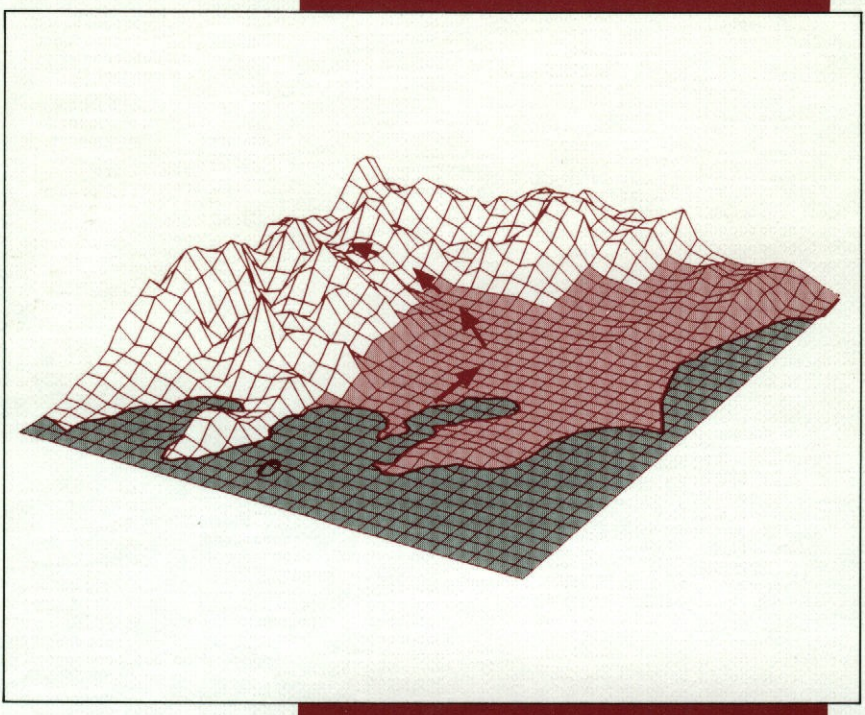
# Environmental Modelling

Editors: P. Melli
and P. Zannetti



Computational Mechanics Publications
Elsevier Applied Science

P. Melli
IBM ECSEC
Via Giorgione, 159
00147 Roma
Italy

P. Zannetti
IBM Semea
Via Pirelli, 18
20124 Milano
Italy

Co-published by

Computational Mechanics Publications
Ashurst Lodge, Ashurst, Southampton, UK

Computational Mechanics Publications Ltd
Sole Distributor in the USA and Canada:

Computational Mechanics Inc.
25 Bridge Street, Billerica, MA 01821, USA

and

Elsevier Science Publishers Ltd
Crown House, Linton Road, Barking, Essex IG11 8JU, UK

Elsevier's Sole Distributor in the USA and Canada:

Elsevier Science Publishing Company Inc.
655 Avenue of the Americas, New York, NY 10010, USA

**British Library Cataloguing-in-Publication Data**

A Catalogue record for this book is available
from the British Library

ISBN 1-85166-681-8 Elsevier Applied Science, London, New York
ISBN 1-85312-120-7 Computational Mechanics Publications, Southampton
ISBN 1-56252-053-9 Computational Mechanics Publications, Boston, USA

Library of Congress Catalog Card Number 91-73784



Printed and bound by Bell & Bain Ltd., Glasgow

# Chapter 11

# Particle Modeling and Its Application for Simulating Air Pollution Phenomena

P. Zannetti*
*IBM Semea, Via Pirelli 18, 20124 Milano, Italy*

* Currently at Failure Analysis Associates, Inc., 149 Commonwealth Drive, Menlo Park, CA 94025, U.S.A.

## 11.1 Introduction

This paper presents the particle modeling method and its application to simulate atmospheric dispersion phenomena. This approach is based upon the Lagrangian theory of atmospheric dispersion. The turbulent motion of air parcels is simulated by recreating atmospheric wind fluctuations and by tracing the motion of fictitious particles. This motion can be realistically simulated using Monte-Carlo techniques, i.e. computer-generated fluctuations.

This method is computationally powerful and particularly suited to simulate fluid dynamics phenomena. Moreover, this technique allows a unique handling of shear effects in the atmosphere flow and provides an impressive visualization of diffusion patterns.

Lagrangian models provide an alternative method for simulating atmospheric diffusion. They are called Lagrangian because they describe fluid elements that follow the instantaneous flow. The "Lagrangian" term was initially used to distinguish the Lagrangian box models from the Eulerian box models. In this case, the difference is manifest, since the Eulerian box does not move, while the Lagrangian box follows the average wind trajectory. The term has, however, been extended to describe all models in which plumes are broken up into "elements", such as segments, puffs, or fictitious particles (see Zannetti, 1990, for further discussion on Lagrangian and Eulerian modeling techniques).

Several efforts have been made to understand and parameterize the relationship between equivalent atmospheric parameters as seen in an Eulerian and a Lagrangian view. Hanna (1979) performed statistical analyses of wind fluctuations and showed that both Lagrangian and Eulerian observations of wind speed fluctuations $u'$ can be simulated by the linear first-order autoregression relationship

$$u'(t + \Delta t) = u'(t)R(\Delta t) + u''(t) \tag{1}$$

Where $R(\Delta t)$ is the autocorrelation coefficient at time lag $\Delta t$ and $u''$ is a random component. Davis (1982) examined various theories that aim at relating the velocity statistics of Lagrangian particles to the statistics of the Eulerian flow in which they move. Novikov (1969) proposed a connection between Lagrangian and Eulerian probabilities that was then generalized (Novikov, 1986) to fluids with variable density. In spite of the above efforts (and others), uncertainties still persist and a fully acceptable theoretical relationship between Eulerian and Lagrangian variables has not yet been developed (or, if it has, has not been fully tested against experimental data; testing is complicated by the fact that reliable Lagrangian measurements are scarce).

## 11.2 The Lagrangian Approach

The fundamental Lagrangian equation for atmospheric dispersion of a single pollutant species is

$$< c(\mathbf{r},t) > \; = \int_{-\infty}^{t} \int p(\mathbf{r},t|\mathbf{r}',t') \; S(\mathbf{r}',t') d\mathbf{r}' \; dt' \tag{2}$$

where the integration in space is performed over the entire atmospheric domain; $<c(\mathbf{r},\mathrm{t})>$ is the ensemble average concentration at $\mathbf{r}$ at time $t$; $\mathrm{S}(\mathbf{r}',\mathrm{t}')$ is the source term (mass volume$^{-1}$ time$^{-1}$); and $p(\mathbf{r},t \mid \mathbf{r}',t')$ is the probability density function (volume$^{-1}$) that an air parcel moves from $\mathbf{r}'$ at $t'$ to $\mathbf{r}$ at t, where, for any $\mathbf{r}'$ and $t > t'$,

$$\int p(\mathbf{r},t|\mathbf{r}',t') \; d\mathbf{r} \leq \; 1 \tag{3}$$

The expression in Equation 3 can be less than one when chemical or deposition phenomena are considered; otherwise, mass conservation always requires the value to be equal to one. For a primary pollutant, $S(\mathbf{r}',t')$ is greater than zero only at points $\mathbf{r}'$ where the pollutant is released (e.g., the exit points of stacks). For a secondary pollutant, $S(\mathbf{r}',t')$ can be nonzero virtually anywhere. For both primary and secondary pollutants, however, Equation 3, which represents mass conservation, must be satisfied.

Since it is often difficult to evaluate the entire emission "history" $S(\mathbf{r}',t')$ for $-\infty \leq t' \leq t$, Equation 2 can be rewritten as the sum of two integral terms

$$\begin{aligned} < c(\mathbf{r},t) > \; &= \int p(\mathbf{r},t|\mathbf{r}',t_o) < c(\mathbf{r}',t_o) > d\mathbf{r}' \\ &+ \int_{t_o}^{t} \int p(\mathbf{r},t|\mathbf{r}',t') \; S(\mathbf{r}',t') \; d\mathbf{r}' \; dt' \end{aligned} \tag{4}$$

in which only the contribution of the sources during $t_o \leq t' \leq t$ needs to be included, since the first integral term accounts for the source contribution before $t_o$. However, Equation 4 requires some estimate of the average concentration $<c>$ at $t_o$ throughout the computational domain.

It must be pointed out that the use of Equation 4 instead of Equation 2 may be incorrect when the exact fractional impact of a specific source (or group of sources) needs to be estimated. In fact, when using Equation 4, known background concentrations can be used to estimate the term $<c(\mathbf{r}', t_o)>$. But, by so doing, the contribution of a specific source to the concentration $<c(\mathbf{r}, t)>$ becomes the sum of two terms: (1) the direct contribution of the second integral of Equation 4, and (2) the indirect contribution of the source to the background concentration $<c(\mathbf{r}', t_o)>$. When these two contributions are of the same order of magnitude, as in long-range regional modeling applications, the fractional impact of a specific source becomes difficult to evaluate because, even when the second integral of Equation 4 is computed correctly, the contribution of a source to background concentrations is difficult to calculate.

The key parameter in the above equations is the probability density function $p$, which, for nonreactive pollutants, is a function of only the meteorology (and the type of pollutant when deposition phenomena are considered). Equations 2 or 4 represent a rigorous description of transport and diffusion processes expressed in a probabilistic notation. The full incorporation of chemical reactions, however, is difficult.

Different assumptions concerning the probability density function $p$ allow the derivation of both Gaussian equations and the K-theory equation. Seinfeld (1975) shows that all Gaussian plume and puff formulas can be derived from the Lagrangian equation (2) under the following simplifying assumptions:

1. Turbulence is stationary and homogeneous; i.e.,

$$p(\mathbf{r}, t|\mathbf{r}', t') = p(\mathbf{r} - \mathbf{r}', t - t') \tag{5}$$

2. $p$ obeys a multidimensional normal distribution; i.e.,

$$p(\mathbf{r} - \mathbf{r}', t - t') = \frac{1}{(2\pi)^{3/2} |\mathbf{P}|^{1/2}} \exp\ [-\boldsymbol{\xi}^T \mathbf{P}^{-1} \boldsymbol{\xi}/2] \tag{6}$$

where each element $P_{ij}$ of the matrix $\mathbf{P}$ is ($i$ and $j$ = 1, 2, or 3)

$$P_{ij} = < \xi_i \xi_j > \tag{7}$$

and the "displacements" $\xi_i$ are

$$\xi_i = |\mathbf{r} - \mathbf{r}'|_i - |< \mathbf{r} - \mathbf{r}' >|_i \tag{8}$$

in which $i$ indicates the space component ($x, y$, or $z$, for $i$ = 1, 2, or 3, respectively).

3. The term $< \mathbf{r} - \mathbf{r}' >$ is the average displacement, which is assumed to be due only to the average (deterministic) wind $\bar{\mathbf{u}}$.

4. $P_{ij} = 0, \text{for } i \neq j$.

Several types of models can be classified as Lagrangian:

- Lagrangian box, or trajectory, models, which are used for photochemical simulations
- Gaussian segmented plume models
- Gaussian puff models
- Particle models

Particle models are presented in Section 11.3 below. See Zannetti (1990) for a presentation of the other modeling techniques.

## 11.3 Particle Models

Particle modeling is the most recent and powerful computational tool for the numerical discretization of a physical system. It has been particularly successful in a wide spectrum of applications (Hockney and Eastwood, 1981), that range from the atomic scale (electron flow in semiconductors, molecular dynamics) to the astronomical scale (galaxy dynamics), with other important applications to plasma and turbulent fluid dynamics. Particle models handle the transport terms, whose correct numerical treatment is very difficult with Eulerian (grid) models, in a straightforward manner. Particles, in fact, have a Lagrangian nature, since they move following the main flow. For this reason, they are often called Lagrangian particles.

Particle models use a certain number of computational (fictitious) particles to simulate the dynamics of a selected parameter (e.g., mass, heat, electrical charge density, etc.). Particle motion can be produced by both deterministic velocities and semirandom pseudovelocities generated using Monte-Carlo techniques. In the latter case, the trajectory of a single particle simply represents a realization from an infinite set of possible solutions. Important characteristics of the diffusion process can be inferred, however, from the computation of average particle ensemble properties, which are not affected by the randomness of the pseudovelocities if enough particles are used.

Three main types of particle models can be defined (Hockney and Eastwood, 1981):

- particle-particle (PP) models, in which all interaction forces (e.g., gravitational or electric forces) between particles are computed at each time step

- particle-mesh (PM) models, in which forces are computed using a field equation (on a grid) for the potential

- PP-PM or ($P^3M$) models, a hybrid approach, in which interparticle forces are split into a short-range component (computed using the PP method) and a slowly varying one (represented in a mesh system by the PM method)

Length and time scales (as in all discretization systems) play an important role in particle models. In particular, the relation between the actual physical particles (or elements) and the computer model simulation particles is an important factor for the interpretation of the simulation results. In general, three possible cases can be found (Hockney and Eastwood, 1981):

- a one-to-one correspondence between actual and simulated particles, as, for example, in molecular dynamics simulation

- a description of fluid elements (position, vorticity) as particles, as, for example, in vortex fluid simulations, where the correspondence to physical particles (molecules) is totally lost

- the use of "superparticles;" i.e., simulation particles representing a cloud of physical particles having similar characteristics

Particle models have mostly been applied to simulate (and understand) the spiral structure of the galaxies, to simulate plasma dynamics and the electron flow in semiconductors, and to obtain realistic representations of turbulence in fluid.

In air pollution applications, using Lagrangian particle methods, emitted gaseous material is characterized by a set of computational particles and each particle is "moved" at each time step by pseudovelocities, which take into account the three basic dispersion components: 1) the transport due to the mean fluid velocity; 2) the (seemingly) random turbulent fluctuations of wind components (both horizontal and vertical); and 3) the molecular diffusion (if not neglible). After the pioneering work of Smith (1968) and Hall (1975), Lamb (1978) simulated vertical turbulent phenomena by assigning to each particle a velocity

$$w = w_d + w_s \tag{9}$$

where the first term $w_d$ was determined by the Eulerian numerical model of Deardorff (1974) and $w_s$ was a stochastic term describing the effect of subgrid fluctuations not included in the numerical model. (The term $w_d$ was generated every eight seconds on a grid with $\Delta z = 50$ m and, therefore, contained a large fraction of the fluctuating turbulent velocities that, in other models, are simulated by the stochastic terms.) Zannetti (1981, 1984) introduced a scheme for the inclusion of the cross correlation

among the velocity fluctuations. Baerentsen and Berkowicz (1984) used two separate equations to describe particle updrafts and downdrafts, under the assumption that the physics of the two phenomena is different.

As illustrated by de Baas et al. (1986), most particle modeling studies of air quality phenomena are numerical solutions of the Langevin stochastic differential equation (Reid, 1979; Gifford, 1982; Sawford, 1984)

$$dw = -(w/T_L)dt + d\mu \tag{10}$$

where $w$ is any component of the Lagrangian particle velocity, $T_L$ is its Lagrangian time scale, and $d\mu$ are random velocity increments. The use of this equation, its limitations and possible improvements are described below in the section on simulation of convective conditions by Monte-Carlo particle models. Several of the concepts introduced above and related to the use of particle modeling in the simulation of atmospheric diffusion will be expanded in the following sections.

## Simulation of Atmospheric Diffusion by Particle Models

Equation 2 or 4 can be solved analytically or numerically. For example, as discussed before, a Gaussian distribution of $p$, together with other simplifying assumptions, allows the derivation of Gaussian plume and puff equations for $<c>$. More complex functions of $p$ require numerical integrations.

An intuitive solution of Equation 2 or 4 can be obtained if a set of dynamic atmospheric trajectories of pollutant mass can be generated. Then, for each trajectory originating from $\mathbf{r}'$ at $t'$, we have $p(\mathbf{r}, t\,|\,\mathbf{r}', t') = 0$ everywhere, except at the exact location $\mathbf{r} = \mathbf{r}^*$, where the trajectory point is located at $t$, thus giving $p(\mathbf{r}, t|\mathbf{r}', t') = \delta(\mathbf{r}^* - \mathbf{r})$. Therefore, if realistic air parcel trajectories can be computed, the simple calculation of the density of the trajectory points provides an estimate of $<c>$. This is the conceptual basis of a "particle" model for atmospheric dispersion, i.e., a model in which a set of "tracers" (or fictitious computational particles) are used to describe the dynamics of the atmosphere.

Particle models can be used to characterize atmospheric dispersion in two simulation modes: the "single-particle" mode, in which the motion of each particle is independent from the others and, therefore, obeys one-particle statistics; and the "two-particle" mode, used to reproduce *relative* dispersion, e.g., the dispersion properties of a single puff in relation to its center of mass. The second approach has been investigated by several authors (Durbin, 1980; Lamb, 1981; Sawford 1983; Lee et al., 1985) and, in particular, by Gifford (1982), who used the simple Langevin Equation 10 to simulate relative dispersion by constraining the initial particle velocity (see Sawford, 1984, for further discussion of this topic). In the rest of this chapter we will discuss "single-particle" models.

In air pollution applications, simulation particles are moved at each time step by a velocity $\mathbf{u}_e$, which is sometimes called a "pseudovelocity" to emphasize that we do not need to follow precisely each molecule in the atmospheric turbulent flow, but only to define an algorithm for particle displacement computation that gives an accurate density distribution, i.e., an ensemble average. In mathematical notation, if a particle is located in $\mathbf{x}(t_1)$ at time $t_1$, its position at time $t_2$ will be

$$\mathbf{x}(t_2) = \mathbf{x}(t_1) + \int_{t_1}^{t_2} \mathbf{u}[\mathbf{x}(t), t]dt \tag{11}$$

where $\mathbf{u}$ is the "instantaneous" wind vector in each point $\mathbf{x}(t)$ of the particle trajectory between $t_1$ and $t_2$.

Atmospheric turbulent properties make $\mathbf{u}$ practically impossible to know, especially due to semirandom components caused by atmospheric turbulent eddies. But the "equivalent" or "effective" wind vector $\mathbf{u}_e$ can be considered

$$\mathbf{u}_e = \int_{t_1}^{t_2} \mathbf{u}[\mathbf{x}(t), t]\, dt/(t_2 - t_1) \tag{12}$$

which moves the particle directly from $\mathbf{x}(t_1)$ to $\mathbf{x}(t_2)$ in the interval $(t_1, t_2)$. The problem is then to estimate $\mathbf{u}_e$ from Eulerian measurements of $\mathbf{u}$, keeping in mind that $\mathbf{u}_e$ must approximate the integral term in Equation 12 only on a particle *ensemble* basis. For example, we can define

$$\mathbf{u}_e = \bar{\mathbf{u}} + \mathbf{u}' \tag{13}$$

where $\bar{\mathbf{u}}$ is the best estimate of the average Eulerian wind vector (transport) at the particle location, and $\mathbf{u}'$ is a "diffusivity velocity". In other words, $\bar{\mathbf{u}}$ (a smoothly variable term) represents our deterministic understanding of the average transport process, based on Eulerian wind measurement interpolation or provided by a meteorological model, while $\mathbf{u}'$ is an artificial numerical perturbation, which is related to the turbulence intensities and characteristics of those smaller eddies that are not included in the $\bar{\mathbf{u}}$ field.

Since, in Equation 13, $\bar{\mathbf{u}}$ is assumed to be known from measurements and/or meteorological model outputs, computing $\mathbf{u}'$ is the key problem of Lagrangian particle modeling. Two fundamental approaches can be followed: the deterministic approach, which represents a numerical procedure for solving the diffusion equation, and the statistical approach, which actually models the randomness of the trajectories of fluid elements. Both approaches are discussed below.

## Deterministic Calculation of $\mathbf{u}'$

A typical example of the deterministic approach is given by the particle-in-cell method (Lange, 1978; Rodriguez et al., 1982), in which, after some manipulation of the $K$-theory diffusion equation, the following relation is obtained

$$\mathbf{u}' = \left(-\frac{K}{c}\right) \nabla c \tag{14}$$

where $K$ is the eddy diffusion coefficient and $c$ the concentration, computed from particle density. This method generally requires partitioning the computational domain into cells in order to calculate $c$. It is able to duplicate $K$-theory dispersion, with the important feature of decreasing the numerical advection errors otherwise produced by finite-difference solutions. Using this method, the motion of a single particle will be affected by the time-varying concentration field $c$, i.e., by the positions of the other

particles.

### Statistical Calculation of u'

The statistical approach (Monte Carlo-type models) seems more flexible and appealing than the deterministic approach. According to the statistical approach, $\mathbf{u}'$ is a semirandom component computed by manipulating computer-generated random numbers. To perform this computation, it has generally been assumed that Eulerian measurements of $\mathbf{u}$ can provide statistical information on $\mathbf{u}'$.

If we accept this assumption, we can use, for the diffusivity velocity $\mathbf{u}'$, a statistical generation scheme based on our understanding (and Eulerian measurements) of $\mathbf{u}$. In particular, Hanna (1979) has shown that it is a plausible assumption to describe both Eulerian and Lagrangian wind vector fluctuations by a simple Markov process (autocorrelation process of the first order). If we extend this assumption to $\mathbf{u}'$, we have[1]

$$\mathbf{u}'(t_2) = \mathbf{R}(\Delta t)\, \mathbf{u}'\,(t_1) \;+\; \mathbf{u}''(t_2) \tag{15}$$

where $\mathbf{R}(\Delta t)$ is a vector containing the autocorrelations with lag $\Delta t = t_2 - t_1$ of the $\mathbf{u}'$ components, and $\mathbf{u}''$ is a purely random vector that will be discussed further below.

Equation 15 is the key formula for statistically computing $\mathbf{u}'$. It is a recursive sum of two terms: the first is a function of the "previous" $\mathbf{u}'$ of the same particle, and the second is purely randomly generated. Since Equation 15 is computed independently for each particle, two eventually coincident particles at $t_1$ will have, in general, different displacements, even if their past "history" is exactly the same. Using this approach, the motion of a particle is not affected by the position of the other particles and, therefore, this numerical algorithm is extremely fast, since no interacting forces need to be computed.

To apply Equation 15, we need the initial $\mathbf{u}'(t_o)$ for each particle at its generation time $t_o$ (often assumed to be a zero vector or random with variance $\boldsymbol{\sigma}^2_{u'}$) and the dynamic computation of $\mathbf{R}$ and $\mathbf{u}''$. $\mathbf{R}$ can be related to Lagrangian turbulence time scales by

$$\mathbf{R}(\Delta t) = \exp\,[-\Delta t/\mathbf{T}_L] \tag{16}$$

where $\mathbf{T}_L$ is a vector containing the two horizontal and the one vertical Lagrangian time scales[2]. Generally, Lagrangian measurements of $\mathbf{T}_L$ are not available, but empirical relations have been proposed (e.g., Hanna, in Nieuwstadt and van Dop, 1982) to estimate $\mathbf{T}_L$ from Eulerian meteorological measurements (see section below on concentration calculations using particle models).

Assuming $\mathbf{u}''$ a purely random vector with zero-mean, normally distributed independent components, we have that $\mathbf{u}''$ is completely characterized by $\boldsymbol{\sigma}_{u''}$; i.e., the standard deviations of its components. By taking the variances of Equation 15, we obtain

[1] In this formula (and the following ones), when vectors appear on both sides of an equation, each component of the vector on the left side is computed using only the corresponding component of each vector in the right side (componentwise notation).

[2] Other equations have been proposed instead of Equation 16. Note, however, that it is essential that R is exponential if results are to be independent of $\Delta t$ (Thompson, personal communication).

$$\sigma_{u''} = \sigma_{u'} \left[1 - \mathbf{R}^2(\Delta t)\right]^{1/2} \tag{17}$$

Equation 17 requires the knowledge of $\sigma_{u'}$, the standard deviations of the components $\mathbf{u}'$, which, again, can be approximated by the standard deviations of available Eulerian wind measurements. Therefore, using the standard deviations $\sigma_{u''}$ computed by Equation 17, it is easy, with commonly available Monte-Carlo computer programs, to generate each particle's $\mathbf{u}''$ term for use in Equation 15.

As discussed below in the section on simulation of convective conditions by Monte-Carlo particle models, it has been established that the development leading to Equation 17 is valid only for stationary, homogeneous and isotropic turbulence. Nevertheless, in situations where meteorological gradients are not too strong, $\mathbf{R}$ and $\sigma_{u'}$ can be considered space and time dependent (but assumed constant between $t_1$ and $t_2$ to derive Equation 17). Therefore, they can fully incorporate, when available, time-varying three-dimensional meteorological input (Eulerian values) and can simulate, with a high degree of spatial and temporal resolution, extremely complex atmospheric diffusion conditions, which are impossible to treat with other numerical schemes. This approach is grid-free, since, even when the meteorological input $\bar{\mathbf{u}}, \mathbf{R}$ and $\sigma_{u'}$ is given at grid points, each particle can move according to meteorological values that can be interpolated exactly at the particle's location. This provides a high degree of resolution, which is controlled only by the number of particles and the length of the time interval $\Delta t$, and not by the spatial discretization of the computational domain.

Hanna (in Nieuwstadt and van Dop, 1982) proposed a set of semi-empirical formulae that, from a limited number of meteorological parameters ($h, L, w_*$, $z_o$, and $u_*$) provide the meteorological input at each particle's elevation $z$ required by Equations 15 through 17. This scheme, in which the subscripts $a$ and $c$ indicate the along-wind and cross-wind horizontal components, respectively, is described below.

- **Unstable Conditions**

In unstable conditions, the horizontal components of $\sigma_{u'}$ are constant, i.e.,

$$\sigma_{u'_a} = \sigma_{u'_c} = u_* \left(12 + 0.5\, h/|L|\right)^{1/3} \simeq \sqrt{0.31}\ w_* \tag{18}$$

while the vertical component varies with $z$ as follows:

$$\sigma_{u'_z} = 0.96\ w_* \left(\frac{3z}{h} - \frac{L}{h}\right)^{1/3} \tag{19}$$

for $z \leq 0.03\ h$;

$$\sigma_{u'_z} = w_*\ \min\left[0.96 \left(\frac{3z}{h} - \frac{L}{h}\right)^{1/3};\ 0.763 \left(\frac{z}{h}\right)^{0.175}\right] \tag{20}$$

for $0.03\ h < z < 0.4\ h$;

$$\sigma_{u'_z} = 0.722\ w_* \left(1 - \frac{z}{h}\right)^{0.207} \tag{21}$$

for $0.4\ h \leq z < 0.96\ h$; and

$$\sigma_{u'_z} = 0.37\ w_* \tag{22}$$

for $0.96\ h < z \leq h$.

The autocorrelations are computed by Equation 16, where the two horizontal components of $\mathbf{T}_L$ are constant, i.e.,

$$T_{L_a} = T_{L_c} = 0.15\ \frac{h}{\sigma_{u'_a}} \tag{23}$$

and the vertical component varies with $z$ as follows:

$$T_{L_z} = 0.1\ \frac{z}{\sigma_{u'_z}[0.55 + 0.38\ (z - z_o)/L]} \tag{24}$$

for $z < 0.1\ h$ and $z - z_o > -L$;

$$T_{L_z} = 0.59\ \frac{z}{\sigma_{u'_z}} \tag{25}$$

for $z < 0.1\ h$ and $z - z_o < -L$; and

$$T_{L_z} = 0.15\ \frac{h}{\sigma_{u'_z}}\left[1 - \exp\left(-\frac{5z}{h}\right)\right] \tag{26}$$

for $z > 0.1\ h$.

- **Stable Conditions**

In stable conditions, $h$ represents the top of the mechanically turbulent layer above the ground and can be evaluated by

$$h_{eq} = const\sqrt{\frac{u_*}{f}L} \tag{27a}$$

or by

$$h = 0.25\ u_*\ L/f \tag{27b}$$

The components of $\boldsymbol{\sigma}_{u'}$ vary with $z$ as follows

$$\sigma_{u'_a} = 2.0\ u_*\left(1 - \frac{z}{h}\right) \tag{28}$$

$$\sigma_{u'_c} = \sigma_{u'_z} = 1.3\ u_*\left(1 - \frac{z}{h}\right) \tag{29}$$

while the autocorrelations are computed by Equation 16 with

$$T_{L_a} = 0.15\ \frac{h}{\sigma_{u'_a}}\left(\frac{z}{h}\right)^{0.5} \tag{30}$$

$$T_{L_c} = 0.07 \ \frac{h}{\sigma_{u'_c}} \left(\frac{z}{h}\right)^{0.5} \tag{31}$$

and

$$T_{L_z} = 0.10 \ \frac{h}{\sigma_{u'_z}} \left(\frac{z}{h}\right)^{0.8} \tag{32}$$

- **Neutral Conditions**
  In neutral conditions, the components of $\boldsymbol{\sigma}_{u'}$ are

$$\sigma_{u'_a} = 2.0 \ u_* \ \exp(-3 \ f \ z/u_*) \tag{33}$$

and

$$\sigma_{u'_c} = \sigma_{u'_z} = 1.3 \ u_* \ \exp(-2 \ f \ z/u_*) \tag{34}$$

while the autocorrelations are computed again by Equation 16 with

$$T_{L_a} = T_{L_c} = T_{L_z} = \frac{0.5 \ z/\sigma_{u'_z}}{1 + 15 \ f \ z/u_*} \tag{35}$$

In addition to the above formulation, it is generally assumed that $\mathbf{u}' = \mathbf{0}$ for $z > h$ and that the particles are totally or partially reflected at the ground and at the top of the PBL – an operation that also requires a change of sign in the "memory" $u'_z$ of each reflected particle.

## The Introduction of the Cross-Correlations

Using Monte-Carlo techniques, it is necessary to simulate realistically the wind fluctuation behavior in a way that is consistent with measured characteristics. Several current models are based on Equation 15, which assumes that the components of $\mathbf{u}'$ are statistically independent. This assumption is in disagreement with wind fluctuation measurements, which indicate the existence of non-zero cross-correlation terms. Therefore, Equation 15 can be, in many cases, an oversimplification of the atmospheric dispersion processes.

Zannetti (1981), Ley (1982) and Legg (1983) have proposed schemes that include the negative correlation $\overline{u'w'}$ between the "along-wind" component $u'$ and the vertical component $w'$. For example, Zannetti (1981, 1984) developed the scheme

$$u'_a(t_2) = \phi_1 \ u'_a(t_1) + u''_a(t_2) \tag{36}$$

$$u'_c(t_2) = \phi_2 \ u'_c(t_1) + u''_c(t_2) \tag{37}$$

$$u'_z(t_2) = \phi_3 \ u'_z(t_1) + \phi_4 \ u'_a(t_2) + u''_z(t_2) \tag{38}$$

in which the vector $\mathbf{u}'$ is seen in a "flux-coordinate" system where

$$\mathbf{u}' = (u'_a, u'_c, u'_z) \tag{39}$$

and $u'_a$ is the horizontal component along the average wind direction, $u'_c$ is the horizontal cross-wind component, and $u'_z$ is the vertical component. Note that, since the average wind direction varies with space and time, each particle will have, in general, its own time-varying reference system, determined by the horizontal direction of $\bar{\mathbf{u}}$, as defined by Equation 13, at the particle's location.

By analytical manipulations of Equations 36 through 38, the parameters $\phi_1, \phi_2, \phi_3, \phi_4, \sigma_{u''_a}, \sigma_{u''_c}$ and $\sigma_{u''_z}$ are calculated by

$$\phi_1 = r_{u'_a}(\Delta t) \tag{40}$$

$$\phi_2 = r_{u'_c}(\Delta t) \tag{41}$$

$$\phi_3 = \frac{r_{u'_z}(\Delta t) - \phi_1 \, r^2_{u'_a u'_z}(0)}{1 - \phi_1^2 \, r^2_{u'_a u'_z}(0)} \tag{42}$$

$$\phi_4 = \frac{r_{u'_a u'_z}(0) \, \sigma_{u'_z} \, [1 - \phi_1 \, r_{u'_z}(\Delta t)]}{\sigma_{u'_a}[1 - \phi_1^2 \, r^2_{u'_a u'_z}(0)]} \tag{43}$$

and

$$\sigma^2_{u''_a} = \sigma^2_{u'_a}(1 - \phi_1^2) \tag{44}$$

$$\sigma^2_{u''_c} = \sigma^2_{u'_c}(1 - \phi_2^2) \tag{45}$$

$$\sigma^2_{u''_z} = \sigma^2_{u'_z}(1 - \phi_3^2) - \phi_4^2 \, \sigma^2_{u'_a} - 2 \, \phi_1 \, \phi_3 \, \phi_4 \, r_{u'_a u'_z}(0) \, \sigma_{u'_a} \, \sigma_{u'_z} \tag{46}$$

where $r_{u'_a}(\Delta t), r_{u'_c}(\Delta t), r_{u'_z}(\Delta t)$ are the autocorrelations, with time lag $\Delta t = t_2 - t_1$, of the components of $\mathbf{u}'$, as defined by Equation 39; $r_{u'_a u'_z}(0)$ is the cross-correlation, with no time lag, between $u'_a$ and $u'_z$; $\sigma_{u''_a}, \sigma_{u''_c}$ and $\sigma_{u''_z}$ are the standard deviations of the components of the vector $\mathbf{u}''$

$$\mathbf{u}'' = (u''_a, u''_c, u''_z) \tag{47}$$

where these components are uncorrelated zero-averaged Gaussian noises (i.e., random numbers); and $\sigma_{u'_a}, \sigma_{u'_c}, \sigma_{u'_z}$ are the standard deviations of the $\mathbf{u}'$ components. The parameters in Equations 40 through 46 can vary with space and time, but are assumed constant between $t_1$ and $t_2$.

The above method is able to generate a time-varying $\mathbf{u}'$ with any theoretically acceptable degree of auto- and cross-correlations, if the meteorological input is known. The meteorological input $r_{u'_a}(\Delta t), r_{u'_c}(\Delta t), r_{u'_z}(\Delta t), \sigma_{u'_a}, \sigma_{u'_c}$, and $\sigma_{u'_z}$ can be obtained

using Hanna's scheme presented in the previous section, while the extra term $r_{u'_a u'_z}(0)$ can be estimated at the ground by

$$[r_{u'_a u'_z}(0)]_{z=0} = -\frac{u_*^2}{\sigma_{u'_a}\,\sigma_{u'_z}} \tag{48}$$

Equation 48 can be linearly interpolated from $z = 0$ to $z = h$, where $[r_{u'_a u'_z}(0)]_{z=h} = 0$, thus giving, at a generic $z$ below $h$,

$$[r_{u'_a u'_z}(0)]_z = -\frac{u_*^2}{\sigma_{u'_a}\,\sigma_{u'_z}}\left(1 - \frac{z}{h}\right) \tag{49}$$

If direct measurements of $\boldsymbol{\sigma}_{u'}$ are available, they can be used directly (e.g., by interpolation at different altitudes) instead of using the semiempirical formulae described above. For example, measurements of $\sigma_\theta$, the standard deviation of the horizontal wind direction, can be used to calculate $\sigma_{u'_c}$, through the relationship

$$\sigma_{u'_c} \simeq \bar{u}\,\sigma_\theta \tag{50}$$

where $\bar{u}$ is the average horizontal wind speed and $\sigma_\theta$ is expressed in radians. If measurements of $\mathbf{u}'$ are performed in a fixed orthogonal system $x,y,z$, i.e.,

$$\mathbf{u}' = (u'_x, u'_y, u'_z) \tag{51}$$

then, the standard deviations $\sigma_{u'_x}$ and $\sigma_{u'_y}$ allow the calculation of $\sigma_{u'_a}$ and $\sigma_{u'_c}$ by using (Zannetti, 1984)

$$\sigma_{u'_a} = \frac{\overline{u}_x^2\,\sigma_{u'_x}^2 - \overline{u}_y^2\,\sigma_{u'_y}^2}{\overline{u}_x^2 - \overline{u}_y^2} \tag{52}$$

$$\sigma_{u'_c} = \frac{\overline{u}_x^2\,\sigma_{u'_y}^2 - \overline{u}_y^2\,\sigma_{u'_x}^2}{\overline{u}_x^2 - \overline{u}_y^2} \tag{53}$$

where $\overline{u}_x$ and $\overline{u}_y$ are the horizontal components of the average wind $\overline{u}$ in the fixed coordinate system. Equations 52 and 53 are derived by assuming that the $u'_a$ and $u'_c$ are not cross-correlated. Also note that these equations are not valid when $|\overline{u}_x| \simeq |\overline{u}_y|$, i.e., when the average wind direction is blowing with an angle of 45 degrees, 135 degrees, 225 degrees, and 315 degrees with respect to the x-axis (in these cases, no alternative equations can be provided; the system orientation simply does not allow discrimination between along-wind and crosswind fluctuations).

Zannetti (1986) expanded the scheme of Equations 36 through 38 to work in a generic fixed orthogonal system $x, y, z$, which requires the incorporation of all three cross-correlations among the $\mathbf{u}'$ components. The system becomes

$$u'_x(t_2) = f_1\,u'_x(t_1) + u''_x(t_2) \tag{54}$$

$$u'_y(t_2) = f_2\, u'_y(t_1) + f_3\, u'_x(t_2) + u''_y(t_2) \tag{55}$$

$$u'_z(t_2) = f_4\, u'_z(t_1) + f_5\, u'_y(t_2) + f_6\, u'_x(t_2) + u''_z(t_2) \tag{56}$$

Again, this system is able to generate a time sequence of $\mathbf{u}'$ values with any theoretically acceptable degree of auto- and cross-correlation, if the meteorological input is specified. For Equations 54 through 56, the meteorological input must include the three cross-correlations $r_{u'_x u'_y}(0), r_{u'_x u'_z}(0)$, and $r_{u'_y u'_z}(0)$. Algebraic manipulations allow the deviation of the parameters $f_1, f_2, f_3, f_4, f_5, f_6$ and the standard derivations $\sigma_{u''_x}, \sigma_{u''_y}, \sigma_{u''_z}$ from the meteorological input, as follows. First, analytical manipulation of Equations 54 and 55 allow the derivation of

$$f_1 = r_{u'_x}(\Delta t) \tag{57}$$

$$\sigma^2_{u''_x} = \sigma^2_{u'_x}\ (1 - f_1^2) \tag{58}$$

$$f_2 = \frac{r_{u'_y}(\Delta t) - r_{u'_x}(\Delta t)\, r^2_{u'_x u'_y}(0)}{1 - r^2_{u'_x}(\Delta t)\, r^2_{u'_x u'_y}(0)} \tag{59}$$

$$f_3 = \frac{r_{u'_x u'_y}(0)\, \sigma_{u'_y}\, [1 - r_{u'_x}(\Delta t)\, r_{u'_y}(\Delta t)]}{\sigma_{u'_x}\, [1 - r^2_{u'_x}(\Delta t)\, r^2_{u'_x u'_y}(0)]} \tag{60}$$

and

$$\sigma^2_{u''_y} = \sigma^2_{u'_y}\ (1 - f_2^2) - f_3^2\, \sigma^2_{u'_x} - 2\, f_1\, f_2\, f_3\, r_{u'_x u'_y}(0)\, \sigma_{u'_x}\, \sigma_{u'_y} \tag{61}$$

Then, the terms $f_4, f_5$ and $f_6$ are computed by solving the linear system

$$\begin{pmatrix} a_{44} & a_{45} & a_{46} \\ a_{54} & a_{55} & a_{56} \\ a_{64} & a_{65} & a_{66} \end{pmatrix} \begin{pmatrix} f_4 \\ f_5 \\ f_6 \end{pmatrix} = \begin{pmatrix} b_4 \\ b_5 \\ b_6 \end{pmatrix} \tag{62}$$

with

$$a_{44} = \sigma_{u'_z} \tag{63}$$

$$a_{45} = f_2\, r_{u'_y u'_z}(0)\ \sigma_{u'_y} + f_3\, f_1\, r_{u'_x u'_z}(0)\ \sigma_{u'_x} \tag{64}$$

$$a_{46} = f_1\, r_{u'_x u'_z}(0)\ \sigma_{u'_x} \tag{65}$$

$$a_{54} = f_1\, r_{u'_x u'_z}(0)\ \sigma_{u'_z} \tag{66}$$

$$a_{55} = f_1\, f_2\, r_{u'_x u'_y}(0)\, \sigma_{u'_y} + f_1\, f_3\, r_{u'_x}(\Delta t)\, \sigma_{u'_x} + f_3\, \sigma_{u'_x}\ (1 - f_1^2) \tag{67}$$

$$a_{56} = f_1\, r_{u'_x}(\Delta t)\ \sigma_{u'_x} + \sigma_{u'_x}(1 - f_1^2) \tag{68}$$

$$a_{64} = f_2\, r_{u'_y u'_z}(0)\ \sigma_{u'_y}\ \sigma_{u'_z} + f_3\, f_1\, r_{u'_x u'_z}(0)\ \sigma_{u'_x}\ \sigma_{u'_z} \tag{69}$$

$$a_{65} = f_2\, r_{u'_y}(\Delta t)\ \sigma^2_{u'_y} + f_3\, r_{u'_x u'_y}(0)\ \sigma_{u'_x}\ \sigma_{u'_y} + \sigma^2_{u''_y} \tag{70}$$

$$a_{66} = f_2\, f_1\, r_{u'_x u'_y}(0)\ \sigma_{u'_x}\ \sigma_{u'_y} + f_3\, \sigma^2_{u'_x} \tag{71}$$

and

$$b_4 = r_{u'_z}(\Delta t)\ \sigma_{u'_z} \tag{72}$$

$$b_5 = r_{u'_x u'_z}(0)\ \sigma_{u'_z} \tag{73}$$

$$b_6 = r_{u'_y u'_z}(0)\ \sigma_{u'_y}\ \sigma_{u'_z} \tag{74}$$

This system allows a numerical solution of Equation 62. (An analytical solution for $f_4, f_5$, and $f_6$ could be derived but is too cumbersome.)

Finally, the last term $\sigma_{u''_z}$ is given by

$$\begin{aligned} \sigma^2_{u''_z} &= \sigma^2_{u'_z}(1 - f_4^2) - f_5^2\ \sigma^2_{u'_y} - f_6^2\, \sigma^2_{u'_x} \\ &- 2\, f_4\, f_5\, [f_2\, r_{u'_y u'_z}(0)\ \sigma_{u'_y}\ \sigma_{u'_z} + f_3\, f_1\, r_{u'_x u'_z}(0)\ \sigma_{u'_x}\ \sigma_{u'_z}] \\ &- 2\, f_4\, f_6\, f_1\, r_{u'_x u'_z}(0)\ \sigma_{u'_x}\ \sigma_{u'_z} - 2\, f_5\, f_6\, r_{u'_x u'_y}(0)\ \sigma_{u'_x}\ \sigma_{u'_y} \end{aligned} \tag{75}$$

It must be pointed out that the methods dealing with the cross-correlations presented above may be inherently inconsistent. At the end of the section below on simulation of convective conditions by Monte-Carlo particle models, however, we propose a simple mechanism to incorporate these methods into an acceptable theoretical frame.

## Simulation of Convective Conditions by Monte-Carlo Particle Models

Some of the most interesting developments of particle modeling have focused on the one-dimensional (i.e., vertical) simulation of convective dispersion conditions, and on the use of the Langevin Equation 10 to simulate the Lagrangian vertical velocity $w$ of each particle. As summarized and clarified by de Baas et al. (1986), Equation 10 can provide different sets of simulation outputs, depending upon the specification of the random velocity increments $d\mu$.

In homogeneous turbulence, we have

$$\overline{d\mu} = 0 \tag{76}$$

$$\overline{(d\mu)^2} = 2\overline{u_z^2}dt/T_L \tag{77}$$

$$\overline{d\mu^3} = 0 \tag{78}$$

where $u_z$ is the vertical wind component. Under these limiting conditions and with $dt = t_2 - t_1$, Equation 10 becomes equivalent to the vertical component of Equation 15. In this case, particle simulations are able to reproduce (e.g., see the simulations of Brusasca et al., 1987, using the MC-LAGPAR model) the theoretical results obtained by Taylor (1921) for homogeneous turbulence as illustrated in Figure 11.1.

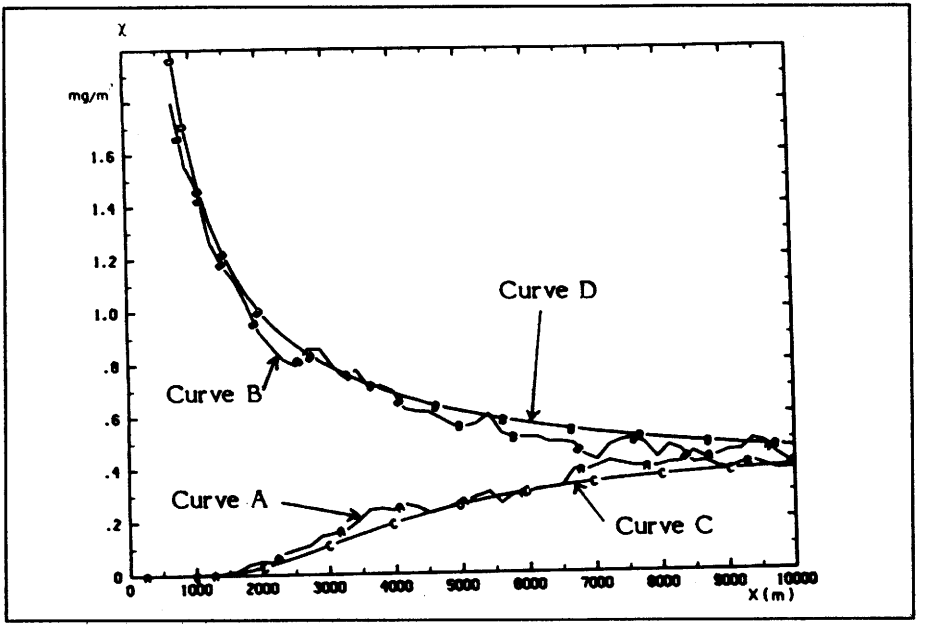



Figure 11.1: Concentration as a function of a downwind distance (from Brusasca et al., 1987). [Reprinted with permission from Computational Mechanics Publications.]

**Curve A:** ground-level concentration simulated by MC-LAGPAR

**Curve B:** centerline plume concentration simulated by MC-LAGPAR

**Curve C:** ground-level concentration computed with the analytical solution of Taylor (1921)

**Curve D:** centerline plume concentration computed with the analytical solution of Taylor (1921)

Convective conditions in the atmosphere, however, are strongly characterized by nonhomogeneous conditions in which, for example, $\overline{u_z^2}$ varies with the height $z$. In this situation, if Equation 10 is used together with Equation 77 and a term $\overline{u_z^2}(z)$ that varies with $z$, particles have a tendency to be trapped, without any physical justifications, in regions with lower $\overline{u_z^2}$. This accumulation is avoided or minimized by

changing Equation 76 into

$$\overline{d\mu} = dt\ \partial\overline{u_z^2}/\ \partial z \tag{79}$$

which represents a nonzero mean random forcing (i.e., a drift velocity) proportional to the vertical gradient of $\overline{u_z^2}(z)$. Legg and Raupach (1982) and Ley and Thompson (1983) proposed the use of Equation 79 and justified its validity by analyzing the Navier-Stokes equations and concluding that a gradient of $\overline{u_z^2}$ induces a mean pressure force that must be incorporated, through Equation 79, into the Langevin Equation 10. Similar considerations and results, using the Fokker-Plank equation (which can be seen as the Eulerian equivalent of the Langevin equation), were obtained by Janicke (1981). Other authors (e.g., Wilson et al., 1981, and Sawford, 1985) proposed drift velocity formulations different from Equation 79. This second group of formulations, however, seems less appropriate than Equation 79, since it contains the instantaneous term $u'_z$ which interferes with the definition of $T_L$ and the correct calculation of the autocorrelation.

A different approach for the treatment of convective conditions by Baerentsen and Berkovicz (1984) and Brusasca et al. (1987) uses two Langevin equations for updrafts and downdrafts, respectively. This allows an explicit, more realistic treatment of the known behavior of air parcel velocities in unstable conditions, in which ascent velocities are stronger than descent velocities, but with shorter duration. For example, Yamamoto et al. (1982) measured average ascent velocities $u'_z$ in the range of 0.5 $w_*$ to 0.6 $w_*$, while Briggs (1975) proposed 0.4 $w_*$ as a suitable average descent velocity $u'_z$. Both Baerentsen and Berkovicz (1984) and Brusasca et al. (1987) allow a probabilistic "jumping" of each particle from an updraft to a downdraft, and vice versa, with probabilities that depend upon the time scales of the two phenomena. These methods implicitly assume that updrafts and downdrafts are not included in the average $\overline{u_z}$ terms of Equation 13 and do not affect the $\sigma_{u'_z}$ terms (otherwise, updrafts and downdrafts terms would be included twice in the calculations).

The most appealing approach for the treatment of convective conditions is the incorporation of an appropriate term $\overline{(d\mu)^3}$ into the Langevin equation, instead of simply using Equation 78. This incorporation has been developed by Thompson (1984) and van Dop et al. (1985) and successfully tested by de Baas et al. (1986). They derived the following expressions for the random forcing function $d\mu$ (instead of Equations 76 through 78)

$$\overline{d\mu} = \Delta t\ \left[\partial\overline{u_z^2(z)}/\partial z\right] \tag{80}$$

$$\overline{(d\mu)^2} = \Delta t\ \left[2\ \overline{u_z^2(z)}/T_L + \partial\overline{u_z^3(z)}/\partial z\right] \tag{81}$$

$$\overline{(d\mu)^3} = \Delta t\ \left[3\ \overline{u_z^3(z)}/T_L + \partial\overline{u_z^4(z)}/\partial z - 3\overline{u_z^2(z)}\ \partial\overline{u_z^2(z)}/\partial z\right] \tag{82}$$

These equations were obtained by Thompson (1984) by imposing the conditions that, for large times, the distribution of particles in the phase space possesses the same distribution as the air.

The use of Equations 80 through 82 requires the generation of non-Gaussian terms $d\mu$ from a skewed distribution function $P(d\mu)$. Baerentsen and Berkovicz (1984) and de Baas et al. (1986) calculate $P(d\mu)$ by choosing $d\mu$ from two Gaussian distributions $P_1 = N(m_1, \sigma_1)$ and $P_2 = N(m_2, \sigma_2)$ with a chance $q$ and $(1 - q)$, respectively. This allows the derivation of the relationships

$$q\, m_1 + (1 - q)\, m_2 = \overline{d\mu} \tag{83}$$

$$q\, (m_1^2 + \sigma_1^2) + (1 - q)\, (m_2^2 + \sigma_2^2) = \overline{(d\mu)^2} \tag{84}$$

and

$$q\, (m_1^3 + 3\, m_1\, \sigma_1^2) + (1 - q)\, (m_2^3 + 3\, m_2\, \sigma_2^2) = \overline{(d\mu)^3} \tag{85}$$

which can be used with the simplifying assumptions of $m_1^2 = \sigma_1^2$ and $m_2^2 = \sigma_2^2$, since the above equations have two degrees of freedom.

The Langevin Equation 10 is approximated by de Baas et al. (1986) in a finite different form, using the explicit, fast and unconditionally stable scheme

$$\begin{aligned} w(t + \Delta t) &= w(t)\, (1 - 0.5\, \Delta t/T_L)(1 + 0.5\, \Delta t/T_L)^{-1} \\ &+ d\mu\, (1 + 0.5\, \Delta t/T_L)^{-1} \end{aligned} \tag{86}$$

$$z(t + \Delta t) = z(t) + 0.5\, \Delta t\, [w(t + \Delta t) + w(t)] \tag{87}$$

where $z$ is the altitude of the particle, $w$ is the Lagrangian vertical velocity of the particle, and the random terms $d\mu$ are computed using Equations 80 through 82 and numerically generated from the two Gaussian distributions $P_1$ and $P_2$, according to Equations 83 through 85.

Further assumptions are made by de Baas et al. (1986) to simulate convective conditions. Using mixed layer scaling they assume the profile of the second moment to be

$$u_z^2(z)/w_*^2 = 1.54\, (z/z_i)^{2/3} \exp\, (-2\, z/z_i) \tag{88}$$

for $z > 0.0025\, z_i$, and

$$\overline{u_z^2} = 0.028\, w_*^2 \tag{89}$$

for $z \leq 0.0025\, z_i$. Also, they assume the following profile for the third moment

$$\overline{u_z^3(z)}/w_*^3 = 1.4\, (z/z_i) \exp\, (-2.5\, z/z_i) \tag{90}$$

and a constant $T_L$

$$T_L = c\, z_i/w_* \tag{91}$$

with $c = 1$, instead of the common assumption of c = 0.24 - 0.55 (Hanna, 1981), which requires some slight adjustments of the profile of $\overline{u_z^3}(z)$ at the top of the PBL, to satisfy the requirement that $\overline{(d\mu)^2} > 0$ at all heights. Finally, they assume the

fourth moment to be

$$\overline{u_z^4(z)} = \alpha \left(\overline{u_z^2(z)}\right)^2 \tag{92}$$

with $\alpha = 3$, since no measurements of the fourth moment are available. Simulations are performed by generating particles at a source height $z = z_s$ with initial (i.e., t = 0) Lagrangian velocities $w$ that obey the relationships

$$\overline{w} = 0 \tag{93}$$

$$\overline{w^2} = \overline{u_z^2(z_s)} \tag{94}$$

and

$$\overline{w^3} = \overline{u_z^3(z_s)} \tag{95}$$

Particles are also reflected at the top ($z = z_i$) and the bottom of the computational domain. All the above assumptions provide particle simulation results that agree well with water tank experiments by Willis and Deardorff (1978, 1981) (see Figure 11.2), wind tunnel experiments of Poreh and Cermak (1984), and field experiments by Briggs (1983).

As noted above, the scheme of Equations 80 through 82 requires the generation of non-Gaussian terms $d\mu$, a generation that asks for some slight adjustments to force the variance of the random numbers to be positive. Alternatively, the terms $d\mu$ could remain Gaussian and the $(-w/T_L)dt$ term in Equation 10 could be modified instead, e.g., by making it nonlinear (Thompson, 1987). This alternative approach avoids the problem met by de Baas et al. (1986) of satisfying the requirement that $\overline{(d\mu)^2} > 0$ at all heights and seems, at least theoretically, a more satisfactory development (Sawford, personal communication).

Schemes such as the one of Equations 80 through 82 for the solution of Equation 10 represent an improvement of the Langevin equation to simulate convective diffusion. However, they complicate the treatment of the cross-correlation terms. These terms, in the section above dealing with the introduction of the cross-correlations, were discussed under the implicit assumption of using the Langevin Equation 10 with the conditions of Equations 76 through 78, which make it equivalent to Equation 15. A simple modification is required to maintain the advantages of both approaches (i.e., the improved Langevin equations for convective simulations and the cross-correlation terms). In fact, either set of Equations 36 through 38 or 54 through 56 can be rewritten in the sequence $u_z', u_c', u_a'$ (Equations 36 through 38) or $u_z', u_y', u_x'$ (Equations 54 through 56), thus allowing the first equation of either scheme, which does not contain any cross-correlation term, to represent the vertical velocities. By doing so, any complex Langevin equation scheme can be used for $u_z'$ while still maintaining all the cross-correlation terms. (Naturally, however, if equations are rewritten in a different sequence, the derivation of the parameters will change accordingly, even though the general form of the solutions will remain the same.)

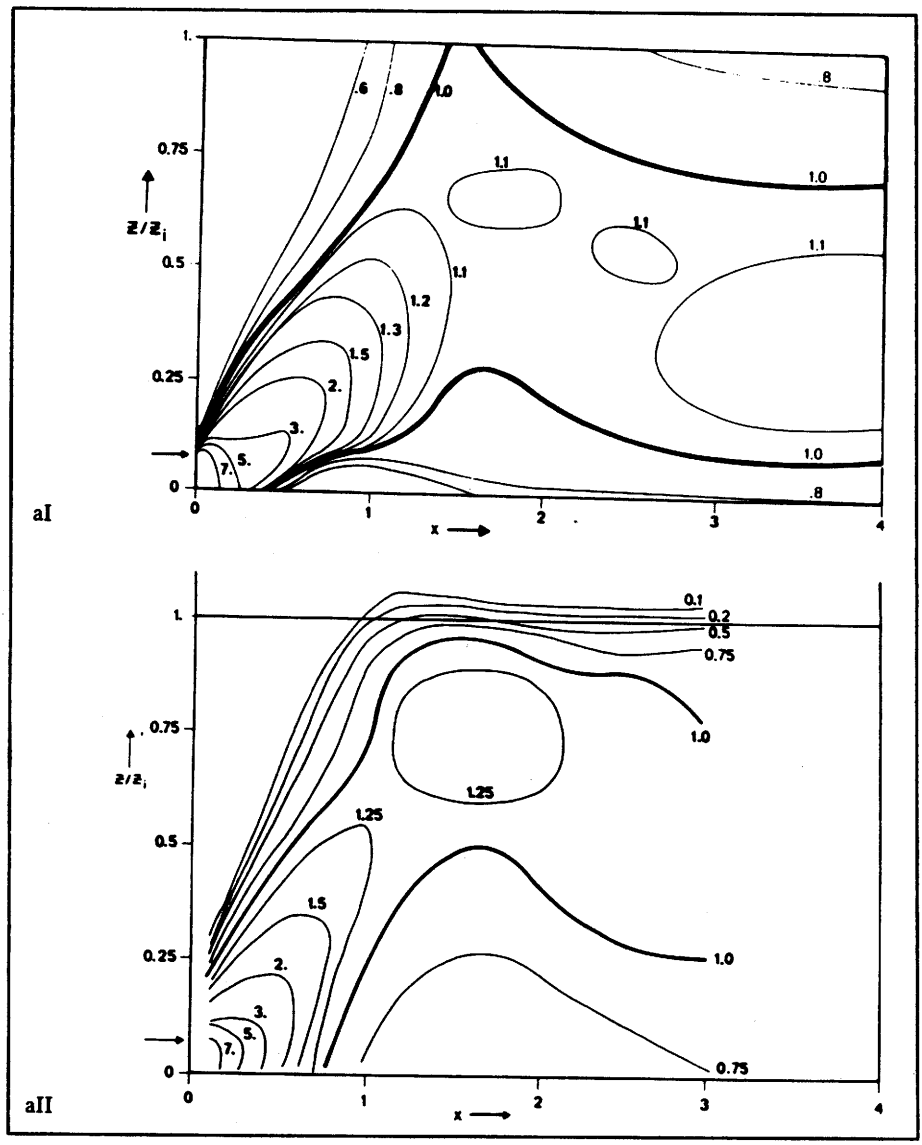



Figure 11.2: Dimensionless concentration contours in the vertical x,z plane. The different plots present the results of the Langevin model (I) and the cross-wind-integrated measurements of Willis and Deardorff (II) for the source heights: (a) $z_s/z_i = 0.067$; (b) $z_s/z_i = 0.24$; (c) $z_s/z_i = 0.49$. Source height is indicated by arrow on ordinate (from de Baas, et al., 1986). [Reprinted with permission from the Royal Meteorological Society.]

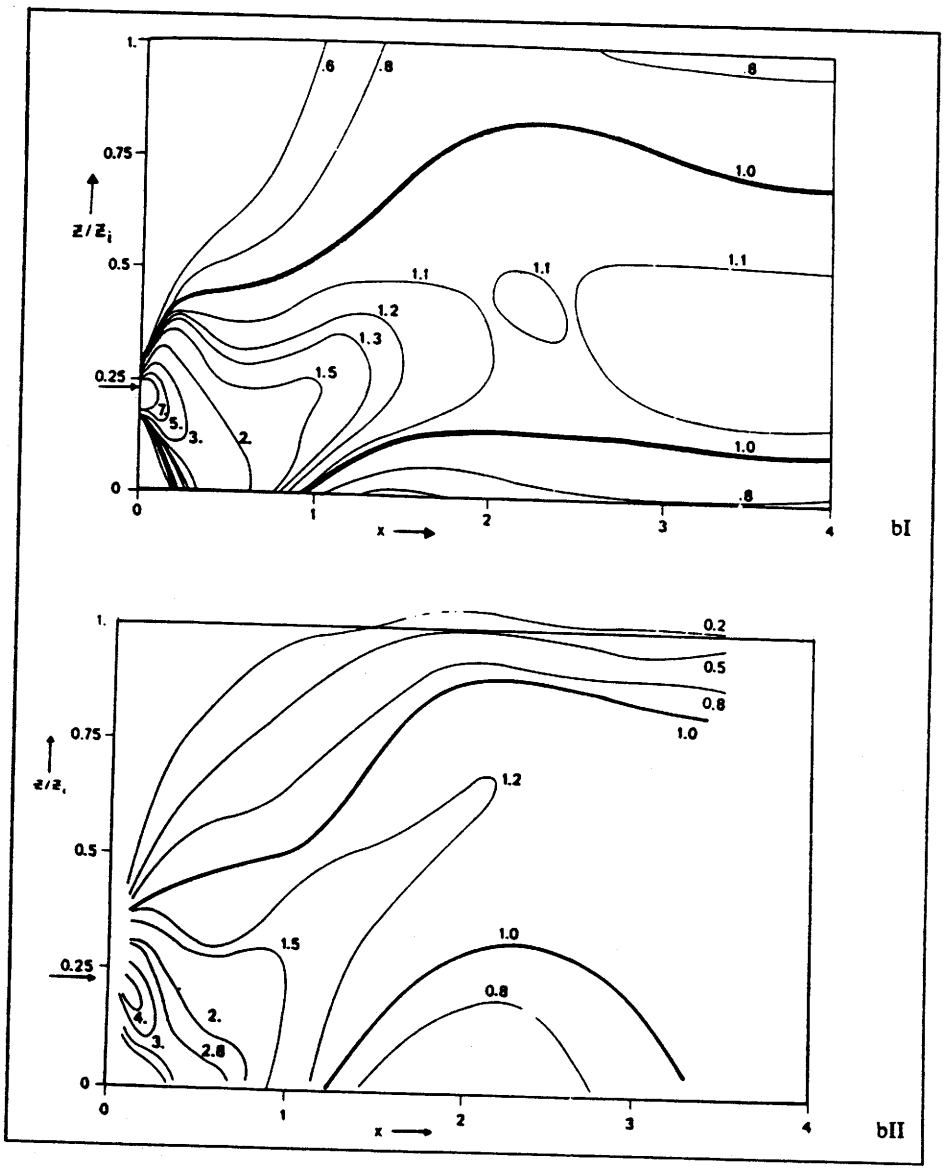



Figure 11.2 (continued)

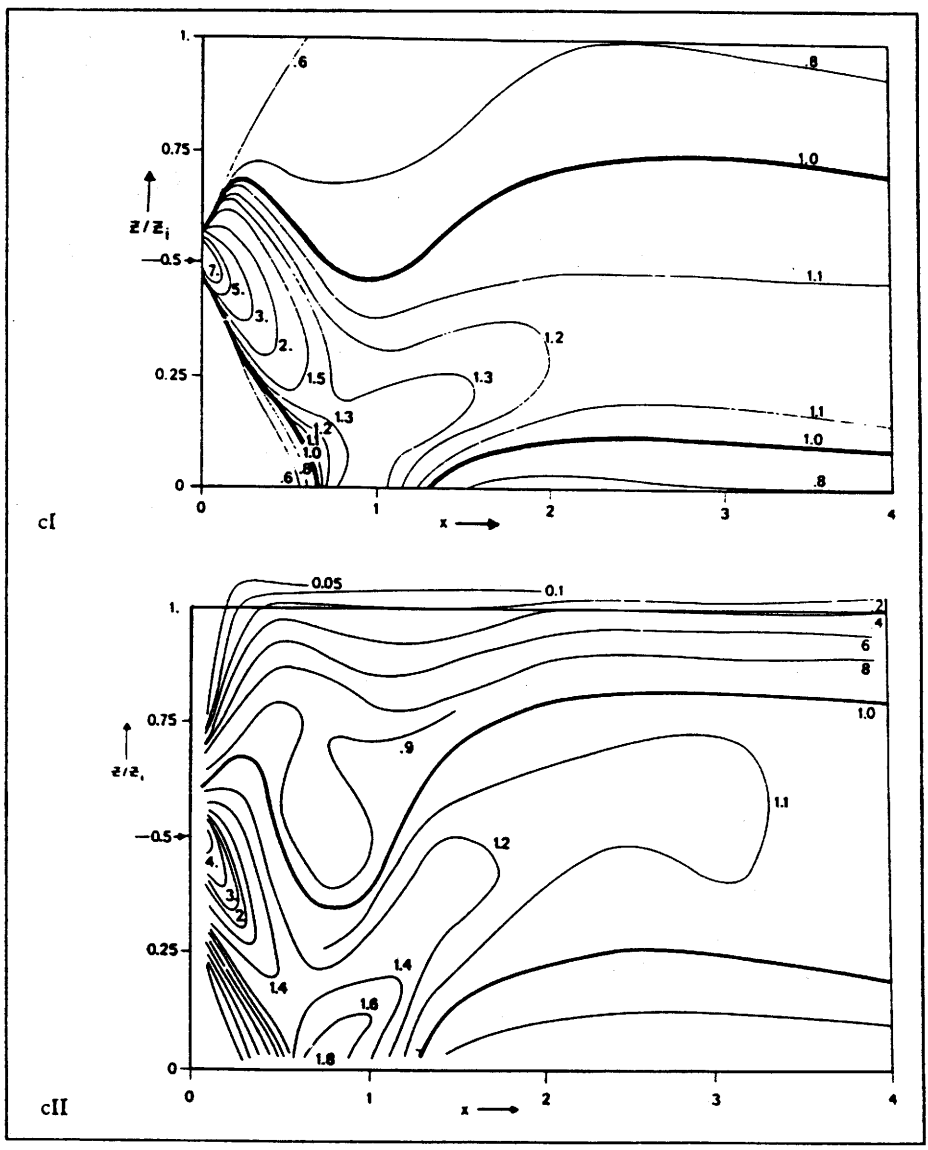



Figure 11.2 (continued)

## Concentration Calculations Using Particle Models

Particle models are a set of algorithms for the generation of realistic trajectories of imaginary, fictitious particles that simulate atmospheric motion. Each particle can be tagged by a mass of pollutant that can be either constant or time-varying to allow loss of mass due to ground deposition and chemical decay phenomena. Therefore, the spatial distribution of particle mass in the computational domain allows the calculation of a three-dimensional mass concentration field, under certain computational assumptions.

For example, the most straightforward assumption is the superimposition, in the computational domain, of a three-dimensional concentration grid. The concentrations are then computed simply by counting the number of particles in each grid cell and accumulating their masses. If concentrations need to be computed only at "receptor" points (e.g., at a ground level), receptor cells can be defined around these points and particles counted only inside those cells. A rigorous concentration calculation, however, should not just add up the particle mass in a given cell at a given time. In fact, the contribution of each particle mass should be weighted by the total time the particle spent inside the cell during each time step (Lamb, 1979b).

One of the great advantages of Monte-Carlo particle models, however, is their "grid-free" characteristics, which allow higher time and space resolution than other simulation techniques. In this respect, grid-free concentration calculations (i.e., calculations that do not require the definition of cells) to maintain this important feature of the model are appealing. "Kernel" methods (Gingold and Monaghan, 1982) allow grid-free concentration calculations that are smooth and efficient. Kernel methods for air quality modeling are discussed by Lorimer (1986). A general form of kernel density estimator is

$$c(\mathbf{r},t) = \frac{A(\mathbf{r})}{l^3} \sum_{i=1}^{n} m_i \, W(\mathbf{r}_i - \mathbf{r}, l) \tag{96}$$

where $c$ is the concentration in $\mathbf{r}$ at time $t$; $l$ is a time-dependent resolution bandwidth (or smoothing length); $m_i$ is the pollutant mass of each particle $i$; $W$ is the smoothing kernel, which is a function of $l$ and the distance $\mathbf{r}_i - \mathbf{r}$ of each particle $i$ from the receptor point. $A(\mathbf{r})$ is a correction term for concentration computations at locations $\mathbf{r}$ close to the boundary of the computational domain $D$, where

$$A(\mathbf{r}) = \frac{l^3}{\int_D W(\mathbf{r}' - \mathbf{r}, l) d\mathbf{r}'} \tag{97}$$

which, for an infinite domain $D$, reduces to $A(\mathbf{r}) = 1$ everywhere.

Several kernel functions $W$ are available in the literature. The most common is the Gaussian kernel, in which

$$\mathbf{d}_i = \mathbf{r}_i - \mathbf{r} \tag{98}$$

and

$$W(\mathbf{d}_i, l) = \frac{1}{(2\pi)^{3/2}} \exp\left(-\frac{1}{2}\frac{|\mathbf{d}_i|^2}{l^2}\right) \tag{99}$$

The choice of $l$ is critical. This term should not be kept constant, as is done in many applications, but should change in relation to a natural length scale. In general, $l$ should be particle dependent and should be related to the mean interparticle separation around $\mathbf{r}$. Only particles with $|\mathbf{d}_i| < l$ give substantial contribution to $c$ (Lorimer, 1986). If $l$ is too small, the spatial distribution of the concentration $c$ is "jagged" with a series of local maxima at each $\mathbf{r}_i$; if $l$ is too large, $c$ becomes overly smooth.

Using a Gaussian kernel, the particle model becomes very similar to the Gaussian puff models. It is important to note, however, that for a puff model, $l$ is substituted by $\sigma_x, \sigma_y$, and $\sigma_z$ (i.e., the standard deviations of the spatial concentration distributions of each puff), and these values are related to the physics of atmospheric diffusion, while, in the kernel method, $l$ should be related only to the density of the particles around $\mathbf{r}$. However, Yamada and Bunker (1988) use a kernel density estimator, for their RAPTAD particle model, which makes it, in reality, a puff model, in which each particle $i$ is associated with time-growing $\sigma_{xi}, \sigma_{yi}$ and $\sigma_{zi}$ values that are estimated based on the homogeneous diffusion theory by Taylor (1921).

### Particle Simulation of Buoyancy Phenomena

One of the main advantages of Monte-Carlo particle models versus other particle methods (such as the particle-in-cell method described above in the section on deterministic calculation of $\mathbf{u}'$) is the cost-effective ability to move each particle independently from the others. A correct treatment of buoyancy phenomena, however, requires the capability of incorporating, into the particle dynamics, extra velocity terms that simulate plume rise effects and heavy gas phenomena, both functions of local space properties related to the local concentration. Therefore, the motion of each particle affected by buoyancy phenomena depends upon the particle concentration, i.e., the dynamics of neighboring particles.

A simple, analytical approach to account for plume rise phenomena was illustrated by Zannetti and Al-Madani (1983). More comprehensive approaches have been proposed by Cogan (1985) and Gaffen et al. (1987).

### Chemistry and Deposition

Since each particle can be tagged by its mass, i.e., the mass of pollutant(s) whose dynamics is represented by that particle, linear chemistry and deposition phenomena can be easily accounted for by properly modifying, in a dynamic way, the mass $m_i$ of each particle. For example, Zannetti and Al-Madani (1983) proposed relationships such as

$$m_i(t + \Delta t) = m_i(t) \exp(-\Delta t/T) \tag{100}$$

to account for dry deposition, wet deposition, and linear chemistry transformation, where $T$ is the appropriate time scale of each phenomenon that can vary with time and

space. Alternatively, any deposition can be computed using the deposition velocity concept, which requires the calculation of particle mass concentration in the layers just above the ground and a consequent dynamic reduction of the mass $m_i$ of the particle to account for the ground deposition mass flux.

If nonlinear chemistry is required, for example, to simulate atmospheric photochemistry and ozone production, two possible methods can be used. With the first method, a concentration grid can be superimposed on the domain and, at each time step, concentrations can be computed in each grid cell. Then, an Eulerian photochemical model can be used to calculate the effects of chemical reactions from all sources at each time step. If the chemical reactions of a single plume must be simulated, a second method can be used, in which each particle can be considered an expanding box representing a section of the plume that grows with time and entrains background air and, possibly, other emissions along its trajectory. Then, the photochemical module of a Lagrangian box model can be used to calculate the effects of chemical reactions inside each box at each time step. It is clear, however, that nonlinear chemistry by particle models is complex and extremely demanding of computational resources. A proper consideration of nontrivial chemical reactions may well be impossible in a Lagrangian framework. In any case, it would at least require a two-particle approach, since, for a second-order reaction, the reaction rate depends on the reactant covariance, which is a second-order concentration statistics (Sawford, personal communication).

Particle models can also be used to simulate the behavior of actual particulate matter in the atmosphere, whose dynamics is affected by atmospheric turbulence and gravitational settling velocity $V_G$, which depends upon the diameter $d_p$ of the aerosol particle. For example, Figure 11.3 shows the velocities with which spherical particles for different particle diameters and densities fall. When these particles hit the ground, permanent deposition can be assumed or, alternatively, probabilistic methods can be used to simulate particle deposition and, when deposited, the possibility of particle resuspension. For example, Zannetti and Al-Madani (1983) use expressions such as

$$q = 1 - \exp(-\Delta t/T) \tag{101}$$

to calculate both the deposition probability of a particle that reaches the ground, and the resuspension probability, where $T$ is an appropriate time scale that depends upon meteorology and surface characteristics.

## Advantages and Disadvantages of Particle Models

Dispersion simulation by Lagrangian particles has been called "natural" modeling. These models do not need the input of artificial stability classes, empirical sigma curves, or diffusion coefficients that are practically impossible to measure. Instead, diffusion characteristics are simulated by attributing a certain degree of "fluctuation" to each particle, using, for example, the computer's capability to generate semirandom numbers.

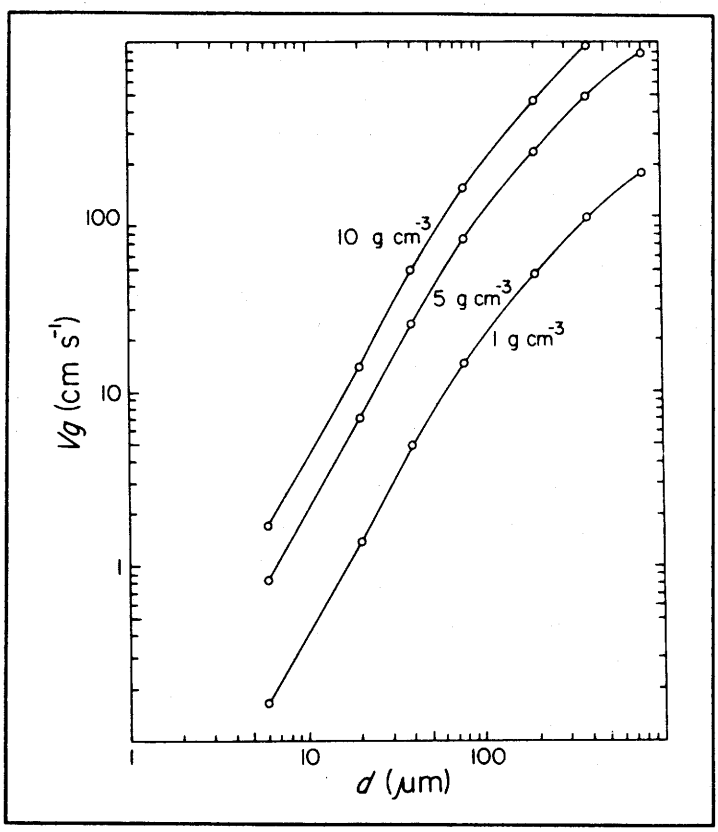



Figure 11.3: Fall velocity of spherical particles as a function of particle diameter and density. (Adapted from Hanna et al., (1982), as presented by Stern et al., (1984).) [Reprinted with permission from Academic Press.]

The basic advantages of this approach (e.g., see Lamb et al., 1979a; Lange, 1978) are:

- Compared with grid models, this method avoids the artificial initial diffusion of point source in the corresponding cell and the advection numerical errors.

- This method is practically free of restricting physical assumptions, since all uncertainties are combined into the correct determination of pseudovelocities.

- Each particle can be tagged with its coordinates, source indicator, mass, activity, species and size, allowing computation of wet and dry deposition, decay, and

particle size distribution.

- The meteorological input required can be directly inferred from measured data. The primary information needed is (Lamb et al., 1979a) the variance of wind velocity fluctuations and the Lagrangian autocorrelation function, which can be estimated from Eulerian measurements.

Potentially, this method is superior in both numerical accuracy and physical representativeness. However, much research is still needed to extract, from the scarce available meteorological measurements and our limited theoretical understanding of turbulence processes, the meteorological input required to run this model (i.e., the pseudovelocities to move each particle at each time step).

## References


1. Baerentsen, J.H., and R. Berkovicz (1984): Monte Carlo simulation of plume dispersion in the convective boundary layer. *Atmos. Environ.,* **18**:701-712.
2. Briggs, G.A. (1975): Plume rise predictions, in *Lectures on Air Pollution and Environmental Impact Analyses.* edited by D.A. Augen, Boston: American Meteorological Society.
3. Briggs, G.A. (1983): Diffusion modeling with convective scaling. AMS specialty conference on Air Quality Modeling of the Urban Boundary Layer, Baltimore.
4. Brusasca, G., G. Tinarelli, D. Anfossi, P. Zannetti (1987): Particle modeling simulation of atmospheric dispersion using the MC-LAGPAR package. *Envir. Software,* **2**:151-158.
5. Cogan, J.L. (1985): Monte Carlo simulations of buoyancy dispersion. *Atmos. Environ.,* **19**:867-878.
6. Davis, R.E. (1982): On relating Eulerian and Lagrangian velocity statistics: Single particles in homogeneous flows. *J. Fluid Mech.,* **114**:1-26.
7. Deardorff, J.W. (1974): Three-dimensional numerical study of the height and mean structure of a heated boundary layer. *Boundary-Layer Meteor.,* **7**:81-106.
8. de Baas, A.F., H. van Dop, and F.T. Nieuwstadt (1986): An application of the Langevin equation for inhomogeneous conditions to dispersion in a convection boundary layer. *Quarterly J. Roy. Meteor. Soc.,* **112**:165-180.
9. Drivas, P.J., M.W. Chan, and L.G. Wayne (1977): Validation of an improved photochemical air quality simulation model. *Proceedings,* AMS joint conference on Applications of Air Pollution Meteorology. Salt Lake City, Utah. November.
10. Gifford, F.A. (1982): Horizontal diffusion in the atmosphere: Lagrangian-dynamical theory. *Atmos. Environ.,* **16**:505-512.

11. Durbin, P.A. (1980): A stochastic model of two-particle dispersion and concentration fluctuations in homogeneous turbulence. *J. Fluid Mech.,* **100**:279-302.

12. Gaffen, D.J., C. Benocci, and D. Olivari (1987): Numerical modeling of buoyancy dominated dispersal using a Lagrangian approach. *Atmos. Environ.,* **21**:1285-1293.

13. Gingold, R.A., and J.J. Monaghan (1982): Kernel estimates as a basis for general particle methods in hydrodynamics. *J. Computational Phys.,* **46**:429-453.

14. Hall, C.D. (1975): The simulation of particle motion in the atmosphere by a numerical random-walk model. *Quarterly J. Roy. Meteor. Soc.,* **101**:235-244.

15. Hanna, S.R. (1979): Some statistics of Lagrangian and Eulerian wind fluctuations. *J. Appl. Meteor.,* **18**:518-525.

16. Hanna, S.R., G.A. Briggs, and R.P. Hosker, Jr. (1982): *Handbook on Atmospheric Diffusion,* edited by J.S. Smith, Washington, D.C.: Technical Information Center, U.S. Department of Energy.

17. Hanna, S.R. (1981): Lagrangian and Eulerian time scale relations in the daytime boundary layer. *J. Appl. Meteor.,* **20**:242-249.

18. Hockney, R.W., and J.W. Eastwood (1981): *Computer Simulation Using Particles.* New York: McGraw-Hill, Inc.

19. Janicke, L. (1981): Particle simulation of inhomogeneous turbulent diffusion. *Proceedings,* 12th International Technical Meeting of the NATO CCMS. Palo Alto. California: Plenum Press.

20. Lamb, R.G. (1978): A numerical simulation of dispersion from an elevated point source in the convective planetary boundary layer. *Atmos. Environ.,* **12**:1297-1304.

21. Lamb, R.G., H. Hogo, and L.E. Reid (1979a): A Lagrangian Monte Carlo model of air pollutant transport, diffusion and removal processes. 4th AMS Symposium on Turbulence, Diffusion and Air Pollution. Reno, Nevada. January.

22. Lamb, R.G., H. Hogo, and L.E. Reid (1979b): A Lagrangian approach to modeling air pollutant dispersion: Development and testing in the vicinity of a roadway. EPA Research Report EPA-600/4-79-023.

23. Lamb, R.G. (1981): A scheme for simulating particle pair motions in turbulent fluid. *J. Computational Phys.,* **39**:329-346.

24. Lange, R. (1978): ADPIC – A three-dimensional particle-in-cell model for the dispersal of atmospheric pollutants and its comparison to regional tracer studies. *J. Appl. Meteor.,* **17**:320.

25. Legg, B.J., and M.R. Raupach (1982): Markov chain simulation of particle dispersion in inhomogeneous flows: The mean drift velocity induced by a gradient in Eulerian velocity variance. *Boundary-Layer Meteor.,* **24**:3-13.

26. Legg, B.J. (1983): Turbulent dispersion from an elevated line source: Markov chain simulations of concentration and flux profiles. *Quarterly J. Roy. Meteor. Soc.,* **109**:645-660.

27. Lee, J.T., G.L. Stone, R.E. Lawson, Jr., and M. Shipman (1985): Monte Carlo simulation of two-particle relative diffusion using Eulerian Statistics. Los Alamos National Laboratory Document LA-UR-85-2008, Los Alamos, New Mexico.

28. Ley, A.J. (1982): A random walk simulation of two-dimensional turbulent diffusion in the neutral surface layer. *Atmos. Environ.,* **16**:2799-2808.

29. Ley, A.J., and D.J. Thompson (1983): A random walk model of dispersion in the diabatic surface layer. *Quarterly J. Roy. Meteor. Soc.,* **109**:847-880.

30. Lorimer, G.S. (1986): The kernel method for air quality modelling; I. Mathematical foundation. *Atmos. Environ.,* **20**:1447-1452.

31. Lurmann, F.W., D.A. Gooden, and H.M. Collins, Eds. (1985): User's guide to the PLMSTAR air quality simulation model. Environmental Research & Technology, Inc. Document M-2206-100, Newbury Park, California.

32. Martinez, J.R., R.A. Nordsieck, and M.A. Hirschberg (1973): User's guide to diffusion/kinetics (DIFKIN) code. General Research Corporation Final Report CR-2-273/1, prepared for the U.S. Environmental Protection Agency.

33. Nieuwstadt, F.T., and H. van Dop, Eds. (1982): *Atmospheric Turbulence and Air Pollution Modeling.* Dordrecht, Holland: D. Reidel.

34. Novikov, E.A. (1969): *Appl. Math. Mech.,* **33**:887.

35. Novikov, E.A. (1986): The Lagrangian-Eulerian probability relations and the random force method for nonhomogeneous turbulence. *Phys. of Fluids,* **29**:3907-3909.

36. Poreh, M., and J.E. Cermak (1984): Wind tunnel simulation of diffusion in a convective boundary layer. *Proceedings,* 29th Oholo Conference on Boundary Layer Structure-Modelling and Application to Air Pollution and Wind Energy. Israel. March.

37. Reid, J.D. (1979): Markov chain simulations of vertical dispersion in the neutral surface layer for surface and elevated releases. *Boundary-Layer Meteor.,* **16**:3-22.

38. Rodriguez, D.J., G.D. Greenly, P.M. Gresho, R. Lange, B.S. Lawver, L.A. Lawson, and H. Walker (1982): User's guide to the MATHEW/ADPIC models. Lawrence Livermore National Laboratory Document UASG 82-16, University of California Atmospheric and Geophysical Sciences Division, Livermore, California.

39. Sawford, B.L. (1983): The effect of Gaussian particle-pair distribution functions in the statistical theory of concentration fluctuations in homogeneous turbulence. *Quarterly J. Roy. Meteor. Soc.,* **109**:339-354.

40. Sawford, B.L. (1984): The basis for, and some limitations of, the Langevin equation in atmospheric relative dispersion modelling. *Atmos. Environ.,* **11**:2405-2411.

41. Sawford, B.L. (1985): Lagrangian simulation of concentration mean and fluctuation fields. *J. Clim. Appl. Meteor.,* **24**:1152-1166.

42. Seinfeld, J.H. (1975): *Air Pollution – Physical and Chemical Fundamentals.* New York: McGraw-Hill.

43. Smith, F.B. (1968): Conditioned particle motion in a homogeneous turbulent field. *Atmos. Environ.,* **2**:491-508.

44. Stern, A.C., R.W. Boubel, D.B. Turner, and D.L. Fox (1984): *Fundamentals of Air Pollution.* Orlando, Florida: Academic Press.

45. Taylor, G.I. (1921): Diffusion by continuous movements. *Proceedings,* London Math. Soc., **20**:196-211.

46. Tran, K. (1981): User's guide for photochemical trajectory model TRACE. Applied Modeling, Inc., Report 81/003. California.

47. Thompson D.J. (1984): Random walk modelling of diffusion in inhomogeneous turbulence. *Quarterly J. Roy. Meteor. Soc.,* **110**:1107-1120.

48. Thompson, D.J. (1987): *J. Fluid Mech.,* **180**:529.

49. van Dop, H., F.T. Nieuwstadt, and J.C. Hunt (1985): Random walk models for particle displacements in inhomogeneous unsteady turbulent flows. *Phys. of Fluids,* **28**:1639-1653.

50. Willis, G.E., and J.W. Deardorff (1978): A laboratory study of dispersion from an elevated source within a modeled convective planetary boundary layer. *Atmos. Environ.,* **12**:1305-1311.

51. Willis, G.E., and J.W. Deardorff (1981): A laboratory study of dispersion from a source in the middle of the convective mixed layer. *Atmos. Environ.,* **15**:109-117.

52. Wilson, J.D., G.W. Thurtell, and G.E. Kidd (1981): Numerical simulation of particle trajectories in inhomogeneous turbulence; III. Comparison of predictions with experimental data for the atmospheric surface layer. *Boundary-Layer Metor.* **12**:423-441.

53. Yamada, T., and S.S. Bunker (1988): Development of a nested grid, second moment turbulence closure model and application to the 1982 ASCOT Brush Creek data simulation. *J. Appl. Meteor.,* **27**:562-578.

54. Yamamoto, S., M. Gamo, and Y. Osayuki (1982): Observational study of the fine structure of the convective atmospheric boundary layer. *J. Meteor. Soc. of Japan,* **60**:882-888.

55. Zannetti, P. (1981): Some aspects of Monte Carlo type modeling of atmospheric turbulent diffusion. *Preprints,* Seventh AMS conference on Probability and Statistics in Atmospheric Sciences. Monterey, California. November, 1978.

56. Zannetti, P., and N. Al-Madani (1983): Simulation of transformation, buoyancy and removal processes by Lagrangian particle methods. *Proceedings,* 14th international technical meeting on Air Pollution Modeling and its Application. Copenhagen, Denmark. September.

57. Zannetti, P. (1984): New Monte Carlo scheme for simulating Lagrangian particle diffusion with wind shear effects. *Appl. Math. Modelling,* **8**:188-192.

58. Zannetti P. (1986): Monte-Carlo simulation of auto- and cross-correlated turbulent velocity fluctuations (MC-LAGPAR II Model). *Environ. Software,* **1**:26-30.

59. Zannetti P. (1990): *Air Pollution Modeling.* Computational Mechanics Publications, Southampton and Van Nostrand Reinhold, New York.