

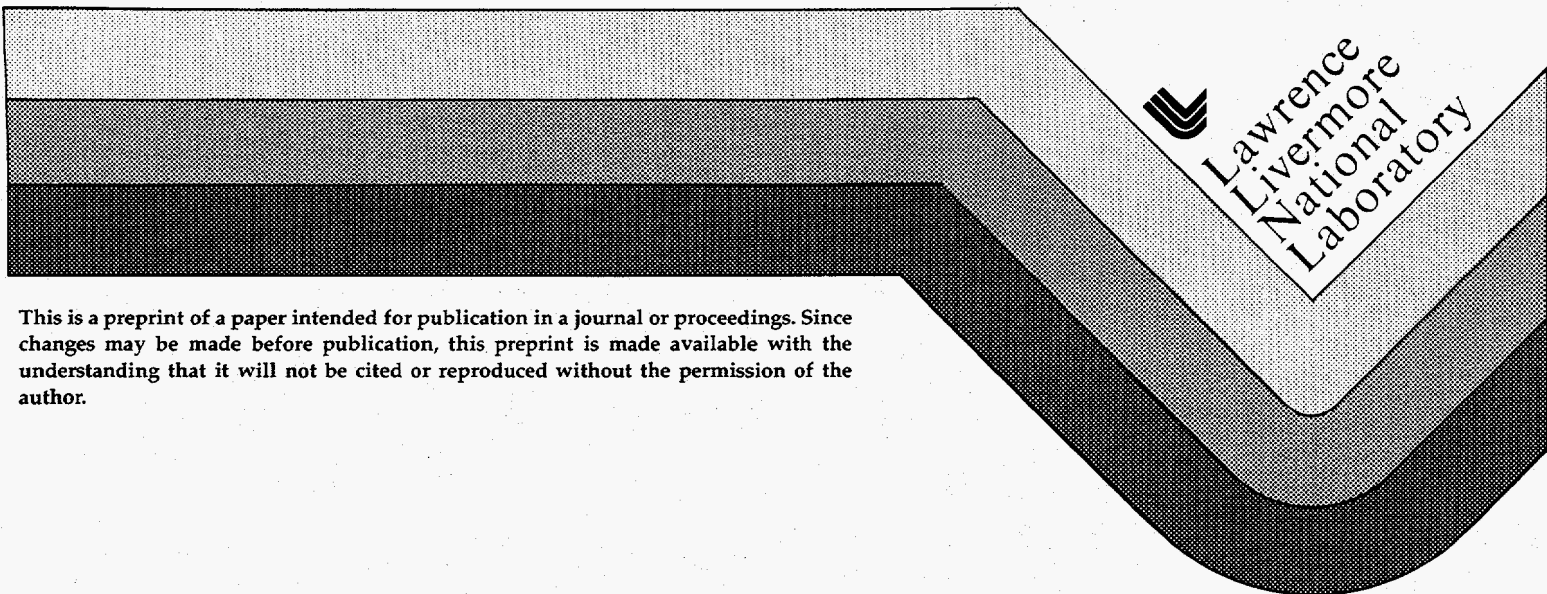
Numerical Simulation of the Langevin Equation for Skewed Turbulence

D.L. Ermak
Lawrence Livermore National Laboratory
Livermore, California 94551
and

J.S. Nasstrom*
EG&G Energy Measurements Inc.
Pleasanton, California 94588

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Donald L. Ermak*

Lawrence Livermore National Laboratory
Livermore, California

John S. Nasstrom

EG&G, Energy Measurements, Inc.
Pleasanton, California

1. INTRODUCTION

Skewness of the vertical velocity probability distribution has been shown to be one of the key factors in the complex process of vertical diffusion of a tracer or pollutant emitted into an unstable, convective boundary layer or CBL (Lamb, 1982). In the CBL, the probability distribution of fluid vertical velocity, $P(w_f)$, is positively skewed and, therefore, non-Gaussian. A typical value of skewness, $S \equiv \overline{w_f^3} / (\overline{w_f^2})^{3/2}$, in the CBL is 0.6 (Wyngaard, 1988). Positive skewness in the inversion-capped CBL is due to solar heating of the surface which generates strong updrafts or thermals over approximately 40% of the horizontal area with compensating weaker downdrafts over about 60% of the area. Compared to unskewed turbulence, dramatic differences in vertical diffusion result. For example, pollutant emitted from an elevated, continuous, non-buoyant source has a higher probability of meeting a downdraft in skewed turbulence, so the locus of maximum time-average concentration decreases in height with downwind distance.

A powerful approach to modeling complex turbulent diffusion processes, such as those in the CBL, is the Lagrangian stochastic simulation technique based on the Langevin equation for the motion of a tracer particle within the surrounding fluid (see reviews by Sawford, 1985 & 1993). The Langevin equation equates the net force on a fluid element associated with the tracer particle to the sum of a deterministic force (such as a damping force) and a random force. Integration of the Langevin equation over time provides a means of calculating successive Lagrangian velocity and displacement increments of the fluid element or particle. From many independent particle trajectories, ensemble-average and (in stationary turbulence) time-average concentrations can be calculated. Thomson (1987) showed that the

most rigorous criterion to date for a Langevin equation model is the "well-mixed condition". That is, a tracer initially well-mixed in a fluid must remain so, and, therefore, the joint probability distribution of position and velocity of a tracer will remain the same as that of the fluid.

Standard Langevin equations use Gaussian random forcing terms, but can be generalized to include non-Gaussian or skewed forcing (van Kampen, 1992). Both types of Langevin equations have been used for skewed turbulence. An approach introduced by Thomson (1987) for treating skewed turbulence in a Langevin model with *Gaussian* random forcing has been the most successful. Using this approach CBL vertical diffusion models have been developed (e.g., Luhar & Britter, 1989 and Weil, 1990). These models meet the well-mixed condition and have achieved good results. Even so, there are some undesirable features of these models. The resulting deterministic forces in these models are complex and difficult to interpret physically. In addition, some models of this type lack generality, in that they don't result in Gaussian random forcing when skewness is zero. Development of Langevin equation models with *skewed* random forcing (see Thomson, 1984; van Dop et al., 1985; de Baas et al., 1986; Sawford, 1986; Sawford and Guest, 1987) have been less successful in meeting the well-mixed condition in skewed, inhomogeneous turbulence. The reasons for these failures appear to be multiple, including difficulties in treating inhomogeneous turbulence and the interaction with boundaries (Sawford and Guest, 1987).

In this paper we present a numerical method for solving the generalized Langevin equation of motion with skewed random forcing for the case of *homogeneous*, skewed turbulence. We begin by showing how the analytic solution to the Langevin equation for this case can be used to determine the relationship between the particle velocity moments and the properties of the skewed random force. We then present a numerical method that uses simple probability distribution functions to simulate the effect of the

* Corresponding author address: Donald L. Ermak, L-262, LLNL, P.O. Box 808, Livermore, CA 94550, USA

random force. The numerical solution is shown to be exact in the limit of infinitesimal time steps, and to be within acceptable error limits when practical time steps are used.

2. LANGEVIN EQUATION MODEL

The Langevin equation generalized to include skewed random forcing can be written as follows (van Kampen, 1992):

$$\frac{dw}{dt} = -w/\tau + \Lambda(t), \quad (1)$$

where w is the particle velocity, t is time, τ is the Lagrangian correlation time scale, and $\Lambda(t)$ is the skewed random force (per unit mass). The deterministic damping force is $-w/\tau$. The random force, $\Lambda(t)$, is not a function of w and has statistical properties defined by the cumulants

$$\langle\langle \Lambda(t_1)\Lambda(t_2)\cdots\Lambda(t_n) \rangle\rangle = \Gamma_n \delta(t_1 - t_2)\delta(t_1 - t_3)\cdots\delta(t_1 - t_n), \quad (2)$$

where $\langle\langle x \rangle\rangle$ denotes the cumulant of a quantity x , $\{\Gamma_n\}$ are coefficients to be determined, and $n=1, 2, \dots$. A cumulant of order n is a function of the moments of order n and lower (Gardiner, 1985). For a Gaussian distribution, $\Gamma_n = 0$ for $n > 2$, so the higher order cumulants ($n > 2$) are a measure of the departure from a Gaussian distribution. The random force, $\Lambda(t)$, is seen to be delta-function correlated in time, which means it is uncorrelated over any time period of interest. We will assume that the random force has zero mean, $\overline{\Lambda(t)} = \Gamma_1 = 0$ (this implies that the steady-state mean fluid velocity is zero).

For homogeneous turbulence, the time scale τ and the set of coefficients $\{\Gamma_n\}$ are constant. Consequently, the Langevin equation (1) can be formally integrated to yield the velocity equation

$$w(t) = w(0)e^{-t/\tau} + \underbrace{\int_0^t e^{(s-t)/\tau} \Lambda(s) ds}_{\equiv R(t)}. \quad (3)$$

This equation is stochastic in nature and yields the velocity of a particle at time t given the initial velocity at time zero. The new velocity is composed of two terms: a deterministic term that is proportional to the initial velocity, $w(0)$, and a random term that is a function of the random force, $\Lambda(t)$. The statistical properties of the random velocity increment, $R(t)$, in terms of its cumulants can be readily calculated using the definition of $R(t)$ presented in (3), the cumulants of $\Lambda(t)$ given in (2), and by carrying out the necessary time integration. The result is

$$\langle\langle R^n(t) \rangle\rangle = \frac{1}{n} \Gamma_n \tau (1 - e^{-n t/\tau}). \quad (4)$$

The moments of $R(t)$, $\overline{R^n(t)}$, can be calculated directly from (4) using the definition of the cumulant. Similarly, the time-dependent particle velocity moments, $\overline{w^n(t)}$, for $n=1, 2, \dots$, can be determined using (3) and the moments of $R(t)$. Note that both $R(t)$ and $w(t)$ are described by the same probability distribution function since they are linearly related.

Now, in the limit of $t \rightarrow \infty$, we see from (3) that the particle velocity is equal to the velocity increment $R(t)$. Consequently, all the respective limiting moments (and cumulants) of $w(t)$ and $R(t)$ are equal, i.e.,

$$\overline{R^n(t)}_{t \rightarrow \infty} = \overline{w^n(t)}_{t \rightarrow \infty} \equiv \overline{w_\infty^n}. \quad (5)$$

For stationary turbulence, the steady-state particle velocity distribution must approach the fluid velocity distribution, so

$$\overline{w_\infty^n} = \overline{w_f^n}, \quad (6)$$

where w_f is the fluid velocity. We can use (4), (5) and (6) to obtain the result, using cumulant notation, that

$$\Gamma_n = n \langle\langle w_f^n \rangle\rangle / \tau. \quad (7)$$

This equation completes the definition of the Langevin equation model system by uniquely relating the random force cumulant coefficients $\{\Gamma_n\}$ to the fluid velocity cumulants.

3. MODEL IMPLEMENTATION

In principle, we have a well-defined model system. Although we have not directly determined the tracer particle velocity probability distribution, $P(w; w_0)$, we have determined all the moments of $w(t)$ in terms of the initial velocity, w_0 , the Lagrangian correlation time scale, τ , and the statistical properties of the fluid expressed by the velocity cumulants $\{\langle\langle w_f^n \rangle\rangle, n=1, 2, \dots\}$. Consequently, we have demonstrated that a unique $P(w; w_0)$ exists, and have derived its statistical properties in terms of the velocity moments. However, before this model can be applied in a numerical simulation two practical difficulties must be addressed.

The first difficulty is related to our ability to accurately define the statistical properties of the stationary fluid or, more specifically, the CBL. In general, the velocity moments of most fluids, are not known with sufficient accuracy to determine the velocity cumulants beyond the first few. This is because the higher moments are highly dependent on the tails of the velocity probability distribution where experimental statistics are poorest. On the other hand, it is only the first few velocity moments that have been shown to have a dominant effect on tracer diffusion in the CBL.

The second obstacle to direct implementation of these equations in a numerical simulation is mathematical in nature. Assuming all of the moments of $R(t)$ are known, it is mathematically difficult (if not impossible) to obtain $P(R)$ from an arbitrary and infinite number of moments. Thus, it does not seem possible to obtain an analytic form for $P(R)$ from which individual values of $R(t)$ can be selected for a numerical simulation.

To address these practical difficulties, we take the following approach to defining the system. We begin by defining the random force coefficients in terms of the known first three* moments of the fluid velocity using (7). Assuming the first three velocity moments are known, and specified to be $\overline{w_f} = 0$, $\overline{w_f^2} = \sigma^2$, and $\overline{w_f^3} = \zeta^3$, then the first three random force coefficients are

$$\Gamma_1 = 0, \quad (8a)$$

$$\Gamma_2 = 2\sigma^2/\tau, \text{ and} \quad (8b)$$

$$\Gamma_3 = 3\zeta^3/\tau. \quad (8c)$$

To specify the remaining random force coefficients, we assume a functional form for $P(R)$ in the limit of $t \rightarrow 0$. As a matter of convenience, we select a known and simple function that can be defined by the first three moments of $R(t)$, which we specify using (8a-c). In this work, we use the "double-block" function described in the Appendix (obviously, this distribution is not unique and we are exploring others). We then use (4) in the $t \rightarrow 0$ limit to define the remaining values of $\{\Gamma_n\}$ where the higher moments of $R(t)$ are calculated from the selected $P(R)$. For the double-block function we have selected, the higher random forcing coefficients are found to be

$$\Gamma_n = \frac{4(3)^{n-2}(\zeta^3)^{n-2}}{(n+1)\tau(\sigma^2)^{n-3}}, \quad n > 3. \quad (8d)$$

Using (7), we can now specify the remaining velocity cumulants (and moments) of the stationary-fluid velocity. These cumulants are

$$\langle\langle w_f^n \rangle\rangle = \frac{4(3)^{n-2}(\zeta^3)^{n-2}}{n(n+1)(\sigma^2)^{n-3}}, \quad n > 3. \quad (9)$$

Again we have a well-defined model where all of the random force coefficients $\{\Gamma_n\}$ are uniquely related to the fluid velocity cumulants. However, rather than use the fluid velocity cumulants to define all of the

* This could probably be extended to four or five. However, only marginal improvement has been reported by Du *et al.* (1994) by including information on the fourth moment in a Langevin equation model.

random force coefficients, we have done so only for the first three coefficients where the fluid velocity moments are accurately known. The remaining random force coefficients have been defined by assuming a functional form for $P(R)$ as $t \rightarrow 0$. As a result, all of the random force coefficients $\{\Gamma_n\}$ are expressed in terms of the first three moments of the fluid velocity: $\overline{w_f} = 0$, σ^2 and ζ^3 .

4. NUMERICAL SIMULATION METHOD

The equation we propose for numerically simulating the Langevin equation model defined by (1), (2) and (8a-d) is

$$w(\Delta t) = w(0)e^{-\Delta t/\tau} + R_a(\Delta t), \quad (10)$$

where $w(\Delta t)$ is the numerically-calculated velocity of a particle after time step Δt given velocity $w(0)$ at the beginning of the time step. $R_a(\Delta t)$ is a random velocity increment with (double-block) probability distribution $P_a(R)$, which has the same functional form as $P(R)$ in the limit of $\Delta t \rightarrow 0$, and the first three moments

$$\overline{R_a(\Delta t)} = 0,$$

$$\overline{R_a^2(\Delta t)} = \sigma^2(1 - e^{-2\Delta t/\tau}) \equiv \sigma_b^2, \text{ and} \quad (11)$$

$$\overline{R_a^3(\Delta t)} = \zeta^3(1 - e^{-3\Delta t/\tau}) \equiv \zeta_b^3.$$

Eqs. (11) completely defines $P_a(R)$ since it has a functional form defined by only the first three moments (see Appendix).

In general, $P_a(R)$ is only an approximation to $P(R)$, although they have the exact same first three moments. The approximate nature of $P_a(R)$ is due to the fact that the functional form of $P_a(R)$ remains constant; namely, the double-block form described in the Appendix. On the other hand, the functional form of $P(R)$ continually changes with time step. As a result, there is numerical error in the higher ($n > 3$) moments of $R_a(\Delta t)$ and these are then passed on to the statistical properties of the velocity $w(t)$.

One can show analytically that in the steady state the first three moments of the velocity ($\overline{w_\infty}$, $\overline{w_\infty^2}$, and $\overline{w_\infty^3}$) using the numerical simulation method of (10-11) are exact. Just as is the case with the random velocity increment $R_a(\Delta t)$, the approximate nature of $P_a(R)$ manifests itself in the higher moments of the steady state velocity (i.e., $\overline{w_\infty^n}$, $n > 3$).

This approach to defining and simulating the Langevin equation model system has several positive aspects. First, we have a well-defined system with specified values for the random force coefficients $\{\Gamma_n\}$ that are in agreement with the known turbulence properties of the actual fluid, namely, the first few

velocity moments. Second, in the limit of very small time steps, the numerical simulation results will approach the exact solution since the functional form of $P_a(R)$ is equal to $P(R)$ in the limit of $t \rightarrow 0$. Finally, the exact values of the velocity moments $\{\overline{w^n(t)}\}$ can be used to quantitatively determine the accuracy of the numerical method for any desired time step.

5. EXAMPLE SIMULATIONS

To demonstrate the accuracy of the numerical method, we performed several computer simulations using (10-11) for several values of fluid velocity skewness, $S \equiv \overline{w_f^3} / (\overline{w_f^2})^{3/2}$, and for several time step values. All the simulations used input values of zero mean fluid velocity, $\overline{w_f} = 0$; unit fluid velocity second moment, $\overline{w_f^2} = 1$; unit time scale, $\tau = 1$; and a sample of 10^6 particles.

5.1 Effect of time step size

To demonstrate the increased accuracy of the method with decreasing time step, simulations were performed with three time step values, $\Delta t/\tau = 0.2, 0.05$ and 0.01 . Using $S = 0.5$, initial particle velocities were generated with the steady-state, fluid velocity distribution. A simulation was then performed for a duration of 4τ . To generate initial velocities with the steady-state distribution, particle velocities were assigned a first-guess value and then simulated using the numerical simulation method (10-11) for a period from $t = -4\tau$ to 0 .

Table 1 compares the first six steady-state particle velocity moments averaged over the time period from $t = 0$ to 4τ with the exact values for the three simulations. (The first three exact fluid moments were input to the simulation and the higher exact moments are given by (9).) These values show that the numerical error in the particle velocity moments does become very

Table 1. Numerically-calculated steady-state particle velocity moments and exact fluid velocity moments and in three simulations using $S = 0.5$ and different time step values.

	Average numerical value $\Delta t/\tau = 0.2$	numerical value 0.05	value 0.01	Exact value
$\overline{w_\infty}$	0.00	0.00	0.00	0.
$\overline{w_\infty^2}$	1.00	1.00	1.00	1.
$\overline{w_\infty^3}$	0.50	0.50	0.50	0.5
$\overline{w_\infty^4}$	3.30	3.41	3.44	3.45
$\overline{w_\infty^5}$	4.91	5.32	5.44	5.45
$\overline{w_\infty^6}$	20.99	23.74	24.60	24.73

small with decreasing time step. In these calculations, the statistical error was kept smaller than the numerical error by using a large sample size (10^6 particles). Consequently, the error in the first three moments is essentially zero.

5.2 Well-mixed condition

To test the numerical simulation method versus the well-mixed condition, initial particle velocities were generated which had the steady-state velocity distribution and which had particle positions, z , uniformly distributed in a bounded region between $z = 0$ and $z = 1$. This was done in the simulation with $\Delta t/\tau = 0.01$ described in section 5.1. Particle positions were calculated using the displacement equation: $z(\Delta t) = z(0) + \frac{1}{2}[w(0) + w(\Delta t)]\Delta t$. A periodic boundary condition was imposed at the two boundaries (e.g., particles leaving the region at $z = 1$ were placed at $z = 0$ with their velocity unchanged). Figure 1 shows the velocity and position distributions at the beginning ($t = 0$) and end of the simulation ($t = 4\tau$) are very close to each other. This indicates that the well-mixed condition is met. Small differences in the tails of the velocity distribution can lead to significant differences in the higher velocity moments, but may not be

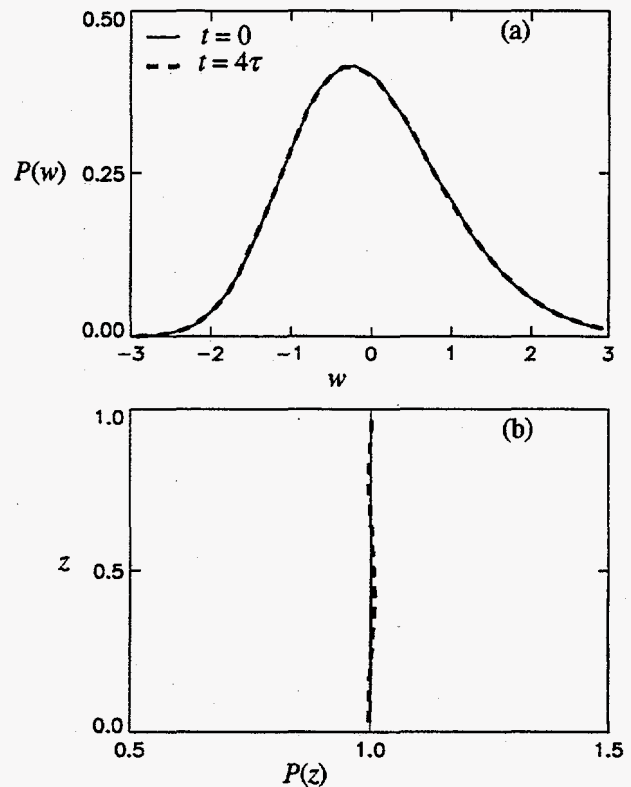


Fig. 1. Well-mixed velocity (a) and position (b) probability density functions at $t = 0$ (solid line) and $t = 4\tau$ (dashed line) from simulation using $S = 0.5$ and $\Delta t/\tau = 0.01$.

distinguishable in Fig. 1. However, the first six velocity moments at the beginning and end of the simulation were the same within the uncertainty associated with the particle number ($\pm 2 \times$ standard error).

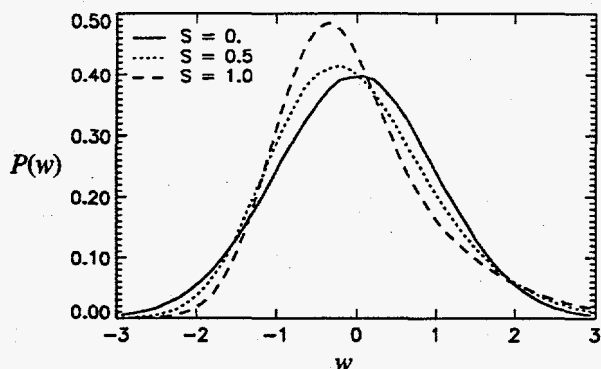


Fig. 2. Steady-state particle velocity probability density functions, $P(w)$, from three numerical simulations with $S = 0.0, 0.5,$ and $1.0,$ respectively, and $\Delta t/\tau = 0.01.$

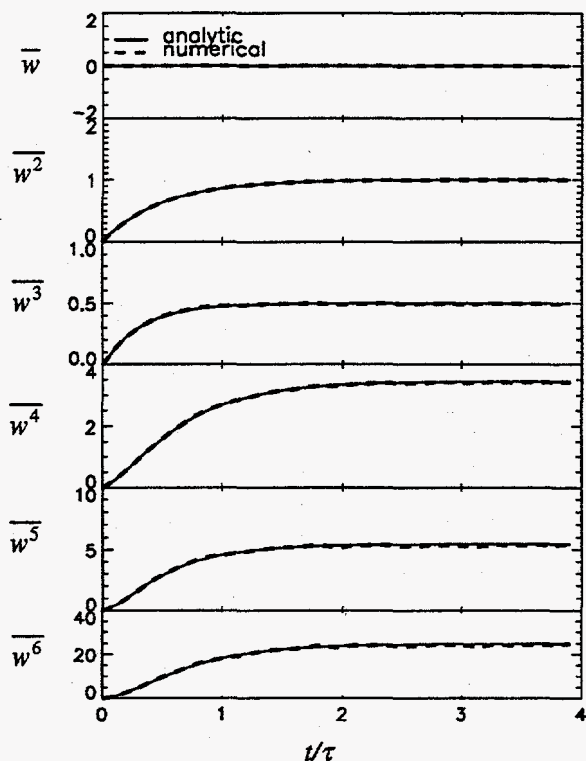


Fig. 3. Exact analytic (solid line) and numerically-calculated (dashed line) first six velocity moments versus time for simulation using $S = 0.5,$ $\Delta t/\tau = 0.01,$ and particles initialized with zero velocity.

5.3 Approach to steady-state for particles with zero initial velocity

To demonstrate the accuracy of the numerical method under non-steady-state conditions and over a range of skewness, three simulations were performed in which all the particles were initialized with zero velocity. These simulations used $S = 0.0, 0.5,$ and $1.0,$ and were all done with $\Delta t/\tau = 0.01$ from $t = 0$ to $10\tau.$ Fig. 2 shows the steady-state numerically-calculated probability density function of particle velocity, $P(w),$ at the end of each simulation. For the $S = 0.5$ case, Fig. 3 shows the time-dependent values of the numerically-calculated first six velocity moments and the corresponding exact analytic expressions derived from (3-4). There is excellent agreement between the numerically-calculated and analytic time-dependent moments.

For the three values of $S,$ Table 2 shows the first six numerical steady-state moment values averaged from $t = 6$ to $10\tau,$ are in excellent agreement with the exact, fluid moment values. When $S = 0,$ Table 2 and Fig. 1 show that the simulated velocity distribution is Gaussian to a very good approximation.

6. SUMMARY

We have presented a numerical method for simulating the Langevin equation with skewed random forcing which is exact in the limit of infinitesimal time step. Simulations using this method (a) yield acceptably small errors for velocity moments when the time step is within practical limits and (b) maintain initially well-mixed velocity and position distributions.

In future work, we hope to explore alternate simple probability distributions for generating random velocity increments, implement physically-realistic boundary conditions, and test the method against CBL diffusion experiments.

7. ACKNOWLEDGMENTS

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8. APPENDIX

The "double-block" probability density function, $P(b),$ for a random variable, $b,$ is defined as a sum of two overlapping uniform probability distributions,

$$P(b) = P_1(b) + P_2(b),$$

where

$$P_1(b) = \begin{cases} P_1, & \text{if } (m_1 - \Delta_1) \leq b \leq (m_1 + \Delta_1) \\ 0, & \text{elsewhere} \end{cases}$$

and

Table 2. Numerical steady-state particle velocity moments and exact fluid velocity moments for three simulations with $S = 0.0, 0.5,$ and $1.0,$ respectively, using $\Delta t/\tau = 0.01$.

	$S = 0.0$		$S = 0.5$		$S = 1.0$	
	Numerical	Exact	Numerical	Exact	Numerical	Exact
$\overline{w_\infty}$	0.00	0.	0.00	0.	0.00	0.
$\overline{w_\infty^2}$	1.00	1.	1.00	1.	1.00	1.
$\overline{w_\infty^3}$	0.00	0.	0.50	0.5	1.00	1.
$\overline{w_\infty^4}$	2.99	3.	3.45	3.45	4.78	4.8
$\overline{w_\infty^5}$	-0.02	0.	5.46	5.45	13.5	13.6
$\overline{w_\infty^6}$	14.9	15.	24.68	24.73	59.39	59.71

$$P_2(b) = \begin{cases} p_2, & \text{if } (m_2 - \Delta_2) \leq b \leq (m_2 + \Delta_2) \\ 0, & \text{elsewhere} \end{cases}$$

where the six parameters which define this distribution are the means, m_1 and m_2 ; half-widths, Δ_1 and Δ_2 ; and probability densities, p_1 and p_2 , of the two uniform distributions, respectively. These six unknown parameters are reduced to four using the following two equations:

$$\Delta_1^2 = m_1^2 + \sigma_b^2 \text{ and}$$

$$\Delta_2^2 = m_2^2 + \sigma_b^2.$$

The remaining four parameters are then determined from the first three moments of b , $\bar{b}=0$, σ_b^2 , and ζ_b^3 , along with the equation for the "zeroth" moment (total probability equal to one). The result is

$$m_{1,2} = \frac{1}{\sigma_b^2} \left[\frac{1}{2} \zeta_b^3 \mp \left(\frac{1}{4} \zeta_b^6 + \frac{1}{2} \sigma_b^6 \right)^{1/2} \right],$$

$$p_1 = \frac{m_2}{2\Delta_1(m_2 - m_1)}, \text{ and}$$

$$p_2 = \frac{-m_1}{2\Delta_2(m_2 - m_1)}.$$

9. REFERENCES

- de Baas, van Dop and Nieuwstadt, 1986: An application of the Langevin equation for inhomogeneous conditions to dispersion in a convective boundary layer, *Quart. J. R. Met. Soc.*, **112**, 165-180.
- Du, S., J.D. Wilson and E. Yee, 1994: Probability density functions for velocity in the convective boundary layer, and implied trajectory models, *Atmospheric Environment*, **28**, 1211-1217.
- Gardiner, C.W., 1985: *Handbook of stochastic methods for physics, chemistry and the natural sciences*, Second edition, Springer-Verlag, N.Y., 442 pp.
- Lamb, R.G. 1982: Diffusion in the convective boundary layer. In *Atmospheric Turbulence and Air Pollution Modelling* (F.T.M. Nieuwstadt and H. van Dop, eds.), pp. 159-229. D. Reidel, Boston.
- Luhar, A.K. and R.E. Britter, 1989: A random walk model for dispersion in inhomogeneous turbulence in a convective boundary layer. *Atmos. Environ.*, **23**, 1911-1924.
- Sawford, B.L., 1985: Lagrangian statistical simulation of concentration mean and fluctuation fields, *J. Climate and Appl. Meteor.*, **24**, 1152-1166.
- Sawford, B.L., 1986: Generalized random forcing in random-walk turbulent diffusion models. *Phys. Fluids*, **29**, 3582-3585.
- Sawford, B.L. and F.M. Guest, 1987: Lagrangian stochastic analysis of flux-gradient relationships in the convective boundary layer, *J. Atmos. Sci.*, **44**, 1152-1165.
- Sawford, B.L., 1993: Recent developments in the lagrangian stochastic theory of turbulent dispersion, *Boundary-Layer Meteorol.*, **62**, 197-215.
- Thomson, D.J., 1984: Random walk modeling in diffusion in inhomogeneous turbulence. *Quart. J.R. Met. Soc.*, **110**, 1107-1120.
- Thomson, D.J., 1987: Criteria for the selection of stochastic models of particle trajectories in turbulent flows. *J. Fluid Mech.*, **180**, 529-556.
- van Dop, H., F.T.M. Nieuwstadt, and J.C.R. Hunt, 1985: Random walk models for particle displacements in inhomogeneous unsteady turbulent flows. *Phys. Fluids*, **28**, 1639-1653.
- van Kampen, N.G., 1992: *Stochastic processes in physics and chemistry*. North-Holland, N.Y., 465 pp.
- Weil, J.C., 1990: A diagnosis of the asymmetry in top-down and bottom-up diffusion using a lagrangian stochastic model, *J. Atmos. Sci.*, **47**, 501-515.
- Wyngaard, J.C., 1988: Structure of the PBL, *Lectures on Air Pollution Meteorology*, A. Venkatram and J.C. Wyngaard (eds.). Am. Meteorol. Soc., Boston., 390 pp..