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"The Exponential Probability Density Function and Concentration Fluctuations in Smoke Plumes"

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THE EXPONENTIAL PROBABILITY DENSITY FUNCTION AND CONCENTRATION FLUCTUATIONS IN SMOKE PLUMES

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Abstract. Observations of 1-s average concentration fluctuations during two trials of a U.S. Army diffusion experiment are presented and compared with model predictions based on an exponential probability density function (pdf). The source is near the surface and concentration monitors are on lines about 30 to 100 m downwind of the source. The observed ratio of the standard deviation to the mean of the concentration fluctuations is about 1.3 on the mean plume axis and 4 to 5 on the mean plume edges. Plume intermittency (fraction of non-zero readings) is about 50% on the mean plume axis and 10% on the mean plume edges. A meandering plume model is combined with an exponential pdf assumption to produce predictions of the intermittency and the standard deviation of the concentration fluctuations that are within 20% of the observations.

1. Introduction

This paper contains an analysis of observations of the standard deviation of concentration fluctuations from U.S. Army experiments during Smoke Week III. In that study, 1-s average concentrations of smoke were observed on lines of monitors about 30 to 100 m downwind of a near-surface source. Many military and environmental applications require knowledge of the total distribution of concentrations in order to answer questions regarding toxicity, flammability, and visibility of smoke plumes. It is not enough to be able to predict the mean concentration, which has been the sole emphasis of most previous studies (e.g., all EPA dispersion models), because the standard deviation is typically at least as large as the mean. In most cases the probability density function of concentration observations may be approximated by a single distribution such as the log-normal or exponential, which implies that the entire distribution can be determined once the mean and standard deviation are known (Larsen, 1970; Barry, 1977).

The past decade has seen a great increase in activity in both monitoring and modeling concentration fluctuations in smoke plumes in the atmosphere. The intensity of short-time average concentration fluctuations, $\sigma_c/\overline{C}$, is consistently observed in field experiments to be of order unity on the mean plume axis and of order 10 on the mean plume edges. The parameters σ_c and $\overline{C}$ are defined to be the standard deviation and mean of the concentration time series at a fixed point. Plume intermittency, I, is defined as the fraction of time that non-zero concentrations are observed. Field studies at small distances downwind show that I is typically 0.5 near the plume axis and less than 0.1 on the mean plume edges.

Several models are available for estimating $\sigma_c/\overline{C}$. These modeling approaches cover

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a wide variety of diffusion theories, ranging from the meandering plume model proposed by Hilst (1957) and Gifford (1959) to the two-particle Lagrangian Monte Carlo model proposed by Durbin (1980) and Sawford (1983). In all cases the results are highly dependent upon proper estimation of the Lagrangian time scale, T_L, since fluctuations are dominated by meandering for travel time t less than T_L and are dominated by internal turbulence for t greater than T_L. Hanna (1984) finds that T_L is not well-known for mesoscale atmospheric turbulence in the boundary layer, often resulting in poor agreement between predicted and observed $\sigma_c/\overline{C}$. If the modeler is allowed to 'tune' T_L, much better agreement is obtained. Another source of model error is a failure to account properly for the initial effects of the finite source, which tend to decrease $\sigma_c/\overline{C}$.

2. Data

During the Smoke Week III Experiment at Eglin Air Force Base, Florida, the U.S. Army tested several types of munitions and sampling devices (Sutherland *et al.*, 1981). We selected two tests in which fog oil was released from a smoke generator at a height of 2 m, and the release rate was constant over a duration of about 5 minutes (R.A. Sutherland, private communication). One-sec average concentrations were observed by aerosol photometers on two lines of monitors, as illustrated in Figure 1. The source was about 100 m from the far line of monitors in Test 2 and about 60 m from the far line of monitors in Test 4. Our analysis emphasizes the far line of monitors since the spacing of the monitors is less and it is better able to resolve the plume than the near line of monitors. The time series of concentration observations at each monitor illustrate both meandering (intermittency) and in-plume fluctuations, as shown in Figure 2. When the plume is present at the monitor, it is highly turbulent. Figures 3 and 4 show the crosswind variation of the concentration statistics over the far line of monitors for Trials 2 and 4, respectively. Because the 'plume' does have a beginning and end in these trials, we consider data only from the time that marks the first significant impact

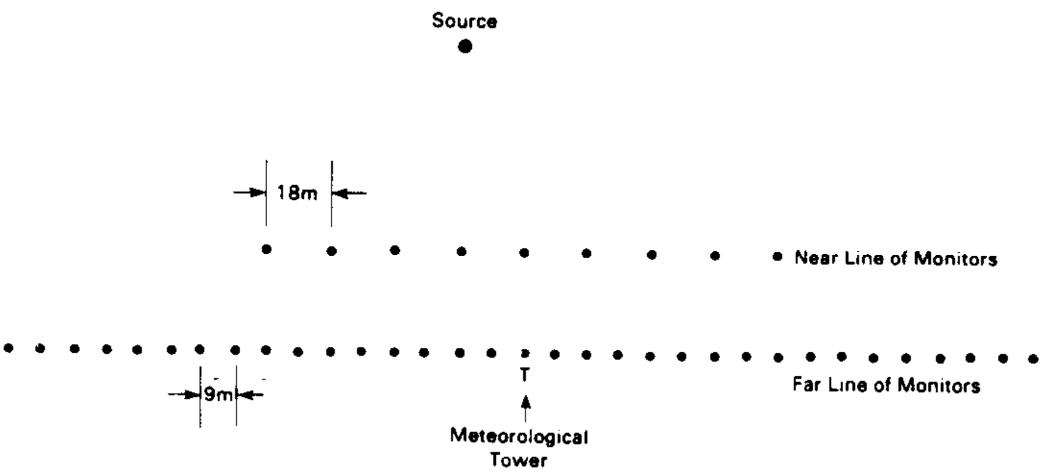

Fig. 1. Monitor array for Smoke Week III experiments. The source location is shown for Trial 2.

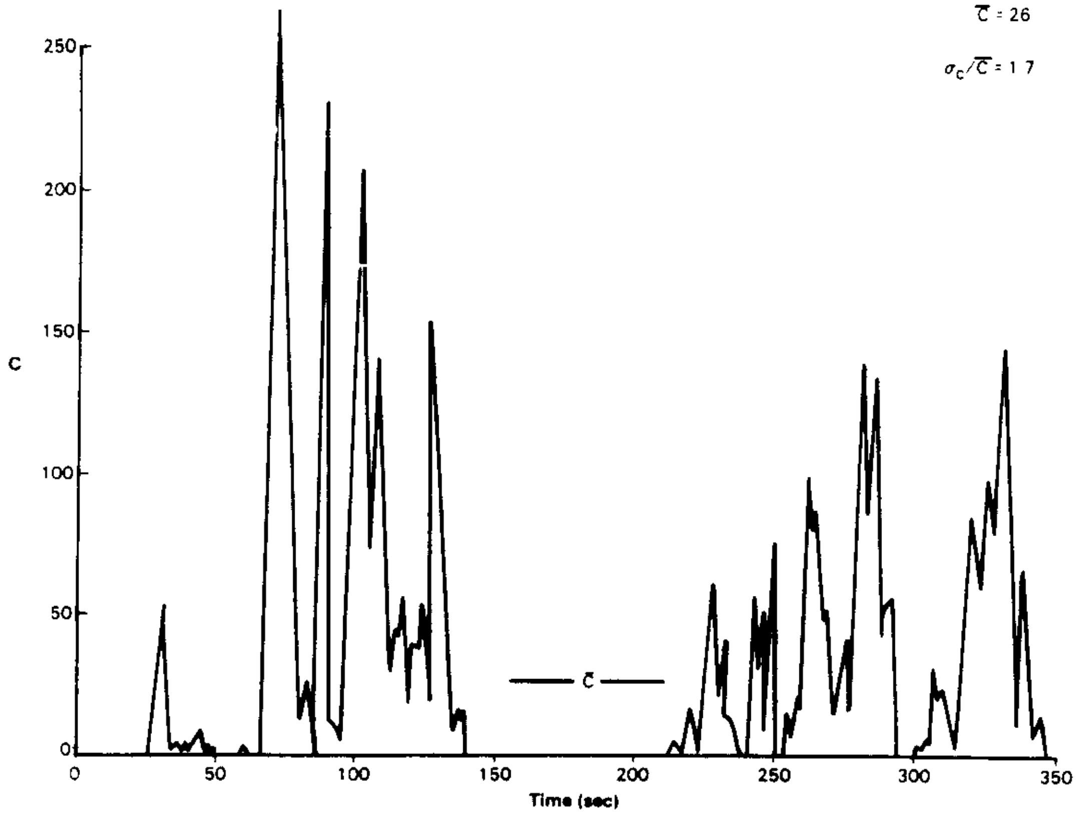

Fig. 2. Time series of concentration for a monitor near the mean plume center in Trial 2.

($C > 15$) anywhere on the arc to the time that marks the last impact. Also, because spurious values of C equal to 1 or 2 appeared randomly during the 'clean air' periods, we defined a threshold of 3 for plume intermittency (I) calculations.

Figures 3 and 4 contain observed values of $\overline{C}$, $\sigma_c/\overline{C}$, and I. It can be seen that the shapes of the curves from the two trials are very similar, with an effective σ_y for the mean concentration curve of about 25 m for Trial 2 and 18 m for Trial 4. It is interesting that intermittency I equals only about 0.5 to 0.6 at the center of the mean plume. That is, the plume is absent almost half the time on the mean plume centerline. At crosswind distances of two standard deviations from the center of the mean plume, the plume is absent over 90% of the time. The variation of concentration fluctuation intensity, $\sigma_c/\overline{C}$, is fairly smooth in both trials, ranging from about 1.3 on the mean plume centerline to about 4 or 5 at crosswind distances of two standard deviations.

The crosswind integrated (CWI) concentrations were also analyzed for these arcs, giving $\sigma_c/\overline{C}$ (CWI) equal to 1.06 for Trial 2 and 0.80 for Trial 4. These numbers are less than those for the point observations because the variation in the y direction has been averaged out.

The following meteorological observations were made on a tower near the middle of the far line of monitors. Sampling times were the same as those for the monitoring data.

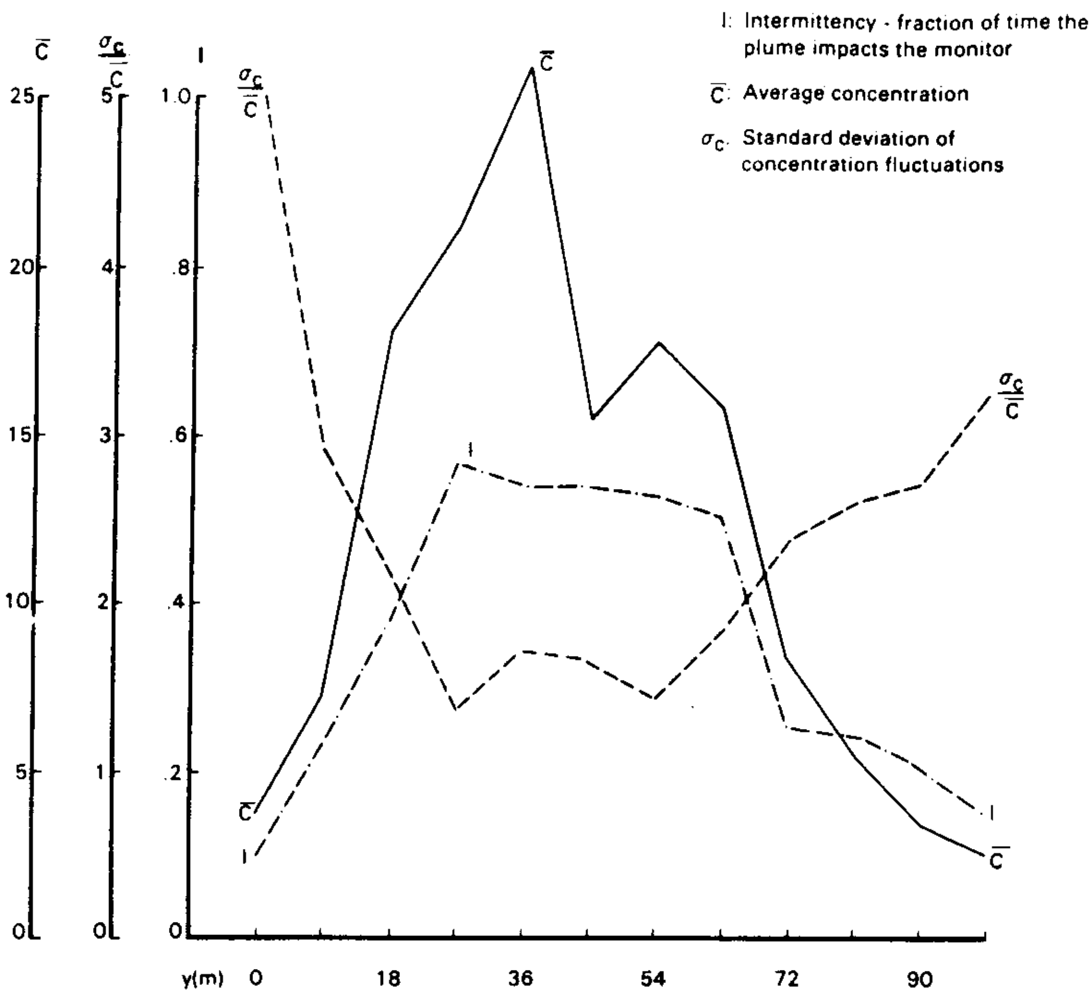

Fig. 3. Trial 2 observations of $\overline{C}$, $\sigma_c/\overline{C}$, and I.

	u(2 m)	σ_θ(2 m)	Pasquill Class
Trial 2	2.7 m s^{-1}	18°	B
Trial 4	4.1 m s^{-1}	13°	C

3. Validity of Exponential PDF

Most atmospheric observations of pollutant or tracer concentrations can be fit by the log-normal (Larsen, 1970) or exponential (Barry, 1977) distributions. Gifford (1972) shows that the log-normal distribution is more likely to be valid in an urban (multiple-source) setting, while the exponential distribution is more likely for isolated point sources. D. J. Wilson (1982) suggests that the log-normal distribution applies to cases

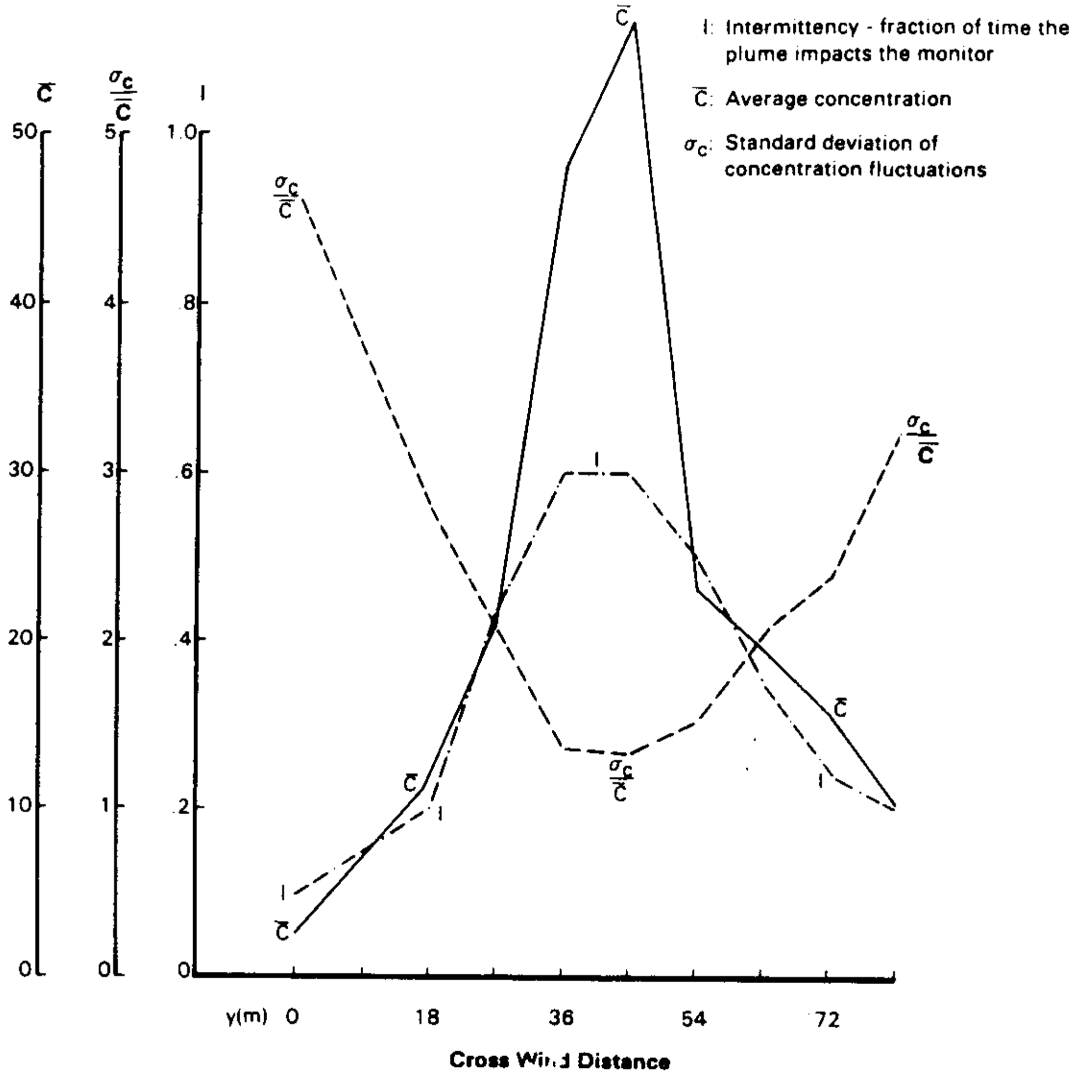

Fig. 4. Trial 4 observations of $\overline{C}$, $\sigma_c/\overline{C}$, and I.

where the concentration observations are well-correlated in time (such as inside a plume), while the exponential distribution applies to cases where the plume more or less randomly passes over a monitor (such as a meandering plume). An exponential distribution would also be expected for hourly-averaged concentrations sampled over a year in the vicinity of an isolated point source. An advantage of the exponential distribution is that it can account explicitly for intermittency, I. Barry (1977) gives the following formulas

$$p(C) = (I^2/\overline{C})\exp(-IC/\overline{C}) + (1 - I)\,\delta(0) \tag{1}$$

$$\sigma_c/\overline{C} = ((2/I) - 1)^{1/2} \tag{2}$$

$$P(C) = 1 - I\exp(-IC/\overline{C}) \tag{3}$$

where p is the probability density function (PDF) and P is the cumulative density function (fraction of data with concentrations equal to or less than C). The mean, $\overline{C}$, and standard deviation, σ_c, include all the zero readings. Intermittency, I, is defined as the fraction of non-zero readings. The parameter $\delta(0)$ is the Dirac delta function, defined such that δ equals infinity at C equal to 0 and equals zero at all other C; and the integral of $\delta(0)$ equals unity at the point C equal to 0.

The function $(1 - P)$ for monitor AP30 in Test 2 is plotted in Figure 5. When C is plotted versus $\log(1 - P)$, an exponential distribution will be evident as a straight line. Despite the curvature, the data in the figure are reasonably close to a straight line over most of their range. The observed ratio $\sigma_c/\overline{C}$ is 1.81, which is within 6% of the predicted ratio $\sigma_c/\overline{C} = 1.71$ from Equation (2), based on an assumed exponential distribution.

An important test of an exponential distribution is whether Equation (2) is satisfied, using observations of $\sigma_c/\overline{C}$ and intermittency I. In Figure 6, observations of $\sigma_c/\overline{C}$ are plotted versus $((2/I) - 1)^{1/2}$ for the monitors in U.S. Army Tests 2 and 4 and for four monitors in an experiment described by Jones (1981). The source height was 1 m in Jones' experiments and the monitors were located at downwind distances of 2, 5, 10, and 15 m. The observed and predicted $\sigma_c/\overline{C}$ values in the figure are in good agreement,

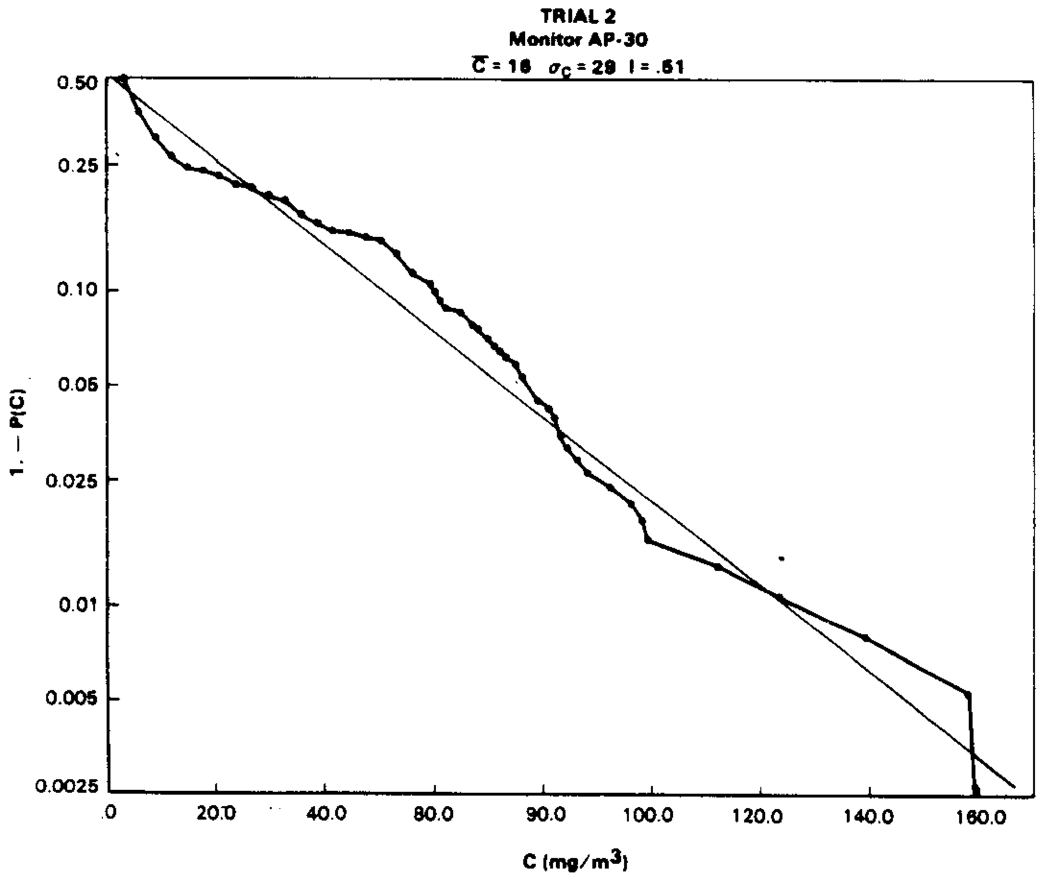

Fig. 5. Observed cumulative distribution function for Trial 2, monitor AP-30. The straight line is fit by eye to the data.

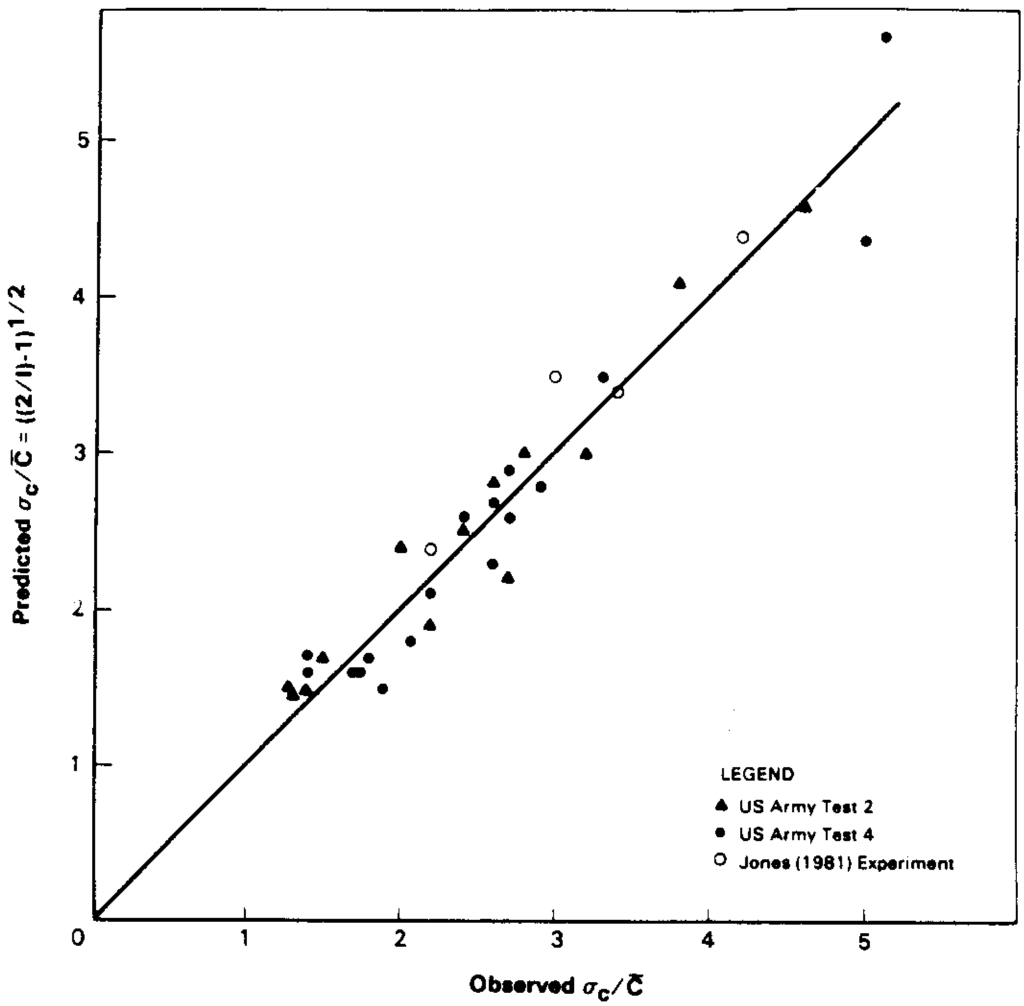

Fig. 6. Comparison of observed $\sigma_c/\overline{C}$ with $\sigma_c/\overline{C}$ predicted from Equation (2) and observations of intermittency I.

with an average bias near zero and a maximum deviation of 20%. There is no systematic difference between the three tests and no variation with the magnitude of observed $\sigma_c/\overline{C}$. We conclude from this analysis that the exponential distribution is valid for short-term concentration fluctuation data, and that this distribution permits simple relations between the intensity of concentration fluctuations and plume intermittency.

4. Models for I and $\sigma_c/\overline{C}$

An extensive review of models for calculating concentration fluctuations was given by Hanna (1984), who showed that the most important parameters for determining intermittency I and concentration fluctuation intensity $\sigma_c/\overline{C}$ at a given travel time t and averaging time T are the source size, σ_0, the Lagrangian time scale, T_L, the Eulerian length scale, L, and the relative crosswind distance from the mean plume axis, y/σ_y. In general, I increases and $\sigma_c/\overline{C}$ decreases as t/T_L increases, as σ_0/L increases, and as y/σ_y decreases. We assume in our analysis that the 1-s averaging time for the U.S. Army data is essentially instantaneous.

If the exponential PDF for C is valid, then $\sigma_c/\overline{C}$ is a one-to-one function of I. We propose separate models for $\sigma_c/\overline{C}$ and I for large and small values of t/T_L. At large values of t/T_L, the plume is no longer bodily moved about by large eddies, and Durbin's (1980) and Sawford's (1983) estimates of $\sigma_c/\overline{C}$ on the mean plume centerline can be combined with Csanady's (1973) expression for the lateral variability of σ_c to give the formula:

$$\sigma_c/\overline{C} = 0.56(L/\sigma_0)^{0.3}\exp(y^2/4\sigma_y^2) \qquad t/T_L > 1 \tag{4}$$

which is valid in the range of L/σ_0 from 14 to 1400. This result is based on calculations for one-dimensional turbulence. From Equations (2) and (4), we can derive the following formula for intermittency, I, at large values of t/T_L:

$$I = 2/(1 + 0.31(L/\sigma_0)^{0.6}\exp(y^2/2\sigma_y^2)) \qquad t/T_L > 1\,. \tag{5}$$

These equations produce plume centerline $\sigma_c/\overline{C}$ in the range from 1.2 to 5 and I in the range from 0.08 to 0.8.

At small values of t/T_L, the plume motion is dominated by eddies larger than the plume, leading to significant meandering. This effect is greatly diminished if the ratio σ_0/L approaches unity or larger. We assume that Hilst's (1957) and Gifford's (1959) meandering plume model is valid, where the instantaneous plume standard deviation is defined as σ_{yI} and the time-mean plume standard deviation is defined as σ_{yT}. For small σ_{yI}/σ_{yT}, the intermittency in one dimension is assumed to be given by the relation:

$$I = (\sigma_{yI}/\sigma_{yI})\exp(-y^2/2\sigma_{yT}^2)\,. \tag{6}$$

This result is obtained through the definition of intermittency by integrating the product of the mean Gaussian plume distribution over y times the instantaneous Gaussian plume distribution centered at a given lateral distance, y, from the mean plume axis. Equation (6) is exact for small σ_{yI}/σ_{yT} but is slightly in error at σ_{yI}/σ_{yT} greater than about 0.5. Substituting I from Equation (6) into Equation (2), we find that the following relation is true for small I:

$$\sigma_c/\overline{C} = (2\sigma_{yT}/\sigma_{yI})^{1/2}\exp(y^2/4\sigma_{yT}^2) \tag{7}$$

which exhibits the same y variation as Csanady's (1973) model employed in Equation (4).

Equation (5) or (6) can be used to estimate the components I_y and I_z of intermittency in a continuous plume. We assume that the total two-dimensional intermittency (as observed by a fixed monitor) is given by the formula

$$I(\text{two dimensional}) = I_y I_z\,. \tag{8}$$

This can be substituted into Equation (2) to give the total $\sigma_c/\overline{C}$.

The only missing piece in this model is a method to estimate σ_{yI}/σ_{yT}. Of the three methods described by Hanna (1984), the simplest one that accounts for initial source size, σ_0, is a formula based on the Langevin equation and suggested by Gifford (1982) and Lee and Stone (1983):

$$\frac{\sigma_{yI}^2}{\sigma_{yI}^2} = \frac{\sigma_0^2 + T' - (1 - e^{-T'}) - 0.5S(1 - e^{-T'})^2}{\sigma_0^2 + T' - (1 - e^{-T'})} \tag{9}$$

where $\sigma_0^2 = \sigma_0^2/2\sigma_v^2 T_L^2$

$$T' = t/T_L$$

$$S(\sigma_0/L) = (L^2/6\sigma_0^2)(3.46(\sigma_0/L) - 1 + \exp(-3.46\,\sigma_0/L)). \tag{10}$$

According to Pasquill (1974), L and T_L are directly related to each other through the formula $T_L = 0.6\,L/\sigma_v$. Analysis of Equation (10) shows that the function S is close to unity for $\sigma_0/L < 0.1$ while σ_{yI}/σ_{yT} is only slightly underestimated by setting $S = 1$. The solutions for seven values of σ_0^2 are plotted in Figure 7, illustrating that the predicted σ_{yI}/σ_{yT} or plume centerline intermittency I for zero source size increases from 0.1 at T' equal to 0.01 to 0.93 at T' equal to 5. A finite source size is seen to have a strong effect on increasing σ_{yI}/σ_{yT} or plume centerline intermittency I at small travel times.

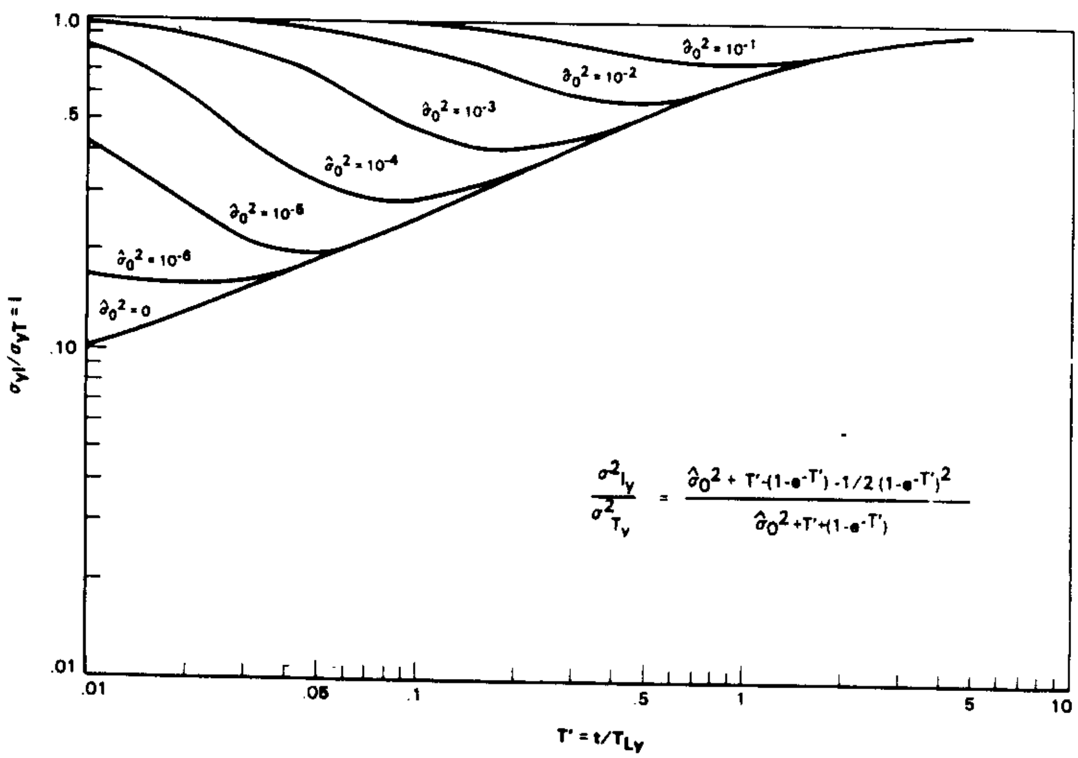

Fig. 7. Solutions to Equation (9) for $S = 1$.

5. Model Application

In order to decide which of the models should be applied to the U.S. Army data, the Lagrangian time scales, T_{Ly} and T_{Lz}, must be estimated. Since both these trials took place during unstable conditions, the following equations are valid (Hanna 1984):

$$T_{Ly} = 0.19\, z_i/w_* \qquad L_{Ey} = 0.3\, z_i \tag{11}$$

$$T_{Lz} = z/w_* \qquad L_{Ez} = z \tag{12}$$

where z_i is the mixing depth and w_* is the convective velocity scale. Since there is not sufficient information to calculate the parameters w_* and z_i directly, we use the identity $w_* = 1.7\, \sigma_v$ and assume that z_i equals its typical climatological value of 1000 m. These assumptions yield the following estimates:

	w_*	T_{Ly}	T_{Lz}	L_{Ey}	L_{Ez}
Trial 2	1.0 m s^{-1}	190 s	1.0 s	300 m	1 m
Trial 4	0.7 m s^{-1}	271 s	1.4 s	300 m	1 m

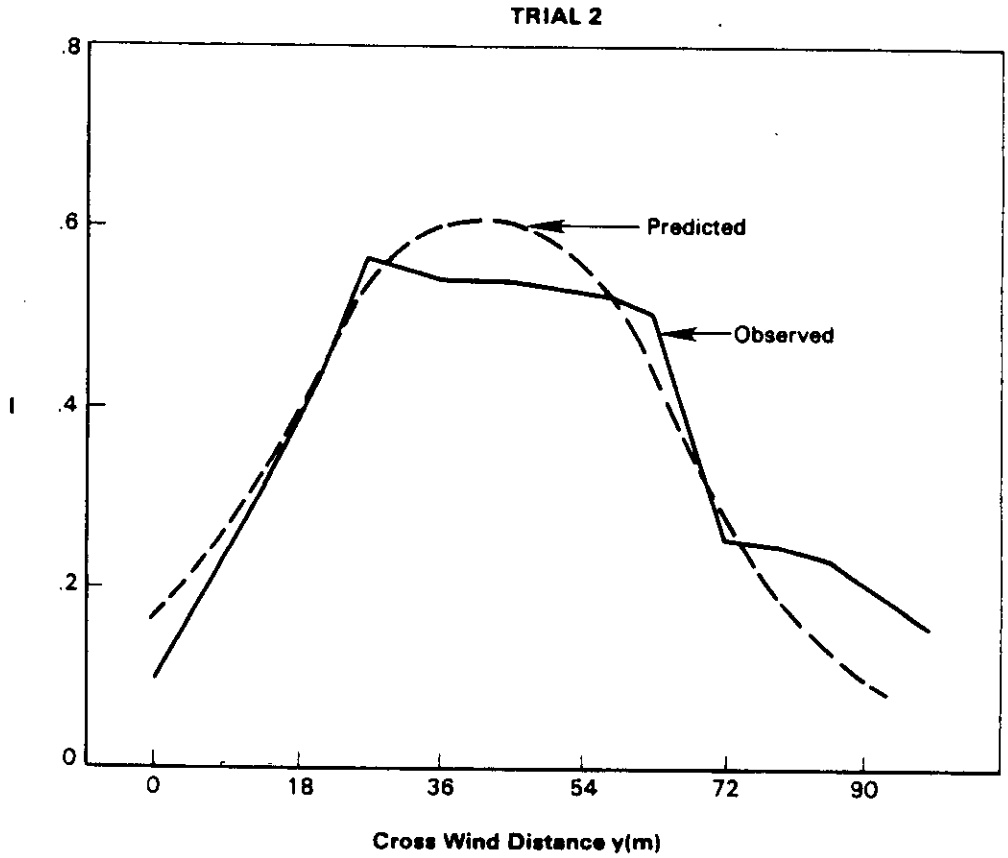

Fig. 8. Comparison of observed and predicted intermittency I for Trial 2.

The period of the lateral eddies is about five times the Lagrangian time scale for an exponential autocorrelogram, or about 15 to 25 min. Because the sampling time is only about 6 min, there is not sufficient time for the full effect of the large lateral eddies to be felt. We arbitrarily redefine the lateral time scale, T_{Ly}, to be 30s for both trials, corresponding to the observed lateral meandering period of about 3 min.

The travel time, t, to the far arc is about 37 s for Trial 2 and 15 s for Trial 4. According to the recommendations in Section 4, the model in Equations (4) through (6) can be used to estimate the vertical component of $\sigma_c/\overline{C}$ (because $t/T_{Lz} \gg 1$), and the meandering plume model (Equations (2), (6), and (9)) can be used to estimate the lateral component (because $t/T_{Ly} \simeq 1$ and averaging time$/T_{Ly} \ll 1$). If it can be assumed that the monitors are located on the centerline of the vertical plume, then Equation (4) predicts that the laterally averaged $\sigma_c/\overline{C}$ (CWI) equals 1.12, where it is assumed that initial plume size, σ_{0z}, equals 10 cm. (R. Sutherland, private communication). The average observed crosswind integrated $\sigma_c/\overline{C}$ (CWI) for the two trials was 0.93, which is within 20% of the prediction. From Equation (5), this value of $\sigma_c/\overline{C}$ (CWI) is consistent with a value of the vertical component of the intermittency, I_z, of 0.89.

For the lateral component, I_y, we use Equations (6) and (9). We estimate that σ_0^2 equals 7.1×10^{-6} for Trial 2 and 6.2×10^{-6} for Trial 4, giving I_y equal to 0.67 on

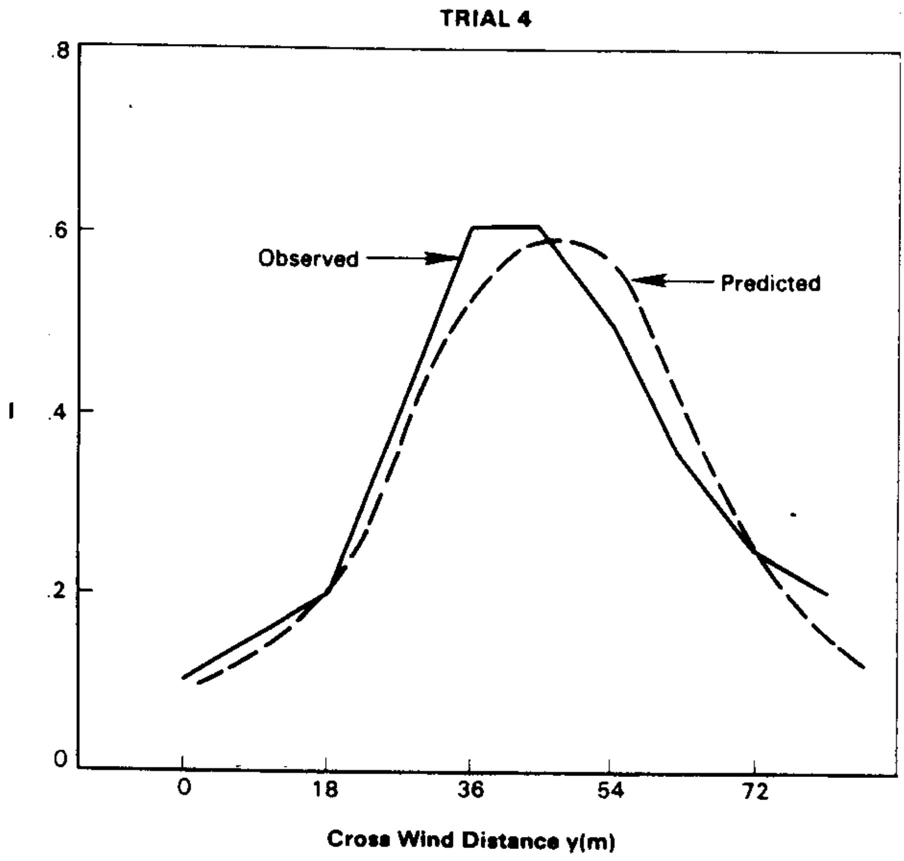

Fig. 9. Comparison of observed and predicted intermittency I for Trial 4.

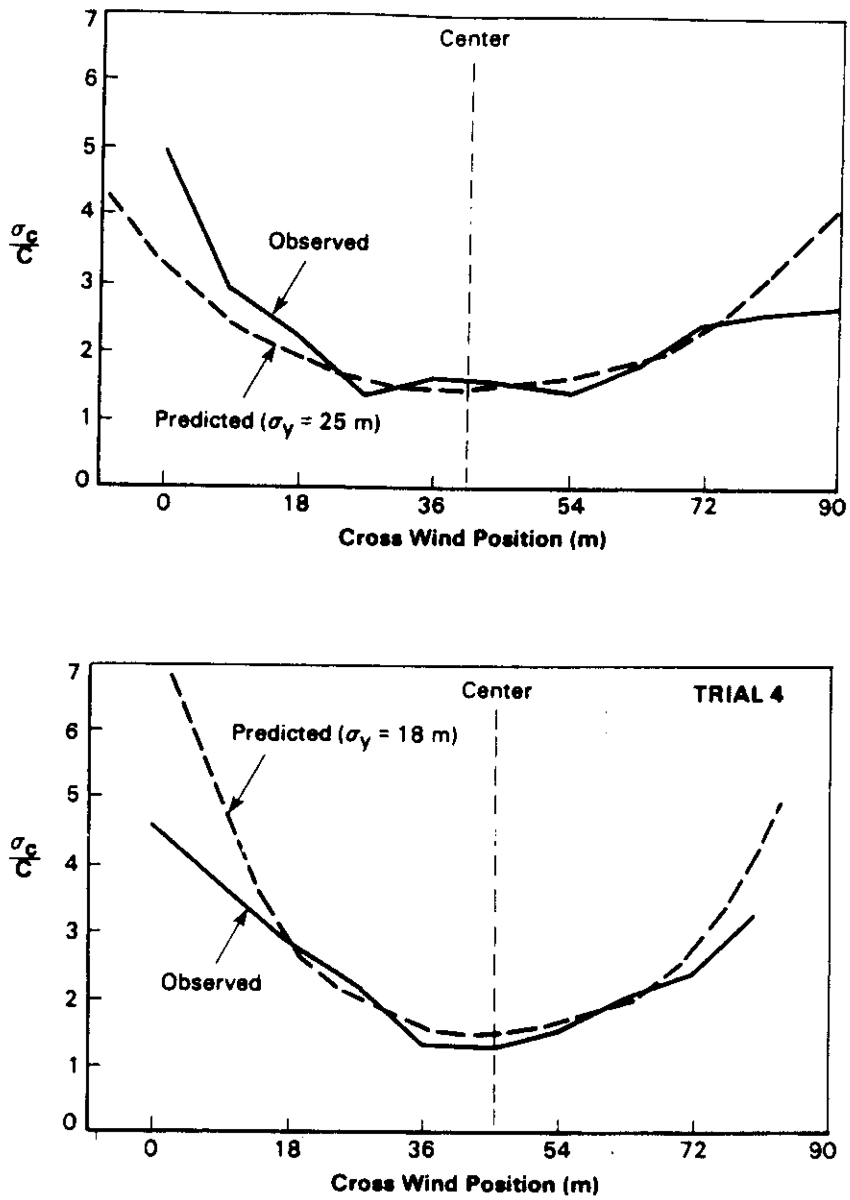

Fig. 10. Comparison of observed and predicted $\sigma_c/\overline{C}$ for Trials 2 and 4.

the mean plume centerline for both trials. The total I and $\sigma_c/\overline{C}$ at any crosswind distance, y, are given by the formulas:

$$I = I_y I_z = 0.60 \exp(-y^2/2\sigma_{yT}^2) \tag{13}$$

$$\sigma_c/\overline{C} = (3.33 \exp(y^2/2\sigma_{yT}^2) - 1)^{1/2}. \tag{14}$$

These predictions are plotted along with the observations in Figures 8 through 10, illustrating surprisingly good agreement in the magnitude and shape of the curves.

6. Variation of Concentration Fluctuations with Averaging Time

The analysis above assume that the averaging time for the data is 1 s. These data can also be used to calculate the integral time scale for the concentration fluctuations, T_I, (which may be different from T_L) and the variation of concentration fluctuations with averaging time. Autocorrelograms were drawn for all the monitors and the results averaged for each experiment, with the result that the best-fit T_I equals 7.5 s in Trial 2 and 10 s in Trial 4. These values are one-fourth to one-third the value of T_L used in Section 5, in agreement with Sykes' (1984) findings that T_I should be less than the Eulerian (and hence the Lagrangian) time scale. However, part of the difference may be explained by the fact that our T_L estimate was for the lateral component of turbulence, and when the vertical component is included, the effective T_L will decrease.

The concentration variances $\sigma_c^2(T)$ for averaging times T ranging from 1 to 60 s were calculated for each monitor. The averaged ratios $\sigma_c^2(T)/\sigma_c^2(1\text{ s})$ over all the monitors at each value of T were then plotted versus averaging time T for each trial, with the results for Trial 2 being given in Figure 11. At each value of T, the mean, and the mean plus and minus the standard deviation of the data from the monitors, are shown. Also

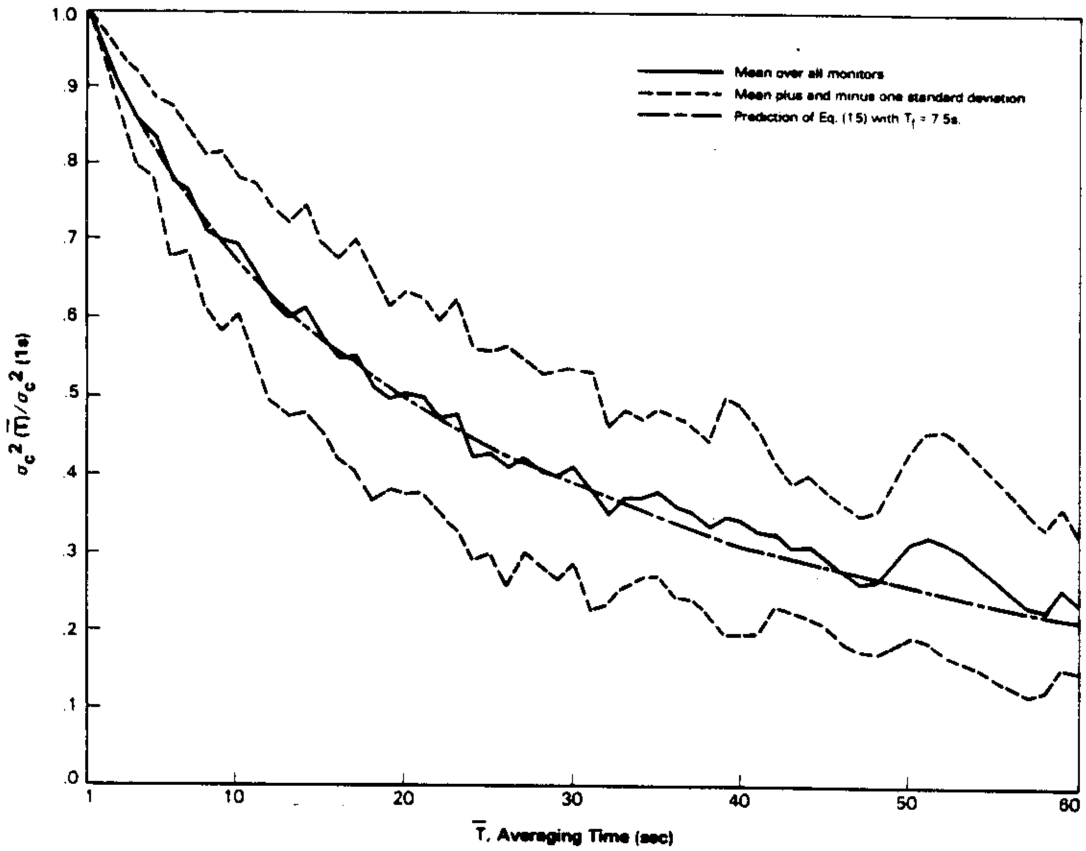

Fig. 11. Variation of concentration fluctuation variance with averaging time for Trial 2. The mean and the standard deviation of the data for all the monitors are shown, as well as the theoretical curve (Equation (15)) with T_I = 7.5 s.

shown is the theoretical curve (Venkatram 1979), which assumes that the autocorrelogram is exponential with T_I equal to 7.5 s.

$$\frac{\sigma_c^2(T)}{\sigma_c^2(1\,s)} = 2\,\frac{T_I}{T}\left(1 - \frac{T_I}{T}\left(1 - \exp\left(-\frac{T}{T_I}\right)\right)\right). \tag{15}$$

This curve clearly agrees very well with the mean observed curve. Similar results were obtained for Trial 4, with T_I equal to 10 s. It is important to stress that these data are from relatively short sampling periods of about 6 min, and that as the sampling time increases, the integral time scales T_I and T_L can also be expected to increase, as larger and larger lateral eddies are included in the data.

7. Conclusions

The Smoke Week III experiment provided data from two diffusion trials suitable for testing models for concentration fluctuations. By adjusting the Lagrangian time scale to agree with the observed frequency of wind meandering, good agreement was obtained between observed and predicted intermittency I and concentration fluctuation intensity $\sigma_c/\overline{C}$. We conclude that the exponential PDF does provide a useful framework for analyzing concentration fluctuations, especially since it explicitly accounts for plume intermittency. In order to refine our models, more detailed experiments are needed with the following characteristics:

– constant source strength with known dimensions; constant meteorological forcing parameters (solar heating, geostrophic wind speed) over the duration of the experiment;

– observations of turbulence over the depth of the plume sufficient to calculate vertical variation of mean winds, turbulence, and integral scales; and

– observations of concentrations with averaging times of 1 s or less, with sufficient numbers of monitors to resolve the plume on at least three arcs such that travel time is less than, equal to, and greater than the Lagrangian time scale.

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References

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

Csanady, G. T.: 1973, *Turbulent Diffusion in the Environment*, D. Reidel Publ. Co., Dordrecht, Holland, pp. 222–248.

Durbin, P. A.: 1980, 'A Stochastic Model of Two Particle Dispersion and Concentration Fluctuations in Homogeneous Turbulence', *J. Fluid Mech.* **2**, 279–302.

Gifford, F. A.: 1959, 'Statistical Properties of a Plume Dispersion Model', *Advan. Geophys.*, **6**, 117–138.

Gifford, F. A.: 1972, 'The Form of the Frequency Distribution of Air Pollution Concentrations', Symposium on Statistical Aspects of Air Quality Data, Chapel Hill, NC, November 9–10, 1972.

Gifford, F. A.: 1982, 'Horizontal Diffusion in the Atmosphere: A Lagrangian-Dynamical Theory', *Atmos. Environ.* **16**, 505–512.

Hanna, S. R.: 1984, 'Concentration Fluctuations in a Smoke Plume', to appear in *Atmos. Environ.*

Hilst, G. R.: 1957, 'The Dispersion of Stack Gases in Stable Atmospheres', *J. Air Poll. Control Assoc.* **7**, 205–210.

Jones, C. D.: 1981, 'Statistics of Concentration Fluctuations in Short Range Atmospheric Diffusion', in C. J. Harris (ed.), *Math. Modeling of Turb. Diff. in Environ.*, Academic Press, New York, pp. 277–300.

Larsen, R. I.: 1970, 'Relating Air Pollutant Effects to Concentration and Control', *J. Air Poll. Control Assoc.* **20**, 214–225.

Lee, J. T. and Stone, G. L.: 1983, 'The Use of Eulerian Initial Conditions in a Lagrangian Model of Turbulent Diffusion', *Atmos. Environ.* **17**, 2477–2481.

Pasquill, F.: 1974, *Atmospheric Diffusion*, John Wiley and Sons, New York, 429 pp.

Sawford, B. L.: 1983, 'The Effect of Gaussian Particle-Pair Distribution Functions on the Statistical Theory of Concentration Fluctuations in Homogeneous Turbulence', *Q. J. Roy. Meteorol. Soc.* **190**, 339–354.

Sutherland, R. A., Hoock, D. W., and Gomez, R. B.: 1981, 'An Objective Summary of U.S. Army Electro-Optical Modeling and Field Testing in an Obscuring Environment', Report No. ASL-TR-0096 from U.S. Army Atmospheric Sciences Laboratory, White Sands Missile Range, NM 88002.

Sykes, R. I.: 1984, 'The Variance in Time-Averaged Samples from an Intermittent Plume, *Atmos. Environ.* **18**, 121–123.

Venkatram, A.: 1979, 'The Excepted Deviation of Observed Concentrations from Predicted Ensemble Means', *Atmos. Environ.* **13**, 1547–1549.

Wilson, D. J., 1982, 'Prediciting Risk of Exposure to Fluctuating Concentrations in Plumes', Alberta Environment, 56–57.