History of Lagrangian Stochastic Models for Turbulent Dispersion

D. J. Thomson

Met Office, Exeter, UK

J. D. Wilson

Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Alberta, Canada

This article briefly summarizes the historical evolution of the modern Lagrangian stochastic (LS) class of models for the calculation of fluid element (or particle) paths in turbulence. The fundamental advantages of a "first-order" LS model relative to alternative descriptions of turbulent dispersion are (1) its ability to correctly describe the concentration field even in the nondiffusive "near field" of sources and (2) its ability to rationally incorporate all available statistical information on the velocity field, even in the case that the latter is nonstationary and inhomogeneous in all directions. There are also advantages of convenience: for example, being grid free, LS models are easy to implement; and because particle paths are computed independently, they are amenable to easy parallelization. LS models are presently used to treat atmospheric transport and dispersion problems on scales ranging from the intercontinental (for which case typically they are "driven" by motion fields from numerical weather models) down to the scale of the atmospheric surface layer (meters to hundreds of meters). Papers at the Chapman Conference on Langrangian modeling, from which this chapter was derived, exhibited many interesting applications.

1. INTRODUCTION

The aim of a Lagrangian stochastic (LS) model is to compute an ensemble of random paths of marked fluid elements through a turbulent flow, based on knowledge of velocity statistics. The simplest class of LS model is the random displacement model (RDM, or zeroth-order LS model), which represents a particle trajectory by a sequence of random increments in position. The more sophisticated "generalized Langevin approach" or first-order LS model (which draws ideas from Langevin's 1908 work on Brownian motion) creates the particle path by integrating a sequence of (damped) random increments in *velocity*, such that the particle position **X** and velocity **U** together constitute a Markovian state variable. To be more specific, the general form of the first-order model is

$$\mathrm{d}U_i = a_i\ \mathrm{d}t + b_{ij}\ \mathrm{d}\xi_j, \tag{1}$$

$$\mathrm{d}X_i = U_i\ \mathrm{d}t \tag{2}$$

where t is time, $a_i = a_i(\mathbf{X}, \mathbf{U}, t)$ is the systematic part of the acceleration, and b_{ij} (normally diagonal) is another coefficient scaling the random Gaussian forcing $\mathrm{d}\xi_j$. Equations (1) and (2) can be integrated numerically by replacing the infinitesimal $\mathrm{d}t$ with a finite time step Δt, whose magnitude may vary along the trajectory in proportion to a local turbulence time scale. Heuristic arguments for the validity of equation (1) as an approximation to the Navier-Stokes equations can be made

Lagrangian Modeling of the Atmosphere
Geophysical Monograph Series 200

10.1029/2012GM001238

[e.g., *van Dop et al.*, 1985], and specification of the coefficients a_i, b_{ij} is the selection problem for LS models. Many interesting and useful models are known, applying to diverse regimes of turbulence spanning from the ideal (unbounded homogeneous, isotropic turbulence) through the everyday (stratified atmospheric surface layer in a horizontally homogeneous state; convective boundary layer (CBL)) to the exotic (three-dimensionally inhomogeneous urban flow with buildings resolved).

However, this article is not intended as a thorough review of the science of modern Lagrangian models, which may be sought elsewhere [*Sawford*, 1985; *Thomson*, 1987; *Wilson and Sawford*, 1996; *Rodean*, 1996]. Rather, its intent is to capture some of the broad trends and developments that have brought us to where we now are. We give some flavor of the antecedent models and of the diverse disciplines, backgrounds, motivations, and styles of early contributors; we illustrate the range and impact of contributions driven by intuition and by rigor; and we note a chronological evolution in the specificity of the turbulence regime addressed and in the degree of connection with (or disconnection from) observations of dispersion. We restrict the focus to LS models where the particles are (conceptually) sampled independently from an ensemble of turbulent flows and so move independently (so called one-particle models), and to nonbuoyant and nonreactive ("passive") particles, in flows for which the turbulence prescription is limited to single-point statistics. Meteorological applications are emphasized.

2. EARLY DEVELOPMENT OF THE LAGRANGIAN PERSPECTIVE ON TURBULENT DISPERSION

Taylor [1921] provided an exact Lagrangian solution for the rate of spread of tracer in unbounded, stationary homogeneous turbulence. Let us take the case where particles are independently released into such a flow at $z = 0$ (here, and generally when we consider dispersion in one dimension only, we take the direction to be the vertical axis). For each realization, i.e., for each trajectory, the clock is reset ($t = 0$) upon release. Taylor showed that the rate of increase in time of the ensemble mean spread (as measured by the variance $\overline{z'^2} \equiv \sigma_z^2$ of displacement along the z axis) is given exactly by (Taylor's equation 17)

$$\frac{\mathrm{d}\sigma_z^2}{\mathrm{d}t} = 2\int_0^t \overline{w(t')w(t'+\xi)}\ \mathrm{d}\xi \equiv 2\sigma_w^2 \int_0^t R_{ww}(\xi)\ \mathrm{d}\xi, \qquad (3)$$

where $R_{ww}(\xi)$ is the Lagrangian velocity autocorrelation function, first introduced by Taylor, and σ_w^2 the velocity variance. (In terms of the eddy diffusion paradigm, the left-hand side of equation (3) is twice the eddy diffusivity). Integrating this result gives

$$\begin{aligned} \overline{z'^2} &= 2\sigma_w^2 \int_0^t \int_0^{t'} R_{ww}(\xi)\ \mathrm{d}\xi\ \mathrm{d}t' \\ &= 2\sigma_w^2 \int_0^t (t-\xi)\, R_{ww}(\xi)\ \mathrm{d}\xi. \end{aligned} \qquad (4)$$

Let $\tau = \int_0^\infty R_{ww}(\xi)\ \mathrm{d}\xi$ be the Lagrangian integral time scale. Equation (4) has asymptotic "near-field" and "far-field" limits

$$\overline{z'^2} = \begin{cases} \sigma_w^2\, t^2, & t \ll \tau \\ 2\sigma_w^2\, \tau t, & t \gg \tau \end{cases} \qquad (5)$$

corresponding to a nondiffusive regime of "memory-dominated" spread during which the release velocity is preserved ($t \ll \tau$), and a long time regime in which the turbulent convection of tracer may legitimately be *represented* as "diffusion," with effective far-field eddy diffusivity $K_\infty = \sigma_w^2 \tau$. Taylor's result proves, then, that the classic "eddy diffusion" paradigm for the evolution of the particle concentration $p = p(z,t)$ in this 1-D (z-) space, namely,

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial z}\left(-K\frac{\partial p}{\partial z}\right) = K\frac{\partial^2 p}{\partial z^2}, \qquad (6)$$

(constant eddy diffusivity K, Fickian diffusion equation) is insufficiently general. Being equivalent to the eddy diffusion treatment (not shown, but see *Monin and Yaglom* [1977, section 10.3] and *Boughton et al.* [1987]), a zeroth-order LS model (i.e., random displacement model or "random walk in position") cannot represent the near field. Conversely, the first-order LS model gives the correct small and large time behavior and, indeed, agrees exactly with Taylor's result at intermediate times in the case where R_{ww} decays exponentially. This represents the *fundamental* advantage of the (first-order) LS models over simpler models, though in practice, this capability is decisive only for a restricted range of problems involving the near field of sources (we expand on this in section 5).

Ever since its derivation, Taylor's result has served to guide turbulent dispersion modeling, and with suitable restriction as to the domain occupied by a puff or plume of dispersing tracer, it can be used in an approximate way for sources in real flows. However, its adequate extension to inhomogeneous and nonstationary turbulence, in the form of today's Lagrangian stochastic models, occurred only after newfound access to computers spurred heuristic experiments in the numerical simulation of particle trajectories and after these experiments, in turn, stimulated the development

during the 1980s of guidance in formulating the models for general flows (see section 4). Of course, there were many ingenious and illuminating developments, both theoretical and experimental, in the period between Taylor's work and the advent of (accessible) computers: for example, *Batchelor's* [1949] reexpression of Taylor's result in terms of a weighted integral

$$\overline{z'^2} = \sigma_w^2 \, t^2 \int_0^\infty S_w^{(L)}(f) \frac{\sin^2(\pi f t)}{(\pi f t)^2} \, \mathrm{d}f \tag{7}$$

of the Lagrangian velocity spectrum (f representing frequency). The transformation from equation (4) to equation (7) is straightforward (the Lagrangian spectrum and the Lagrangian autocorrelation function constituting a Fourier transform pair). The low-pass spectral filter $\sin^2(\pi f t)/(\pi f t)^2$ expresses the (intuitive) fact that for small travel times t, all eddies contribute to spread, while, with increasing t, increasingly only "slow" (small f) eddies dominate.

As already noted, and as testified by its prominence in textbooks [e.g. *Sutton*, 1953; *Pasquill and Smith*, 1983], Taylor's Lagrangian paradigm proved preeminent, either explicitly or indirectly, in subsequent efforts to deepen the theoretical framework and provide useable real-world dispersion models, one example of the latter being *Sutton's* [1953, equation 8.31] model for dispersion from a continuous ground-level point source in the atmospheric surface layer [see also *Monin and Yaglom*, 1977, section 10.5]. For several decades one of the influences of Taylor's work could be found in theoretical papers that attempted to interrelate Eulerian and Lagrangian statistics in idealized flow regimes, for "the relation between Lagrangian and Eulerian correlation functions is basic to the understanding of turbulent diffusion" [*Weinstock*, 1976]. Summing up a symposium at Oxford University, *Sutton* [1959, p. 438] stated:

> It is most appropriate that, with Sir Geoffrey Taylor in our midst, so much of the work has been founded on his famous paper of 1921 on the random walk. I have given up counting the number of times that celebrated equation connecting the Lagrangian correlation coefficient with the standard deviation of the particles has been written out on the blackboard.

Many influential scientists (including S. Corrsin, J. L. Lumley, R. H. Kraichnan, J. R. Philip, and P. G. Saffman) participated in this prolonged effort to relate Lagrangian to Eulerian statistical properties, and following a widely cited contribution by *Hay and Pasquill* [1959] delivered at the above-mentioned symposium, namely, a practicable method for short-range air pollution calculations involving the ratio β of Lagrangian to Eulerian integral time scales, a particular focus was the provision of theoretical values for that ratio [e.g., *Corrsin*, 1963; *Philip*, 1967; *Smith*, 1968]. It is interesting to remark that a modern LS model, if it respects the well-mixed condition [*Thomson*, 1987] for a given regime of flow having a specific (postulated) Eulerian velocity pdf, must "produce" the long sought for Lagrangian statistics, though in numerical rather than analytic form, and with the time scales determined using turbulence phenomenological relations (e.g., a parameterization of the energy dissipation rate) and hinging on the specified value of a dimensionless constant "C_0" (that we define and discuss later). Thus, the LS model can be said to "solve" the problem of relating Lagrangian to Eulerian statistics, albeit in a restricted sense: for the LS approach achieves this outcome by virtue of adopting the (plausible, but nonrigorous) Markovian framework that (possibly) these scientists might have considered a too sweeping simplification.

Taylor's result may easily be obtained by an analysis of *Langevin's* [1908] equation, which had been developed to describe Brownian motion and represents "the first example of a stochastic differential equation" [*Lemons*, 2002; *Gardiner*, 2004]; indeed, as a precursor to his main result, Taylor himself gave an alternative analysis breaking a trajectory into a sequence of discrete steps whose magnitudes were correlated from one to the next. Representing Lagrangian variables in upper case, and translating Langevin's coefficients into a notation appropriate to our ends, Langevin's equation may be written

$$\mathrm{d}W = -\frac{W}{\tau}\mathrm{d}t + \sqrt{\frac{2\sigma_w^2}{\tau}}\,\mathrm{d}\xi, \tag{8}$$

where $\mathrm{d}\xi$ represents an uncorrelated sequence of Gaussian random numbers having vanishing mean and variance $\mathrm{d}t$. Increments in velocity over intervals $\mathrm{d}t$ comprise a deterministic component (usually, as here, having the effect of damping the excursions in W on a time scale τ) and a purely random component. In stationary homogeneous turbulence, equation (8) reproduces Taylor's result for the special case of an exponential correlation function.

In modern parlance, a *generalized* Langevin equation is taken as the framework for developing Lagrangian models and typically is written

$$\mathrm{d}U_i = a_i(\mathbf{X}, \mathbf{U}, t)\,\mathrm{d}t + b_{ij}(\mathbf{X}, \mathbf{U}, t)\,\mathrm{d}\xi_j, \tag{9}$$

where the random forcing $\mathrm{d}\xi_i$ is Gaussian, with $\overline{\mathrm{d}\xi_i} = 0$ and $\overline{\mathrm{d}\xi_i \mathrm{d}\xi_j} = \mathrm{d}t\,\delta_{ij}$. The selection problem for first-order LS models amounts to the prescription of the systematic part of the acceleration a_i and the scaling coefficient b_{ij}, and we return to this later.

For the comprehensibility of what is to follow, this is an appropriate point to introduce the Fokker-Planck (FP) equation corresponding to equation (9). Let $p(\mathbf{x},\mathbf{u},t)$ represent the

joint probability density (at time t) for the position and velocity of "particles" of tracer in a flow, and let $\mathcal{V}$ represent the domain of the position-velocity phase space. Because (**X**,**U**) is Markovian, one may write

$$p(\mathbf{x}_2, \mathbf{u}_2, t_2) = \iint_{\mathcal{V}} p_T(\mathbf{x}_2, \mathbf{u}_2, t_2 | \mathbf{x}_1, \mathbf{u}_1, t_1) p(\mathbf{x}_1, \mathbf{u}_1, t_1)\, \mathrm{d}\mathbf{x}_1 \mathrm{d}\mathbf{u}_1, \tag{10}$$

where $p_T(\mathbf{x}_2,\mathbf{u}_2,t_2|\mathbf{x}_1,\mathbf{u}_1,t_1)$ is the transition probability density for going from state $\mathbf{x}_1,\mathbf{u}_1$ to (a region around) $\mathbf{x}_2,\mathbf{u}_2$ over the time interval $t_2 - t_1$ and is related to the posited model of motion, i.e., equation (9). The FP equation is derived from this integral equation, and (adopting the transition density implied by equation (9) for small $t_2 - t_1$) it reads

$$\frac{\partial p}{\partial t} = -\frac{\partial}{\partial x_i}(u_i p) - \frac{\partial}{\partial u_i}(a_i p) + \frac{1}{2}\frac{\partial^2}{\partial u_j \partial u_j}(b^2 p) \tag{11}$$

(where we have anticipated the restriction $b_{ij} = b\delta_{ij}$).

Returning to the chronological line of development, *Obukhov* [1959] realized the appropriateness to high Reynolds number turbulence of models formulated in position-velocity space. He noted "the conditional distribution function (for velocity and position) is the principal characteristic of the motion in Lagrangian variables" and assumed "the evolution of the state of the selected particle in time forms a Markov process and can be described by the Fokker-Planck equation in the space (of position and velocity)" [*Obukhov*, 1959, p. 113]. He then invoked the Kolmogorov-Obukhov inertial subrange similarity theory [*Kolmogorov*, 1941a, 1941b; *Obukhov*, 1941] to identify the coefficient b with the turbulent kinetic energy dissipation rate ε, namely,

$$b = \sqrt{C_0 \varepsilon}, \tag{12}$$

where C_0 is a dimensionless coefficient that the original theory regarded as universal. It is common to express this coefficient b in terms of a decorrelation time scale τ, e.g.,

$$b = \sqrt{C_0 \varepsilon} = \sqrt{\frac{2\sigma_w^2}{\tau}} \tag{13}$$

[*Tennekes*, 1979].

Obukhov's paper spurred other efforts. *Lin* [1960a, 1960b] aimed to provide a theoretical foundation for Richardson's law of relative dispersion, accounting for anisotropy (but not inhomogeneity). He considered his work to be based on less restrictive assumptions than Obukhov's and envisaged the possibility that C_0 might vary with the turbulence Reynolds number, as argued more recently by *Sawford* [1991] and *Poggi et al.* [2008]. Lin recognized that the class of models under consideration should be regarded as representing the motion only on time scales that are large compared to the acceleration time scale of the turbulence. Acknowledging guidance from Lin and others, *Chadam* [1962, pp. iii and 3] noted

> It appears that in the Lagrangian sense, the motion of a particle in a turbulent fluid is governed by a Langevin equation... fluid elements are moving almost freely and are subjected to small random accelerations.

Considering diffusion in homogeneous, isotropic turbulence, Chadam extracted Taylor's results from the Langevin equation and noted that if the process is Markovian in (the position-velocity) phase space, the joint pdf for position and velocity satisfies the Fokker-Planck equation. *Krasnoff and Peskin* [1971] also studied Lin's model, noting that it "necessarily reproduces G.I. Taylor's theory of diffusion," and focused on relating parameters of the Langevin model (for stationary, homogeneous, and isotropic turbulence) in a rigorous way to observable properties of the turbulence; in particular, they focused on evaluating (what in modern terms would be) $C_0\varepsilon$ and used their formulation to deduce some Lagrangian statistics, such as "the Lagrangian time microscale." Several other contributions could be listed, some bearing on relative (rather than absolute) diffusion, all grounded in the theory of stochastic processes, but none of them yielding a model applicable to even the simplest regime of inhomogeneous atmospheric turbulence (except to the extent that the inhomogeneity can be neglected). Many such examples can be traced from a review by *Yaglom* [1973].

3. EARLY HEURISTIC COMPUTATIONAL LS MODELS

By the late 1960s, access to digital computers had penetrated to the level of meteorological offices and (many) universities. An era ensued of what might be termed "playful" attempts to numerically mimic atmospheric dispersion along Lagrangian lines in realistic atmospheric turbulence. *Thompson* [1971] suggested that in treating atmospheric dispersion problems, rather than adopt a "deterministic method" (such as solving an advection-diffusion equation), "it may often be more useful to simulate the original physical situation directly" and that doing so "also turns out to be simpler." Thompson's treatment falls into the first-order LS category, for "each particle is tracked by integrating the forces on it, such as wind and buoyancy, to get its velocity, and integrating that to get its position." The time constant τ, as in many of the early papers, was independent of position. Interpolating from the description given, independent stochastic variables "p_i" were computed by way of (three) Markov chains (each with Gaussian forcing, and a memory term specified to achieve the nominated autocorrelation time scale τ), and these p's were scaled to provide the Lagrangian velocity fluctuations. Several idealized regimes of turbulence were studied,

e.g., dispersion in a constant-K (Ekman) layer with a simple formulation of the mean winds.

With the objective of studying "diffusion downwind of a low-level source in a thermally neutral atmosphere," *Hall* [1975] adopted a Markov chain model

$$W(t + \mathrm{d}t) = \alpha W(t) + \beta r, \tag{14}$$

$$\alpha = 1 - \mathrm{d}t/\tau \approx \exp(-\mathrm{d}t/\tau), \tag{15}$$

$$\beta = \sigma_w \sqrt{1 - \alpha^2}, \tag{16}$$

for the vertical velocity (r is a standardized Gaussian random variate, and the Markov chain formulation is equivalent to the Langevin model with finite steps). Hall incorporated a streamwise velocity fluctuation by randomly adding $\pm 2.2u_*$ to the mean velocity upon each time step (u_* being the friction velocity and $2.2u_*$ an approximate value for the root-mean-square (r.m.s.) along wind velocity σ_u). This was, in most respects, an adequate treatment of dispersion in an ideal neutral surface layer, and adjustments to the unstably stratified case were offered by way of diabatically corrected profiles (of mean velocity $\bar{u}$ and velocity standard deviations σ_u, σ_w); however, Hall's specification $\tau \propto z/\bar{u}$ for the time scale did not equip his model with the correct (effective) diffusivity. Hall's choice implies $K_\infty \propto \sigma_w^2 z/\bar{u}$, whereas in a wall shear layer (where the eddy viscosity $\nu_T = k_v u_* z$, with $k_v = 0.4$ the von Karman constant) one should have $K_\infty \propto \sigma_w z$. These simulations were compared only very schematically with observations.

In a pioneering paper, *Bullin and Dukler* [1974] implemented a sophisticated first-order LS model on a hybrid analog/digital computer. These authors, Chemical Engineers, adopted "the generalized Langevin equation," giving an explicit rationalization for doing so, and painstakingly evaluated all needed free parameters in their model by relating them to specific measurements: in their own words, "all statistical terms in the equations are related to measurable Eulerian meteorological or fluid mechanical conditions." Very good simulations of laboratory and field dispersion experiments were reported; for example Figure 1 is their comparison of their model with a vertical concentration profile observed in Project Prairie Grass. As an aside, these authors [see also *Lee and Dukler*, 1976] based their simulation on a rescaled Lagrangian velocity (in their terminology, $\widehat{M}_L$) that, upon multiplication by the local velocity standard deviation, gave the true velocity; and they generated the needed random forcing by filtering electrical white noise. These techniques were carried over, or perhaps adopted independently, by Thurtell and Kidd (see below).

Joynt and Blackman [1976] used a form of the random displacement model to simulate atmospheric dispersion on

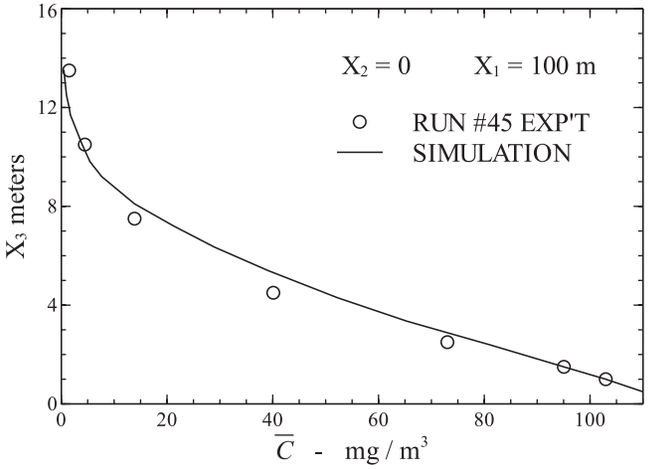

Figure 1. *Bullin and Dukler's* [1974] first-order LS simulation of the vertical profile of concentration at a radial distance of (X_1 =) 100 m from a continuous point source in the stratified atmospheric surface layer (Project Prairie Grass run 45, for which, according to *van Ulden* [1978], the friction velocity $u_* = 0.39$ m s^{-1} and the Obukhov length $L = -87$ m). The specification $X_2 = 0$ presumably indicates the data stem from that one of the six profile masts that was best centered within the plume. The turbulent velocity standard deviation was assigned a constant measured value, and a *measured* Eulerian integral time scale was converted to an effective Lagrangian time scale by assuming the ratio of Lagrangian to Eulerian time scales was 4.0. That time scale was implemented by adjusting the cutoff frequency of an electronic white noise generator. Transcribed, with permission, from *Bullin and Dukler* [1974, Figure 15]. Copyright 1974 American Chemical Society.

the mesoscale, specifically the dispersion of sulfur dioxide near Melbourne, Australia. In each time step, a particle was advected by the mean velocity and displaced "to a point randomly chosen on the surface of an ellipsoid" whose axes were matched to the velocity standard deviations so that the variance of the vertical displacement was $2K_\infty \Delta t \equiv 2\sigma_w^2 \tau \Delta t$, with τ specified as z/σ_w and the time step Δt constant (and larger than τ). Particles were reflected off ground, and their motion was damped at the height of the capping inversion. This heuristic RDM did not compensate for vertical gradients in $K_\infty = \sigma_w^2 \tau$, nor was the specification for τ appropriately tuned for the entire CBL (though it is of the correct order of magnitude near ground), but for its time, this paper represented an ambitious application of a Lagrangian stochastic methodology.

Perhaps the most comprehensive early Lagrangian simulations of dispersion on the (smaller) scale of the atmospheric surface layer were those initiated at the University of Guelph by G.W. Thurtell. As a doctoral student at the University of Wisconsin, which at that time had a major role in the U.S. army's micrometeorology program, Thurtell and other

students had participated in discussions with H. Lettau of what were (at the time) considered to be errors in measured canopy wind and concentration profiles (the measurements, sponsored by the U.S. Army Signal Corps out of Fort Huachuca, had been made in Thailand). Thurtell later realized that near-field effects around leaves might explain the apparent anomalies and began work on numerical simulations. In 1976, while visiting CSIRO Division of Environmental Mechanics in Canberra (the "Pye Lab," headed by J.R. Philip), Thurtell participated in measurements by E.F. Bradley, O.T. Denmead and others of fluxes of heat, water vapor, and carbon dioxide in the Uriarra pine forest. These measurements, eventually published by *Denmead and Bradley* [1985], dramatically showed countergradient vertical fluxes in the canopy (see Figure 2), and Thurtell, understanding the cause, continued to develop a Lagrangian framework for their interpretation. Intuition led him to take the step of transforming a layer of sheared, vertically inhomogeneous turbulence in (x, z, t) space into a layer of sheared but homogeneous turbulence in (x, z_*, t_H) space, where the rate at which t_H advances along the particle trajectory depends on the particle position (this idea is consistent with, but did not derive from, *Batchelor's* [1957, 1964] Lagrangian similarity theory). The properties of the scaled turbulence were specified as those appropriate to the real world at an arbitrary reference height H and were generated by passing a white noise analog voltage signal through a low-pass RC filter. (Another aside: the RC time constant of this filter had to be chosen as a function not only of the wanted reference time scale τ_H, but also as a function of the real-time iteration rate

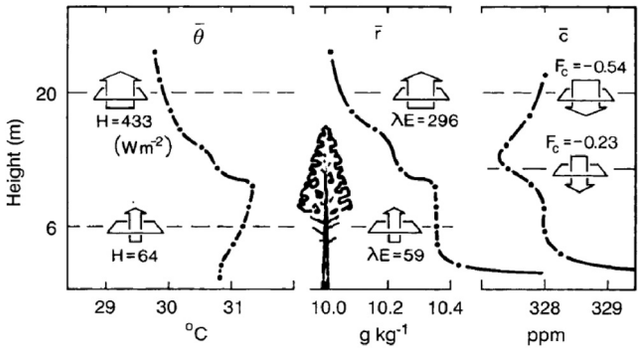

Figure 2. Countergradient vertical eddy flux of heat (H, W m^{-2}) beneath the crown of a pine forest, and of carbon dioxide F_c within the crown. An upward latent heat flux (λE, W m^{-2}) occurs beneath the crown where the associated mean vertical gradient of mixing ratio vanishes. By virtue of providing a valid treatment of the near field of nearby sources, a first-order LS model correctly reproduces the flux-gradient relationship. Reproduced from *Denmead and Bradley* [1985], with kind permission of Springer Science + Business Media B.V.

of the algorithm, which was written in assembly language and ran on a DEC PDP-11 with 16 K-bytes of 8 bit memory in Thurtell's field trailer. A digital-to-analog convertor provided signals $x(t_H)$, $z_*(t_H)$, which were displayed on an oscilloscope. G. Kidd, an electrical engineer, played a key role in this work.)

The Thurtell algorithm imposed whatever vertical profiles of velocity variance, mean velocity $\bar{u}(z)$, and time scale τ were wanted and preserved the essential property that in traveling a streamwise distance $\bar{u}(z)t$ at height z, a particle would (on average) experience the correct number of independent vertical velocity "choices" (i.e., the proper ratio $dt/\tau(z)$ was encoded; the real world time step dt equivalent to dt_H varied along the trajectory so as to keep dt/τ constant and small). This scheme could be attuned to simulate dispersion in interesting regimes of turbulence for which analytic solutions existed and was shown to reproduce those solutions [*Wilson et al.*, 1981a] and (upon suitable tuning of the autocorrelation time scale τ) to provide an excellent simulation of the Project Prairie Grass dispersion trials [*Wilson et al.*, 1981c]. However, the most consequential aspect of the work under Thurtell's scheme was the realization [*Wilson et al.*, 1981b] that when the algorithm was applied to dispersion in the sort of turbulence system characterized by plant canopy flow, i.e., in which the turbulent velocity scale σ_w varies with height, particles would accumulate in the bottom of the canopy where σ_w was small. A heuristic upward drift velocity $\bar{w}_L = \sigma_w \tau \partial \sigma_w / \partial z$ had to be added to the scheme, and only by virtue of that step could this phenomenon, loosely reminiscent of the high density of a gas where its temperature (proportional to the kinetic energy of molecules) is low, be circumvented (*Faller and Mignerey* [1982] passingly allude to having experienced the same difficulty). By simulating the case of an elevated area source extending far upstream, and treating the surface as a perfect reflector, Wilson et al. focused on the mean concentration profile in the canopy layer beneath the source, where the mean vertical *flux* was (effectively) zero: a criterion for the adequacy of the bias velocity was that it should ensure that, away from the immediate region of the source, the mean concentration gradient should vanish wherever the mean flux did.

Legg and Raupach [1982], in one of a series of contributions from the CSIRO Pye Lab. group relating to this history, argued that since in steady state, horizontally homogeneous flow, a vertical gradient $\partial \sigma_w^2 / \partial z$ in vertical velocity variance is accompanied by (i.e., balanced by) a vertical gradient in the mean pressure departure from the hydrostatic reference state, one must include in the Langevin equation a deterministic acceleration equal to the pressure force per unit mass, namely, $\partial \sigma_w^2 / \partial z$. Legg and Raupach adopted the Langevin equation (here slightly rephrased)

$$dW = \left(-\frac{W}{\tau} + \frac{\partial \sigma_w^2}{\partial z}\right) dt + \sqrt{\frac{2\sigma_w^2}{\tau}}\, d\xi, \qquad (17)$$

which can be thought of as giving the particles a mean drift velocity $2\sigma_w \tau \partial \sigma_w / \partial z$, i.e., *twice* the value used by *Wilson et al.* [1981b]. The discrepancy was reconciled by *Wilson et al.* [1983] who showed that it lay in the distinction between a formulation of the Markov chain (or Langevin equation) in terms of the velocity W [*Legg and Raupach*, 1982] or in terms of the ratio $p = W/\sigma_w$ [*Wilson et al.*, 1981b]. However, *Wilson et al.* [1983] demonstrated that *neither* formulation is successful in a turbulence regime with rapidly changing $\sigma_w \tau \partial \sigma_w / \partial z$ and gave yet a third formulation (labeled WTK″ and also arrived at by *Durbin* [1983], see his footnote page 63) that subsequently was proven [*Thomson*, 1984, 1987] to correspond to the unique well-mixed, 1-D LS model for Gaussian inhomogeneous turbulence.

Several other papers in the early 1980s continued to develop the first-order LS model in one way or another. For instance, *Ley* [1982] treated the neutral surface layer using a Markov chain model with constant time step, incorporating the $\overline{u'w'}$ covariance and comparing her simulations with neutral Project Prairie Grass experiments (by now, LS models were being "tuned" to respect whatever reliable empirical relationships had been established for the turbulent flow under consideration, e.g., the needed time scale τ can be related to the surface layer eddy viscosity/diffusivity, etc.). As another example, *Legg* [1983] simulated dispersion experiments performed in a boundary-layer wind tunnel, his Markov chain model providing correlated vertical and along-stream fluctuations, whose statistics could be contrived to be non-Gaussian (motivation for this being that velocity distributions within and above a plant canopy are highly skewed). Legg calculated not only mean concentrations but also (and he was perhaps the first to have done so) the turbulent mass fluxes, which could be compared with measurements.

Thomson [1984] sought to formulate a Langevin model in a way that would ensure "the correct steady state distribution of particles in (*position-velocity*) phase space." He presupposed a model framework of the form

$$\Delta W = -\frac{W}{\tau}\Delta t + \mu \qquad (18)$$

and deduced the necessary statistics of the random forcing μ. The restricted model form, with the linear damping term, led to a requirement (in general) for non-Gaussian forcing. Thomson also analyzed models formulated in terms of rescaled velocity (W/σ_w), showing that such a model is satisfactory even with Gaussian forcing, if the Eulerian velocity pdf is Gaussian.

We close this section by considering the Markov chain simulation by *Baerentsen and Berkowicz* [1984] of dispersion in the convective boundary layer (CBL). The CBL represents a particularly challenging regime of turbulence, and more recent LS simulations of the CBL will feature below. Setting a precedent for much subsequent work, Baerentsen and Berkowicz approximated the pdf for vertical velocity in the CBL as a linear combination of two Gaussians, one representing the velocity within updrafts and the other within the (areally more dominant) downdrafts, leading to an overall velocity distribution that was skewed. They separated their Markov chain simulation into "two parallel schemes with different statistics, one for particles in updrafts and one for particles in downdrafts," the particles having an assigned probability of jumping during any given step from updraft to downdraft or vice versa. The random forcing was Gaussian, a term in their Markov chains compensated (as was by now known to be necessary) for the vertical inhomogeneity of the updraft (or downdraft) velocity variance, and particle paths were reflected at the top and bottom of the CBL. Like *Thomson* [1984], but in this case by performing random flight simulations, Baerentsen and Berkowicz examined whether a cloud of particles that were initially uniformly mixed on the vertical axis would remain uniformly mixed, and whether their velocity pdf would remain consistent with the assumed (skew) Eulerian pdf, an idea that was to be important for subsequent developments. Shortly afterward, *de Baas et al.* [1986] also performed an LS simulation of the CBL, but using "a single Langevin equation to describe all particle velocities." The random forcing in their Langevin equation (which was of the form of equation (18)) made use of the prescription of *Thomson* [1984], i.e., it was non-Gaussian. Similar simulations, but using an improved description of the Lagrangian time scale, were carried out by *Sawford and Guest* [1987].

4. NEW CRITERIA FOR LS MODELS AND THE WELL-MIXED CONDITION

The previous section covered contributions whose basis was, to differing degrees, heuristic. Overlapping with and stemming from those contributions, there commenced roughly in the early 1980s a search for more rigorous criteria to guide the specification of trajectory models in arbitrarily complex turbulent flows, for it had become evident that inhomogeneities (most familiarly in the atmospheric context, $\partial \sigma_w^2 / \partial z$) needed to be carefully handled. A spate of new contributions exploited the existing theory of stochastic processes and spurred the analysis of LS models in terms of properties of the Fokker-Planck equation to which they were equivalent, and in terms of their relationship to the Navier-Stokes and/or Reynolds equations. At the risk of excessive

abbreviation, the main points of attack were (1) acceptability of a first-order model's small time limit for spread from a source [*Durbin*, 1980a; *van Dop et al.*, 1985]; (2) acceptability of the zeroth-order model (i.e., "diffusion" model) implied (asymptotically, in the limit $t/\tau \to \infty$) by a first-order LS model [e.g., *Durbin*, 1983, 1984]; (3) admissibility of non-Gaussian random forcing of the generalized Langevin equation [*Thomson*, 1984; *van Dop et al.*, 1985; *Novikov*, 1986; *Sawford*, 1986; *Thomson*, 1987]; (4) justifiability of the generalized Langevin equation on the basis of the Navier-Stokes equations [e.g., *van Dop et al.*, 1985; *Haworth and Pope*, 1986]; (5) reconciliation of the Eulerian closure assumptions implied by a generalized Langevin equation with the Reynolds equations [e.g., *van Dop et al.*, 1985; *Haworth and Pope*, 1986; *Pope*, 1994]; (6) equivalence of forward and reverse formulations of dispersion in the context of incompressible flows [e.g., *Egbert and Baker*, 1984]; (7) proposed requirement [*Pope*, 1987] that driving velocity statistics should be "physical," i.e., consistent with the Navier-Stokes (and Reynolds) equations (from the subsequent perspective of the well-mixed condition, one adopts whatever field of flow statistics is plausible and convenient, and one infers a consistent LS model); and (8) requirement that models produce the correct steady state distribution of particles in position-velocity phase space [e.g., *Janicke*, 1983; *Thomson*, 1984; *Novikov*, 1986; *Sawford*, 1986; *Thomson*, 1987].

Space does not permit to do justice to this body of work, which led to the realization that many of these criteria are equivalent to or implied by the "well-mixed condition," i.e., the condition that if the particles are well mixed in velocity-position space, they should remain so [*Thomson*, 1987]. We shall arbitrarily pick some few salient contributions.

Durbin [1980a] proposed to model dispersion by way of a Markov chain for velocity, noting that in the limit $\Delta t \to 0$ (infinitesimal time step), this reduces to a continuous Langevin equation whose solution in homogeneous turbulence for $t \gg \tau$ is equivalent to the outcome of a random walk in position

$$\mathrm{d}Z = \sqrt{2\sigma_w^2 \tau}\ \mathrm{d}\xi \tag{19}$$

(where $\mathrm{d}\xi$ is Gaussian with variance $\mathrm{d}t$, and $\sigma_w^2 \tau$ is, of course, Taylor's far-field diffusivity). In extending this to inhomogeneous turbulence with $\tau = \tau(z)$, Durbin envisaged that particles would "tend to drift up gradients of τ" and proposed to account for this in his random displacement model by adding to equation (19) a term $W_L \mathrm{d}t$, where

$$W_L = \sigma_w^2 \frac{\mathrm{d}\tau}{\mathrm{d}z}. \tag{20}$$

This he proved to be consistent with Batchelor's formula for the drift of the center of mass of a tracer cloud in a neutral surface layer ($\mathrm{d}\langle Z\rangle/\mathrm{d}t = bu_*$). Although envisaging that the Markov chain formulation must, in general, be solved numerically, Durbin put an emphasis on the formal basis of the model(s) and extracted analytic solutions (to the random displacement model, i.e., "Markov limit" of the Langevin equation) for special cases. Later, *Boughton et al.* [1987] wrote the RDM in a more general form as

$$\mathrm{d}Z = \frac{\mathrm{d}K}{\mathrm{d}z}\mathrm{d}t + \sqrt{2K}\ \mathrm{d}\xi \tag{21}$$

($\mathrm{d}\xi$ having variance $\mathrm{d}t$), where the term in $\partial K/\partial z$ corrects for vertical inhomogeneity [see also *Monin and Yaglom*, 1977, section 10.3].

van Dop et al. [1985] set out to "develop a generalized form of the Langevin equation, suitable for inhomogeneous and unsteady turbulent flows, which has a simple physical explanation" and suggested a model of the form

$$\mathrm{d}W = (-W/\tau + a_1(Z,t))\ \mathrm{d}t + a_2^{1/2}\mathrm{d}\omega_t, \tag{22}$$

where the forcing $\mathrm{d}\omega_t$ was not restricted to be Gaussian (this general approach was extended by *Sawford* [1986]). The coefficients a_1 and a_2 were determined by considering the small t behavior and by comparison with the Eulerian moment equations. Note that equation (22) excludes models in which the systematic part of the acceleration is nonlinear in the particle velocity, a feature also of the class of generalized Langevin models prescribed by *Haworth and Pope* [1986].

Thomson [1987] took equation (9) as the most general framework for a first-order LS model and using results from the theory of stochastic processes established that the forcing $\mathrm{d}\xi_j$ necessarily must be Gaussian if the velocity is to evolve continuously in time without jumps. Accordingly, the joint density function $p(\mathbf{x},\mathbf{u},t)$ is governed by the Fokker-Planck equation (11). Thomson considered the implication of the requirement that an LS model should provide the correct steady state particle distribution in position-velocity phase space. For simplicity, we suppose the flow is stationary and of constant density ρ, and we let $g_a(\mathbf{u};\mathbf{x})$ be the probability density function for the Eulerian velocity $\mathbf{u}$ at position $\mathbf{x}$. Suppose further that the model is applied to the motion of particles whose distribution at time $t = 0$ is $p(\mathbf{x},\mathbf{u},0) \propto \rho g_a(\mathbf{u};\mathbf{x})$. As these particles are already well mixed, an acceptable model must ensure $p(\mathbf{x},\mathbf{u},t) \propto \rho g_a(\mathbf{u};\mathbf{x})$ for arbitrary $t > 0$. It follows that $g_a(\mathbf{u};\mathbf{x})$ must be a steady state solution of equation (11), providing a single constraint on the model's formulation. For a 1-D model, and once b (or C_0) has been specified (see equation (12)), that single constraint suffices to yield the unique well-mixed, first-order model; for a multi-dimensional model, the well-mixed condition selects a class of acceptable models. Thomson showed that, if the

well-mixed condition is satisfied, then most of the other then known selection criteria are also fulfilled.

Weil [1990] pointed out that the treatment of boundaries could lead to violations of the well-mixed condition, even if away from the boundaries the terms in the stochastic differential equation (9) were consistent with it. Treating homogeneous *skew* turbulence in the context of an examination of the CBL, Weil reasoned that using perfect reflection would be inconsistent with the assumed Eulerian velocity pdf, and introduced a more complex reflection scheme. *Hurley and Physick* [1993] also addressed the problem of modeling the CBL using homogeneous skewed turbulence, with a view to designing an efficient applied model. They proposed a number of reflection algorithms, including one which relates the reflection velocity W_r to the incident velocity W_i using

$$\int_{W_i}^{0} w g_a(w)\ \mathrm{d}w + \int_{0}^{W_r} w g_a(w)\,\mathrm{d}w = 0. \qquad (23)$$

This was later shown by *Thomson and Montgomery* [1994] to ensure the well-mixed condition is satisfied (in the limit of small time step) and was extended by *Thomson et al.* [1997] to the case where there is a discontinuity in turbulence levels across an interface such as the boundary-layer top.

In the balance of this article, we shall attempt two tasks: first, to give some flavor of the immediate impact of the ideas of this section and of the range of subsequent applications; and second, to indicate what progress has occurred subsequently to these ideas in terms of the ongoing attempt to define a unique formulation of the first-order LS model in the multidimensional case.

5. APPLICATIONS OF WELL-MIXED LS MODELS

In choosing a few examples to illustrate the span of applications of the modern Lagrangian stochastic model, we select those which seem best to indicate the unique capabilities of the approach, namely, (1) ability to rationally utilize known velocity statistics, whether these be Gaussian or otherwise; (2) correct treatment of the near field; (3) applicability to arbitrarily complex, 3-D turbulence; and (4) universality of the LS approach for treating unresolved (or "turbulent") scales of motion across the range of scales of application, i.e., from micrometeorology to planetary meteorology.

Dispersion in the CBL is difficult to model accurately because of the strongly inhomogeneous and non-Gaussian character of the flow (see Figure 3). As a result, the CBL became the application that arguably best demonstrates the advantages of the Lagrangian stochastic technique, namely, its ability to accommodate non-Gaussian flow statistics (and more broadly, whatever statistical information on the flow is available). We have covered above the early LS treatments of the CBL by *Baerentsen and Berkowicz* [1984], *de Baas et al.* [1986], and *Sawford and Guest* [1987]. Soon after the developments summarized in section 4, *Luhar and Britter* [1989] treated vertical dispersion in the CBL with the 1-D LS model implied by the well-mixed constraint when the Eulerian velocity pdf is a linear combination of two Gaussians. Adopting plausible profiles of velocity variance and skewness, Luhar and Britter produced simulations that nicely matched the striking features of dispersion from near-ground and elevated sources in the CBL (see Figure 4). Subsequent LS models of the CBL differ in details concerning the parameterization of velocity statistics [e.g., *Weil*, 1990; *Du et al.*, 1994] but not in their fundamentals.

The validity of the first-order LS model in the near field of sources is of key significance for modeling turbulent transport in plant canopies. Vertical profiles of mean temperature, humidity, and carbon dioxide concentration within a canopy are strongly influenced by heat, water vapor, and carbon dioxide fluxes to or from nearby leaves [*Raupach*, 1987, 1989], and a treatment of the interaction of a crop with its environment must detail the mutual feedback on one another of the canopy source distribution and the airstream properties. That is to say, models of crop productivity must, in some

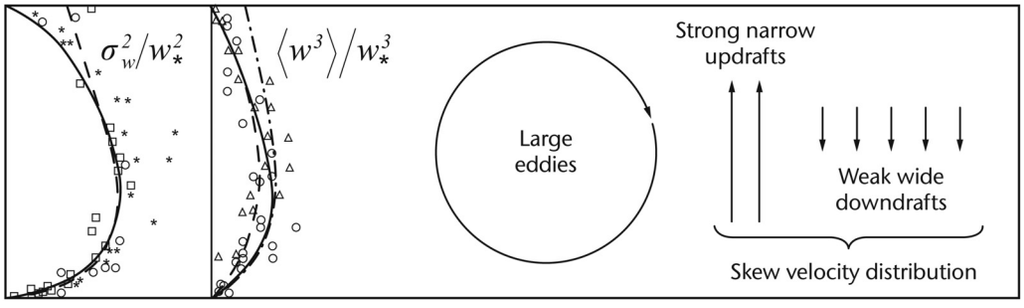

Figure 3. Schematic view of statistics of vertical motion in a convective boundary layer. The plots of σ_w^2/w_*^2 and $\langle w^3 \rangle / w_*^3$ are adapted from *Sawford and Guest* [1987]. Copyright American Meteorological Society, used with permission.

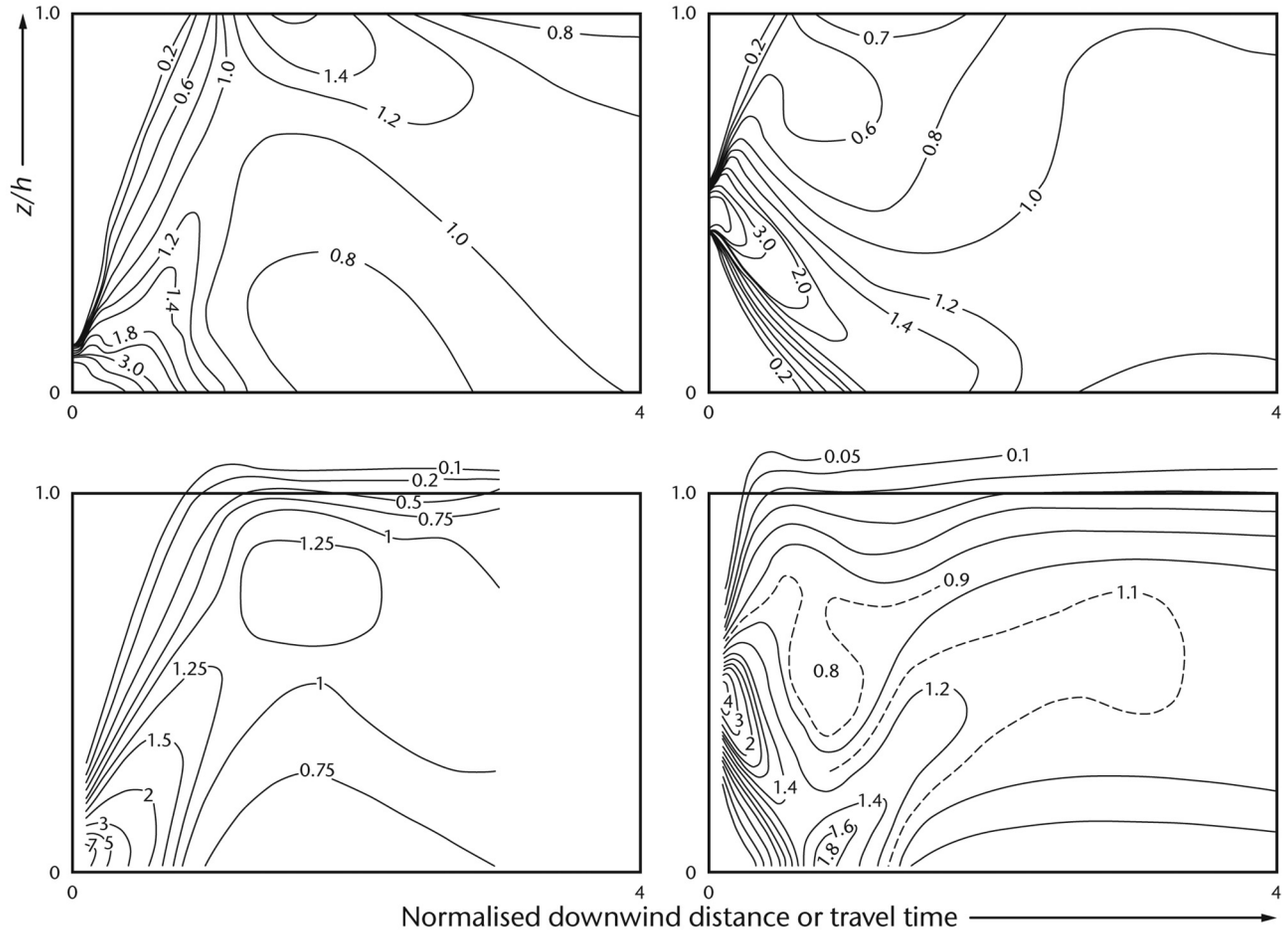

Figure 4. Dispersion in the CBL (mixed layer depth h) from a continuous source at (left) $z/h = 0.07$ or at (right) $z/h = 0.5$. Vertical (height) axis ranges from ground to the top of the CBL. (top) The LS simulations of *Luhar and Britter* [1989], reprinted from *Luhar and Britter* [1989] with permission from Elsevier. (bottom) The *Willis and Deardorff* [1976, 1981] convection tank experiments. Adapted from *Willis and Deardorff* [1976] with permission from John Wiley and Sons and from *Willis and Deardorff* [1981] with permission from Elsevier.

manner, parameterize the canopy micrometeorology. Starting at least as early as the 1950s, crop modelers parameterized canopy transport by way of K theory (*Legg and Long* [1975] performed an interesting experimental study that attempted to determine the eddy diffusivity in a canopy by way of tracer dispersion experiments) or, more typically, a transfer resistance network (which is the spatially aggregated equivalent of a K theory model). For some ends, this may be satisfactory [e.g., *Dolman and Wallace*, 1991]. However, *Leuning et al.* [2000], who used the Localized Near Field theory of *Raupach* [1987] to estimate the source distributions of heat, water vapor, carbon dioxide, and methane in a rice canopy, reported that it was essential to account for the near-field contributions of sources to the canopy concentration profile. The most compelling sign that this is so is the potential occurrence (depending in detail on the source distribution) of countergradient eddy fluxes [*Denmead and Bradley*, 1985, see Figure 2]. Correct description of the flux-gradient relationship within plant canopies is a unique capability of the first (or higher)-order LS model or computational simplifications thereof [e.g., *Raupach*, 1989; *Warland and Thurtell*, 2000; *Siquiera et al.*, 2000; *Hsieh et al.*, 2003].

Irrespective of the connection with crop modeling (or more generally, soil-vegetation-atmosphere coupling), the regime of turbulence in a canopy is scientifically challenging in that it is acutely (vertically) inhomogeneous, velocity statistics are highly non-Gaussian, and the r.m.s. velocity fluctuations exceed the mean velocity in much of the canopy (necessitating adoption of a multidimensional turbulence model).

Canopy flow is dominated by penetrating gusts (or sweeps) of high-momentum air originating from above the canopy, alternating (by no means regularly) with ejections of sluggish (and by day, warm and humid) air from the canopy [*Finnigan*, 2000]. Throughout much of the canopy, the dominant eddy size is of the order of the canopy depth h, and the canopy and the ground beneath constitute a vertically distributed source (or sink) for heat, momentum, water vapor, and carbon dioxide. As a corollary of the dominating role of large eddies (e.g., quadrant analysis shows that a large stress fraction is transported in a small time fraction by the sweeps and ejections), turbulent transport terms in the Reynolds equations are non-negligible, e.g., the turbulent kinetic energy budget is far from being in local equilibrium.

This complexity would seem to suggest that a useful Lagrangian model for canopy dispersion must be comparably intricate. In view of the known skewness of the pdf's for streamwise and vertical velocity, *Flesch and Wilson* [1992] constructed a well-mixed 2-D LS model derived by using the approximation that the (skewed) velocity fluctuation pdf $g_a(u', w';z)$ within the canopy is a superposition of two joint Gaussians and by introducing a hypothesis concerning the effect of the probability current ϕ whose components appear in the systematic part (a_i) of the acceleration and whose divergence is constrained by the well-mixed condition [*Thomson*, 1987, equation 9b], namely, that ϕ acts to conserve the direction of the Lagrangian velocity fluctuation vector. The unexpected finding was that this model performed no better in regard to simulations of the mean tracer distribution than did models based on a simplification to Gaussian pdf's (namely, the model provided by *Thomson* [1987] or the alternative closure of Flesch and Wilson). Most subsequent Lagrangian simulations of canopy transport [e.g., *Poggi et al.*, 2006; *Cassiani et al.*, 2007; *Postma et al.*, 2011] have adopted the approximation of Gaussian pdf's. In general, in such acutely inhomogeneous flows, it appears the effects of velocity skewness are secondary, except very close to the source(s).

The so-called "urban canopy" shares all of the flow characteristics of a natural canopy flow and is further complicated by the fact that the excluded space (i.e., buildings) may represent a substantial fraction of the urban canopy volume. Along with population drift to urban centers and the growth of megacities, comes an increasing interest from meteorologists and others in a capability to diagnose or even anticipate the patterns of wind and wind transport in cities. Many scientific approaches are possible, each being appropriate for some subclass of urban dispersion problems. The extreme of simplicity would be represented by merely assigning a city an appropriately large roughness length and computing flow and dispersion from sources in the same manner as elsewhere. At the other extremity, the most rigorous approach is to compute the flow itself, typically by solving the Reynolds or large eddy simulation (LES) equations on a grid that resolves buildings and perhaps with the exterior boundary conditions supplied by a numerical weather prediction model; having thereby estimated the velocity statistics, one may then compute wind transport from sources within the flow. *Naslund et al.* [1994] and *Lee and Naslund* [1998] may have been the first to implement this approach, computing urban dispersion around resolved buildings using a first-order LS model, driven by a computed 3-D field of wind statistics. Taking the same approach, *Wilson et al.* [2009, 2010] found that the first-order LS model yielded skillful predictions of dispersion from a continuous near-ground point source in downtown Oklahoma City to detectors kilometers away, with over 50% of predicted concentrations being within a factor of two of the corresponding observations. Of course, in this context, model performance hinges not only on the fidelity of the LS model itself, but also on the quality of the computed flow statistics (computing the flow is the more difficult part of the overall problem). Not surprisingly, the same level of skill had been reported for an Eulerian simulation of the same experiment (the $k - \varepsilon$ flow model that computed the flow also solved the mass conservation equation, and the detectors were all in the far field of the source). There are, nonetheless, some secondary advantages of the Lagrangian approach. For instance, the simulations showed explicitly that tracer particles emitted in the urban canyon in close proximity to the wall of a building were entrained into a recirculation eddy and ascended the wall.

Inverse (or backward) LS simulations have assumed an important role for those interested in identifying and quantifying sources or sinks of atmospheric trace gases, whether on the micrometeorological, regional, continental, or planetary scale. *Flesch et al.* [1995] introduced (and later tested [*Flesch et al.*, 2004]) a "backward Lagrangian stochastic" method for inverse dispersion on the micrometeorological scale, i.e., to infer the emission rate Q of a source from the measured concentration rise it causes. This method, which is covered in more detail by *Wilson et al.* [this volume], has subsequently been widely applied, e.g., to deduce agriculture-related emissions of methane or ammonia (as recent examples, see *Todd et al.* [2011], *Laubach et al.* [2012], and *McGinn and Beauchemin* [2012]). The inverse dispersion approach based on LS models is commonly applied, too, on much larger scales (see below).

Our final category of applications concerns Lagrangian models (of first or zeroth, or even mixed order) capable of tracking particle motion throughout and even above the troposphere, from one continent to another, by virtue of their being coupled to flow fields calculated by numerical weather prediction models. Examples are NAME [*Maryon et al.*,

1999; *Jones et al.*, 2007], FLEXPART [*Stohl et al.*, 2005], STILT [*Nehrkorn et al.*, 2010], MLDP0/MLDP1 [*D'Amours et al.*, 2010; *Malo et al.*, 2011], and HYSPLIT [*Draxler and Hess*, 1997, 1998]. An important motivation for the development of these models was the Chernobyl reactor accident in 1986, which highlighted the need to be able to estimate long-range transport and dispersion in a timely and flexible way. Lagrangian models of this type are now used to estimate the dispersion of a wide variety of materials including radionuclides, chemicals, volcanic ash, airborne diseases, and mineral dust, and in inverse (backward) mode, they can be used for "top-down" diagnosis of surface-atmosphere gas exchange [e.g., *Thompson et al.*, 2011; *Manning et al.*, 2011] with high spatial resolution. Such models may include parametrizations of dry and wet deposition and, in some cases, are coupled to full atmospheric chemistry calculations to estimate the air quality resulting from many individual sources. Vertical transport by (unresolved) deep convection is an important element of such models. Approaches have been developed for treating deep convection that are somewhat different in approach to the turbulent dispersion models that are the main focus of this article [e.g., *Collins et al.*, 2002; *Forster et al.*, 2007]. These schemes often make use of estimates of the upward and downward mass fluxes in the convective clouds and, in the case of Forster et al., make use of the well-mixed condition, although in a different context to that considered above.

6. OUTSTANDING PROBLEMS AND CRITERIA SUPPLEMENTING THE WELL-MIXED CONDITION

As noted above, the criteria developed for designing Lagrangian models do not lead to a unique multidimensional model. *Sawford and Guest* [1988] were the first to show explicitly two distinct models, both satisfying the well-mixed condition for the same flow and giving significantly different dispersion predictions. Here the nonuniqueness arose from different expressions for the terms that depend on the gradients of flow statistics and which are required by the well-mixed condition. Nonuniqueness can arise even in homogeneous turbulence (although not if it is isotropic), and *Borgas et al.* [1997] considered Lagrangian models appropriate to homogeneous axisymmetric turbulence without reflectional symmetry, in which case the systematic part of the acceleration in equation (9) can contain a term of the form $\varepsilon_{ijk}\Omega_j U_k$ where $\boldsymbol{\Omega}$ points along the axis of symmetry. They found that the extra term can lead to spiraling of the velocity vector and, hence, of the trajectories, in the plane perpendicular to $\boldsymbol{\Omega}$. This is associated with oscillations in the Lagrangian velocity correlation function and a reduced rate of dispersion.

Wilson and Flesch [1997] considered the mean rate of rotation of the velocity vector in the context of a 2-D model. For dispersion in a plant canopy, they showed that models where the mean rate of rotation was zero gave best agreement with wind tunnel measurements, and they speculated that this might be a good criterion for selecting models more generally. However, this criterion does not lead to a unique model, and *Reynolds* [1998] has shown that the remaining nonuniqueness can mean that different "zero spin" models can give significantly different dispersion predictions. Reynolds also argued that for some flows, the zero spin models were not optimal. *Sawford* [1999] introduced a more tractable and general way to quantify the tendency of the velocity vector to rotate, taking the average of the cross product of particle velocity and acceleration (which is related to the rate at which area is swept out by the velocity vector). Sawford suggested that for the better-performing models, this quantity is zero. Although these studies have shed useful light on the nonuniqueness problem, a completely satisfactory solution has yet to be found. It is not certain that a universally best approach, valid for all flows, necessarily exists.

A second outstanding issue, and (operationally) a second form of nonuniqueness, is the problem of determining the correct (or optimal) value of the constant C_0 in equation (12). By virtue of the way C_0 is introduced to the LS model, it equates to the constant in the Kolmogorov-Obukhov model for the second-order Lagrangian velocity structure function. However, while being (in its origin) an attribute of the *inertial subrange* of scales of motion in turbulence, in LS models C_0 modulates the (far more important) effective *integral* time scale of the model (see equation (13); *Wilson et al.* [2009] cover this in some detail). It follows that even *if* C_0 were universal, and even *if* its value were known, that putative constant value might not necessarily give the best estimate of dispersion, which is more sensitive to the integral time scale than to the inertial subrange part of the velocity correlation. To make matters worse, despite the appealing simplicity of the original Kolmogorov hypotheses, evidence suggests they may be a too drastic simplification [e.g., *Sreenivasan and Antonia*, 1997; *Sreenivasan*, 1999]; if so, then C_0 may not (in reality) be universal. Furthermore, *Sawford* [1991] showed that in first-order LS models, the optimal value of C_0 should be expected to depend on the Reynolds number of the flow of interest (Sawford introduced a second-order LS model, i.e., a model for which the Markovian state variable includes the particle acceleration $\mathbf{A}$, which is modeled by way of a Langevin equation), so that (for instance) LS models of dispersion within an engineering wall shear layer (with relatively low Reynolds number) might have a different optimal C_0 than LS models of the atmospheric surface layer. Reported values for C_0, arising from several

possible approaches to its determination (e.g., velocity-difference statistics from direct numerical simulations or the fitting of Lagrangian models to observations) cover the range from about 2 to 10 [*Du et al.*, 1995], presumably reflecting the above complexities and the following further complication.

In atmospheric turbulence, at least within the boundary layer, it is reasonable to assume that the Reynolds number is effectively infinite, eliminating one source of uncertainty. Then one may hope simply to compare an LS model with an authoritative experiment (such as Project Prairie Grass) and infer the optimal value for C_0 (for that class of flow). A complicating factor here, however, is that the optimal value for C_0 will depend on the *dimensionality* of the LS model one adopts (it will also depend, and in a sense more fundamentally, on one's choice of a particular model from within the well-mixed class; for instance, it is probably significant whether or not the chosen model induces a mean rotation of the velocity fluctuation vector). This was first shown by *Sawford and Guest* [1988], who following the suggestion of Durbin derived the diffusion limit of *Thomson's* [1987] multidimensional model for Gaussian inhomogeneous turbulence and showed that the implied (effective) eddy diffusivity (for vertical dispersion) is

$$K = \frac{2(\sigma_w^4 + \overline{u'w'}^2)}{C_0\varepsilon}, \tag{24}$$

where $\overline{u'w'}(=-u_*^2)$ is the velocity covariance. As an aside, albeit one that hints at yet another nonuniqueness, note the very large sensitivity of the effective eddy diffusivity (and thus optimal C_0) to one's parametrization of σ_w: taking specifically the neutral surface layer as an example, whether one chooses $\sigma_w/u_* = 1.25$ or 1.3 is consequential, and indeed, observations of this rather primary observable (σ_w/u_*, in the neutral limit) are more scattered than a casual reading of the textbooks might suggest. Be that as it may, the implication of equation (24) is that optimal C_0 will depend on whether the LS model does or does not include the Lagrangian alongwind velocity fluctuation U', or more precisely, whether or not one includes its *correlation* with the vertical velocity. Based on this result, *Wilson et al.* [2009] deduced (by reference to Project Prairie Grass neutral runs) that for a multidimensional model with the $\overline{u'w'}$ correlation included (and neutral $\sigma_w/u_* = 1.3$), $C_0 \simeq 4.8$, while if the covariance is neglected (or alongwind fluctuations entirely dropped), then $C_0 \simeq 3.6$ [see also *Sawford*, 2001]. Wilson et al. found that the performance of simulations of urban dispersion with/without covariance was consistent with the above reasoning, but in view of the large uncertainties surrounding the quality of (computed) velocity statistics driving the LS models, this is not an entirely convincing confirmation. In any case, to what extent should these optimal values for C_0 generalize, given that they stem from tuning the LS model to the Project Prairie Grass observations of surface-layer dispersion? In view of the above discussion, it seems unlikely these results necessarily should apply to (for instance) the CBL. In their pioneering LS simulation of the CBL, *Luhar and Britter* [1989] chose $C_0 \simeq 2$ and showed that their simulation "reproduces the experimental concentration contours qualitatively and quantitatively quite well."

7. CONCLUSION

The science of what are now called Lagrangian stochastic models of dispersion has matured, but there remains room for fundamental progress, particularly as regards further selection criteria for the multidimensional case. LS models are being used to good effect on scales from the individual farmer's field or the city block, to the intercontinental, and indeed, they have assumed a vital social role, nicely exemplified by their application to predict the hazards of volcanic ash clouds [e.g., *Stohl et al.*, 2011; *Webster et al.*, 2012] or to locate or quantify sources of air pollutants or greenhouse gases [e.g., *Hirdman et al.*, 2010; *Manning et al.*, 2011].

Our short history has excluded many variants of the LS approach to turbulent dispersion, notably multiparticle models capable of predicting higher moments of concentration [*Durbin*, 1980b; *Thomson*, 1990; *Borgas and Sawford*, 1994]; models of higher order, based on a generalized Langevin equation for acceleration [*Sawford*, 1991]; the class of first-order models that add a further state variable, e.g., temperature may be added in order to model stably stratified flows or buoyant plumes [*Csanady*, 1964; *Pearson et al.*, 1983; *van Dop*, 1992; *Das and Durbin*, 2005], while gas concentration can be added as a state variable in an alternative approach to modeling concentration fluctuations (the so-called micromixing models) [e.g. *Pope*, 1998; *Cassiani et al.*, 2005; *Sawford*, 2004; *Luhar and Sawford*, 2005]; models for heavy particles [e.g., *Reid and Crabbe*, 1980; *Wilson*, 2000] or heavy gases [e.g., *Anfossi et al.*, 2010]; models with chemistry [e.g., *Chock and Winkler*, 1994a, 1994b; *Stevenson et al.*, 1998]; and models that compute the statistics of the flow field [e.g., *Pope*, 1994; *Bakosi et al.*, 2009]. As meteorologists, we have preferred to focus on LS modeling of the atmosphere. Of course parallel efforts exist in other disciplines, e.g., engineering and oceanography, but on the whole the field of research has remained reasonably well integrated across scientific communities.

In documenting the history of ideas in any field there is the certainty of making omissions. The authors were discomfited

to uncover, in the course of preparing this summary, pertinent papers of which they had been entirely unaware. No doubt others have gone unmentioned, and we perhaps have simplified the lines of development in a way that largely reflects the incidental factor of our own involvement in this field. We apologize to anyone whose contributions we have overlooked, and we thank the organizers of the Chapman Conference on Lagrangian models for the opportunity to present this retrospective on the subject, however uneven and incomplete it may be.

REFERENCES

Anfossi, D., G. Tinarelli, S. Trini-Castelli, M. Nibart, C. Olry, and J. Commanay (2010), A new Lagrangian particle model for the simulation of dense gas dispersion, *Atmos. Environ.*, *44*, 753–762.

Baerentsen, J. H., and R. Berkowicz (1984), Monte Carlo simulation of plume dispersion in the convective boundary layer, *Atmos. Environ.*, *18*, 701–712.

Bakosi, J., P. Franzese, and Z. Boybeyi (2009), Joint PDF modelling of turbulent flow and dispersion in an urban street canyon, *Boundary Layer Meteorol.*, *131*, 245–261.

Batchelor, G. K. (1949), Diffusion in a field of homogeneous turbulence. I. Eulerian analysis, *Aust. J. Sci. Res.*, *2*, 437–450.

Batchelor, G. K. (1957), Diffusion in free turbulent shear flows, *J. Fluid Mech.*, *3*, 67–80.

Batchelor, G. K., (1964), Diffusion from sources in a turbulent boundary layer, *Arch. Mech. Stosow.*, *3*, 661–670.

Borgas, M. S., and B. L. Sawford (1994), A family of stochastic models for two-particle dispersion in isotropic homogeneous stationary turbulence, *J. Fluid Mech.*, *279*, 69–99.

Borgas, M. S., T. K. Flesch, and B. L. Sawford (1997), Turbulent dispersion with broken reflectional symmetry, *J. Fluid Mech.*, *332*, 141–156.

Boughton, B. A., J. M. Delaurentis, and W. E. Dunn (1987), A stochastic model of particle dispersion in the atmosphere, *Boundary Layer Meteorol.*, *40*, 147–163.

Bullin, J. A., and A. E. Dukler (1974), Stochastic modeling of turbulent diffusion with hybrid computer, *Environ. Sci. Technol.*, *8*, 156–165.

Cassiani, M., P. Franzese, and U. Giostra (2005), A PDF micromixing model of dispersion for atmospheric flow. Part I: Development of the model, application to homogeneous turbulence and to neutral boundary layer, *Atmos. Environ.*, *39*, 1457–1469.

Cassiani, M., A. Radicchi, and J. Albertson (2007), Modelling of concentration fluctuations in canopy turbulence, *Boundary Layer Meteorol.*, *122*, 655–681.

Chadam, J. (1962), *On a Theory of Turbulent Diffusion*, Air Force Cambridge Res. Lab., Bedford, Mass.

Chock, D. P., and S. L. Winkler (1994a), A particle grid air quality modeling approach, 1. The dispersion aspect, *J. Geophys. Res.*, *99*, 1019–1031.

Chock, D. P., and S. L. Winkler (1994b), A particle grid air quality modeling approach, 2. Coupling with chemistry, *J. Geophys. Res.*, *99*, 1033–1041.

Collins, W. J., R. G. Derwent, C. E. Johnson, and D. S. Stevenson (2002), A comparison of two schemes for the convective transport of chemical species in a Lagrangian global chemistry model, *Q. J. R. Meteorol. Soc.*, *128*, 991–1009.

Corrsin, S. (1963), Estimates of the relations between Eulerian and Lagrangian scales in large Reynolds number turbulence, *J. Atmos. Sci.*, *20*, 115–119.

Csanady, G. T. (1964), Turbulent diffusion in a stratified fluid, *J. Atmos. Sci.*, *21*, 439–447.

D'Amours, R., A. Malo, R. Servranckx, D. Bensimon, S. Trudel, and J.-P. Gauthier-Bilodeau (2010), Application of the atmospheric Lagrangian particle dispersion model MLDP0 to the 2008 eruptions of Okmok and Kasatochi volcanoes, *J. Geophys. Res.*, *115*, D00L11, doi:10.1029/2009JD013602. [Printed 116(D2), 2011.]

Das, S. K., and P. A. Durbin (2005), A Lagrangian stochastic model for dispersion in stratified turbulence, *Phys. Fluids*, *17*, 025109, doi:10.1063/1.1849184.

de Baas, A. F., H. van Dop, and F. T. M. Nieuwstadt (1986), An application of the Langevin equation for inhomogeneous conditions to dispersion in a convective boundary layer, *Q. J. R. Meteorol. Soc.*, *112*, 165–180.

Denmead, O. T., and E. F. Bradley (1985), Flux-gradient relationships in a forest canopy, in *The Forest-Atmosphere Interaction*, edited by B. A. Hutchison and B. B. Hicks, pp. 421–442, D. Reidel, Dordrecht, The Netherlands.

Dolman, A. J., and J. S. L. Wallace (1991), Lagrangian and K-theory approaches in modelling evaporation from sparse canopies, *Q. J. R. Meteorol. Soc.*, *117*, 1325–1340.

Draxler, R. R., and G. D. Hess (1997), Description of the HYSPLIT_4 modeling system, *NOAA Tech. Memo. ERL ARL-224*, NOAA Air Resour. Lab., College Park, Md.

Draxler, R. R., and G. D. Hess (1998), An overview of the HYSPLIT_4 modeling system of trajectories, dispersion, and deposition, *Aust. Meteorol. Mag.*, *47*, 295–308.

Du, S., J. D. Wilson, and E. Yee (1994), Probability density functions for velocity in the convective boundary layer, and implied trajectory models, *Atmos. Environ.*, *28*, 1211–1217.

Du, S., B. L. Sawford, J. D. Wilson, and D. J. Wilson (1995), Estimation of the Kolmogorov constant (C_0) for the Lagrangian structure function, using a second-order Lagrangian model of grid turbulence, *Phys. Fluids*, *7*, 3083–3090.

Durbin, P. A. (1980a), A random flight model of inhomogeneous turbulent dispersion, *Phys. Fluids*, *23*, 2151–2153.

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

Durbin, P. A. (1983), Stochastic differential equations and turbulent dispersion, *NASA Ref. Publ. 1103*, 73 pp., NASA Glenn Res. Cent., Cleveland, Ohio.

Durbin, P. A. (1984), Comments on Papers by Wilson et al. (1981) and Legg and Raupach (1982), *Boundary Layer Meteorol.*, *29*, 409–411.

Egbert, G. D., and M. B. Baker (1984), Comments on paper 'The effect of Gaussian particle-pair distribution functions in the statistical theory of concentration fluctuations in homogeneous turbulence' by B. L. Sawford (Q. J. April 1983, 109, 339–353), *Q. J. R. Meteorol. Soc.*, *110*, 1195–1199.

Faller, A. J., and P. C. Mignerey (1982), On one-dimensional turbulent dispersion in convergent and divergent flows, *Phys. Fluids*, *25*, 1306–1316.

Finnigan, J. J. (2000), Turbulence in plant canopies, *Annu. Rev. Fluid Mech.*, *32*, 519–571.

Flesch, T. K., and J. D. Wilson (1992), A two-dimensional trajectory-simulation model for non-Gaussian, inhomogeneous turbulence within plant canopies, *Boundary Layer Meteorol.*, *61*, 349–374.

Flesch, T. K., J. D. Wilson, and E. Yee (1995), Backward-time Lagrangian stochastic dispersion models, and their application to estimate gaseous emissions, *J. Appl. Meteorol.*, *34*, 1320–1332.

Flesch, T. K., J. D. Wilson, L. A. Harper, B. P. Crenna, and R. R. Sharpe (2004), Deducing ground-air emissions from observed trace gas concentrations: A field trial, *J. Appl. Meteorol.*, *43*, 487–502.

Forster, C., A. Stohl, and P. Seibert (2007), Parameterization of convective transport in a Lagrangian particle dispersion model and its evaluation, *J. Appl. Meteorol. Clim.*, *46*, 403–422.

Gardiner, C. W. (2004), *Handbook of Stochastic Methods for Physics, Chemistry and the Natural Sciences*, 3rd ed., 415 pp., Springer, New York.

Hall, C. D. (1975), The simulation of particle motion in the atmosphere by a numerical random-walk model, *Q. J. R. Meteorol. Soc.*, *101*, 235–244.

Haworth, D. C., and S. B. Pope (1986), A generalized Langevin model for turbulent flows, *Phys. Fluids*, *29*, 387–405.

Hay, J. S., and F. Pasquill (1959), Diffusion from a continuous source in relation to the spectrum and scale of turbulence, *Adv. Geophys.*, *6*, 345–364.

Hirdman, D., H. Sodemann, S. Eckhardt, J. F. Burkhart, A. Jefferson, T. Mefford, P. K. Quinn, S. Sharma, J. Ström, and A. Stohl (2010), Source identification of short-lived air pollutants in the Arctic using statistical analysis of measurement data and particle dispersion model output, *Atmos. Chem. Phys.*, *10*, 669–693.

Hsieh, C.-I., M. Siqueira, G. Katul, and C.-R. Chu (2003), Predicting scalar source-sink and flux distributions within a forest canopy using a 2-D Lagrangian stochastic dispersion model, *Boundary Layer Meteorol.*, *109*, 113–138.

Hurley, P. J., and W. L. Physick (1993), A skewed, homogeneous Lagrangian particle model for convective conditions, *Atmos. Environ., Part A*, *27*, 619–624.

Janicke, L. (1983), Particle simulation of inhomogeneous turbulent diffusion, in *Air Pollution Modelling and Its Application II*, edited by C. de Wispelaere, pp. 527–535, Plenum Press, New York.

Jones, A. R., D. J. Thomson, M. Hort, and B. Devenish (2007), The U.K. Met Office's next-generation atmospheric dispersion model, NAME III, in *Air Pollution Modeling and its Application XVII*, edited by C. Borrego and A.-L. Norman, pp. 580–589, Springer, New York.

Joynt, R. C., and D. R. Blackman (1976), A numerical model of pollutant transport, *Atmos. Environ.*, *10*, 433–442.

Kolmogorov, A. N. (1941a), The local structure of turbulence in incompressible viscous fluid for very large Reynolds numbers, *Dokl. Akad. Nauk SSSR*, *30*(4), 301–305. [Reprinted in *Proc. R. Soc. London, Ser. A*, *434*, 9–13, 1991. Also reprinted in Turbulence and Stochastic Processes: Kolmogorov's Ideas 50 Years On, Cambridge Univ. Press, Cambridge, U. K., 1991.]

Kolmogorov, A. N. (1941b), Dissipation of energy in the locally isotropic turbulence, *Dokl. Akad. Nauk SSSR*, *31*(1), 19–21. [Reprinted in *Proc. R. Soc. Lond., Ser. A*, *434*, 15–17, 1991. Also reprinted in *Turbulence and Stochastic Processes: Kolmogorov's Ideas 50 Years On*, Cambridge Univ. Press, Cambridge, U. K., 1991.]

Krasnoff, E., and R. L. Peskin (1971), The Langevin model for turbulent diffusion, *Geophys. Fluid Dyn.*, *2*, 123–146.

Langevin, P. (1908), Sur la théorie du mouvement brownien, *C. R. Hebd. Seances Acad. Sci.*, *146*, 530–533. [Reprinted in *An Introduction to Stochastic Processes in Physics*, John Hopkins Univ. Press, Baltimore, Md., 2002.]

Laubach, J., A. Taghizadeh-Toosi, R. R. Sherlock, and F. M. Kelliher (2012), Measuring and modelling ammonia emissions from a regular pattern of cattle urine patches, *Agric. Forest Meteorol.*, *156*, 1–17.

Lee, N., and A. E. Dukler (1976), Lagrangian simulation of dispersion in turbulent shear flow with a hybrid computer, *AIChE J.*, *22*, 449–455.

Lee, R. L., and E. Naslund (1998), Lagrangian stochastic particle model simulations of turbulent dispersion around buildings, *Atmos. Environ.*, *32*, 665–672.

Legg, B. J. (1983), Turbulent dispersion from an elevated line source: Markov chain simulations of concentration and flux profiles, *Q. J. R. Meteorol. Soc.*, *109*, 645–660.

Legg, B. J., and I. F. Long (1975), Turbulent diffusion within a wheat canopy: II. Results and interpretation, *Q. J. R. Meteorol. Soc.*, *101*, 611–628.

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 Meteorol.*, *24*, 3–13.

Lemons, D. S. (2002), *An Introduction to Stochastic Processes in Physics*, 110 pp., John Hopkins Univ. Press, Baltimore, Md.

Leuning, R., O. T. Denmead, A. Miyata, and J. Kim (2000), Source/sink distributions of heat, water vapour, carbon dioxide and methane in a rice canopy estimated using Lagrangian dispersion analysis, *Agric. Forest Meteorol.*, *104*, 233–249.

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

Lin, C. C. (1960a), On a theory of dispersion by continuous movements, *Proc. Natl. Acad. Sci. U. S. A.*, *46*, 566–570.

Lin, C. C. (1960b), On a theory of dispersion by continuous movements, II. Stationary Anisoptropic Processes, *Proc. Natl. Acad. Sci. U. S. A.*, *46*, 1147–1150.

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.

Luhar, A. K., and B. L. Sawford (2005), Micromixing modelling of concentration fluctuations in inhomogeneous turbulence in the convective boundary layer, *Boundary Layer Meteorol.*, *114*, 1–30.

Malo, A., R. Servranckx, D. Bensimon, and R. d'Amours (2011), A brief overview of atmospheric transport and dispersion models in the Environmental Emergency Response Section, Environ. Canada, New Brunswick, Canada.

Manning, A. J., S. O'Doherty, A. R. Jones, P. G. Simmonds, and R. G. Derwent (2011), Estimating UK methane and nitrous oxide emissions from 1990 to 2007 using an inversion modeling approach, *J. Geophys. Res.*, *116*, D02305, doi:10.1029/2010JD 014763.

Maryon, R. H., D. B. Ryall, and A. L. Malcolm (1999), The NAME 4 dispersion model: Science documentation, *Turbulence and Diffusion Note 262*, Met Off., Devon, U. K.

McGinn, S. M., and K. A. Beauchemin (2012), Dairy farm methane emissions using a dispersion model, *J. Environ. Qual.*, *41*, 73–79.

Monin, A. S., and A. M. Yaglom (1977), *Statistical Fluid Mechanics*, vol. 1, 769 pp., MIT Press, Cambridge, Mass.

Naslund, E., H. C. Rodean, and J. S. Nasstrom (1994), A comparison between two stochastic diffusion models in a complex three-dimensional flow, *Boundary Layer Meteorol.*, *67*, 369–384.

Nehrkorn, T., J. Eluszkiewicz, S. C. Wofsy, J. C. Lin, C. Gerbig, M. Longo, and S. Freitas (2010), Coupled weather research and forecasting-stochastic time-inverted Lagrangian transport (WRF-STILT) model, *Meteorol. Atmos. Phys.*, *107*, 51–64.

Novikov, E. A. (1986), The Lagrangian-Eulerian probability relations and the random force method for nonhomogeneous turbulence, *Phys. Fluids*, *29*, 3907, doi:10.1063/1.865778.

Obukhov, A. M. (1941), On the distribution of energy in the spectrum of turbulent flow, *Dokl. Akad. Nauk SSSR*, *32*(1), 22–24.

Obukhov, A. M. (1959), Description of turbulence in terms of Lagrangian variables, *Adv. Geophys.*, *6*, 113–116.

Pasquill, F., and F. B. Smith (1983), *Atmospheric Diffusion*, 3rd ed., 487 pp., Ellis Horwood Ltd., Chichester, U. K.

Pearson, H. J., J. S. Puttock, and J. C. R. Hunt (1983), A statistical model of fluid-element motions and vertical diffusion in a homogeneous stratified turbulent flow, *J. Fluid Mech.*, *129*, 219–249.

Philip, J. R. (1967), Relation between Eulerian and Lagrangian statistics, *Phys. Fluids*, *10*, S69–S71.

Poggi, D., G. Katul, and J. Albertson (2006), Scalar dispersion within a model canopy: Measurements and three-dimensional Lagrangian models, *Adv. Water Res.*, *29*, 326–335.

Poggi, D., G. G. Katul, and M. Cassiani (2008), On the anomalous behaviour of the Lagrangian structure function similarity constant inside dense canopies, *Atmos. Environ.*, *42*, 4212–4231.

Pope, S. B. (1987), Consistency conditions for random-walk models of turbulent dispersion, *Phys. Fluids*, *30*, 2374–2379.

Pope, S. B. (1994), On the relationship between stochastic Lagrangian models of turbulence and second-moment closures, *Phys. Fluids*, *6*, 973–985.

Pope, S. B. (1998), The vanishing effect of molecular diffusivity on turbulent dispersion: Implications for turbulent mixing and the scalar flux, *J. Fluid Mech.*, *359*, 299–312.

Postma, J. V., J. D. Wilson, and E. Yee (2011), Comparing two implementations of a micromixing model. Part II: Canopy Flow, *Boundary Layer Meteorol.*, *140*, 225–241.

Raupach, M. R. (1987), A Lagrangian analysis of scalar transfer in vegetation canopies, *Q. J. R. Meteorol. Soc.*, *113*, 107–120.

Raupach, M. R. (1989), A practical Lagrangian method for relating scalar concentrations to source distributions in vegetation canopies, *Q. J. R. Meteorol. Soc.*, *115*, 609–632.

Reid, J. D., and R. S. Crabbe (1980), Two models of long-range drift of forest pesticide aerial spray, *Atmos. Environ.*, *14*, 1017–1025.

Reynolds, A. M. (1998), On trajectory curvature as a selection criterion for valid Lagrangian stochastic dispersion models, *Boundary Layer Meteorol.*, *88*, 77–86.

Rodean, H. C. (1996), *Stochastic Lagrangian Models of Turbulent Diffusion*, *Meteorol. Monogr.*, vol. 48, 84 pp., Am. Meteorol. Soc., Boston, Mass.

Sawford, B. L. (1985), Lagrangian statistical simulation of concentration mean and fluctuation fields, *J. Clim. Appl. Meteorol.*, *24*, 1152–1166.

Sawford, B. L. (1986), Generalized random forcing in random-walk turbulent dispersion models, *Phys. Fluids*, *29*, 3582–3585.

Sawford, B. L. (1991), Reynolds number effects in Lagrangian stochastic models of turbulent dispersion, *Phys. Fluids A*, *3*, 1577, doi:10.1063/1.857937.

Sawford, B. L. (1999), Rotation of trajectories in Lagrangian stochastic models of turbulent dispersion, *Boundary Layer Meteorol.*, *93*, 411–424.

Sawford, B. L. (2001), Project Prairie Grass – A classic atmospheric dispersion experiment revisited, paper presented at 14th Australasian Fluid Mechanics Conference, Adelaide Univ., South Australia, Australia.

Sawford, B. L. (2004), Micro-mixing modelling of scalar fluctuations for plumes in homogeneous turbulence, *Flow Turbul. Combust.*, *72*, 133–160.

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., and F. M. Guest (1988), Uniqueness and universality of Lagrangian stochastic models of turbulent dispersion, paper presented at 8th Symposium on Turbulence and Diffusion, Am. Meteorol. Soc., San Diego, Calif.

Siqueira, M., C.-T. Lai, and G. Katul (2000), Estimating scalar sources, sinks, and fluxes in a forest canopy using Lagrangian, Eulerian, and hybrid inverse models, *J. Geophys. Res.*, *105*, 29,475–29,488.

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

Sreenivasan, K. R. (1999), Fluid turbulence, *Rev. Mod. Phys.*, *71*, S383–S395.

Sreenivasan, K. R., and R. A. Antonia (1997), The phenomenology of small scale turbulence, *Annu. Rev. Fluid Mech.*, *29*, 435–472.

Stevenson, D. S., W. J. Collins, C. E. Johnson, and R. G. Derwent (1998), Intercomparison and evaluation of atmospheric transport in a Lagrangian model (STOCHEM), and Eulerian model (UM), using ^{222}Rn as a short-lived tracer, *Q. J. R. Meteorol. Soc.*, *124*, 2477–2491.

Stohl, A., C. Forster, A. Frank, P. Seibert, and G. Wotawa (2005), Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, *Atmos. Chem. Phys.*, *5*, 2461–2474.

Stohl, A., et al. (2011), Determination of time- and height-resolved volcanic ash emissions and their use for quantitative ash dispersion modeling: The 2010 Eyjafjallajokull eruption, *Atmos. Chem. Phys.*, *11*, 4333–4351.

Sutton, O. G. (1953), *Micrometeorology: A Study of Physical Processes in the Lowest Layers of the Earth's Atmosphere*, 333 pp., McGraw-Hill, Columbus, Ohio.

Sutton, O. G. (1959), Introductory remarks by session chairman, *Adv. Geophys.*, *6*, 437–439.

Taylor, G. I. (1921), Diffusion by continuous movements, *Proc. London Math. Soc.*, *s2-20*, 196–212.

Tennekes, H. (1979), The exponential Lagrangian correlation function and turbulent diffusion in the inertial subrange, *Atmos. Environ.*, *13*, 1565–1567.

Thompson, R. (1971), Numeric calculation of turbulent diffusion, *Q. J. R. Meteorol. Soc.*, *97*, 93–98.

Thompson, R. L., C. Gerbig, and C. Rödenbeck (2011), A Bayesian inversion estimate of N_2O emissions for western and central Europe and the assessment of aggregation errors, *Atmos. Chem. Phys.*, *11*, 3443–3458, doi:10.5194/acp-11-3443-2011.

Thomson, D. J. (1984), Random walk modelling of diffusion in inhomogeneous turbulence, *Q. J. R. Meteorol. 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.

Thomson, D. J. (1990), A stochastic model for the motion of particle pairs in isotropic high-Reynolds-number turbulence, and its application to the problem of concentration variance, *J. Fluid Mech.*, *210*, 113–153.

Thomson, D. J., and M. R. Montgomery (1994), Reflection boundary conditions for random walk models of dispersion in non-Gaussian turbulence, *Atmos. Environ.*, *28*, 1981–1987.

Thomson, D. J., W. L. Physick, and R. H. Maryon (1997), Treatment of interfaces in random walk dispersion models, *J. Appl. Meteorol.*, *36*, 1284–1295.

Todd, R. W., N. A. Cole, M. B. Rhoades, D. B. Parker, and K. D. Casey (2011), Daily, monthly, seasonal, and annual ammonia emissions from southern high plains cattle feedyards, *J. Environ. Qual.*, *40*, 1090–1095.

van Dop, H. (1992), Buoyant plume rise in a Lagrangian framework, *Atmos. Environ., Part A*, *26*, 1335–1346.

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 Ulden, A. P. (1978), Simple estimates for vertical diffusion from sources near the ground, *Atmos. Environ.*, *12*, 2125–2129.

Warland, J. S., and G. W. Thurtell (2000), A Lagrangian solution to the relationship between a distributed source and concentration profile, *Boundary Layer Meteorol.*, *96*, 453–471.

Webster, H. N., et al. (2012), Operational prediction of ash concentrations in the distal volcanic cloud from the 2010 Eyjafjallajökull eruption, *J. Geophys. Res.*, *117*, D00U08, doi:10.1029/2011JD016790.

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.

Weinstock, J. (1976), Lagrangian-Eulerian relation and the independence approximation, *Phys. Fluids*, *19*, 1702–1711.

Willis, G. E., and J. W. Deardorff (1976), A laboratory model of diffusion into the convective planetary boundary layer, *Q. J. R. Meteorol. Soc.*, *102*, 427–445.

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.

Wilson, J. D. (2000), Trajectory models for heavy particles in atmospheric turbulence: Comparison with observations, *J. Appl. Meteorol.*, *39*, 1894–1912.

Wilson, J. D., and T. K. Flesch (1997), Trajectory curvature as a selection criterion for valid Lagrangian stochastic dispersion models, *Boundary Layer Meteorol.*, *84*, 411–425.

Wilson, J. D., and B. L. Sawford (1996), Lagrangian stochastic models for trajectories in the turbulent atmosphere, *Boundary Layer Meteorol.*, *78*, 191–210.

Wilson, J. D., G. W. Thurtell, and G. E. Kidd (1981a), Numerical simulation of particle trajectories in inhomogeneous turbulence. I. Systems with constant turbulent velocity scale, *Boundary Layer Meteorol.*, *21*, 295–313.

Wilson, J. D., G. W. Thurtell, and G. E. Kidd (1981b), Numerical simulation of particle trajectories in inhomogeneous turbulence. II. Systems with variable turbulent velocity scale, *Boundary Layer Meteorol.*, *21*, 423–441.

Wilson, J. D., G. W. Thurtell, and G. E. Kidd (1981c), Numerical simulation of particle trajectories in inhomogeneous turbulence. III. Comparison of predictions with experimental data for the atmospheric surface-layer, *Boundary Layer Meteorol.*, *21*, 443–463.

Wilson, J. D., B. J. Legg, and D. J. Thomson (1983), Calculation of particle trajectories in the presence of a gradient in turbulent-velocity variance, *Boundary Layer Meteorol.*, *27*, 163–169.

Wilson, J. D., E. Yee, N. Ek, and R. d'Amours (2009), Lagrangian simulation of wind transport in the urban environment, *Q. J. R. Meteorol. Soc.*, *135*, 1586–1602.

Wilson, J. D., E. Yee, N. Ek, and R. d'Amours (2010), Erratum to "Lagrangian simulation of wind transport in the urban environment," *Q. J. R. Meteorol. Soc.*, *136*, 552.

Wilson, J. D., T. K. Flesch, and B. P. Crenna (2012), Estimating surface-air gas fluxes by inverse dispersion using a backward Lagrangian stochastic trajectory model, in *Lagrangian Modeling of the Atmosphere*, *Geophys. Monogr. Ser.*, doi:10.1029/2012GM 001269, this volume.

Yaglom, A. M. (1973), Semi-empirical equations of turbulent diffusion in boundary layers, *Fluid Dyn. Trans.*, *7*(II), 99–144.

D. J. Thomson, Met Office, FitzRoy Road, Exeter EX1 3PB, U.K. (david.thomson@metoffice.gov.uk)

J. D. Wilson, Department of Earth and Atmospheric Sciences, 1-26 Earth Sciences Building, University of Alberta, Edmonton, AB T6G2E3, Canada. (jaydee.uu@ualberta.ca)