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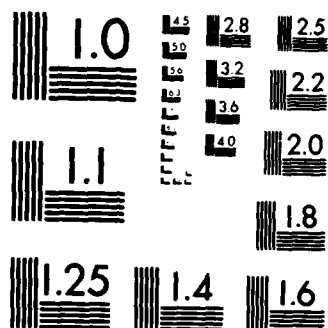
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TIME AND SPACE VARIABILITY OF  
TURBULENT CONCENTRATION FLUCTUATIONS  
FINAL REPORT

by  
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DECEMBER 31, 1984

U.S. ARMY RESEARCH OFFICE  
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## 1. STATEMENT OF PROBLEM STUDIED

Observed and modeled concentration fluctuations in smoke plumes in the atmosphere are quite large, with the standard deviation  $\sigma_c$  at least as large as the mean  $\bar{C}$ . There is interest in concentration fluctuations because of their importance in assessing environmental and toxicological effects, in evaluating models for predicting mean concentrations, and in determining the response of remote sensors pointed towards smoke plumes. During the past ten years there has been a large increase in research on this subject because of the concerns listed above, because of advances in the development of instruments that can measure short-term concentration fluctuations, and because of increases in speed and storage capabilities of computers that some researchers are using to run models of concentration fluctuations.

The purpose of the research performed under this contract is to review existing models and data sets on concentration fluctuations, and use this information to develop and test a simplified model. Emphasis is on the use of U.S. Army data to evaluate the model. As a result of the model evaluation, research required for model improvement is recommended and steps taken to begin some of this new research.

## 2. SUMMARY OF THE MOST IMPORTANT RESULTS

This research project can be divided into three tasks:

- Review of existing models and data,
- Development of simple model and evaluation with U.S. Army data.
- Development of strand model.

The first two tasks were completed during the first 20 months of the project and most of the results are reported in the publications that are listed in Section 3. Abstracts of the journal articles are given here, and the reader is referred to the original articles for details.

The last task has been underway since September, 1984, and is not completed. It is briefly reviewed below and a more detailed progress report is given as Appendix A.

## 2.1 Review of Existing Models and Data

### CONCENTRATION FLUCTUATIONS IN A SMOKE PLUME

Steven R. Hanna

Atmospheric Environment, 18 (6), 1091-1106

(First received 5 September 1983 and received  
for publication 17 January 1984)

Abstract - Previous research results are reviewed and used to derive a new set of analytical formulas for predicting concentration fluctuations in smoke plumes. The meandering plume approach and the internal plume approach are compared. Some simple models are tested with field and laboratory data sets, showing that several aspects of the data (e.g. the concentration fluctuations on the plume axis) are reasonably well simulated. However there is much room for improvement, since the models have some fundamental disagreements and must more testing with data should take place. In particular, a good knowledge of the Lagrangian time scale is essential for predicting concentration fluctuations.

## 2.2 Development of Simple Model and Evaluation with U.S. Army Data

### THE EXPONENTIAL PROBABILITY DENSITY FUNCTION AND CONCENTRATION FLUCTUATIONS IN SMOKE PLUMES

Steven R. Hanna

Boundary Layer Meteorology, 29, 361-375.

(Received in final form 31 May, 1984)

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 100m 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.

In addition to the above paper that was published in Boundary Layer Meteorology, three other papers have been prepared in which the simple model is compared with observations. Two of these can be found in conference proceedings (CRDC Conference, AMS Conference), and the third is to be published soon in Atmospheric Environment. The first two present additional statistics from the U.S. Army Smoke Week III experiment, including observed probability density functions and moments of the distributions of instantaneous concentration, plume width, plume centroid, and cross-wind integrated concentration. The latter paper compares the new model with observations of ground level concentrations resulting from tracer releases from a tall stack in Deardorff and Willis' convective tank.

### 2.3 Development of Strand Model

The strand model is a new concept that is under development and has not yet been published in a journal or report. A preliminary description of the model is in Appendix A. The purpose of this research is to explicitly account for concentration fluctuations at very small scales. This model assumes that the source emits strands of polluted material, which diffuse due to molecular motions and split when their diameters double. If polluted strands are close enough together, they may entrain each other when they split. The whole packet of strands also diffuses at a rate comparable to that of



an instantaneous Gaussian puff. When combined with a model for the meander of the instantaneous plume, the model gives predictions in agreement with wind tunnel data of Fackrell and Robins. The model produces a probability density function of the concentration fluctuations at a point, and can account for the effects of source and sampler size and sampling and averaging time.

### 3. LIST OF PUBLICATIONS

Hanna, S.R., 1984. Concentration Fluctuations in a Smoke Plume.

Atmos. Environ., 18, 1091-1106.

Hanna, S.R., 1984. The Exponential Probability Density Function and Concentration Fluctuations in Smoke Plumes. Bound. Lay.

Meteorol., 29, 361-375.

Hanna, S.R., 1984. Observed and Modeled Concentration Fluctuations in a Small Smoke Plume. Proceedings of Fourth Joint Conference on Applications of Air Pollution Meteorology. Am. Meteorol. Soc., 128-131.

Hanna, S.R., 1984. Comments on Deardorff and Willis' (1984) Paper Entitled "Ground-Level Concentration Fluctuations from a Buoyant and a Non-Buoyant Source within a Laboratory Convectively-Mixed Layer." to be published in Atmos. Environ.

Hanna, S.R. and J. Pleim, 1984. Characteristics of Observed Concentration Fluctuations During Smoke Week III. Proceedings of 1984 CRDC Scientific Conference on Obscuration and Aerosol Research, Aberdeen Proving Ground, MD.

#### 4. LIST OF MEETINGS ATTENDED

Feb. 20-21, 1983, White Sands Missile Range.

Concentration fluctuation data sets were discussed with W. Ohmstede, R. Sutherland, and others at the Atmospheric Science Laboratory. A seminar on models of concentration fluctuations was given. Plans were made for a possible smoke experiment to take place this fall.

June 15-17, 1983, ARO Quail Roost Workshop on Aerosol Dispersion in the Atmospheric Surface Layer.

At this workshop current U.S. Army research projects were discussed and recommendations made for future research priorities.

November 17, 1983, Aberdeen Proving Ground, MD.

A paper entitled "Concentration fluctuations in smoke plumes from near surface releases" was presented at the 1983 Scientific Conference on Chemical Defense Research. This research project was discussed with D. Sloop and R. Saucier of CSL.

December 5 & 6, 1983 and February 3, 1984, Boulder, CO.

Dr. Hanna attended a meeting of the Large Eddy Simulation Working Group, sponsored by ARO. Research plans for this project were discussed with W. Bach of ARO and W. Ohmstede of ASL.

June 25, 1984, Aberdeen Proving Ground, MD.

Dr. Hanna attended the 1984 CRDC Scientific Conference on Obscuration and Aerosol Research at Aberdeen, Proving Ground, MD, where he presented a paper entitled "Characteristics of Observed Concentration Fluctuations during Smoke Week III.

October 19, 1984, Portland, OR

Dr. Hanna presented a paper entitled "Observed and Modeled Concentration Fluctuations in a Small Smoke Plume" at the AMS/APCA Fourth Joint Conference on Applications of Air Pollution Meteorology.

October 23-25, 1984, Kiawah Island, SC.

Dr. Hanna attended the DOE/AMS Workshop on Model Evaluation in Kiawah Island, SC, where he presented a talk on the results of this project as they affect model evaluation, and discussed progress with Dr. Bach of ARO.

November 7, 1984, Raleigh, NC

Dr. Hanna visited Dr. Bach and Dr. Flood at ARO in RTP, NC.

December 5, 1984, White Sands Missile Range, NM.

Dr. Hanna visited ASL at White Sands Missile Range, NM, where he presented a seminar on his research and discussed this project with R. Meyers and other ASL scientists.

##### 5. LIST OF PARTICIPATING SCIENTIFIC PERSONNEL

Dr. Steven R. Hanna  
Dr. Robert J. Yamartino  
Mr. Jonathan Pleim

No degrees were awarded.

APPENDIX A  
A STRAND THEORY OF CONCENTRATION FLUCTUATIONS

by  
R.J. Yamartino  
and  
S.R. Hanna

STATUS REPORT

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## 1. Introduction

The notion that pollutants may be emitted, dispersed and described mathematically as though the material were confined to small packets of size  $\lambda$  has been described by Csanady (1973), Chatwin and Sullivan (1979), Venkatram (1983), and recently by Chatwin (1984). The prime motivation for such a formulation is that higher moments of the concentration distribution, including the variance, can be readily computed since the problem reduces to that of "counting" statistics.

The size of the packets,  $\lambda$ , is generally chosen to correspond to the conduction length scale where molecular diffusion is the only operative dilution mechanism; however, most treatments then either ignore the effects of molecular diffusion or assume that packet growth by molecular diffusion results only in the entrainment of clean air.

In this paper we assume that pollutants emerge from a source in long thin strands, rather than small three-dimensional packets, and that these strands subsequently grow by diffusive processes until a time  $t_d$ , whereupon they split into two strands that then repeat the above mentioned process. Consideration of strand entrainment of both clean and polluted air leads to a differential equation for the time development of the spatial density of polluted strands. The solution of the resulting first-order, non-linear, differential equation combined with counting statistics then permits a determination of ensemble average concentration statistics as would be measured by a finite aperture detector possessing a time response that is extremely rapid compared with all relevant time scales.

Model predictions for plume intermittency are then compared with the wind tunnel observations of Fackrell and Robins (1982). Further assumptions about the statistical nature of the strands then permits total and conditional (i.e.,  $C > 0$ ) concentration statistics to be evaluated. Additions to the theory to include plume meander, time averaging, and finite response detectors are discussed.

## 2. Strand Theory

### 2.1 Development of the Governing Equations

Rather than postulating a non-linear differential equation for the concentration variance, as is done by Lewellen and Sykes (1983), we begin with the simple notion of pollutants confined to thin strands of diameter  $\lambda$  that are dispensed by turbulent eddies and also grow in size until shear forces tear the strand in two. Such an approach leads naturally to the computation of higher moments of the observed concentration distribution including the variance. The actual size of these strands will correspond to the conduction length scale, which Chatwin and Sullivan (1979) estimate to be of order  $10^{-3}$  meter; however, specific choice of a numerical value for  $\lambda$  is not a prerequisite for development of the formalism.

Consider a source of diameter  $d_s$  emitting at rate  $Q$  (mass/time) into a flow field of velocity  $u$ . Assuming that the emissions are well mixed across the source aperture, one might imagine (Figure 1) that the pollutant emerges from the source in thin strands of diameter  $\lambda$  and containing a concentration  $C_o$ . In such a case the number of strands,  $N_s$ , emerging would be

$$N_s = (d_s / \lambda)^2 \quad (1)$$

and the concentration in the strands would be

$$C_o = Q / (\frac{\pi}{4} d_s^2 u) \quad (2a)$$

or

$$C_o = Q / [2\pi \sigma_y(0) \sigma_z(0) u] \quad (2b)$$

in the more familiar notation of plume modeling. Instantaneous concentrations  $C(x,y,z)$  downwind could then be expressed as

$$C(x,y,z) = C_o D(x,y,z) \quad (3)$$

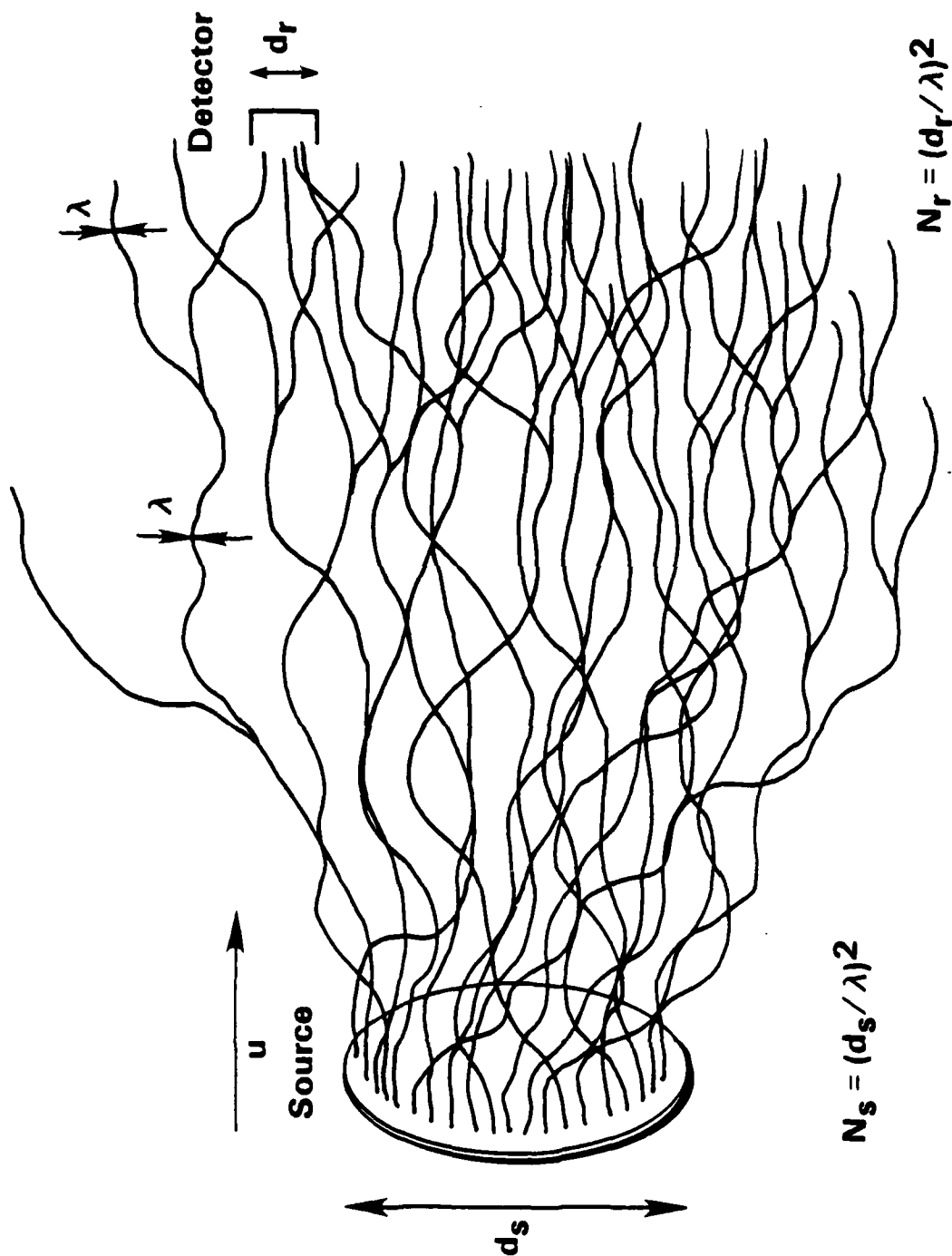


Figure 1 Visualization of the Strand Concept.  $N_s$  Strands are Emitted and Subsequently Experience Dispersion among "Clean Air" Strands (not shown). Individual Strands Undergo Diffusive Growth and Split. A Receptor then Samples a Cluster of  $N_r$  Strands; a Fraction  $\rho$  of which Contain Pollution.

where the instantaneous plume dilution function  $D(x,y,z)$  is defined such that at the source  $D(x,y,z) = D(0,0,0) = 1$ . The dilution function is assumed to be known, through means of a numerical or analytic model, but is given for the Gaussian plume model as

$$D(x,y,z) = \frac{\sigma_y(0) \sigma_z(0)}{\sigma_y(x) \sigma_z(x)} \exp \left[ -\frac{1}{2} (y/\sigma_y(x))^2 - \frac{1}{2} (z/\sigma_z(x))^2 \right], \quad (4)$$

where  $\sigma_y(x)$  and  $\sigma_z(x)$  are the horizontal and vertical instantaneous plume width standard deviations respectively as a function of downwind distance  $x$ . Substituting Eqs. (2b) and (4) into (3) gives us back a Gaussian plume expression and indicates that nothing unusual results from this re-grouping of terms.

Ignoring the minor "defect" introduced by the assumed Gaussian shape at the source, earlier assumed to be uniform, we note that when the dilution function takes on a maximal value of unity all available strands are populated with pollutant, as is consistent with our definition of the number of polluted strands  $N_s$  by Eq. (1). Temporarily ignoring diffusive breakup or splitting of the strands, these entities would simply disperse relative to each other, so that at some downwind receptor sampling strands of air, a fraction  $D(x,y,z)$  would contain pollutant and a fraction  $1-D$  would contain only "clean ambient air". Thus, we may think of  $D(x,y,z)$  as either the usual diffusion function or as the polluted strand density function representing that fraction of locally available strands that are polluted. With this information in hand, one could immediately write down expressions for mean square concentration, concentration variance, and other moments of the distribution; however, the results would be in poor agreement with experiments as the all-important diffusive growth of the strands and subsequent strand breakup has been ignored.



If the strands experience diffusive forces specified by the molecular diffusivity  $\nu$ , they will double in areal size in a time,  $t_d$ , given as

$$t_d = \lambda^2 / (4\pi\nu) \quad (5a)$$

whereupon the strand is sheared apart by turbulent eddies into two strands of diameter  $\lambda$ . Given molecular diffusivities of order  $10^{-5} \text{ m}^2/\text{sec}$  and  $\lambda \approx 10^{-3} \text{ m}$ , one computes  $t_d$  in the  $10^{-2} - 10^{-3}$  second range; however, as with  $\lambda$ , the precise numerical value of  $t_d$  is not critical at this point except to note that it will be sub-second and thus relatively short in comparison to many time scales relevant to atmospheric dispersion. More generally we may merely say that growth in the number of strands is associated with a time scale  $\tau$  without relating  $\tau$  to  $t_d$  via a rigorous relation such as

$$\tau = t_d / \ln 2 \quad (5b)$$

that would result if we took the doubling concept literally. Allowing such a growth in the number of polluted strands creates an interesting dilemma that will prove quite useful in developing a differential equation for the number density of polluted strands. Imagine that a long pipe with diameter  $d_s$  were connected to the source described earlier and that the flow speed  $u$  was such that the travel time in the pipe was much greater than  $\tau$ .  $N_s$  strands of concentration  $C_0$  would go into the pipe and, since the pipe is completely filled with strands,  $N_s$  strands of concentration  $C_0$  would come out the far end. What if anything, happened during the journey along the pipe? Diffusion and strand splitting did in fact occur, but since the pipe was already as full as it could possibly be with a polluted strand density of  $D = 1$ , the growing strands had no other choice but to devour existing strands with the result that no net growth,  $g$ , occurred in the polluted strand density. Can we then write down a plausible differential equation describing the time evolution of the effective polluted strand density,  $\rho(t)$ , defined as

$$\rho(t) \equiv g(t) D(t) \quad (6)$$

where the dilution function  $D(x,y,z)$  has been re-expressed as  $D(t) = D(ut,y,z)$  for compactness? Knowing also that the number of strands will grow freely as  $g \sim \exp(t/\tau)$  if the polluted strand density is low (i.e.,  $\rho \ll 1$ ) we may write the differential equation

$$\rho' = \rho(1-\rho)/\tau + g D' \quad , \quad (7a)$$

where the prime indicates the time derivative operator, as the simplest form that possesses the three necessary properties:

- a) exponential growth of polluted strand fraction,  $\rho$ , with time scale  $\tau$  for  $\rho$  small
- b) termination of growth as  $\rho$  approaches unity via the term  $(1-\rho)$ , and
- c) forced dilution created by changes in  $D(t)$  in time.

Applying the chain rule to Eq. (6) and dividing through by  $D$ , Eq. (7a) may be alternatively expressed as

$$g' = g(1-\rho)/\tau \quad (7b)$$

which, because the factor  $(1-\rho)$  corrects for new strands consuming or recombining with existing strands, elucidates the meaning of the term "net growth" function being applied to  $g(t)$ .

Equation (7a) for the polluted strand density or fraction is the principal result given to us by this simple, physically-based model of strand behavior, and this equation may be used in conjunction with any dispersion model, analytic or numerical, giving rise to a diffusion function  $D$ . It is also worth noting that since  $\rho(t)$  is closely related to the intermittency factor  $\gamma$  discussed extensively by Wilson et. al. (1984), many useful expressions could be written down by inspection; however, we shall first explore the theory further.

## 2.2 Closed Form Solution

Without the forced dilution function  $D'$ , Equation (7a) is identical to that used by population biologists to describe population

growth in a world with a fixed and finite food supply and is easily solved analytically by means of the substitution  $y = 1/\rho$ . (Kaplan, 1958). Using this hint it quickly follows that a solution<sup>+</sup> to Equation (7b) is

$$g(t) = \left\{ e^{-t/\tau} + \frac{1}{\tau} \int_0^t dt' e^{(t'-t)/\tau} D(t') \right\}^{-1} \quad (8)$$

where  $t'$  is simply a dummy variable for the time between release (i.e.,  $t = 0$ ) and present time  $t$ . The only thing disturbing about Eq. (8) is that the path between source and end point at time  $t$  is not specified. Figure 1 shows how many different paths, equivalent to the strand trajectories, could end up at a specific point and that some weighted consideration of all possible paths (and thus strand density time developments) should be involved in the correct solution of Eq (7b). A plausible solution might consider the straight line path as the mean solution pathway with the reciprocity theorem suggesting that  $D(t')$  in Eq. (8) be replaced by

$$D(t') = \frac{1}{2\pi\sigma_y(t-t')\sigma_z(t-t')} \int_{-\infty}^{+\infty} dy' dz' D(y'_m, z'_m) \cdot \exp\left\{-\frac{1}{2} \frac{(y_m - y')^2}{\sigma_y^2(t-t')} - \frac{1}{2} \frac{(z_m - z')^2}{\sigma_z^2(t-t')}\right\} \quad (9)$$

where  $y_m = yt'/t$  and  $z_m = zt'/t$  specify the mean path and  $y', z'$  are dummy integration variables. While Eq. (9) may represent a significant theoretical point, it appears to be of secondary importance in most applications. In addition, we note that Eq. (9) can be cumbersome and numerically problematic to evaluate even for a single mean path; thus, we turn to numerical integration of the more well-conditioned Eq. (7b).

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<sup>+</sup>Non-linear differential equations may have multiple solutions.

### 2.3 Numerical Solution

Numerical integration of Eq. (7b), subject to the initial condition  $g(0)=1$ , must recognize that the potentially explosive growth (i.e., exponential) in the solution could lead to rapid divergence of the numerical solution from the true solution. For this reason we adopt a second-order accurate, time-centered (i.e., Crank-Nicolson) procedure to provide a reasonable compromise between accuracy and stability. The finite difference form of (7b) then becomes

$$g_{n+1} = g_n + \delta (g_{n+1} + g_n) \left( 1 - \frac{1}{2} D_{n+1} g_{n+1} - \frac{1}{2} D_n g_n \right) \quad (10)$$

where  $\delta = \frac{1}{2} \Delta t / \tau$  and  $\Delta t$  is the time step. The time advanced value,  $g_{n+1}$ , appears on both sides of the equation, and solution of the resulting quadratic equation for  $g_{n+1}$  yields the familiar

$$g_{n+1} = \frac{-b + (b^2 - 4ac)^{1/2}}{2a} \quad (11)$$

where  $a = \frac{1}{2} \delta D_{n+1}$ ,  
 $b = 1 - \delta \left[ 1 - \frac{1}{2} g_n (D_n + D_{n+1}) \right]$ ,  
 and  $c = -g_n \left[ 1 + \delta \left( 1 - \frac{1}{2} g_n D_n \right) \right]$ .

Equation (11) is then marched in time using a time step chosen as the lesser of  $0.1\tau$  and  $0.1 D/D'$ .

Figure 2 shows several crosswind profiles of the resulting  $\rho$  function at several values of downwind distance  $x=ut$  using parameters relevant to the wind tunnel studies of Fackrell and Robins (1982). The most interesting feature of these curves is the variation in crosswind profile shape as one moves to different downwind distances. Profiles with a fairly small plume-axis value of  $\rho$ , denoted  $\rho_0$ , look Gaussian in shape while those with  $\rho_0 \approx 1$  show a broad flat-top region with a precipitous drop at large  $y/\sigma_y$ . As it will prove important in the inclusion of plume meander effects to know this

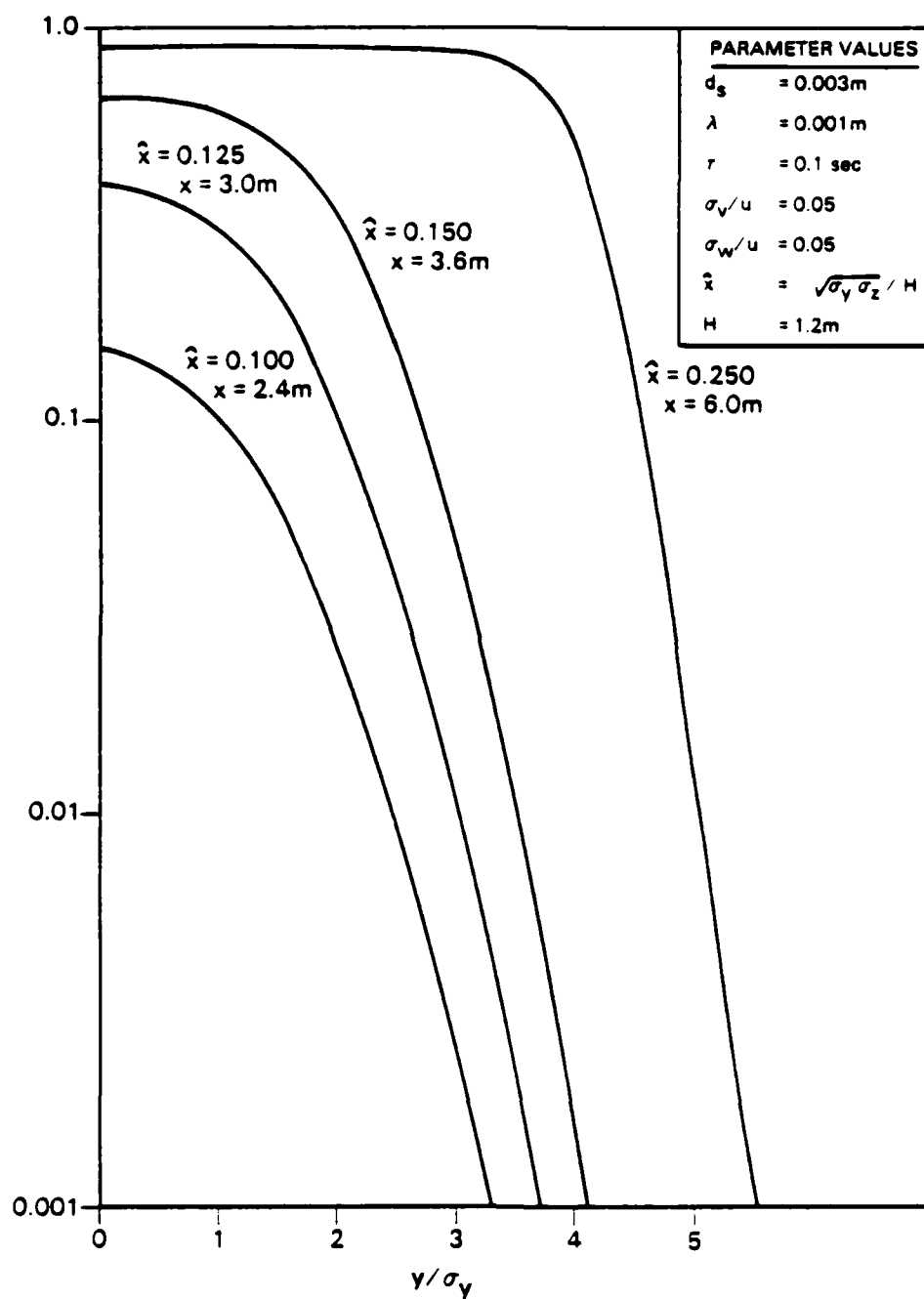


Figure 2 Crosswind  $\sigma$  Profiles for Several Downwind Distances. Distances are given in Meters and in Normalized Distance Assuming a Boundary Layer Depth of 1.2m.

crosswind profile function, the crosswind behavior of the analytic solution, Eq. (8), was considered assuming that the on-axis values were available from the numerical integration procedure described above. For the particular trajectory that follows the path  $y = \epsilon_y \sigma_y$ ,  $z = \epsilon_z \sigma_z$ , it can be shown that

$$\rho(ut, y, z) / \rho_0 = \frac{S(ut) \cdot \exp[-\frac{1}{2} (\epsilon_y^2 + \epsilon_z^2)]}{1 + \exp[-\frac{1}{2} (\epsilon_y^2 + \epsilon_z^2)] [S(ut) - 1]} \quad (12)$$

where  $\rho_0 = \rho(ut, 0, 0)$

and  $S(ut) = e^{t/\tau} / g(t) = D(ut, 0, 0) e^{t/\tau} / \rho_0$ . The function  $S(ut)$  is just the ratio of what  $g$  could have been (i.e.,  $e^{t/\tau}$ ) if its growth were unhindered to what it actually is on the plume centerline and hence it will be referred to as the "growth suppression factor"  $S$ . Equation (12) provides a reasonable description of the crosswind profiles shown in Figure 2 with the exception that falloff is too rapid beyond when  $\rho/\rho_0 \approx 10^{-2}$ , but this may not be a serious drawback since this plume fringe regime should not dominate most practical problems.

## 2.4 Statistical Measures

Once the mean probability  $\rho$  is known at the receptor, the binomial distribution (Ross, 1972) gives the probability  $P(j)$ , that  $j$  of the  $N_r$  detected strands contain pollutant, as

$$P(j) = \frac{N_r!}{(N_r - j)! j!} \rho^j (1 - \rho)^{N_r - j} \quad j = 0, 1, \dots, N_r$$

where  $N_r = (d_r / \lambda)^2$  for a receptor having an aperture  $d_r$ . Thus, the probability distribution for detecting  $j$  polluted strands is identical to those used for predicting the probabilities in die ( $\rho = 1/6$ ) and coin ( $\rho = 1/2$ ) toss games.

Since, there is always the finite probability

$$P(0) = (1-\rho)^{N_r}$$

that none of the strands contain pollutant, the "intermittency" factor

$$\gamma = 1-P(0), \quad (15)$$

discussed extensively by Wilson et al. (1984), will be less than one even for an ideal, zero concentration threshold detector and in the complete absence of plume meander, as presently assumed here.

Figure 3 shows the intermittencies predicted by Eq. (15) for plausible strand properties of  $\tau \approx 0.2$  sec and  $\lambda \approx 0.4$  mm and superimposed onto wind tunnel measurements of Fackrell and Robins (1982). The curves, one for each of the five elevated source diameters considered, demonstrate that the strand theory approach can

- i) yield predictions that exhibit reasonable qualitative behavior in terms of source size and downwind distance dependence and
- ii) span the range of observed values.

However, as important ingredients (e.g., plume meander, finite detector thresholds and averaging times, strand concentration distribution assumptions) are still missing from the formulation, results depicted should only be taken as grounds for "cautious optimism."

The moment generating function for the binomial distribution gives the ensemble average number of polluted strands sampled as

$$\bar{j} = N_r \rho, \quad (16)$$

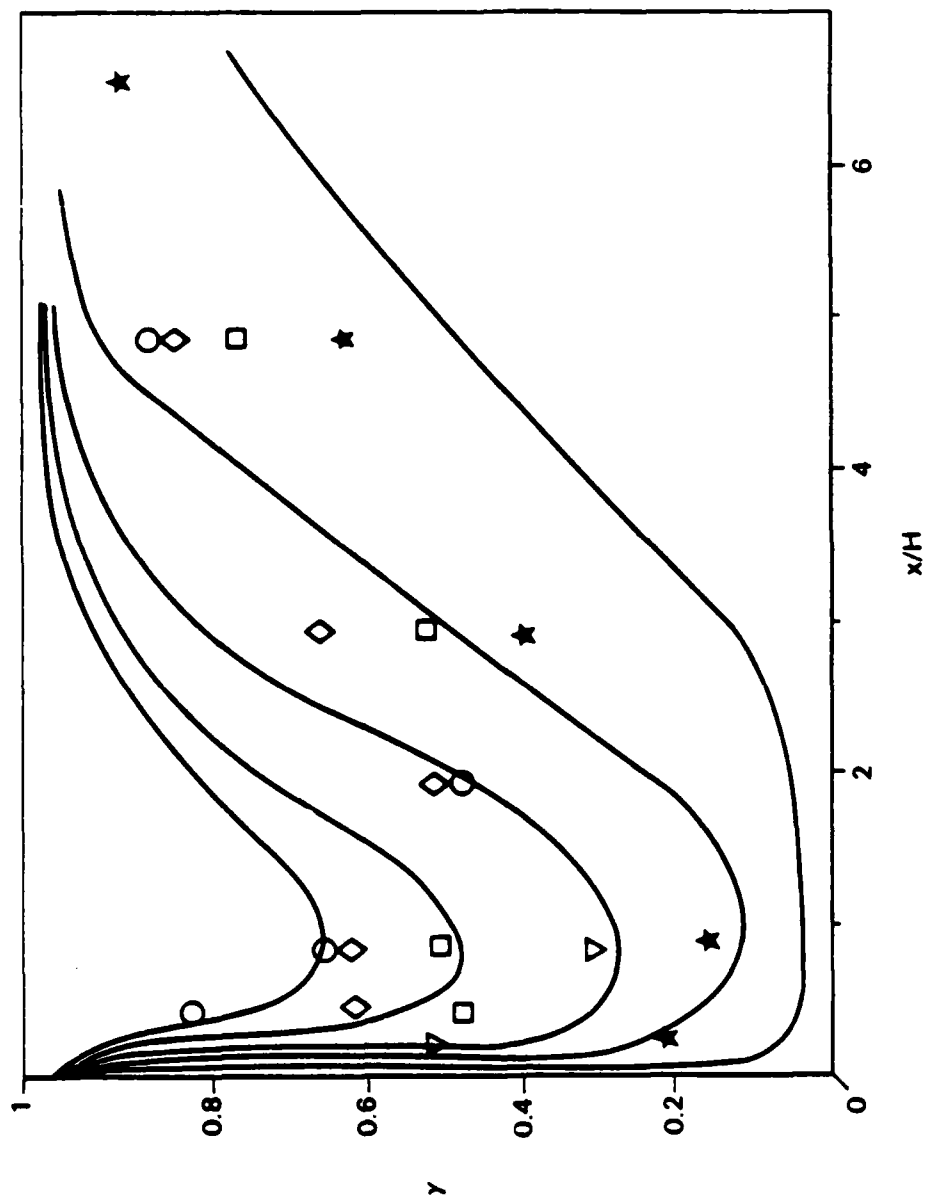


Figure 3 Plume-axis ( $x, 0, z_s$ ) Intermittency Factors Measured in the Fackrell and Robins (1982) Wind Tunnel Study. Solid Curves are Eq. (15) Predictions for the Five Elevated Source Diameters of 0.30, 0.85, 1.50, 2.50 and 3.50 Centimeters. (The lowest curve corresponds to the smallest source).



the ensemble mean square number of polluted strands as

$$\overline{j^2} = N_r(N_r-1)\rho^2 + N_r\rho \quad , \quad (17)$$

as well as any higher order measure  $\overline{j^n}$ . Thus, the ensemble variance in the number of polluted strands is just

$$\sigma_j^2 = \overline{j^2} - \bar{j}^2 = N_r\rho(1-\rho) \quad , \quad (18)$$

so that, even if at each point all strands have the same concentration, a finite ensemble concentration variance would be observed.

Hypothesizing that at the receptor each of the  $j$  polluted strands has the same concentration,  $C_s$ , the particular sample will have concentration

$$C(j) = C_s(j/N_r) \quad (19)$$

and the ensemble mean concentration will be

$$\bar{C} = \sum_j P(j)C(j) = C_s(\bar{j}/N_r) = C_s\rho \quad (20)$$

However, for this result to be consistent with Eq. (3) it must be true that

$$C_s = C_o/g(t) \quad . \quad (21)$$

Similarly, the squared concentration

$$C^2(j) = [C_s(j/N_r)]^2 = j^2 [C_o/(gN_r)]^2 \quad (22)$$

results in the ensemble mean square concentration

$$\overline{C^2} = \sum_j P(j) C^2(j) = [(1-1/N_r)\rho^2 + \rho/N_r] (C_o/g)^2 \quad (23)$$

and variance of

$$\sigma_C^2 = \overline{C^2} - \bar{C}^2 = \frac{1}{N_r} \rho(1-\rho) (C_o/g)^2 \quad (24)$$

The total concentrations fluctuation intensity,  $i$ , can then be expressed as

$$i^2 = \sigma_C^2 / \bar{C}^2 = \frac{1}{N_r} (1/\rho - 1) \quad (25)$$

Wilson et al. (1984) discuss the logic and convenience of decomposing the fluctuation intensity,  $i$ , into two parts: the part arising from detecting positive concentrations and the part due to samples where the detector sees no material. The resulting decomposition is

$$i^2 = i_p^2 / \gamma + (1-\gamma) / \gamma \quad , \quad (26)$$

where

$$i_p^2 = \sigma_{Cp}^2 / \bar{C}_p^2 \quad , \quad (27)$$

and the subscript p refers to that fraction  $\gamma$  of the samples where the receptor conditionally sees positive (i.e., finite non-zero) signals. This decomposition into conditional quantities, denoted by the subscript p, is also convenient for consideration of the effects of allowing strands to have a distribution of concentrations.

## 2.5 The Exponential Strand Concentration Distribution

In the previous section, all strands at a given point in space were assumed to have the same concentration  $C_s$ ; however, a receptor at some downwind location will detect strands that have experienced a spectrum of histories as opposed to the simplistic notion that each strand will have suffered  $g(t)-1$  bifurcations. In developing a theory of strand "ageing" as a sequence of bifurcations, analogy (Ross, 1972) with other decay distribution examples (e.g., radioactive decay, probabilistic failure analysis) suggests that the strands will have an exponential age distribution

$$P(r) = \frac{1}{\alpha} e^{-r/\alpha} \quad (28)$$

where  $r$  is the age parameter and  $\alpha$  represents the average strand age. As increasing strand age corresponds to a greater number of bifurcations, yielding successively lower strand concentrations, it is reasonable to conjecture that these concentrations also will be distributed exponentially as

$$C_s(r) = C_o e^{-r/\delta} \quad (29)$$

where  $\delta$  is a constant<sup>+</sup> describing the effect on concentration of a single bifurcation. The average strand concentration, which Eq. (21) tells us must be  $C_o/g(t)$ , is then

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<sup>+</sup>A value of  $\delta = \ln 2$  would yield  $C_s(r) = C_o (\frac{1}{2})^r$  implying that each effective bifurcation reduced the strand concentration in half, but such a strong assumption is not necessary.

$$\bar{C}_s = \frac{C_0}{\alpha} \int_0^{\infty} dr \exp[-r(1/\alpha + 1/\delta)] \quad (30)$$

$$= C_0 / (1 + \alpha/\delta) = C_0 / g(t)$$

which therefore constrains the ratio  $\alpha/\delta$  to be

$$(\alpha/\delta) = g(t) - 1 \quad (31)$$

and leads to the same ensemble mean concentrations of

$$\bar{C} = C_0 \rho / g \quad (32)$$

obtained assuming uniform concentration strands. Perhaps, not surprisingly, the average age parameter  $\alpha$  has a time dependence proportional to  $g-1$ . At the source  $g=1$  and the distribution becomes an infinite spike at  $r=0$  (i.e., the delta function  $\delta(r)$ ) corresponding to the emission of unaged strands of concentration  $C_0$ . It is also noteworthy that, even at great distances from the source, there will be a finite probability of detecting an undiluted strand with the original concentration  $C_0$ .

Computation of higher moments of the observed concentration must recognize that for any one sample only  $j$  of the  $N_r$  strands are populated with pollutant. Thus, the mean square concentration for  $j$  polluted strands involves the  $j$ -dimensional integral

$$\begin{aligned} \overline{C^2(j)} &= \frac{C_0^2}{N_r^2 \alpha^j} \int_0^{\infty} dr_1 \int_0^{\infty} dr_2 \dots \int_0^{\infty} dr_j [e^{-r_1/\delta} + e^{-r_2/\delta} + \dots + e^{-r_j/\delta}]^2 \\ &\quad \cdot e^{-(r_1 + r_2 + \dots + r_j)/\alpha} \end{aligned} \quad (33)$$

$$= \frac{C_0^2}{N_r^2} \left[ \frac{j}{1 + 2\alpha/\delta} + \frac{j^2 - j}{(1 + \alpha/\delta)^2} \right]$$

and the ensemble mean square concentration

$$\overline{C^2} = \sum_j P(j) \overline{C^2(j)}$$

involving the sum over  $j$  from zero to  $N_r$  becomes

$$\overline{C^2} = \frac{C_0^2}{N_r^2} \left[ \frac{N_r \rho}{1+2\alpha/\delta} + \frac{N_r(N_r-1)\rho^2 + N_r\rho - N_r\rho}{(1+\alpha/\delta)^2} \right]$$

after invoking the expressions for mean  $j$  and  $j^2$  given by Eqs. (16) and (17). Cancelling  $N_r\rho$  contributions in the second term and replacing  $\alpha/\delta$  with  $g-1$  then yields the final results

$$\overline{C^2} = \frac{C_0^2}{N_r} \left[ \frac{\rho}{2g-1} + \frac{(N_r-1)\rho^2}{g^2} \right] \quad \text{and} \quad (34)$$

$$\sigma_C^2 = \frac{C_0^2}{N_r} \left[ \frac{\rho}{2g-1} - (\rho/g)^2 \right] \quad (35)$$

for the ensemble mean square concentration and concentration variance respectively. The corresponding conditional estimates can then be immediately written down as  $\gamma^{-1}$  times the corresponding ensemble mean values given by Eqs. (32) and (34), since excluding the  $j = 0$  terms from the sum requires renormalization of the probabilities  $P(j)$  by the factor  $\gamma^{-1} = (1-P(0))^{-1}$  to ensure that the conditional probabilities  $P_c(j)$  obey the constraint

$$\sum_j P_c(j) = 1 \quad j = 1, 2, \dots, N_r$$

where  $P_c(j) = P(j)/\gamma$ . The equivalent conditional variance is then just

$$\sigma_{cp}^2 = \frac{c_0^2}{\gamma N_r} \left[ \frac{\rho}{2g-1} - (\rho/g)^2 (N_r(1/\gamma-1)+1) \right] \quad (36)$$

and the conditional concentration fluctuation intensity,  $i_p$ , given by

$$i_p^2 = \frac{\gamma}{N_r} \left[ \frac{g}{D(2g-1)} - N_r(1/\gamma-1)-1 \right] \quad (37)$$

Far from the source  $g \gg 1$  and  $D \ll 1$  so that the first term dominates and  $i_p \approx [\gamma/(2N_r D)]^{1/2}$ , revealing the increasing fluctuation intensity as one moves away from the plume centerline.

### 3. Inclusion of Additional Effects

The theory development to this point has focussed on the problem of determining ensemble statistical measures. Experimental evaluation of these measures would require repeated sampling at fixed points relative to the centerline of the instantaneous plume. While perhaps not an impossible experimental feat, it is of greater relevance to adapt the theory to the case of a plume meandering back and forth across a receptor fixed in space and accepting samples without regard to the plume's position.

In the case of the Gaussian plume model example we are able to build upon the meandering plume formalism developed by Gifford (1959). Defining a coordinate system where the receptor is located at a point  $(y, z)$  relative to the mean centerline position of the plume, the position of the receptor relative to the plume centerline at any instant is  $(y-y', z-z')$  and occurs with probability

$$P(y', z') = \frac{1}{2\pi\sigma_{yH}(x)\sigma_{zH}(x)} \exp\left[-\frac{1}{2}(y'/\sigma_{yH}(x))^2 - \frac{1}{2}(z'/\sigma_{zH}(x))^2\right],$$

neglecting the influence of ground reflections. Referring back to Eq. (32) and recalling that  $D = \rho/g$  we may immediately recompute the mean concentration as

$$\bar{C} = C_0 \int_{-\infty}^{+\infty} \int dy' dz' P(y', z') D(x, y-y', z-z') = C_0 D_T(x, y, z) \quad (39)$$

where  $D$  is given by Eq. (4) and  $D_T$  has the same form as  $D$  but uses the total sigma,  $\sigma_T = (\sigma^2 + \sigma_M^2)^{1/2}$  in place of the instantaneous sigma measures  $\sigma$ , referred to as  $\sigma_y$  and  $\sigma_z$  throughout this paper.

Computation of the mean square concentration involves a similar integral averaging of Eq. (34) and can be written

$$\overline{C^2} = \frac{C_0^2}{N_r} \int_{-\infty}^{+\infty} \int dy' dz' P(y', z') \left[ \frac{\rho}{2g-1} + (N_r-1)D^2 \right] \quad (40)$$

where

$$\begin{aligned} D &= D(x, y-y', z-z') \quad , \\ \rho &= \rho(x, y-y', z-z') \quad , \\ g &= \rho/D \end{aligned}$$

and  $\rho(x, y-y', z-z')$  can be approximated with the help of Eq. (12). Given the complexity of Eq. (12), evaluation of Eq. (40) will either require additional approximations or numerical integration.

Inclusion of the effects of a finite detector concentration threshold,  $C_T$ , is another complexity that may require numerical evaluation in most cases. Referring back to Eq. (33), the generalized form of the nth moment may be expressed as

$$\overline{C^n(j)} = (C_0/N_r)^{n-j} \int_0^\infty dr_1 \int_0^\infty dr_2 \dots \int_0^\infty dr_j [e^{-r_1/\delta} + e^{-r_2/\delta} + \dots + e^{-r_j/\delta}] \quad (41)$$

$$\cdot e^{-(r_1+r_2+\dots+r_j)/\alpha} \theta(e^{-r_1/\delta} + e^{-r_2/\delta} + \dots + e^{-r_j/\delta} - C_T N_r / C_0)$$

where the theta function  $\theta(x)$  is defined as

$$\theta(x) = \begin{cases} 0 & \text{for } x < 0 \\ 1 & \text{for } x \geq 0 \end{cases}$$

Need for the  $\theta$ -function disappears as  $C_T \rightarrow 0$  and can be replaced by altered integration limits for  $N_r = 1$  or 2, but we have not yet developed a general, analytic solution for Eq. (41); however, integrals of this type are relatively common in statistical mechanics. Once accomplished, one could compute the weighted sum over  $j$  followed by the integral averaging suggested by Eq. (40).

Another important effect of the  $\theta$  function in Eq. (41) is that the  $n = 0$  normalization moment is less than unity so that the probabilities given by Eq. (13) are modified and become

$$P'(j) = P(j) \overline{C(j)} \quad (42)$$

This has the immediate effect of reducing the value of  $\gamma$  from that given by Eq. (15) to

$$\gamma = 1 - P(0) - \sum_{j=1}^{N_r} P(j) [1 - \overline{C^0(j)}] \quad (43)$$

thus, indicating the significance of instrumental "details" on the outcome of concentration fluctuation experiments.



Perhaps the last refinement that would be considered essential for application to experimental data is the correction for averaging time. For very small averaging times,  $\tau_a$ , much smaller than the Lagrangian time scale,  $\tau_L$ , associated with the principal, meander driving eddies, it is thought that the temporal smearing is equivalent to an increased receptor aperture diameter  $d_r'$  defined as

$$d_r' = d_r + (\sigma_v \sigma_w)^{1/2} \tau_a, \quad (44)$$

where  $\sigma_v$  and  $\sigma_w$  are the relevant lateral and vertical turbulent velocities. This then has the effect of increasing the number of strands received by the receptor to

$$N_r' = (d_r'/\lambda)^2 = N_r [1 + (\sigma_v \sigma_w)^{1/2} \tau_a / d_r]^2 \quad (45)$$

which in turn has the effect of lowering the magnitude of the concentration fluctuations as given by Eq. (35) for example.

Extension of the theory to longer time averaging periods remains to be worked out in detail, but may require further assumptions about the form of the auto-correlation function and  $\tau_a/\tau_L$ . Venkatram (1979) provides results of assuming an exponential form for this function and Hanna (1984) discusses other treatments of this problem.

Finally, it should be noted that the strand theory can easily be extended to multiple species. This extension could be an important step in modeling the influence of sub-grid scale inhomogeneities on processes involving non-linear chemical reactions.

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