction of more parameters to specify the PDF than can be achieved presently using even the most advanced atmospheric dispersion models [e.g., higher-order closure models (Sykes et al., 1984), or stochastic models for the motion of particle pairs (Durbin, 1980)].

## References

Barry, P. J.: 1977, 'Stochastic Properties of Atmospheric Diffusivity', in *Sulfur and its Inorganic Derivatives in the Canadian Environment*, National Research Council of Canada, pp. 313–358.

Borgas, M. L. and Sawford, B. L.: 1994, 'A Family of Stochastic Models for Two-Particle Dispersion in Isotropic Homogeneous Stationary Turbulence', *J. Fluid Mech.* **279**, 66–99.

Chatwin, P. C. and Sullivan, P. J.: 1990, 'A Simple and Unifying Physical Interpretation of Scalar Fluctuation Measurements from Many Turbulent Shear Flows', *J. Fluid Mech.* **212**, 533–556.

Derksen, R. W. and Sullivan, P. J.: 1990, 'Moment Approximations for Probability Density Functions', *Combust. Flame* **81**, 378–391.

Dinar, N., Kaplan, H., and Kleiman, M: 1988, 'Characterization of Concentration Fluctuations of a Surface Plume in a Neutral Boundary Layer', *Boundary-Layer Meteorol.* **45**, 157–175.

Durbin, P. A.: 1980, 'A Stochastic Model of Two-Particle Dispersion and Concentration Fluctuations in Homogeneous Turbulence', *J. Fluid Mech.* **100**, 279–302.

Hanna, S. R.: 1984, 'The Exponential Probability Density Function and Concentration Fluctuations in Smoke Plumes', *Boundary-Layer Meteorol.* **29**, 361–375.

Kendall, M. G. and Stuart, A.: 1977, *The Advanced Theory of Statistics, Vol. 1, Distribution Theory*, Charles Griffin, London.

Lewellen, W. S., and Sykes, R. I.: 1986, 'Analysis of Concentration Fluctuations from LIDAR Observations of Atmospheric Plumes', *J. Clim. and Appl. Meteorol.* **25**, 1145–1154.

Lewis, D. M. and Chatwin, P. C.: 1995, 'A New Model Probability Density Function for Contaminants Dispersing in the Atmosphere', *Environmetrics* **6**, 583–593.

Mole, N. and Clarke, E. D.: 1995, 'Relationships between Higher Moments of Concentration and of Dose in Turbulent Dispersion', *Boundary-Layer Meteorol.* **73**, 35–52.

Mole, N. and Jones, C. D.: 1994, 'Concentration Fluctuation Data From Dispersion Experiments Carried Out in Stable and Unstable Conditions', *Boundary-Layer Meteorol.* **67**, 41–74.

Mylne, K. R. and Mason, P. J.: 1991, 'Concentration Fluctuation Measurements in a Dispersing Plume at a Range of up to 1000 m', *Quart. J. Roy. Meteorol. Soc.* **117**, 177–206.

Sawford, B. L.: 1987, 'Conditional Concentration Statistics for Surface Plumes in the Atmospheric Boundary Layer', *Boundary-Layer Meteorol.* **38**, 209–223.

Sullivan, P. J. and Ye, H.: 1995, 'A Prognosis for the Sudden Release of Contaminant in an Environmental Flow', *Environmetrics* **6**, 627–637.

Sykes, R. I., Parker, S. F., Henn, D. S., and Lewellen, W. S.: 1993, 'Numerical Simulation of ANATEX Tracer Data Using a Turbulent Closure Model for Long-Range Dispersion', *J. Appl. Meteorol.* **32**, 929–947.

Sykes, R. I., Lewellen, W. S., and Parker, S. F.: 1986, 'A Gaussian Plume Model of Atmospheric Dispersion Based on Second-Order Closure', *J. Clim. and Appl. Meteorol.* **25**, 322–331.

Sykes, R. I., Lewellen, W. S., and Parker, S. F.: 1984, 'A Turbulent-Transport Model for Concentration Fluctuations and Fluxes', *J. Fluid Mech.* **139**, 193–218.

Thomson, D. J., 1990: 'A Stochastic Model for the Motion of Particle Pairs in Isotropic High-Reynolds-Number Turbulence, and its Application to the Problem of Concentration Variance', *J. Fluid Mech.* **210**, 113–153.

Ye, H.: 1995, 'A New Statistic for the Contaminant Dilution Process in Turbulent Flows', Ph.D thesis, University of Western Ontario.

Yee, E.: 1990, 'The Shape of the Probability Density Function of Short-Term Concentration Fluctuations of Plumes in the Atmospheric Boundary Layer', *Boundary-Layer Meteorol.* **51**, 269–298.

Yee, E., Chan, R., Kosteniuk, P. R., Chandler, G. M., Biltoft, C. A., and Bowers, J. F.: 1995a, 'Measurements of Level-Crossing Statistics of Concentration Fluctuations in Plumes Dispersing in the Atmospheric Surface Layer', *Boundary-Layer Meteorol.* **73**, 53–90.

Yee, E., Chan, R., Kosteniuk, P. R., Chandler, G. M., Biltoft, C. A., and Bowers, J. F.: 1995b, 'The Vertical Structure of Concentration Fluctuation Statistics in Plumes Dispersing in the Atmospheric Surface Layer', *Boundary-Layer Meteorol.* **76**, 41–67.

Yee, E., Chan, R., Kosteniuk, P. R., Chandler, G. M., Biltoft, C. A., and Bowers, J. F.: 1994, 'Experimental Measurements of Concentration Fluctuations and Scales in a Dispersing Plume in the Atmospheric Surface Layer Obtained Using a Very Fast-Response Concentration Detector', *J. Appl. Meteorol.* **33**, 996–1016.

Yee, E., Kosteniuk, P. R., Chandler, G. M., Biltoft, C. A., and Bowers, J. F.: 1993, 'Statistical Characteristics of Concentration Fluctuations in Dispersing Plumes in the Atmospheric Surface Layer', *Boundary-Layer Meteorol.* **65**, 69–109.

#503190