SIMULATING CONCENTRATION FLUCTUATION TIME SERIES WITH INTERMITTENT ZERO PERIODS AND LEVEL DEPENDENT DERIVATIVES

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Abstract. Intermittent concentration fluctuation time series were produced with a stochastic numerical model derived from the assumption that the concentration fluctuations at a fixed receptor in a point-source plume can be modelled as a first order Markov process. The time derivative of concentration was assumed to be level-dependent and constrained by a stationary lognormal probability density function. The input parameters required to reconstruct the intermittent time series are the intermittency factor γ, the conditional fluctuation intensity i_p^2, and the time scale T_c. A clipped lognormal probability distribution was used to describe the fluctuation time series. Good agreement between the stochastic simulation and experimental water-channel data was demonstrated by comparing the time derivative of concentration and the upcrossing rates over a range of intermittency factors γ = 0.7 to 0.01 and fluctuation intensities i_p^2 = 2.2 to 7.5.

Keywords: Time series, Time derivative of concentration, Concentration fluctuations, Stochastic simulation.

1. Introduction

The inhomogeneous mixing of a point-source plume of contaminant produces large fluctuations in contaminant concentration at a fixed receptor. The instantaneous concentrations can range from zero (background) concentration to more than 20 times the mean concentration. Two examples of typical time series of concentration fluctuations are shown in Figure 1.

Toxicity, flammability and odour effects have a strong non-linear dependence on concentration. For example, ten Berge et al. (1986) analyzed animal experiments for 20 different acutely toxic gases and found that fatalities are a function of concentration C^n where $n = 1.0$ to 3.5. Most chemicals had an exponent n value in the range of 2.0 to 3.0. The Center for Chemical Process Safety of the American Institute of Chemical Engineers (1989) recommends similar non-linear models for predicting acute toxicity from common industrial chemicals.

If the exponent $n = 1$ then high concentrations are no more important than low concentrations and the only variable determining toxicity is the mean concentration C. However, if $n > 1$, as it is for many substances, then concentration fluctuations increase the toxicity of the exposure because high concentrations become

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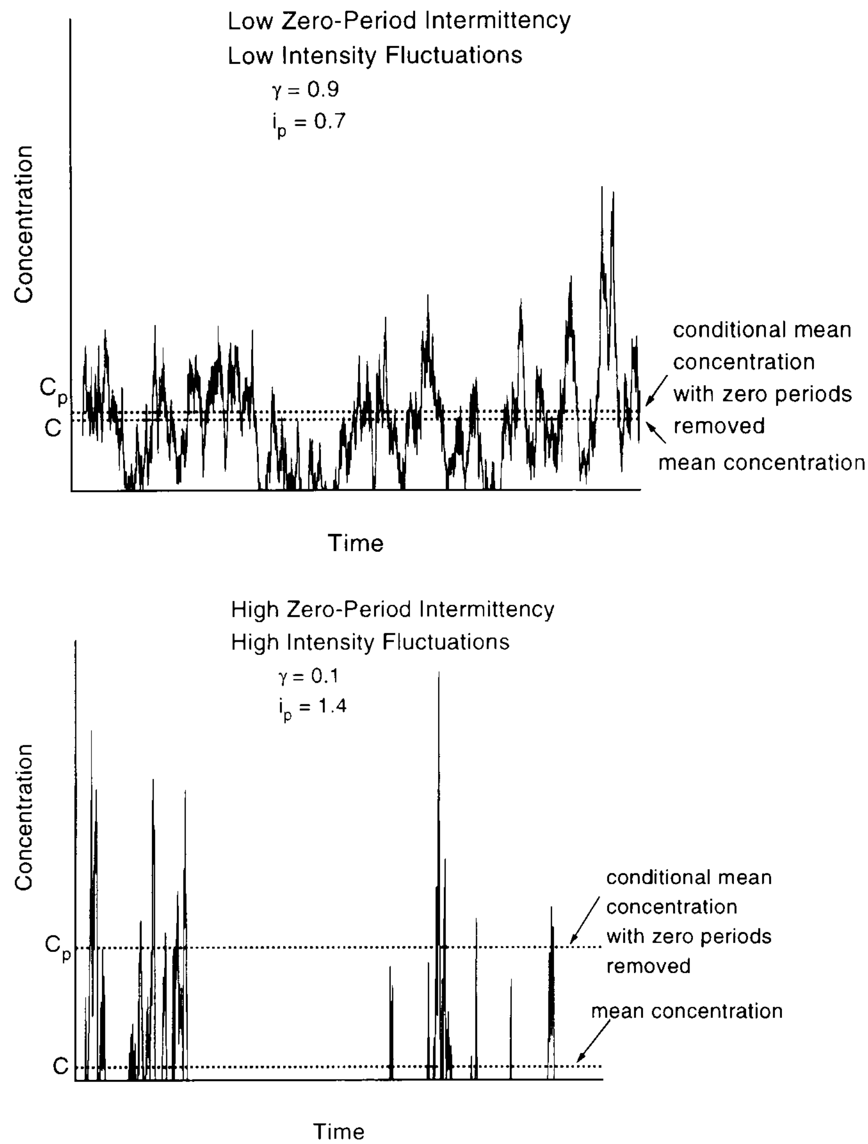

Figure 1. Typical intermittent concentration fluctuation time series with low intermittency (γ = 0.9) and high intermittency (γ = 0.1).

much more important than low concentrations. In a typical point-source gas plume, where the fluctuation standard deviation, c', is often several times larger than the mean concentration, C, fluctuations have a significant effect on the predicted toxic hazard.

The current body of work on concentration fluctuations, reviewed in Wilson (1995), approaches the problem both experimentally and with theoretical models to predict the variance, skewness, kurtosis and other statistics of the fluctuations. Recent work by Yee et al. (1993a, 1994, 1995) recognizes that the evaluation of toxic hazards from a release requires information on concentration level recurrence time intervals, intermittency, and level-crossing statistics in addition to the probability distributions and higher order concentration moments. The stochastic time series reconstruction proposed in the present study can be used to evaluate all of these statistical measures from user specified values of the mean, variance, intermittency and fluctuation time scale.

In this study, time series of intermittent concentration fluctuations are reconstructed as a first-order 'memoryless' Markov process using an input intermittency factor, fluctuation intensity, time scale of fluctuations and a specified probability density function. The stochastic model used to produce these intermittent time series is an extension of the non-intermittent model by Du et al. (1999). Our objective is to generate an ensemble of realistic random time series of intermittent concentration fluctuations that can be applied directly to a hazard model. This direct approach allows the user to examine the uncertainty and variability of hazardous effects between realizations in an ensemble, and to apply complex hazard models that cannot be used if only the overall statistics of the fluctuations are known.

2. Probability Distributions of Intermittent Time Series

A probability distribution, in the form of a probability density function (pdf), is a key input to the proposed stochastic reconstruction. The pdf constrains the fluctuating concentrations to ensure the correct mean, variance, and intermittency in the reconstructed time series.

Probability distributions of intermittent concentration fluctuations usually focus on the conditional in-plume concentrations with the intermittent periods described by a delta function at zero concentration. Wilson (1995) examined several different distributions and recommended the lognormal as the best fit to a wide variety of data. The choice of the lognormal pdf for the present study is supported by the water-channel concentration data analyzed by Yee et al. (1993). Zelt (1992) found that the shape of the pdf evolves with plume travel time, and that lognormal, gamma, and clipped normal pdfs may be used as approximations as the plume evolves. Yee et al. (1995) analyzed full scale atmospheric concentration fluctuation measurements for a fast response detector with an upper frequency cutoff of several hundred hertz and concluded that the gamma pdf gave a better fit than the lognormal. Yee and Chan (1997) recommended a clipped gamma pdf for intermittent concentration fluctuations.

In the present study, a clipped lognormal pdf is used to describe the intermittent concentration fluctuations and to meet the requirements for the stochastic model used to reconstruct intermittent time series.

2.1. Shifted and Clipped Lognormal PDFs

The primary restriction on the choice of pdf is that it must describe both in-plume concentrations and zero concentration intermittent periods. The probability of obtaining a non-zero concentration must be equal to the intermittency factor, γ, and the probability of obtaining a zero concentration must be equal to $(1-\gamma)$. A simple delta function cannot be used to account for the zero periods because the stochastic model becomes mathematically trapped in a delta function and the time series of concentration will not go above zero after hitting zero concentration.

To meet these pdf requirements, the stochastic model is expressed in pseudo-concentration coordinates c_+ where the subscript '+' denotes parameters related to these pseudo-concentrations. Step 1 in Figure 2 shows the pseudo-concentration c_+ time series and pdf. In c_+ coordinates, the concentration fluctuations are represented by a complete lognormal distribution with only positive concentrations and an intermittency factor $\gamma_+ = 1.0$. There are no intermittent periods in c_+.

After a time series is generated, the concentrations are shifted by a value of c_{base} to give positive and negative concentrations $\tilde{c}$ where:

$$\tilde{c} = c_+ - c_{\text{base}}. \tag{1}$$

In $\tilde{c}$ coordinates, the probability of obtaining a positive concentration is equal to the intermittency factor γ and the probability of obtaining a negative concentration is $(1-\gamma)$. The magnitude of the negative concentration is interpreted physically as being inversely related to the likelihood of obtaining a positive concentration in the next time step. The shifted lognormal is shown in Step 2 of Figure 2 for a typical time series and the corresponding pdf.

Negative concentrations are clearly unrealistic, so the final step in interpreting the simulated time series is to clip all of the negative concentrations and replace them with zeros. This gives a pdf with a delta function at zero concentration. As previously discussed, a delta function pdf cannot be used directly in the stochastic simulation, but it is required to make use of the simulated intermittent time series. The final result is an intermittent time series with $c \geq 0$ and a clipped lognormal pdf as shown in Step 3 of Figure 2.

2.2. Physical Interpretation of the Shifted Lognormal

A physical interpretation of the shifted lognormal is that concentration fluctuations and intermittent periods are produced by the same physical mixing process as shown in Figure 3. Eddies with some positive concentration of contaminant flow by a point and cause non-zero concentration fluctuations. Similarly, eddies of clean air flowing by the same point cause intermittent periods. The positive concentrations of the shifted lognormal describe contaminated eddies while the negative concentrations describe clean air eddies. The magnitude of the negative shifted concentration $\tilde{c}$ in Figure 3 is inversely related to the likelihood that the

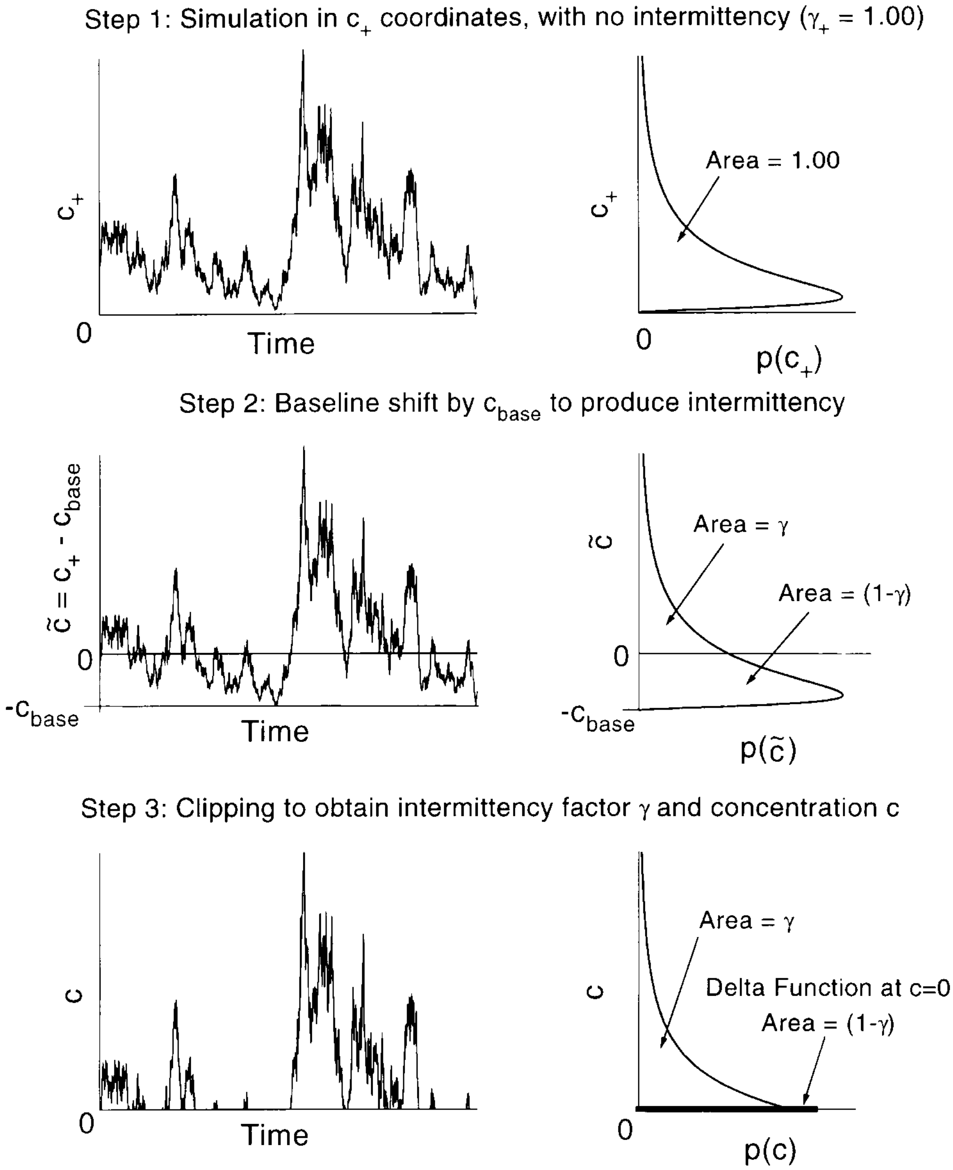

Figure 2. Clipping procedure to produce an intermittent time series.

concentration will be non-negative in the next time step. The larger the negative concentration $\tilde{c}$, the larger the eddy of clean air and the less likely that a concentration greater than zero will occur in the next time step. With this interpretation the stochastic model requires only a single integral time scale T_c to define both the zero and the non-zero concentrations.

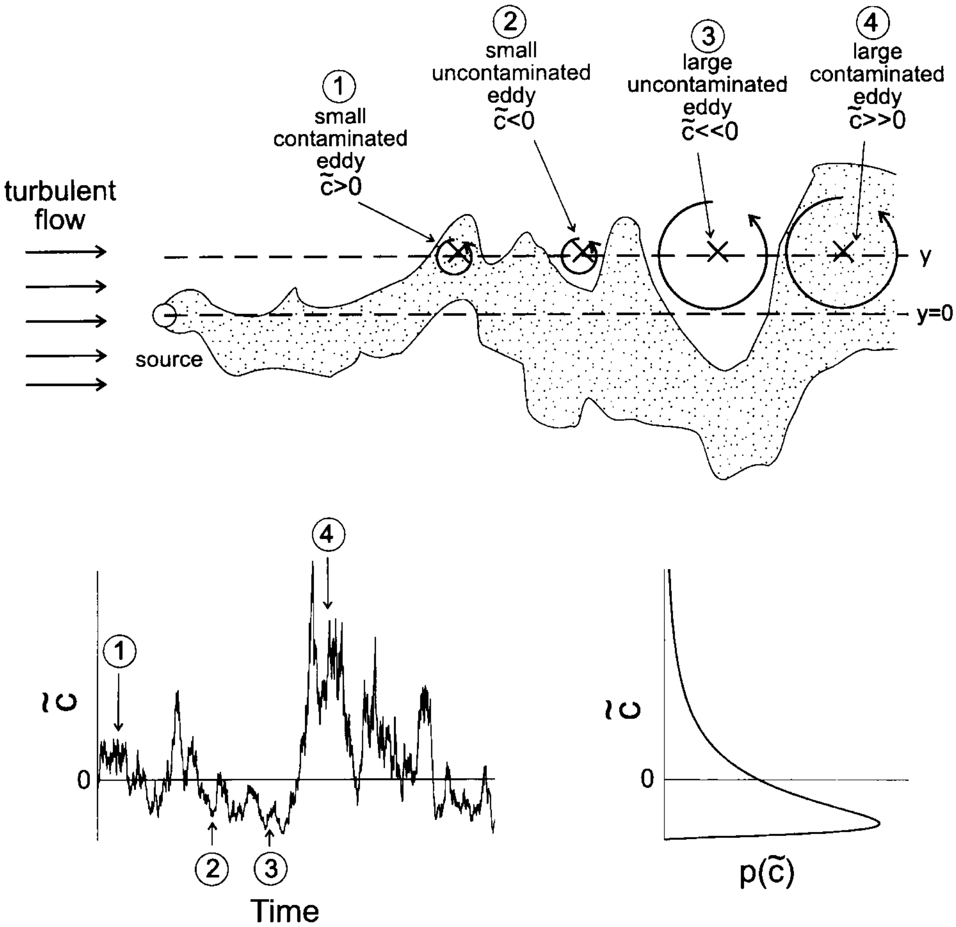

Figure 3. Physical model for interpreting the pdf of negative concentrations as intermittent periods of clean air (zero concentration).

After the shifting is complete, all of the negative concentration values are converted to zero concentration intermittent periods and the result is a clipped lognormal pdf of concentration. Clipped distributions have been used in the past to describe concentration fluctuations, but not with this interpretation. For example, Lewellen and Sykes (1986) used a clipped normal pdf to describe intermittent plumes from a power plant, but their interpretation did not attach any physical significance to the magnitude of the negative concentrations and simply replaced them by a delta function at zero concentration. As pointed out by Bara et al. (1992) and Wilson (1995), the clipped normal is not appropriate for general use because its functional form can only produce in-plume fluctuation intensities less than 1.0. Typical in-plume fluctuation intensities in a point-source plume range from 0.5 to 3.0, with most values greater than unity, making the clipped normal an inappropriate choice.

3. Clipped Lognormal Distribution

The properties of the lognormal pdf are well documented in Aitchison and Brown (1957), and Crow and Shimizu (1988), who also include some information on clipped or truncated lognormals. The clipped lognormal is not widely used and some discussion of its basic statistics and application to the stochastic model is required. A lognormal pdf in c_+ coordinates is used for the stochastic simulation as explained in Section 2, but the clipped lognormal in c coordinates must have the correct statistics for the intermittent time series.

There are two sets of statistics that can be considered for an intermittent time series. Conditional statistics exclude intermittent periods of zero concentration and are denoted by a subscript 'p'. Total statistics include zero periods as well as non-zero concentrations and have no subscript. For example, C is the mean concentration including the zero periods, while C_p is the conditional mean concentration that excludes the zero periods. Uppercase symbols denote mean values.

To use the clipped distribution in the stochastic model, the c_+ lognormal distribution must be chosen so that after it is clipped by c_{base}, as shown in Figure 2, the desired intermittency factor γ and the conditional fluctuation intensity i_p^2 are obtained. Here, the intermittency factor γ is defined as the fraction of the total time during which the concentration is greater than zero. The conditional fluctuation intensity i_p^2 is:

$$i_p^2 = \frac{c_p'^2}{C_p^2}, \tag{2}$$

where $c_p'^2$ is the conditional variance of the non-zero concentrations and C_p is the conditional mean excluding the zeros.

3.1. Normalized Concentrations

The clipped lognormal is simplified by normalizing all concentrations by the median concentration, c_{50}, that includes all of the zero concentrations. The dimensionless concentrations will be denoted by ϕ, where $\phi = c/c_{50}$ is the instantaneous dimensionless concentration, $\Phi = C/c_{50}$ is the mean dimensionless concentration, $\Phi_p = C_p/c_{50}$ is the conditional (in-plume) mean dimensionless concentration, $\phi' = c'/c_{50}$ is the standard deviation of the dimensionless concentration, and $\phi'_p = c'_p/c_{50}$ is the conditional standard deviation of the dimensionless concentration excluding the zero periods.

In pseudo-concentration '+' coordinates all concentrations are normalized by the median concentration c_{50+} of the pseudo-concentration time series. Here $\phi_+ =$

c_+/c_{50+} is the instantaneous dimensionless pseudo-concentration, $\Phi_+ = C_+/c_{50+}$ is the mean dimensionless pseudo-concentration, and $\phi'_+ = c'_+/c_{50+}$ is the standard deviation of the dimensionless pseudo-concentration.

3.2. Lognormal distribution

For the non-intermittent lognormal distribution in pseudo-concentration '+' coordinates, the probability density function (pdf) is:

$$p(\phi_+) = \frac{1}{\sqrt{2\pi}\,\sigma_{l+}\phi_+} \exp\left(-\frac{\ln^2\left(\frac{\phi_+}{\phi_{50+}}\right)}{2\sigma_{l+}^2}\right), \tag{3}$$

with mean

$$\Phi_+ = \exp\left(\frac{\sigma_{l+}^2}{2}\right). \tag{4}$$

and variance

$$\phi_+'^2 = \Phi_+^2(\exp(\sigma_{l+}^2) - 1). \tag{5}$$

3.3. Intermittency factor of clipped lognormal

To produce the specified intermittency factor γ, the pseudo-concentration lognormal distribution is shifted by ϕ_{base} to transform the simulated time series from strictly positive concentrations ϕ_+ to $\tilde{\phi}$ with positive and negative concentrations, where the negative values represent the intermittent periods of zero concentration,

$$\tilde{\phi} = \phi_+ - \phi_{\text{base}}. \tag{6}$$

The shift ϕ_{base} (see Figure 2) must be chosen so that the probability of observing a concentration greater than zero is equal to the intermittency factor γ, using

$$\gamma = \int_{\phi_{\text{base}}}^{\infty} p(\phi_+)d(\phi_+), \tag{7}$$

with Equation (3) and solving for γ in terms of the two unknowns ϕ_{base} and σ_{l+}

$$\gamma = \frac{1}{2}\left(1 - \text{erf}\left(\frac{\ln(\phi_{\text{base}})}{\sqrt{2}\sigma_{l+}}\right)\right). \tag{8}$$

3.4. Fluctuation Intensity of Clipped Lognormal

In addition to the intermittency factor γ, the conditional fluctuation intensity i_p^2 must be specified to fully describe the clipped lognormal. After shifting the non-intermittent pseudo-concentrations ϕ_+ by ϕ_{base}, the negative $\tilde{\phi}$ concentrations are transformed in Step 3 of Figure 2 to a delta function at zero concentration with probability $(1-\gamma)$. This leaves only positive and zero concentrations for the normalized dimensionless concentration ϕ.

The intermittent time series mean Φ of the clipped lognormal is:

$$\Phi = \frac{\exp\left(\frac{\sigma_{l+}^2}{2}\right)}{2}\left(1-\operatorname{erf}\left(\frac{\ln\phi_{\text{base}}-\sigma_{l+}^2}{\sqrt{2}\sigma_{l+}}\right)\right) - \frac{\phi_{\text{base}}}{2}\left(1-\operatorname{erf}\left(\frac{\ln\phi_{\text{base}}}{\sqrt{2}\sigma_{l+}}\right)\right), \tag{9}$$

with total second moment $\overline{\phi^2}$

$$\overline{\phi^2} = \frac{\exp(2\sigma_{l+}^2)}{2}\left(1-\operatorname{erf}\left(\frac{\ln\phi_{\text{base}}-2\sigma_{l+}^2}{\sqrt{2}\sigma_{l+}}\right)\right) - \phi_{\text{base}}\exp\left(\frac{\sigma_{l+}^2}{2}\right)\left(1-\operatorname{erf}\left(\frac{\ln\phi_{\text{base}}-\sigma_{l+}^2}{\sqrt{2}\sigma_{l+}}\right)\right) + \frac{\phi_{\text{base}}^2}{2}\left(1-\operatorname{erf}\left(\frac{\ln\phi_{\text{base}}}{\sqrt{2}\sigma_{l+}}\right)\right). \tag{10}$$

The conditional moments (denoted by a subscript 'p'), including only non-zero in-plume concentration periods, are related by the definition of the intermittency factor γ to the intermittent time series moments by:

$$\gamma\overline{\phi_p^n} = \overline{\phi^n}. \tag{11}$$

By definition, the second moment $\overline{\phi^2}$ is related to the mean Φ and the second moment about the mean ϕ'^2 by:

$$\overline{\phi^2} = \Phi^2 + \phi'^2. \tag{12}$$

The conditional fluctuation intensity i_p^2 is defined as

$$i_p^2 = \frac{\phi_p'^2}{\Phi_p^2}, \tag{13}$$

with Equations (11), (12) and (13):

$$i_p^2 = \frac{\gamma \overline{\phi^2}}{\Phi^2} - 1. \tag{14}$$

Using the definitions of the moments $\overline{\phi^2}$ from Equation (10) and Φ from Equation (9) in Equation (14) relates the input i_p^2 to the unknowns ϕ_{base} and σ_{l+}.

3.5. Lognormal in c_+ Coordinates

Equations (8) and (14) can be used to calculate the zero shift ϕ_{base} and the log standard deviation σ_{l+} in terms of the specified fluctuation intensity i_p^2 and intermittency factor γ. Because these are implicit relationships, ϕ_{base} and σ_{l+} were found numerically using a simple iterative bisection method.

The numerical solution for Equations (8) and (14) does not converge for all possible values of i_p^2 and γ. Figure 4 shows the lower boundary of solutions possible for given i_p^2 and γ combinations. Although the vertical axis in Figure 4 ends at $i_p^2 = 3.0$, values as high as $i_p^2 = 10^4$ were checked and found to have solutions. Fortunately, the presence of this lower boundary of solutions does not cause any problem because real plumes are well within the valid range. Figure 4 also shows an empirical relationship suggested by Wilson (1995) for typical i_p^2 versus γ values in atmospheric plumes.

Using the solution for σ_{l+} and ϕ_{base}, the value of the dimensionless conditional mean Φ_p can be calculated. The value of the median concentration c_{50+} can be found from the definition of Φ_p where $c_{50+} = C_p/\Phi_p$ with the specified conditional mean concentration C_p. Using this value of c_{50+} the value of $c_{\text{base}} = \phi_{\text{base}} c_{50+}$. The log standard deviation σ_{l+} is the same in both dimensionless ϕ_+ coordinates and dimensioned c_+ coordinates.

The c_{50+} and σ_{l+} values are required parameters for the c_+ lognormal pdf:

$$p(c_+) = \frac{1}{\sqrt{2\pi}\sigma_{l+} c_+} \exp\left(-\frac{\ln^2\left(\frac{c_+}{c_{50+}}\right)}{2\sigma_{l+}^2} \right), \tag{15}$$

and the c_{base} value is used after the simulated time series is generated to clip negative values off the series and pdf.

4. Stochastic Model for Fluctuations

Du et al. (1999) developed a numerical stochastic model to predict upcrossing rates of conditional (in-plume) concentration fluctuation time series. The limitation of this model is that it does not account for intermittent periods of zero (background)

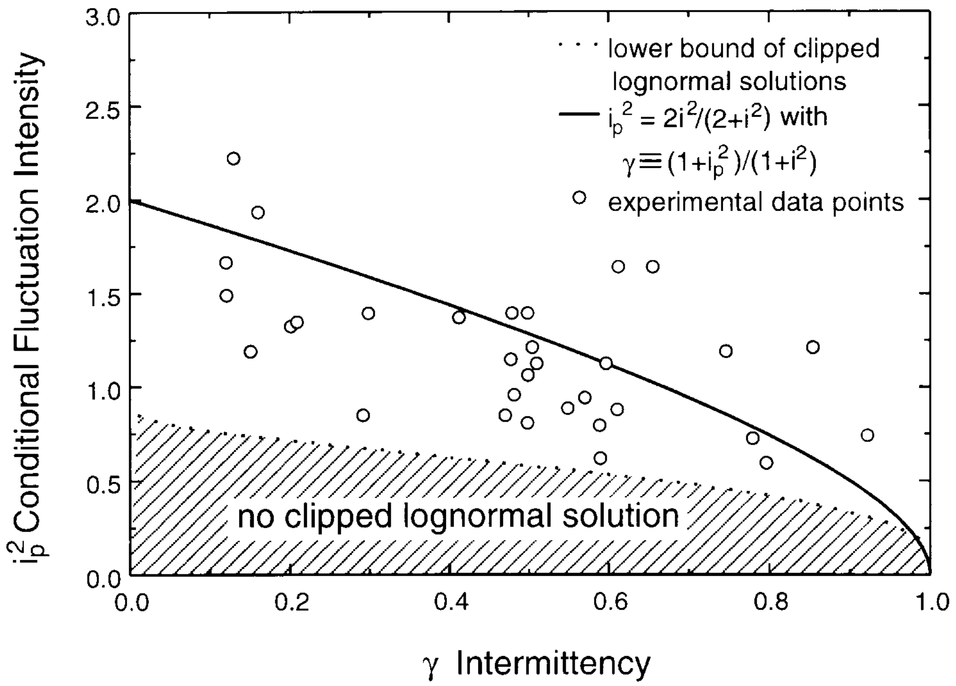

Figure 4. Boundary of clipped lognormal solutions in terms of the intermittency γ and the conditional fluctuation intensity i_p^2. The solid line is the empirical relationship between i_p^2 and γ suggested by Wilson (1995) after considering the experimental data points from full scale and laboratory scale experiments. Experimental data points from Mylne and Mason (1991) and Fackrell and Robins (1982).

concentration that are observed experimentally. In the present study, this model is extended to include the simulation of the intermittent periods.

The basic assumptions of the Du et al. (1999) model are that:

- Eulerian concentration fluctuations are produced by a first order Markov process that can be described by a stochastic differential equation, and by the equivalent Fokker–Planck equation for the time dependent evolution of the concentration probability distribution.
- the derivative of concentration is dependent on the current instantaneous concentration.
- concentration fluctuations are statistically stationary.

A lognormal distribution in non-intermittent pseudo-concentration c_+ coordinates is used for the stochastic model so that all simulated concentrations are greater than zero as discussed in Section 2 and shown in Figure 2. All parameters calculated from this time series of pseudo-concentrations are denoted by the subscript ‘$+$’.

4.1. Stochastic differential equation

We assume that concentration fluctuations at a fixed location in a dispersing plume can be described as a first-order Markov (memoryless) process given by the one-dimensional stochastic differential Langevin equation:

$$\mathrm{d}c_+ = a(c_+, t)\,\mathrm{d}t + b(c_+, t)\,\mathrm{d}\zeta, \tag{16}$$

where $a(c_+, t)$ is the deterministic portion of the time derivative, dependent on concentration c_+ and time t; and $b(c_+, t)\,\mathrm{d}\zeta$ is a random forcing function where $\mathrm{d}\zeta$ is a Gaussian random number with a mean of zero, variance $\mathrm{d}t$ and no dependence on the instantaneous level of c_+.

The Langevin equation, discussed in detail by Gardiner (1983) and Durbin (1983), has been used to describe a wide variety of continuous stochastic processes. Originally, the equation was developed to describe Brownian particle motion, see Gardiner (1983). It has also been applied to modelling concentration fluctuations in the moving Lagrangian frame of reference by tracking the random flights of particles emanating from a point, see Wilson and Sawford (1996). Here, we apply the Langevin equation in a fixed Eulerian frame of reference by assuming that the concentration time series at a point can be modelled as a continuous memoryless Markov process.

4.2. Fokker–Planck constraint

If we assume that the pdf of concentration c_+ is stationary, a useful constraint between the a and b terms can be derived by writing a differential equation for the time evolution of the pdf, and then setting $\partial p/\partial t = 0$. Durbin (1983) provides a detailed derivation of the deterministic relationship for the time evolution of the pdf in a Markov process. The basic procedure is to consider a pdf of concentration c_+ at time t. By definition, a Markov process has no memory of previous states and this requires that the pdf at time t depends only on the pdf at time $(t - \mathrm{d}t)$ and some transition probability between concentration c_+ at time t and concentration $(c_+ - \mathrm{d}c_+)$ at time $(t - \mathrm{d}t)$. Integrating over all possible $\mathrm{d}c_+$ values, and substituting moments calculated for the stochastic process from Equation (16), produces a deterministic equation describing the time evolution of the probability density function $p(c_+)$, as given in Du et al. (1999)

$$\frac{\partial p}{\partial t} = -\frac{\partial(ap)}{\partial c_+} + \frac{1}{2}\frac{\partial^2(b^2 p)}{\partial c_+^2}. \tag{17}$$

Equation (17) is the one-dimensional Fokker–Planck equation. Additional discussion can be found in Gardiner (1983). The Fokker–Planck equation constrains the evolution of the probability distribution of concentration with the relationship between the a and b terms.

Stationarity requires that a, b and the pdf p do not change with time. With this assumption $\partial p/\partial t = 0$ and Equation (17) can be integrated once to yield:

$$\frac{\mathrm{d}(b^2 p)}{\mathrm{d}c_+} = 2ap + K, \tag{18}$$

where K is a constant of integration. Following Du et al. (1999) integrating (18) to solve for b^2 in terms of a produces

$$b^2 = \frac{2}{p} \int_{c_+}^{\infty} -ap \, \mathrm{d}c_+, \tag{19}$$

where the integration constant K from Equation (18) must be zero to prevent b^2 from becoming infinite at all concentration levels. Equation (19) is a deterministic relationship between the pseudo concentration time series generation parameters a and b in Equation (16) and the probability density function p specified by the user.

5. Generating Stochastic Time Series

The stochastic differential Equation (16) was solved numerically by using a forward difference:

$$c_{+(n+1)} = c_{+(n)} + a_n \Delta t + b_n \sqrt{\Delta t} N_n, \tag{20}$$

where $c_{+(n+1)}$ is the instantaneous c_+ concentration at time t_{n+1}, $c_{+(n)}$ is the instantaneous concentration at time t_n, Δt is the time increment, and N_n is a Gaussian random number with zero mean and unity variance. The $\sqrt{\Delta t}$ in Equation (20) arises from the definition of the random fluctuation process in Equation (16). The Gaussian random number $\mathrm{d}\zeta$ has a mean of zero and a variance $\mathrm{d}t$ and the units of $\mathrm{d}\zeta$ are $\sqrt{\text{time}}$. In Equation (20) the Gaussian random number is normalized by $\sqrt{\Delta t}$ because it is more convenient to generate random numbers with mean 0 and variance 1. This leaves a $\sqrt{\Delta t}$ in the numerator of the second term of Equation (20).

A uniform distribution of random numbers was generated with a shift register sequence generator, see Maier (1991) and Carter (1994). This random number generator was used because it has a period of approximately 9×10^{74} before repeating. A large period generator is required to avoid repetition in the random numbers that would cause invalid stochastic simulation results. The Box–Muller (1958) transformation was used to obtain a Gaussian distributed random number N_n from the uniform distribution.

Each run was started at the median concentration c_{50+} and then allowed to run for $10T_{c+}$ to eliminate the effects of picking the same starting point for each time series. The random number generator was seeded based on the clock time at which the run started.

The stochastic simulation ran for a total of at least 5000 integral time scales to allow a sufficient length of time to produce the rare peak concentrations that define the tail of the pdf of concentration where $c_+ \gg C_+$. The following guideline was used to determine an appropriate number of time scales, n_T, needed for each realization generated for this paper,

$$n_T \simeq \frac{5000 i_p}{(\gamma + 0.1)}. \tag{21}$$

The number of time scales n_T varies inversely with γ because as the intermittency factor decreases there are fewer excursions above the zero concentration level in a given length of time, and so a longer duration is required to ensure that a sufficient amount of non-zero data are generated. Similarly, as i_p increases, the fluctuations become larger and additional time is required to capture all of the rare events.

5.1. Time Step for Numerical Model

If we could solve the stochastic differential equation exactly we would find that the time derivative of concentration goes to infinity in the limit as $\Delta t \rightarrow 0$. However, in an actual concentration fluctuation time series the root-mean-square (rms) derivative is finite because molecular diffusion smears out the small-scale structure that produces high frequency derivatives. In the stochastic simulation we remove the high frequencies in two ways: first, the finite time steps automatically cut off derivatives that are faster than the time step, and second, we apply a low pass filter to match the probe response in the experimental data that further limits the maximum time derivative. This filtering will be discussed later in the paper.

The stochastic model produces a time series of concentration fluctuations that has been sampled at a frequency of $1/\Delta t$. Since the purpose of the simulation is to simulate atmospheric releases, we checked that this sampling frequency did not exceed the frequencies that are possible in the real atmosphere due to limits imposed by diffusion and viscosity.

In the atmosphere, the mass diffusivity D of most gases is about the same as the molecular viscosity ν of air, the Schmidt number Sc ≈ 1, and the Kolmogorov microscale of concentrations is approximately equal to the Kolmogorov microscale for turbulence kinetic energy dissipation $\eta_c \simeq \eta$. A typical value is $\eta \simeq 0.001$ m that gives a cutoff wavenumber of $k_{\text{cut}} = 1/\eta = 1000$ m^{-1}. With $k_{\text{cut}} = 2\pi f_{\text{cut}}/U$ and a typical windspeed of $U = 2$ m s^{-1} this corresponds to a cutoff frequency of about $f_{\text{cut}} = 300$ Hz with frequency rolloff beginning at about $0.2 f_{\text{cut}} = 60$ Hz.

In this study, the time step, Δt, was set to $0.01 T_{c+}$, which is short enough to resolve all of the necessary fluctuations and ensures that the values of a and b are not very different at $c(t)$ and $c(t + \Delta t)$. For a typical atmospheric time scale of $T_c = 100$ seconds, the sampling rate is only 1 Hz, about two orders of magnitude below the Kolmogorov cutoff. No physical limitation on fluctuation frequency was present in the simulated concentration fluctuations.

5.2. Functional Relations for a and b

The a term governs the deterministic part of the fluctuation process. In the absence of random fluctuations, a determines the behaviour of the concentration derivative. Experimental evidence from Yee et al. (1993a) and water channel data presented later in this paper show that the rms concentration derivative $\dot{c}'$ increases with the concentration level at which it is measured. That is, large derivatives are observed at extreme concentrations relative to the mean, while small derivatives generally occur near the mean.

The model proposed by Du et al. (1997) for the non-zero (conditional) part of the concentration time series showed that for a stationary process the a term must always be a linear return to mean if we require the same a to work for any concentration pdf p_{c+}. Therefore, a is

$$a = \frac{C_+ - c_+}{T_{c+}}, \tag{22}$$

where T_{c+} is the integral autocorrelation time scale of the pseudo-concentration fluctuations, see Du et al. (1999). The fluctuating time series produced by the stochastic model is inertialess, so T_{c+} may be rescaled to any value without changing the physical basis of the time series. The T_{c+} value used to generate the time series is nominally set to unity, and the value adjusted to the true time scale T_{c+} by rescaling the time axis by multiplying all time steps by T_{c+}.

The pdf $p(c_+)$ must be specified to complete the model. In this study, $p(c_+)$ is a lognormal distribution as discussed in Section 2 and given by Equation (15). The value of the b term is calculated by substituting the pdf and the definition of a from Equation (22) into Equation (19), viz.

$$\begin{aligned} b^2 \;=\; & \frac{C_+\sqrt{2\pi}\,\sigma_{l+} c_+}{T_{c+}} \exp\left(\frac{\ln^2\left(\frac{c_+}{c_{50+}}\right)}{2\sigma_{l+}^2}\right) \\ & \times \left(\operatorname{erf}\left(\frac{\ln\left(\frac{c_+}{c_{50+}}\right)}{\sqrt{2}\sigma_{l+}}\right) - \operatorname{erf}\left(\frac{\ln\left(\frac{c_+}{c_{50+}}\right) - \sigma_{l+}^2}{\sqrt{2}\sigma_{l+}}\right)\right). \end{aligned} \tag{23}$$

The parameters c_{50+} and σ_{l+} are calculated from the user input intermittency factor, γ, and the input conditional fluctuation intensity, i_p^2, as discussed in Section 3.

For a highly intermittent time series, the pseudo-mean, C_+, can be a negative concentration after the shifting necessary to produce intermittent periods. With this in mind, C_+ should be interpreted as a representative concentration that includes the effects of both intermittent zero periods and non-zero fluctuations. If C_+ is less than zero concentration after shifting, it implies that the uncontaminated, $c = 0$, clean air eddies dominate the fluctuation process.

The conditional mean C_{p+} and time scale T_{c+} were arbitrarily set to unity to generate fully normalized time series. Because the model is an inertialess Markov process, the resulting time series is not sensitive to the particular value of the time scale and the output of the simulation can be scaled to match the time scale and mean concentration of any desired time series.

5.3. Time Scales of Intermittent and Non-Intermittent Time Series

The time scale of the non-intermittent pseudo-concentration fluctuations was set to $T_{c+} = 1.0$ for the stochastic simulation. However, the time scale T_c of the actual fluctuations of intermittent concentration c is not equal to 1.0 after clipping. The clipping process removes all fluctuations below c_{base} and replaces them with zero periods. These intermittent zero periods are a defining characteristic of the time series and are included in all analyses. Figure 5 shows the variation in the time scales T_c calculated from the clipped intermittent time series as compared to the T_{c+} time scale.

In general, the time scale T_c of the intermittent concentration fluctuations is shorter than the time scale T_{c+} of the non-intermittent concentrations. This relationship can be approximated as a function of the intermittency factor $T_c/T_{c+} = 0.78 + 0.23\gamma$. The variability between realizations is quite large as demonstrated by the range bars in Figure 5. Fortunately, this is not a difficult problem to deal with since the inertialess nature of the Markov simulation allows the time scale T_c to be rescaled to match the time scale of any real process. It is important to use the actual T_c when rescaling the time series and not T_{c+} because there can be as much as a 40% difference between the two values.

6. Concentration Fluctuation Measurements

The water channel facility in the Mechanical Engineering Department at the University of Alberta was used to collect concentration fluctuation data. The experimental time series used for verification and development of the stochastic model were measured by Wilson et al. (1991). The measurement equipment and technique is documented in the Ph.D. Thesis by Zelt (1992) and discussed in Yee et al. (1993b).

6.1. Water Channel and Conductivity Probes

The data were collected by Wilson et al. (1991) in the water channel shown schematically in Figure 6. Measurements were taken at three downstream positions x/h_s = 9.0, 19 and 29 and various positions y across the width of the plume at each

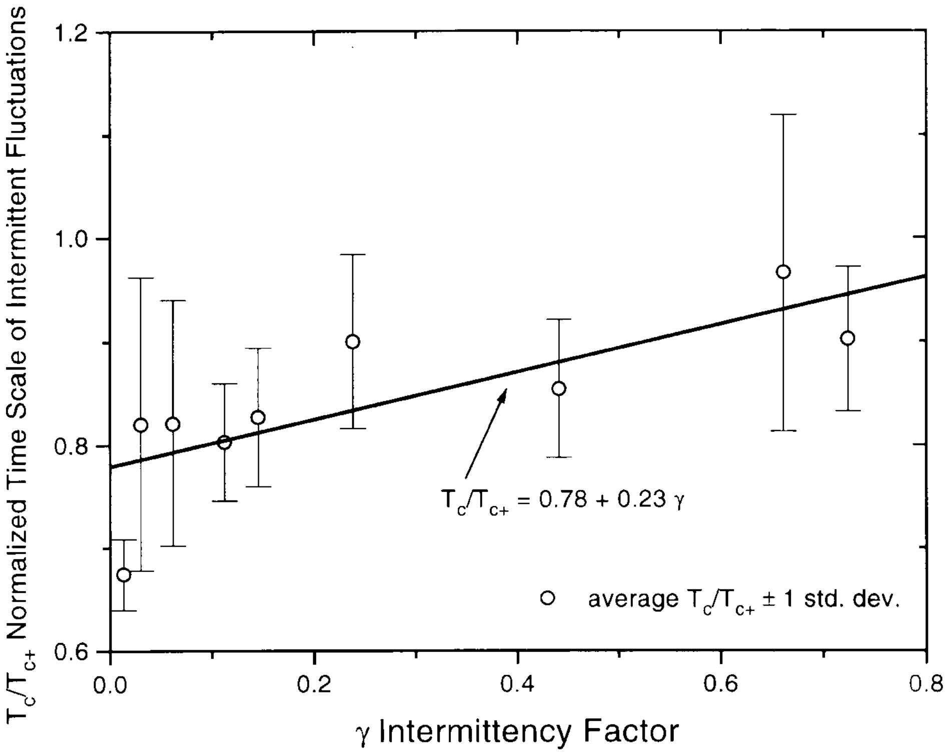

Figure 5. Time scale of intermittent concentration fluctuations T_c normalized by the time scale of the pseudo-concentration time series T_{c+} for a range of intermittency factors, γ, and conditional fluctuation intensities, i_p, that match the water channel experimental data sets.

of these downstream locations. In the present study, only data taken at the source height $z = h_s$ with intermittency factors $\gamma > 0.01$ were used. The restriction on the intermittency factor ensured that there were a sufficient number of non-zero data points to generate reasonable probability distribution histograms.

For short range dispersion ($\leq$ 1 km) in a neutrally stable atmosphere, the crosswind plume spread, σ_y, is well approximated by a linear function of downwind distance x. In the water channel the crosswind plume spread was also linear, so the scale factor can be chosen from a range of possibilities since a linear spread is the same in any scale. In the present study we chose a scale factor of 1000 : 1 based on the thickness of the surface log law velocity-profile layer. In the water channel this logarithmic layer is about 100 mm thick, while in the atmosphere it is about 100 m.

The mean velocity profile of the shear layer in the water channel followed the log law profile:

$$\frac{U}{u_*} = \frac{1}{\kappa} \ln \left(\frac{z - d_0}{z_0} \right). \tag{24}$$

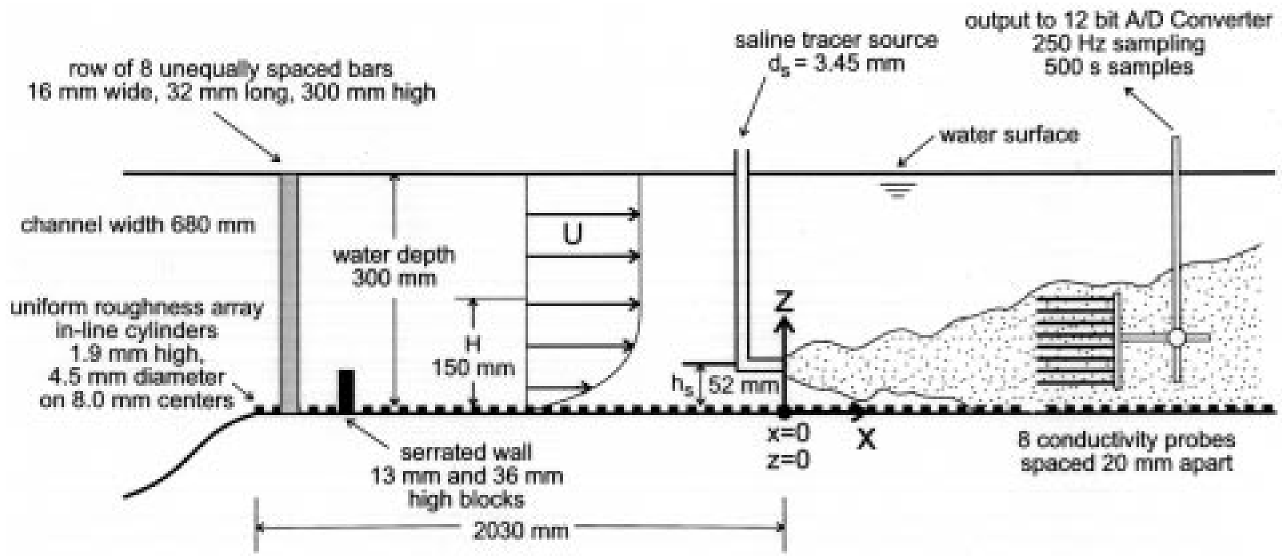

Figure 6. Schematic diagram of the water channel shear layer generator test section.

where U is the mean flow velocity, $u_* = 14.6$ mm s^{-1} is the friction velocity, $\kappa = 0.4$ is the von Karman constant, $d_0 = 1.5$ mm is the displacement height of the surface roughness, $z_0 = 0.15$ mm is the roughness length scale and z is the height above ground level.

A downstream facing isokinetic source from a 3.45 mm internal diameter tube at a position 2030 mm downstream from the beginning of the channel emitted saline tracer solution at a height $h_s = 52$ mm above the bottom of the channel into a mean velocity, U_h of 210 mm s^{-1} at the source height. The tracer was a neutrally buoyant mixture of water, ethanol and 50 g l^{-1} salt emitted with a flow rate of 2.0 ml s^{-1}.

The concentration fluctuation data were obtained with a rake of eight electrical conductivity probes mounted with the probe tips 20 mm apart. Each probe had a spatial resolution of about 1 mm. Throughout the experiments the probe response was corrected for background buildup and electronic drift in the output. Each fluctuation time series was sampled at 250 points per second. At 1000 : 1 scale this corresponded to samples spaced about 1 m apart in the wind direction. The total sample time for each experiment was 500 seconds (about 2000 Eulerian integral time scales) to give 125,000 data points per run. This long sample time was necessary to ensure repeatable fluctuation statistics.

After the data set was collected it was processed to correct for the probe response time constant that produced a −3 dB rolloff frequency of 35 Hz. Deconvolution of the digital signals from the probes with the inverse of a first-order impulse response function enhanced the effective frequency response to 105 Hz.

A zero concentration threshold was set at 8.0 times the background noise level to eliminate the effect of noise on the measured intermittency. This threshold corresponds to about $0.09C_p$ and was determined empirically by analyzing time series with no source emission and adjusting the threshold to produce the required $\gamma = 0$ intermittency.

6.2. Frequency Spectrum of Experimental Data Compared to the Simulation

As Wilson et al. (1991) note, for salt in water the mass diffusivity D is much smaller than the molecular viscosity of the fluid ν and the Schmidt number $\mathrm{Sc} = \nu/D \gg 1$. The effect of this large Schmidt number is that velocity-driven straining of the concentration field decays before molecular diffusion smears out the concentration fluctuations. This can be seen in the spectrum of concentration fluctuations shown in Figure 7 where water channel data follow the viscous-convective Batchelor spectrum with $F_c \propto f^{-1}$ at high frequency f. In contrast, the shape of the concentration spectrum of the stochastic simulation is determined by the assumption of a first-order Markov process that produces $F_c \propto f^{-2}$ at high frequency. The spectrum of concentration fluctuations in the atmosphere where $D \approx \nu$, hence $\mathrm{Sc} \approx 1$, has $F_c \propto f^{-5/3}$ at high frequencies, see Wilson (1995).

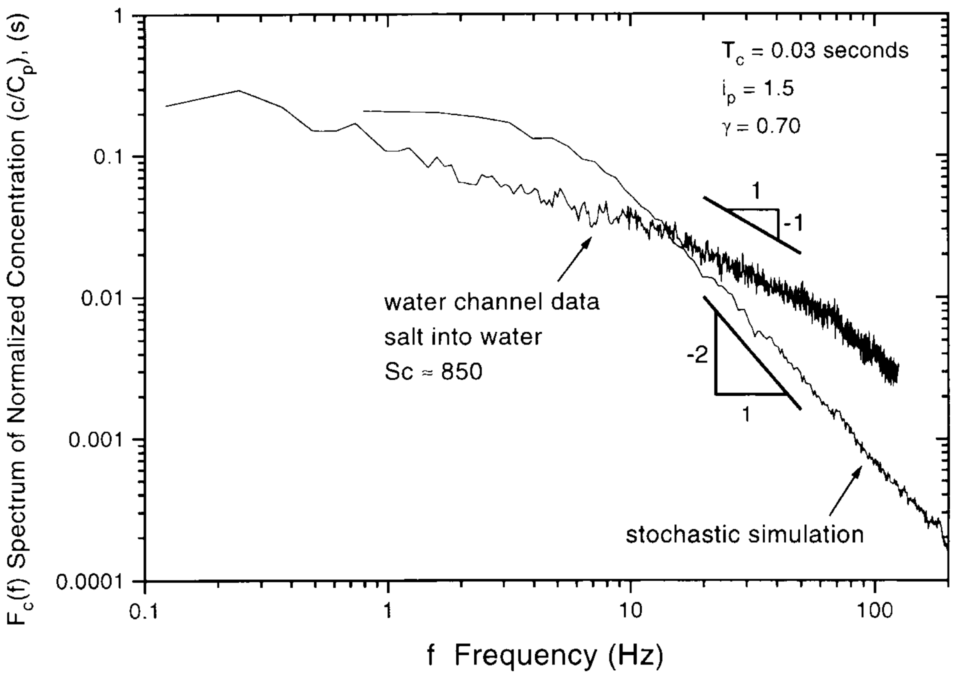

Figure 7. Typical spectra of normalized concentration fluctuations for the experimental data and for the stochastic simulation of that experiment.

The important implication of this spectral mismatch between the stochastic model and experimental data is that there should be more high frequencies in the water channel data than in the stochastic simulation, even if the concentration fluctuation intensities are the same. Higher peak concentrations might be expected in the water channel data. Comparison of the experimental data and the stochastic simulation indicated that the effect of this spectral mismatch is small. When the stochastic model is used for atmospheric concentration fluctuations the effect should be even smaller, since the first-order Markov spectrum $F_c \propto f^{-2}$ provides a much closer match to the $F_c \propto f^{-5/3}$ spectrum expected in the atmosphere.

7. Comparison of Experimental Data and Clipped Lognormal pdf

A key input to the stochastic model is the probability density function (pdf) of concentration. In the previous sections it was assumed that a clipped lognormal distribution could be used to describe intermittent concentration fluctuations. To demonstrate that this assumption is reasonable, the experimental water channel concentration probability distribution was compared to the clipped lognormal probability distribution. For this comparison only non-zero (in-plume) concentrations were considered, so all concentrations were normalized by the conditional mean concentration C_p. The theoretical clipped lognormal distributions were given the same intermittency factor γ and conditional fluctuation intensity i_p^2 as the experimental data sets.

In most hazardous releases high concentrations greater than the mean are the most important, so most relevant information is contained in exceedance probability plots rather than cumulative probability plots. Figures 8 and 9 show the normalized conditional exceedance probability $E_p(c/C_p)$ defined as the probability of finding a concentration greater than c/C_p

$$E_p\left(\frac{c}{C_p}\right) = \int_{\frac{c}{C_p}}^{\infty} p_p\left(\frac{c}{C_p}\right) d\left(\frac{c}{C_p}\right). \tag{25}$$

Agreement with the experimental data set is within a few percent up to $E_p = 0.1$, that is the 90th percentile concentration. At the 99th percentile concentration or $E_p = 0.01$, the error is within a factor of two, even when the intermittency factor γ is very small, with non-zero concentrations only 1% of the total time. All of the plots extend to $E_p = 0.00001$ at which point there are only 1 or 2 events in the entire experimental data set that exceeded that concentration. It is difficult to evaluate the fit at these high concentration, low probability values, because only 125,000 data points were recorded in each experimental time series.

8. Stochastic Model Performance

To determine if the stochastic model accurately reconstructs intermittent concentration fluctuation time series, the output of the stochastic simulation was compared to data from the water channel experiments. The correct distribution of concentration values is guaranteed by the input clipped lognormal pdf that matches the statistics of the data set, but the derivative of concentration and the upcrossing rate are recurrence statistics that will agree with the experimental data only if the reconstructed time series has the correct fluctuations.

8.1. Filtering the Stochastic Simulation

To compare time derivatives and upcrossing rates, the simulated time series and the experimental time series must have the same high frequency cutoff. Each reconstructed stochastic time series was generated with 100 time steps per time scale T_c. When this time step was adjusted to match the experimental data time scale of about $T_c = 0.027$ seconds, the effective sampling rate of the simulation was about 3700 Hz. Because the frequency response of the experimental probes was only 105 Hz, the stochastic simulation was filtered with a sixth-order Butterworth digital filter to limit the high frequency components in the simulation to 105 Hz.

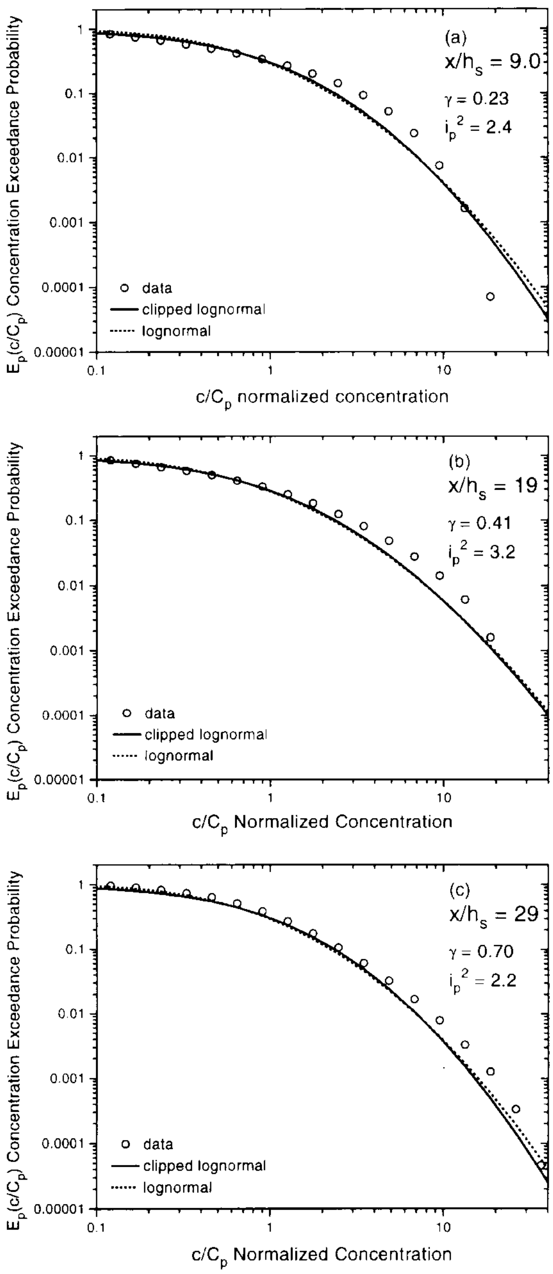

Figure 8. Conditional exceedance probability E_p for moderately intermittent fluctuations of the concentration c/C_p near the centreline of the plume at three downstream positions x/h_s. Cross stream distance from source (a) $y/\sigma_y = 0.3$, (b) $y/\sigma_y = 0.25$, (c) $y/\sigma_y = 0.18$.

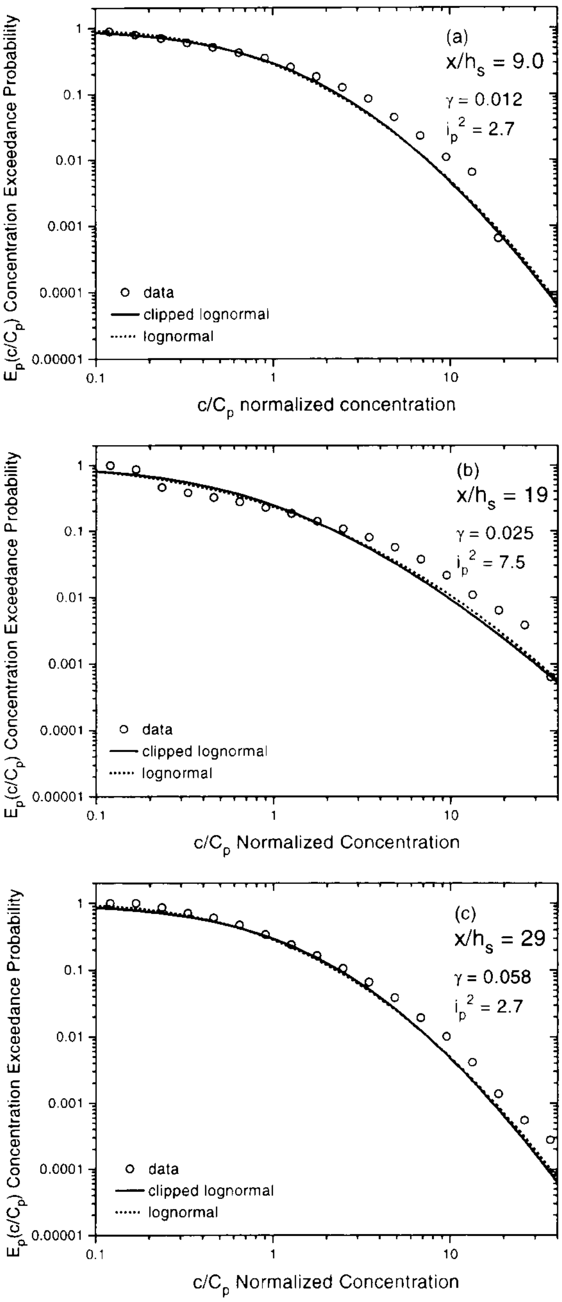

Figure 9. Conditional exceedance probability E_p for highly intermittent fluctuations of the concentration c/C_p on the outside edge of the plume at three downstream positions x/h_s. Cross stream distance from source (a) $y/\sigma_y = 4.1$, (b) $y/\sigma_y = 4.8$, (c) $y/\sigma_y = 2.8$.

The filter was applied to the simulated time series after it was clipped by setting all of the negative concentration values to zero. This approximates the way the experimental probes filter the concentration fluctuations in the water channel data.

Filtering has a strong effect on the length of the intermittent periods as it removes the short zero periods and causes the intermittency factor $\gamma \rightarrow 1.0$ as the cutoff frequency decreases. To recover the intermittent periods in the experimental data a 'zero' cutoff was defined by Wilson et al. (1991) at eight standard deviations above the background noise level. The experimental intermittency is an input to the stochastic model, so the agreement between the measured and experimental intermittency factors was maintained by applying an equivalent zero cutoff of $0.09C_p$ to the stochastic time series after filtering to match the experimental data frequency response. All comparisons between experimental data and the stochastic simulated time series are made after applying the low pass filter and the zero cutoff to the simulated time series.

8.2. Time derivative of concentration

One important assumption in the stochastic model is that the magnitude of the derivative of concentration is dependent on the concentration level. This level dependence in the deterministic term $a(c_+, t)$ in Equation (16) is imposed and as a consequence the b term is also level dependent to maintain a stationary pdf. Figures 10 and 11 show that the water channel measurements confirm the field observations of Yee et al. (1993a), with the rms concentration deviation $\dot{c}'$ exhibiting a strong dependence on concentration level c/C_p.

Figures 10 and 11 compare the reconstructed time series rms time derivative of concentration $\dot{c}'$ with the total rms time derivative of concentration for the water channel data. The time derivative of concentration was calculated as the change in concentration over one time step. The normalized rms time derivative of concentration $\dot{c}'T_c/C_p$ is plotted against the concentration level c/C_p for a range of downstream positions. The stochastic simulation line is the ensemble average of 10 independent time series realizations. Figure 10 shows points near the centreline of the plume and Figure 11 shows points near the outside edge of the plume.

There is evidence in the experimental data of a gradual evolution of the rms derivative, $\dot{c}'$, profiles with downstream distance that does not occur in the stochastic model. Close to the source at $x/h_s = 9.0$ the stochastic simulation overestimates the derivative of concentration. At a downstream distance of $x/h_s = 19$ the stochastic simulation derivative is close to the experimental data and at the far downstream location with $x/h_s = 29$ the derivative is underestimated.

Some of the difference between the experimental data and the stochastic model can be accounted for by the different frequency rolloff rates of the spectrum in the experiments versus the simulation, see Figure 7. The derivatives at high concentration levels $c/C_p > 10$ are underestimated by the stochastic model. This would be

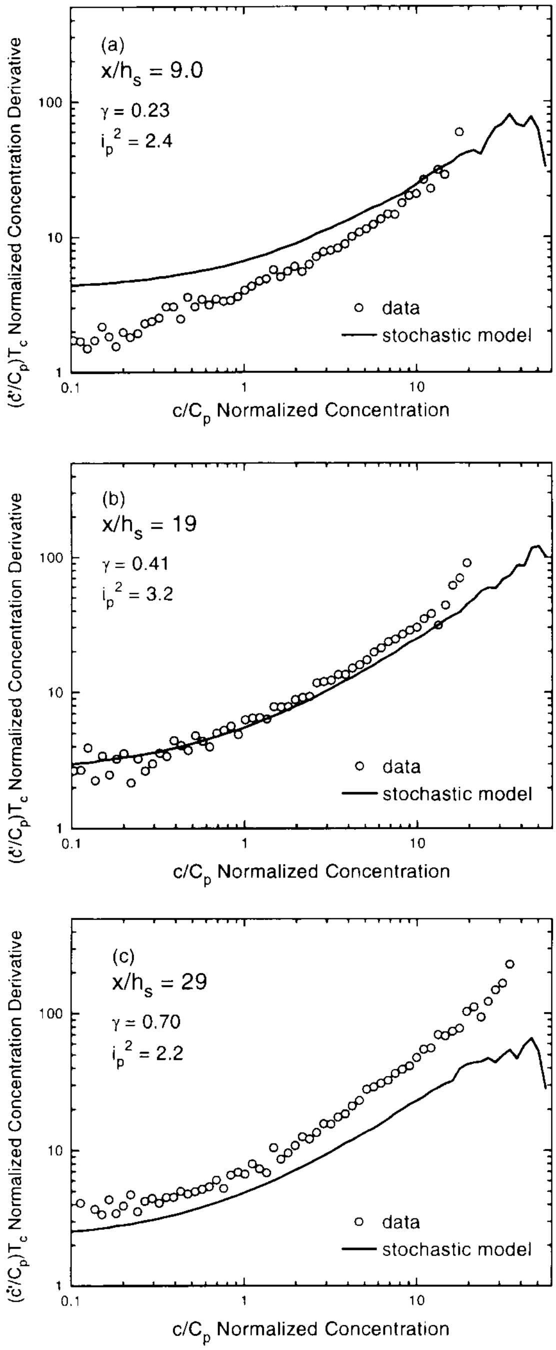

Figure 10. Normalized root-mean-square time derivative of concentration $(\dot{c}'/C_p)T_c$ at the concentration level c/C_p near the centreline of the plume at three downstream positions x/h_s. Cross stream distance from source (a) $y/\sigma_y = 0.30$, (b) $y/\sigma_y = 0.25$, (c) $y/\sigma_y = 0.18$.

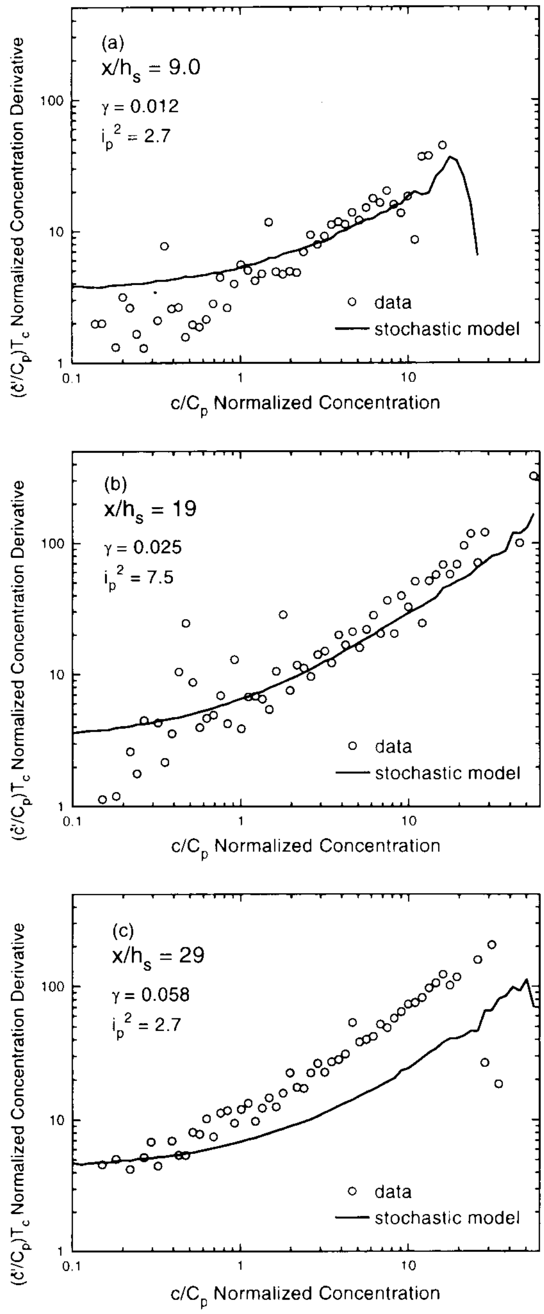

Figure 11. Normalized root-mean-square time derivative of concentration $(\dot{c}'/C_p)T_c$ at the concentration level c/C_p on the outside edge of the plume at three downstream positions x/h_s. Cross stream distance from source (a) $y/\sigma_y = 4.1$, (b) $y/\sigma_y = 4.8$, (c) $y/\sigma_y = 2.8$.

expected because the experimental data have more high frequency fluctuations and higher peak concentrations than the reconstructed time series.

8.3. Concentration level upcrossing rates

An upcrossing is counted each time the concentration in the time series crosses a threshold level while the derivative of concentration is positive (i.e., concentration is increasing). The upcrossing rate is a measure of the average frequency of fluctuations that exceeds a particular concentration level. The upcrossing rate n^+ is not an input to the model, so producing the correct upcrossing rate demonstrates that the simulation produces the correct frequencies of fluctuations for each concentration level. The Du et al. (1999) model estimated the upcrossing rates for conditional time series of concentration fluctuations, ignoring intermittent periods. In the present study, the total upcrossing rate was simulated, including the intermittent periods of zero concentration.

In Figures 12 and 13 the results from a single experimental water channel run are shown. The stochastic simulation line is the mean of 10 realizations. Since this is a stochastic model each realization is different and the dotted lines on the plot indicate a two standard deviation multiplicative range of run-to-run variation calculated as follows:

$$\begin{aligned} \text{upper bound} &= \mu\left(1+2\frac{s}{\mu}\right) \\ \text{lower bound} &= \frac{\mu}{\left(1+2\frac{s}{\mu}\right)}, \end{aligned} \tag{26}$$

where μ is the mean of the upcrossing rate n^+T_c and s is the standard deviation of the 10 simulated time series. The upcrossing rates match quite well for the wide range in intermittencies and fluctuation intensities.

As in the case of the time derivatives of concentration, there is some evidence of the spectral mismatch in the rolloff rate between the data and the stochastic simulation and there is also evidence of evolution of the experimental plume with downstream distance. As expected, there are more high frequency fluctuations in the experimental data and the upcrossing rates at high concentrations $c/C_p > 10$ are underestimated by the simulation. At the far downstream location $x/h_s =$ 29 the stochastic simulation consistently underestimates the upcrossing rate. At the locations closer to the source for x/h_s = 9.0 and 19, the upcrossing rate is overestimated at low concentrations and underestimated at high concentrations.

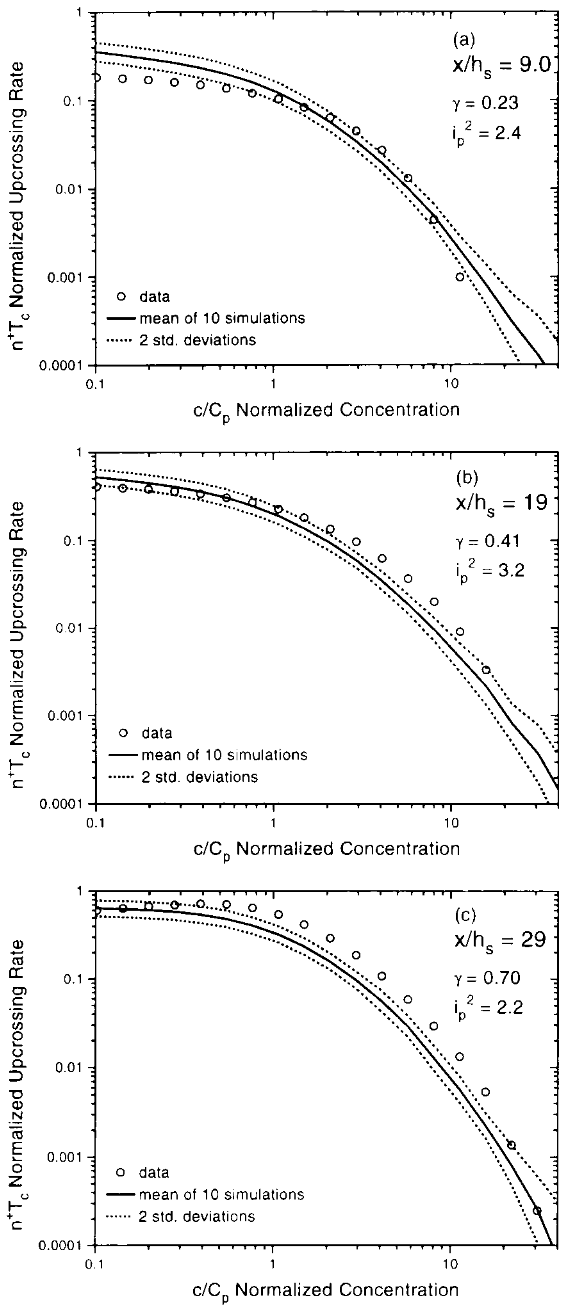

Figure 12. Normalized upcrossing rate n^+T_c for moderately intermittent time series versus the normalized instantaneous concentration c/C_p near the centreline of the plume at three downstream positions x/h_s. Cross stream distance from source (a) $y/\sigma_y = 0.30$, (b) $y/\sigma_y = 0.25$, (c) $y/\sigma_y = 0.18$.

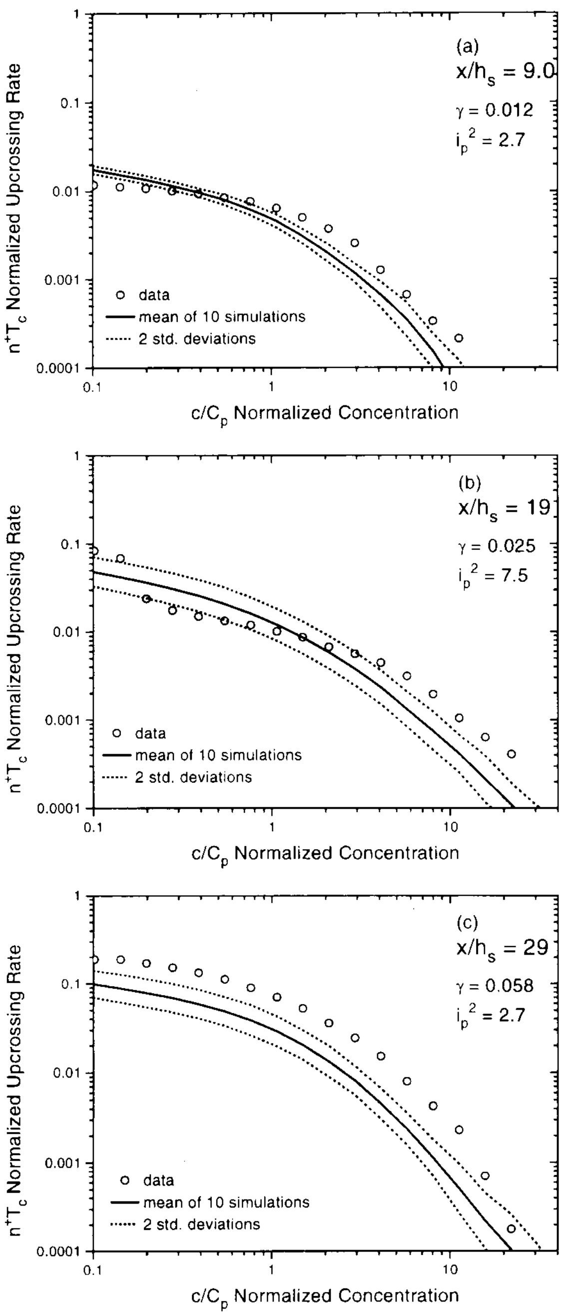

Figure 13. Normalized upcrossing rate n^+T_c for highly intermittent time series versus the normalized instantaneous concentration c/C_p near the outside edge of the plume at three downstream positions x/h_s. Cross stream distance from source (a) $y/\sigma_y = 4.1$, (b) $y/\sigma_y = 4.8$, (c) $y/\sigma_y = 2.8$.

9. Summary and Conclusions

In this study we have demonstrated that intermittent concentration fluctuation time series at a fixed receptor in a dispersing point source plume can be simulated as a first-order memoryless Markov stochastic process. The input parameters required to reconstruct the intermittent time series are the intermittency factor γ, the conditional fluctuation intensity i_p^2, and the probability distribution $p(c)$.

The stochastic simulation cannot generate intermittent time series directly, so some manipulation and interpretation of the reconstructed time series is required to produce the necessary intermittency. The key assumption is that intermittent periods of zero concentration are part of the same physical mixing process as the periods of non-zero concentration. This assumption allows the simulated non-intermittent time series to be shifted by a concentration c_{base} to give positive and negative concentrations $\tilde{c}$. The positive concentrations are interpreted as actual fluctuations while the negative concentrations are interpreted as periods of zero concentration where the magnitude of the negative concentration represents the likelihood of obtaining a non-zero concentration in the next time step.

The clipped lognormal was tested with experimental data from the Wilson et al. (1991) water-channel experiments and found to provide a good fit to these data. There was some evidence of evolution of the experimental plume with downstream distance that does not occur in the stochastic model, but the effect was small. Intermittency, fluctuation intensity, and cross-stream distance did not have an effect on the shape of the pdf.

There was remarkably good agreement between the experimental data and the stochastic model, considering that the spectral rolloff of the experimental data was proportional to frequency f^{-1}, while the simulation had a f^{-2} rolloff. The agreement was demonstrated by comparing the first time-derivative of concentration and the upcrossing rate statistics with experimental data over a range of intermittency factors γ = 0.7 to 0.01 and fluctuation intensities i_p^2 = 2.2 to 7.5. The stochastic model produces the correct rms derivative of concentration, $\dot{c}'$, and the mean upcrossing rate, n^+, within about a factor of two. This confirms that the linear assumption for the a term in the stochastic differential equation, in conjunction with the assumption of a clipped lognormal, is a reasonable model for the time derivatives of concentration. The agreement also suggests that low frequency fluctuations are the dominant feature determining the upcrossing rate and rms derivative. These simulated low frequency fluctuations are forced to agree with the experiment by imposing the measured integral time scale T_c on the simulation.

The advantage of the stochastic model is that it can be used to reconstruct time series with any realistic combination of intermittency and fluctuation intensity to generate large ensembles of random time series with identical means, variances, and intermittencies. Each time series represents an individual realization of the event and complex hazard models can be time stepped through simulated releases to observe the effects of realization-to-realization variability as well as large ensemble averages.

In its present form, the stochastic model is useful for generating time series to evaluate hazardous effects. However, additional investigation is required to determine if the stochastic model, with its single time scale, is adequate for release and receptor positions that are at different heights in a shear flow.

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