REVIEW OF LAGRANGIAN STOCHASTIC MODELS FOR TRAJECTORIES IN THE TURBULENT ATMOSPHERE

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Abstract. We review the theoretical basis for, and the advantages of, random flight models for the trajectories of tracer particles in turbulence. We then survey their application to calculate dispersion in the principal types of atmospheric turbulence (stratified, vertically-inhomogeneous, Gaussian or non-Gaussian turbulence in the surface layer and above), and show that they are especially suitable for some problems (e.g., quantifying ground emissions).

1. Introduction

The purpose of this brief review is to survey up-to-date Lagrangian models for transport and mixing in atmospheric turbulence. A "Lagrangian stochastic" model describes the paths of particles in a turbulent flow, *given* a knowledge (i.e., statistical description) of the random velocity field. It is the natural and most powerful means to describe many interesting atmospheric processes (e.g., the dispersion of pollen, or of air pollutants), and with the aid of such models we can expect eventually to develop better strategies for, as an example, the application of aerial sprays.

That said, we will restrict this review to models of "passive" material, neutrally-buoyant, and non-reactive tracer "particles," or marked fluid elements. We exclude buoyant particles, because their treatment is more difficult, and lacks the guidance recently provided for models of passive tracer transport. We consider then, the short range transport (order 100 km or less) of passive tracer in the atmospheric boundary layer (ABL), where the turbulence is inhomogeneous (in the vertical direction z, if not in x and y), possibly non-stationary, and characterised by having a large Reynolds number, $\mathrm{Re} = UD/\nu$ (where U is a characteristic velocity, D is the ABL depth, and ν is the kinematic viscosity of air). We shall avoid duplicating Sawford's (1985) review, much of which might usefully be read in parallel with this one, and concentrate on subsequent developments. Sawford (1993) has given a selective overview of some modern developments, and a more detailed review which emphasises mathematical developments is forthcoming (Rodean, 1996).

Taylor (1921) initiated the Lagrangian description of turbulent transport, but considered only homogeneous turbulence. Early attempts (in the computer age) to mimic the far more complex case of atmospheric turbulence (e.g., Wilson *et al.*, 1981a,b) were heuristic, but much clarification has recently occurred. In particular, Thomson (1987) provided extremely helpful criteria for models of neutral tracer,

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resolving many of the difficulties with preceding models. *Eulerian* (and therefore measurable) information, on the velocity probability distribution function (pdf) of the background flow, can now be rigorously exploited to build a *Lagrangian* velocity time series. A Lagrangian Stochastic (LS) model for a given atmospheric flow can therefore be derived from an economical set of principles, the assumptions being few and explicit (we do not say "the LS model", for in many cases we have as yet no unique model). Such trajectory models have already been shown to capture in impressive detail many, even most, features of the mean concentration field from atmospheric tracer experiments.

2. Theory of Lagrangian Stochastic Models

2.1. Lagrangian versus Eulerian

Most fluid dynamicists are used to treating fluid flow and the transport of scalar material in an Eulerian reference frame in which properties such as the fluid velocity, $\mathbf{u}(\mathbf{x}, t)$, and the concentration of scalar material, $c(\mathbf{x}, t)$, are defined at a fixed point at time t. The evolution of the velocity and concentration are then described by the Navier–Stokes equations and the scalar conservation equation:

$$\frac{\partial c}{\partial t} + u_i \cdot \nabla c = \kappa \nabla^2 c \tag{1}$$

where κ is the molecular diffusivity (we have ignored internal source terms).

For turbulent flows usually we are interested in statistics of the velocity and concentration fields. The problem with these Eulerian equations is that they contain nonlinear advection terms and so the evolution equations for the *mean* velocity $\langle \mathbf{u} \rangle$ and *mean* concentration $\langle c \rangle$ are not closed; i.e., they involve unknown higher order statistics such as the Reynolds stresses $\langle \mathbf{u}'_i \mathbf{u}'_j \rangle$ and scalar flux densities, $\langle \mathbf{u}'_i c' \rangle$ (note: $\langle\ \rangle$ denotes mean values, and primes fluctuations about the mean). Closure approximations introduced to overcome this insuperable problem depend on the concentration field itself, and so are not uniformly valid. For example, the gradient transfer hypothesis, in which the flux density $\langle \mathbf{u}'_i c' \rangle$ is assumed a linear function of the mean gradient $\partial_j \langle c \rangle$, leads to the diffusion equation, but requires that the length scale of the turbulence be small compared with that of the contaminant distribution. Thus, as is well known, the diffusion equation breaks down close to point sources. For more detail on fundamental limitations of Eulerian methods, see Deardorff (1978).

The mass conservation Equation (1) has a simpler Lagrangian form

$$\frac{\mathrm{d}c}{\mathrm{d}t} = \kappa \nabla^2 c \tag{2}$$

which suggests an alternative approach: to describe the concentration (and flow) fields in a reference frame which follows a point moving with the fluid. The position $\mathbf{X}$ and velocity $\mathbf{U}$ of that point, at any time t, are in general functions of its position $\mathbf{x}_0$ at some earlier labelling time t_0 and of the labelling time itself. At each instant of time the "fluid point" or "fluid particle" moves with the Eulerian velocity at the point at which it is located, $\mathbf{U} = \mathbf{u}[\mathbf{X}(\mathbf{x}_0(t_0), t_0), t]$; capitals denote Lagrangian velocity and position. The main advantage of working in a Lagrangian framework is that the time derivative following the motion includes the non-linear advection terms implicitly, without approximation. Thus in the case of the velocity, the closure approximations we will introduce do not involve these non-linear terms. For the scalar concentration, in the limit of infinite Reynolds number, molecular diffusion can be neglected. The conservation equation for scalar concentration is then trivial, and merely states that the fluid particle retains its original concentration as it moves through the fluid; i.e., $\mathrm{d}c/\mathrm{d}t = 0$. This leads to the idea of "marked fluid particles". Since these marked particles conserve their concentration, changes in the concentration field occur solely due to their redistribution throughout the fluid. In particular, concentration statistics of the tracer material are directly related to displacement statistics of the marked particles. For example, the ensemble mean concentration is (e.g., Tennekes and Lumley, 1972).

$$\langle c(\mathbf{x}, t)\rangle = \int_{-\infty}^{t} \int_V P(\mathbf{x}, t; \mathbf{x}', t') S(\mathbf{x}', t')\, \mathrm{d}\mathbf{x}'\, \mathrm{d}t' \tag{3}$$

where: V denotes the entire volume of the fluid; $P(\mathbf{x}, t; \mathbf{x}', t')$ is the probability density for the position $\mathbf{X}$ at time t of those particles which were at $\mathbf{x}'$ at time t'; and $S(\mathbf{x}', t')$ is the source distribution of the material in question.

In this Lagrangian approach, $P(\mathbf{x}, t; \mathbf{x}', t')$ is usually determined by modelling the Lagrangian velocity. In general this does not avoid the need for approximations – the closure problem still remains. However, any approximations made now involve only the velocity field. The calculation of the concentration field through (3) is a completely separate and essentially exact process. The assumptions made in Lagrangian modelling are thus independent of the concentration field.

2.2. Markov Assumption

Lagrangian methods are now perhaps most often associated with stochastic models of dispersion in which fluid particle trajectories are generated numerically ("Random Flights"), in order to estimate concentration statistics such as those in (3). We assume the Eulerian flow statistics are known; indeed, for our purposes these Eulerian velocity statistics define the type of flow with which we deal. The problem is to generate particle trajectories, and hence displacement statistics and concentration statistics, in a physically realistic and self-consistent way.

The starting point for a modern Random Flight model is the postulate that the "state" of a particle evolves as a Markov process. In a "zeroth-order" RF model the "state" is simply the particle position $\mathbf{X}$ while in a first-order model the position and velocity jointly $(\mathbf{X}, \mathbf{U})$ are presumed Markovian. In the high Reynolds number turbulence of the atmosphere, the acceleration of fluid particles is autocorrelated only over times of order of the Kolmogorov time scale t_η, which is characteristic of the scales on which viscous processes convert turbulence kinetic energy (TKE) into heat (see e.g., Borgas and Sawford, 1991), and much smaller than the time scale of the energy containing eddies, T_L. Thus although the hypothesis of a Markovian process for the (joint) evolution of $(\mathbf{X}, \mathbf{U})$ can not be rigorously justified, it is a reasonable modelling assumption. Given the Markov assumption for $(\mathbf{X}, \mathbf{U})$, the most general equation which can be used to describe the velocity is the stochastic differential equation (SDE)

$$\mathrm{d}U_i = a_i(\mathbf{X}, \mathbf{U}, t) + b_{ij}(\mathbf{X}, \mathbf{U}, t)\,\mathrm{d}\omega_j \tag{4}$$

where $\mathrm{d}\omega_j$ is a component of Gaussian white noise, which is uncorrelated with other components and is uncorrelated in time; i.e.,

$$\langle \mathrm{d}\omega_j(t)\,\mathrm{d}\omega_i(t+\tau)\rangle = \delta_{ij}\delta(\tau)\,\mathrm{d}t\,\mathrm{d}\tau. \tag{5}$$

Particle position is obtained by integrating the velocity.

As a point of detail here, we note that the use of a stochastic differential equation is only appropriate if the Markov process for the velocity is continuous; i.e. if the velocity along a trajectory is a continuous function of time. Discontinuous or step Markov processes have also been used to model dispersion in turbulence (Smith, 1982; Smith and Thomson, 1984; Underwood, 1991; Wang and Stock, 1992). However, Sawford and Borgas (1994) have shown that such discontinuous processes are inconsistent with Kolmogorov's theory and with experimental observations in homogeneous turbulence.

The problem now is to determine the functions $\mathbf{a}$ and $\mathbf{b}$, known as the drift and diffusion terms respectively, for a particular turbulent flow field for which the Eulerian flow statistics are given. Following Thomson (1987), we achieve this by implementing two fundamental consistency conditions.

2.3. Consistency with Kolmogorov's Similarity Theory

The SDE (4) prescribes the Lagrangian velocity increment $\mathrm{d}U_i$ over an infinitesimal time increment $\mathrm{d}t$ in terms of a deterministic component and a random component. According to Kolmogorov's similarity theory for locally isotropic turbulence, for time increments within the inertial sub-range, the statistics of this velocity increment have a universal form, which depends only on the time increment and the mean rate of dissipation of turbulence kinetic energy, ε. In particular, the Lagrangian velocity structure function is of the form

$$\langle \mathrm{d}U_i\,\mathrm{d}U_j\rangle = \delta_{ij}C_0\varepsilon\,\mathrm{d}t \tag{6}$$

where C_0 is a universal constant (Monin and Yaglom, 1975, p. 358). This structure function can be evaluated directly for small times from (4, 5) and is consistent to $O(\mathrm{d}t)$ with (6) provided we choose

$$b_{ij} = \sqrt{C_0\varepsilon}\ \delta_{ij}. \tag{7}$$

Thus the coefficient of the random term in (4) is determined by the universal small-scale properties of turbulent flows, and is independent of the large-scale properties which determine the nature of the flow (i.e., boundary layer, jet etc.).

2.4. Eulerian Consistency

The difference between Eulerian and Lagrangian statistics is merely one of sampling. Eulerian statistics at $(\mathbf{x}, t)$ are determined from an unbiased sample of all trajectories passing through $\mathbf{x}$ at time t. On the other hand Lagrangian statistics at some time t are determined from all trajectories which emanate from some reference point. In principle it is possible to calculate both sorts of statistics from trajectories generated from the SDE (4). Thus, since we assume that the Eulerian statistics are known, they represent a constraint on the form of the SDE. This constraint is most easily implemented through the Fokker-Planck equation which is implied by (and equivalent to) the SDE (Gardiner, 1983). Details are given in Thomson (1987) and Sawford and Guest (1988). The outcome is a condition on the divergence in $\mathbf{u}$-space of the drift term $\mathbf{a}$,

$$\frac{\partial a_i P_E}{\partial u_i} = -\frac{\partial P_E}{\partial t} - \frac{\partial u_i P_E}{\partial x_i} + \frac{1}{2}C_0\varepsilon\frac{\partial^2 P_E}{\partial u_i \partial u_i} \tag{8}$$

where P_E is the Eulerian velocity probability density function, which we have assumed is known (or can be approximated). In general this equation does not have a unique solution for $\mathbf{a}$; an arbitrary rotational vector function $\phi(\mathbf{u})$ can be added to $\mathbf{a}P_E$ without altering the constraint equation (8). For complex flows there may be many Markov models consistent with the specified Eulerian velocity pdf. Specific solutions illustrating this non-uniqueness for Gaussian inhomogeneous turbulence are given by Sawford and Guest (1988), and Borgas *et al.* (1995) have shown that information *in addition to P_E* is in general required to resolve non-uniqueness (Section 3.1.3).

3. LS Models for Particular Flows

3.1. Homogeneous Turbulence

Much of the established theory of turbulence is restricted in its applicability to an idealised region of stationary, homogeneous and isotropic turbulence. Such turbulence is never observed (consideration of the mechanisms of turbulent kinetic energy generation and decay explain this), nevertheless approximations to it, e.g., decaying homogeneous turbulence in the wake of a wind-tunnel or water-channel grid, remain the simplest realisable turbulence in which to test developing theories. Observations show that the Eulerian velocity pdf in such simple flows is close to Gaussian.

3.1.1. *Isotropic Case*

If the turbulence is isotropic, then it can be shown that there is a unique solution to (8); i.e., $\phi = \mathbf{0}$ (Borgas and Sawford, 1994a, b). In the special case of Gaussian turbulence (i.e., $P_E\ (\mathbf{u}; \mathbf{x}, t)$ is a Gaussian) this unique solution is a Langevin equation for each of three independent, (i.e., uncoupled) velocity components,

$$\mathrm{d}U_i = -\frac{C_0\varepsilon}{2\sigma_u^2} U_i\,\mathrm{d}t + \sqrt{C_0\varepsilon}\,\mathrm{d}\omega_i \tag{9}$$

where σ_u is the standard deviation of the velocity fluctuations. In this case, the Lagrangian time scale

$$T_L = \frac{2\sigma_u^2}{C_0\varepsilon} \tag{10}$$

can be identified with the (Lagrangian) integral time scale (defined from the velocity autocorrelation; Tennekes, 1979). Note that uniqueness is a consequence of isotropy and the linearity (in $\mathbf{u}$) is a consequence of the Gaussian form of P_E.

For non-stationary but isotropic and homogeneous turbulence, an approximation which is often applied in theoretical treatments of grid turbulence (Sawford and Borgas, 1994), the solution is still unique. It was given by Sawford and Guest (1988),

$$a_i = \left(-\frac{1}{2}\frac{C_0\varepsilon}{\sigma_u^2} - \frac{1}{\sigma_u}\frac{\partial \sigma_u}{\partial t} \right) U_i. \tag{11}$$

Anand and Pope (1983) have given an analytical solution to (11) for the decaying turbulence analogue of grid turbulence; i.e., grid turbulence for which the Taylor transformation $x = Ut$, where U is the mean wind speed, is used to replace the downstream coordinate by time, thus transforming the stationary inhomogeneous turbulence of measurement to the decaying homogeneous turbulence of theory. Details of the solution for both the velocity statistics and the dispersion are also given by Sawford and Borgas (1994). The solution has been compared with wind-tunnel data for the dispersion of heat downstream of a line source. For the data of Warhaft (1984) and Stapountzis *et al.* (1986), Anand and Pope find good agreement in the far field using $C_0 = 2.1$, while for the data of Sawford and Tivendale (1992), Sawford and Borgas (1994) find $C_0 = 3$ gives the best fit.

3.1.2. *Uniform Mean Shear*

Wilson *et al.* (1993; WFS) constructed an analytical, well-mixed, 2-D LS model for trajectories in non-isotropic, stationary, Gaussian homogeneous turbulence having uniformly-sheared mean velocity field $\langle u\rangle(z) = U_0(1 + \alpha z)$. Their model was a heuristic generalisation of the Langevin equation; conditional mean acceleration a_i was assumed to be $a_i = -(U_i - \langle u_i\rangle)/T_L$. The b_{ij} were constrained by the well-mixed condition, such that inter-component velocity correlations were forced to arise through the random accelerations. In consequence, the model did not have the correct small time Lagrangian structure function (did not satisfy Equation (6)). WFS showed that at large times from release, streamwise spread is dominated not by alongwind "diffusion," which would involve the streamwise velocity variance σ_u^2, but by the joint action of vertical turbulent convection and differential advection in the mean shear. This result had been given much earlier, but is overlooked by those who would parameterise alongwind or crosswind spread by means of an eddy diffusivity. In the atmosphere, crosswind (y) spread may be largely due to turning of the mean wind with height, rather than to the fluctuations v' alone.

The WFS solution is of course invalid in inhomogeneous turbulence. However the near-source plume in complex turbulence can be regarded (approximately, and only sufficiently close to the source) as occupying a region of *locally*-homogeneous turbulence. WFS showed that their model will (sometimes) give a better prediction of the near field in inhomogeneous turbulence than does Taylor's (1921) solution.

3.1.3. *Skew Dispersion in an Idealised Anisotropic Turbulence*

Borgas *et al.* (1995; BFS) considered dispersion in homogeneous turbulence in the state of minimal departure from isotropy. Turbulence properties are taken to involve a special direction (Ω), with respect to which the turbulence is axisymmetric: statistics are symmetric when reflected in planes containing Ω, but reflections in the plane to which Ω is normal are not symmetric, so that when a mirror is held normal to Ω a change in flow screw-sense properties is observed. BFS gave a (non-unique) well-mixed LS model for this flow, and derived the implied Lagrangian velocity covariance function $\langle U_i(t)U_j(0)\rangle$ and the pattern of dispersion $\langle X_i(t)X_j(t)\rangle$. They revealed a remarkable suppression of the rate of dispersion in directions normal to the axis of symmetry, qualitatively explained by the tendency of trajectories to spiral around that axis. It is an implication of this work that non-uniqueness within a class of models that are well-mixed within Thomson's criteria can not be resolved unless flow properties *in addition to* P_E are respected.

3.2. GAUSSIAN INHOMOGENEOUS TURBULENCE

With the exception of canopy turbulence and turbulence in the convective boundary layer, it is usual to treat boundary-layer turbulence in the atmosphere (which here and elsewhere we will assume to be horizontally homogeneous) as Gaussian, but vertically-inhomogeneous. And except very close to the ground (or within a canopy; a case we consider later), the mean velocity is large with respect to the typical fluctuations (u'): thus dispersion in the streamwise direction is frequently ignored, or at least U is treated as independent of W. This leads to a focus on 1-component models for the vertical velocity, W.

3.2.1. *The Atmospheric Surface Layer*

The (unique) well-mixed, 1-component (here W) model for stationary Gaussian turbulence (Thomson, 1987) is given by (4, 5, 7) with $a_1 = a_2 = 0$, $a_3 = a_w$, $b_{11} = b_{22} = 0$ and $b_{33} = b_w$ where

$$a_w = -\frac{C_0\varepsilon(z)}{2\sigma_w^2(z)}W + \frac{1}{2}\frac{\partial\sigma_w^2}{\partial z}\left(\frac{W^2}{\sigma_w^2} + 1\right) \tag{12}$$

and

$$b_w = (C_0\varepsilon(z))^{1/2}. \tag{13}$$

A Lagrangian timescale $T_L(z)$ can still be defined through (10) for inhomogeneous turbulence, but can no longer be interpreted as an integral time scale. In the neutral surface layer, where $\sigma_w = 1.3u_*$, inhomogeneity arises solely through the TKE dissipation rate.

It can be shown that the model compared in detail with the Project Prairie Grass (PPG) field observations by Wilson *et al.* (1981a; WTK) is the discrete-time implementation of (12, 13). Wilson and Flesch (1993) studied the consequences of boundary treatment and time discretisation: any demixing of an initially well-mixed tracer field was attributed to the "Δt-bias error" that inevitably arises when trajectories in an inhomogeneous system are simulated with discrete time steps Δt. Using a time scale parameterisation equivalent to $C_0 = 3.1$, which value accords with latest estimates from simpler flows (Du *et al.*, 1995), WTK showed this LS model to be in excellent agreement with the observations.

They also obtained good agreement with unstable and stable PPG runs, using the parameterisations (which are consistent with $C_0 = 3.1$ in neutral conditions):

$$\frac{2\sigma_w^2}{C_0\varepsilon} = T_L(z) = \sigma_w^{-1}\frac{1}{2}z\left(1 - 6\frac{z}{L}\right)^{1/4}, \quad L < 0 \tag{14}$$

$$\frac{2\sigma_w^2}{C_0\varepsilon} = T_L(z) = \sigma_w^{-1}\frac{1}{2}z\left(1 + 5\frac{z}{L}\right)^{-1}, \quad L > 0 \tag{15}$$

where L is the Obukhov length. These formulae can be used to parameterise the compound variable $C_0\varepsilon$ in (modern) LS models of the equilibrium atmospheric surface layer.

We have focused here on *vertical* dispersion, and ignored the important question of the impact of the fluctuating alongwind component u', which is correlated with the vertical velocity, and of the lateral turbulence component. The inclusion of u' and/or v' presents a difficult problem. There is firstly the difficulty that as yet we do not have a *unique* well-mixed 2- or 3-dimensional LS model, even if we are prepared to accept Gaussian velocity pdfs. Particular 3-D solutions for **a** in Gaussian inhomogeneous turbulence, well-mixed (i.e., satisfying 8), have been given by Thomson (1987) and by Borgas (see Sawford and Guest, 1988), who compared calculations for a neutral surface layer with the wind-tunnel dispersion data of Legg (1983).

The second difficulty of including U, V, a difficulty not peculiar to LS modelling, is that u' and v' contain energy at much lower frequencies than w' – wind direction and speed change on a continuous range of scales, from the almost-microscopic (Kolmogorov) scale, up through scales of order 10 min (cloud/meso scale), to hourly and daily scales (changing large-scale pressure patterns). Therefore no matter what our choice of averaging time, there is always the likelihood of an *irregular* and possibly multi-modal distribution of material *along the horizontal direction(s)*. In general the (mean) mass distribution in x, y cannot be sharply predicted unless we can forecast the mesoscale fluctuations in wind direction. Practical means of accomplishing this involve prediction of the mesoscale wind variations with a prognostic mesoscale wind-field model and coupling these to a LS model for the turbulence. Such models are discussed in Section 5.1.

3.2.2. *Neutral Planetary Boundary Layer*

Davis (1983) used an LS model to study vertical dispersion in the neutral planetary boundary layer (NBL). Lagrangian vertical velocity was calculated using the algorithm of Legg and Raupach (1982), subsequently proven not to be a well-mixed model – however its application to the NBL was probably not seriously in error, since away from the ground, vertical inhomogeneity in σ_w is weak (inhomogeneity index $I_s = \sigma_w^{-1} T_L \partial \sigma_w^2 / \partial z \ll 1$).

Davis specified an effective Lagrangian timescale T_L^W for the vertical velocity above the surface layer, from published estimates derived from dispersion data, and specified T_L^U by considering the (observed) longitudinal velocity spectrum (inclusion of the correlated alongwind fluctuation U' had less than a 5% effect on the mean plume height and plume width). He calculated the effective plume width $\sigma_z(x)$, knowledge of which (usually empirical) parameter is critical to the ubiquitous Gaussian Plume Model. Results were in reasonable agreement with earlier-published $\sigma_z(x)$ curves for neutral stratification, that originated from tuning the Gaussian Plume model to the observed concentration field in dispersion experiments.

3.2.3. *The Stable Boundary Layer*

LS models, along with other aspects of turbulence and dispersion, are less well developed under strongly stable conditions. Luhar and Rao (1993) developed and tested three LS models for dispersion in katabatic flows assuming the turbulence to be Gaussian and inhomogeneous. One was the 1-D model (12); they also considered a 2-D model in which stream-wise dispersion was modelled explicitly, and a random displacement model (position of the particle modelled as a Markov process) was used to model vertical displacement. This latter model was chosen as the diffusion limit of (12), see Durbin (1983): vertical displacement $Z(t)$ is given by

$$\mathrm{d}Z = \left(\langle w \rangle + \frac{\partial K_z}{\partial z} \right) \mathrm{d}t + \sqrt{2K_z(z)}\, \mathrm{d}\omega(t) \tag{16}$$

where, to be consistent with (12) in the diffusion limit, the vertical diffusivity is

$$K_z = \sigma_w^2 T_L = \frac{2\sigma_w^4}{C_0 \varepsilon}. \tag{17}$$

Eulerian turbulence statistics required to drive the LS models were derived from the 2-D katabatic flow model of Nappo and Rao (1987), which is based on a TKE closure. For example, both the TKE dissipation rate and the velocity variances are diagnosed from the TKE and shear-production terms. Minor differences in the predictions of the three models were found.

3.3. Non-Gaussian Turbulence

3.3.1. *The Convective Boundary Layer*

The convective boundary layer (CBL) can be considered as comprising a fractional area (A) of "updrafts" (wherein the *mean* velocity is upward, but the instantaneous velocity need not be) and a complementary fractional area ($B = 1 - A$), considered the (predominantly) subsiding environmental region. In uniform terrain and under suitable restrictions, it is possible to define a "horizontally homogeneous" CBL and to again focus on *vertical* dispersion. The pdf $P_E(w; z)$ of the Eulerian vertical velocity in the CBL is non-Gaussian (as well as vertically inhomogeneous). Over much of the CBL, the skewness $Sk(z) = \langle w'^3 \rangle / \sigma_w^3$ is roughly 1/2.

Baerentsen and Berkowicz (1984; hereafter BB) constructed a skewed pdf $P_E(w; z)$ for the CBL as a linear combination of Gaussians for the updraft and downdraft regions,

$$P_E(w; z) = AG(w_A, \sigma_A) + BG(w_B, \sigma_B) \tag{18}$$

where $G(w, \sigma)$ represents a Gaussian pdf with mean w, and standard deviation σ. This form of pdf has been used in all subsequent LS studies of the CBL, except Du *et al.* (1994a; discussed later).

BB recognised their model should have the property that a well-mixed concentration field remains well-mixed. However the mathematical implications as expounded by Thomson (1984, 1987) were unknown: they constructed a heuristic LS algorithm which we will not detail, except to say that the random forcing was

Gaussian, and an effort was made to correct the algorithm for the effects of vertical inhomogeneity in the velocity statistics. They obtained good agreement with the laboratory data of Willis and Deardorff (1976, 1978, 1981), successfully simulating the "lift-off" from ground of the locus of maximum concentration downstream from a ground-level source, and the near-source decline (with increasing x) downstream from an elevated source (effects that are a consequence of the skewed velocity pdf).

Thomson (1984) examined a generalised Langevin model,

$$\mathrm{d}W = -\frac{W}{T}\,\mathrm{d}t + \mu. \tag{19}$$

He posed the question, "What is the probability density (for μ) required to ensure that the steady-state distribution of particles in (z, w) space has the same density function as the density function $P_E(w, z)$ of the air?" By determining that density function $f(\mu)$, Thomson showed for the first time how to obtain an exactly well-mixed model. However, as he put it, "it is not possible in practise to generate a random variable with all its moments specified in advance," and "in cases of strong inhomogeneity ... the values of higher moments of μ become important, making the model difficult to apply." Thomson showed that μ (if properly tailored to ensure the well-mixed property) must have a *skew* distribution even in *Gaussian* (but inhomogeneous) turbulence; but that if one formulates a stochastic equation

$$\mathrm{d}r = -\frac{r}{T}\,\mathrm{d}t + \mu \tag{20}$$

for the ratio $r = W/\sigma_w(z)$, a procedure introduced by Wilson *et al.* (1981b, 1983), then the correct forcing (μ) in Gaussian inhomogeneous turbulence is *Gaussian*.

de Baas *et al.* (1986) formulated an LS model (in W) with non-Gaussian forcing for the CBL. Guided by Thomson (1984), they attempted to tailor μ so as to account for the mean, variance, and skewness of the Eulerian velocity field (to do so necessitated some juggling in the choice of parameterisations for the vertical profiles of the Eulerian statistics). Generally good agreement was obtained with the available measurements, but despite its derivation from criteria designed to ensure the well-mixed property, this model seriously failed that condition. It is uncertain whether this was due to the use of perfect reflection at the boundaries, or to failure to exactly meet Thomson's specification for the statistics of the non-Gaussian forcing (μ).

Noting that the moments of any practicable non-Gaussian forcing can only be approximately those that guarantee the asymptotic well-mixed state, and that Thomson's stochastic equation in W/σ_w gave a better simulation of Gaussian inhomogeneous turbulence than did the corresponding model for W, Sawford and Guest (1987) formulated a stochastic equation in $W/\sigma_w(z)$, with non-Gaussian forcing. As hoped, their model was much closer to being "well-mixed" than that of de Baas *et al.* They obtained excellent agreement with the Willis-Deardorff observations, and by calculating tracer fluxes, revealed regions of counter-gradient transport (negative effective eddy diffusivity).

Subsequently Thomson (1987) showed that non-Gaussian forcing is incorrect. Following his provision of a general basis and selection criteria for LS models, it was natural to derive a well-mixed model (with Gaussian forcing) for the CBL. Given the Eulerian pdf, the well-mixed condition (as newly-enunciated and exploited mathematically) selects a unique 1-D model, and so 1-dimensional models were promptly formulated by Luhar and Britter (1989; LB) and by Weil (1990). In both cases the BB pdf (superposition of Gaussians) was retained, with minor differences in the specification of the fitting coefficients; and perfect reflection was applied at $z = 0$, and at the top of the boundary layer $z = z_i$. Parameterisations for the Eulerian velocity statistics differed markedly near the boundaries.

Wilson and Flesch (1993) studied these (very similar) models with an interest in the means by which particle containment was achieved, calculating the evolution of an initially well-mixed distribution of particles. In both cases, the well-mixed distribution was retained for a sufficiently small choice of the timestep. In the LB simulation, Wilson and Flesch noted that (for small enough timestep) the boundaries were unattainable (reflection never occurred). On the other hand Weil's simulation *required* the reflection algorithm, no matter how small Δt (reducing $\Delta t/\tau$ did not reduce the frequency of occurrence of reflection).

3.3.2. *Non-Gaussian Turbulence: Canopy Dispersion*

Turbulent velocity statistics within a crop or forest canopy are non-Gaussian, and extremely vertically inhomogeneous (inhomogeneity scale $I_s = O[1]$). Much of the vertical exchange is accomplished by intermittent large eddies, whose origin possibly owes to an instability with respect to the inflexion point in the wind profile. The pdf of the vertical velocity $P_E(w)$ is highly skewed, $Sk_w \approx -1$, and in much of the canopy space, turbulence intensity is large ($\sigma_{u,v} > U$).

We expect K-theory to be useful when the turbulence is "fine-grained" relative to the length scale over which there is curvature in the concentration distribution (Corrsin, 1974; for an extended explanation of the failure of K-theory in a canopy – see Wilson, 1989). Such is not (usually) the case in a canopy – a dramatic demonstration (that K-closure is unworkable) was given by Denmead and Bradley (1985), who by simultaneous measurement of mean profiles and eddy fluxes, demonstrated the routine occurrence of counter-gradient heat and vapour fluxes in a pine forest. Using a Lagrangian model, Raupach (1987) showed that in a canopy a substantial proportion of the scalar concentration (of heat or vapour) is contributed by the non-diffusive "near field" wakes of the nearby leaves, shedding heat and vapour to the airstream. It is natural, then, to turn to a Lagrangian description – irrespective of any wish to account for the highly non-Gaussian turbulence.

The first LS models were heuristic, not linked to the form of $P_E(w)$, and accounted only for the variance $\sigma_w^2(z)$. When applied to the extremely vertically-inhomogeneous canopy flow, these failed miserably the (as yet not clearly stated) well-mixed constraint: particles were found to spuriously accumulate in regions of small turbulent velocity scale (Wilson *et al.*, 1981b). These difficulties spurred ad hoc "drift corrections," until the clarification provided by Thomson (1984, 1987).

Since w-skewness is the signature of the intermittent large gusts, and since those gusts have been proven by quadrant analysis (e.g., Shaw *et al.*, 1983) to dominate the transport process, and furthermore since accounting for skewness had improved LS predictions of the CBL, one naturally expected an LS model properly derived from the skew Eulerian pdf ought to best handle canopy dispersion. Flesch and Wilson (1992) constructed such a model. Of course, if this was to simulate dispersion in a canopy well, it must include both the vertical and the alongwind fluctuations. Two problems then arise: (a) even given the joint pdf $P_E(u, w)$, Thomson's criteria do not provide a unique multi-dimensional LS model; and (b) given only the lower-order moments of u and w, what is the p.d.f.? Concerning (a), non-uniqueness was resolved by postulating that ϕ (as defined in Section 2.4) should act to conserve the *direction* of the Lagrangian velocity *fluctuation* vector. Concerning (b), following the idea of Baerentsen and Berkowizc, Flesch and Wilson (1992) superposed two *joint* Gaussians. Means and covariances of the individual joint-Gaussians were tailored to reproduce the lowest five moments of w, the lowest four moments of u, and the covariance $\langle u'w' \rangle$. This was quite arbitrary, and entailed the hazard that correctly fitting the low moments might constrain other moments to unrealistic values.

Flesch and Wilson simulated experiments (Legg *et al.*, 1986; Coppin *et al.*, 1986) in which heat was released as a tracer, from line and area sources, within a model crop in a turbulent wind-tunnel boundary layer. The surprising outcome of the work was that the more complex LS model which accounted for the known skewness and kurtosis of the canopy velocity statistics performed *worse* than Thomson's (1987) multi-dimensional model based on Gaussian Eulerian statistics! The authors suspected the cause was their adoption of an ad hoc pdf: the "best" (or least biased) choice would have been the maximum missing information pdf (see Section 4.3). However it may also be that the extreme vertical inhomogeneity of canopy flow (I_s order 1) in contrast to CBL flow ($I_s \ll 1$) implies a less critical dependence (of canopy dispersion) on the velocity skewness. It is pertinent here to quote Dr. J. Hunt*: "I believe that inhomogeneity will dominate over the niceties of third and fourth moments."

* Informal comment, 35^{th} Oholo Conference, Israel Institute for Biological Research, Eliat, Israel, 1991.

4. Details of Numerical Implementation

4.1. Discreteness of Time and Imposition of Boundaries

Except in cases that permit analytic solution, an LS model will be implemented with finite timestep Δt ($t_\eta \ll \Delta t \ll T_L$), and it is usual to tack onto the model an algorithm that ensures particles remain in the computational domain (not necessarily only because the timestep is finite). The best available criterion remains the well-mixed condition, whose satisfaction may be checked simply by calculating (numerically) the evolution of an initially well-mixed tracer distribution.

Proceeding more formally to investigate these numerical "issues," Wilson and Flesch (1993) showed how to write a discrete-time LS model that is "complete" (contains reflection; unambiguous order of operations) and how to deduce the implied transition probability density $P(z_2, w_2, t_2; z_1, w_1, t_1)$, the (z, w) phase space analogue of the displacement pdf in (3). By that means they proved that perfect reflection is *exactly* valid, if applied to bound Gaussian homogeneous turbulence (multi-dimensional models that include velocity covariance need to reverse both normal velocity W and the correlated alongwind velocity fluctuation U' upon reflection.) But (according to Wilson and Flesch) *no* reflection scheme can exactly satisfy the well-mixed condition when applied at a location where the pdf for the normal velocity is asymmetric, or locally-inhomogeneous. Since near a boundary there will usually be a region where the statistical character of the flow is anyway unknown, one is free to choose profiles of the flow statistics at the boundary that rebound to the success of a reflection algorithm.

The fact that there is no rigorous reflection algorithm to bound skew and/or inhomogeneous turbulence does not prohibit the existence of reflection algorithms that, for suitably small (but finite) Δt, are acceptable in practise. Suppose a particle crosses $z = 0$ with "incoming" velocity W^-. We wish to determine what outgoing velocity $W^+ = W^+(W^-)$ is correct, or at least, optimal. We require a 1-1 mapping from W^- to W^+, assumed continuous variables with pdf's P_E^-, P_E^+ that satisfy $P_E^-(W^-)\,\mathrm{d}W^- = P_E^+(W^+)\,\mathrm{d}W^+$. Since $W^- \le 0$, and $W^+ \ge 0$, we may require $P_E^-(W^-) = P_E(W^-)/\int_{-\infty}^0 P_E(W)\,\mathrm{d}W$ (and a similar expression for P_E^+). This implies that the $W^+(W^-)$ functional relationship must solve:

$$\frac{P_E(W^+)\,\mathrm{d}W^+}{\int_0^\infty P_E(W)\,\mathrm{d}W} = \frac{P_E(W^-)\,\mathrm{d}W^-}{\int_{-\infty}^0 P_E(W)\,\mathrm{d}W}. \tag{21}$$

Weil (1990) gave a (different) relationship $W^+(W^-)$ that, though not "correct," may better retain the proper pdf asymmetry in skew turbulence at the wall than does perfect reflection. Hurley and Physick (1993) reported an anonymous suggestion that the correct reflection scheme is

$$\int_{-\infty}^{W^-} W P_E(W)\,\mathrm{d}W = \int_{W^+}^{\infty} W P_E(W)\,\mathrm{d}W \tag{22}$$

4.2. Partially-Absorbing Boundaries

Uptake at the ground is usually parameterised in terms of a "deposition velocity", defined as the ratio $w_d = F/C_0$ of the flux density at the surface to a mean concentration C_0 measured at an arbitrary reference location (w_d is not necessarily independent of the choice of that location). Wilson *et al.* (1989) derived and tested an approximate means to achieve the correct effective deposition velocity, by *partial* reflection at the sink. For given w_d, the corresponding reflection probability R (they found) is:

$$\frac{1-R}{1+R} = \sqrt{\frac{\pi}{2}}\frac{w_d}{\sigma_w}. \tag{23}$$

The discrete-time statistical formalism (e.g., Wilson and Flesch, 1993) has yet to be applied to examine this strategy of partial absorption.

4.3. Implication of *Partial* Knowledge of the Flow

Since Thomson's (1987) provision of selection criteria for LS models, "known flow" has come to mean that the single-point pdf (P_E) of the Eulerian velocity field is a mathematically-prescribed function of position. But for any real flow, one has available only partial information, usually in the form of a few low-order velocity moments: the LS model must be built from *partial* information.

Du *et al.* (1994a) took the view that from the available information, the most rational course of action is to form what is called the "maximum missing information" (mmi) pdf (Jaynes, 1957). If the information given is an ordered set of moments $\mu_j (j = 1, 2, \ldots, N)$, the mmi pdf is

$$p(x) = \exp\left(-\sum_{k=0}^{N} \lambda_k x^k\right) \tag{24}$$

where the λ's are determined by the given moments. Du *et al.* constructed an mmi pdf for vertical velocity in the CBL, and derived the implied 1-D, well-mixed model for vertical dispersion. Differences from predictions of the earlier LS models based on a pdf formed by superposition of Gaussians were slight, and not unambiguously attributable to the alternate pdf. However, elsewhere Du *et al.* (1994b) demonstrated that forming the well-mixed model derived from an mmi pdf is the best solution yet provided to the problem of building an LS model from partial flow specification. And it is possible that the non-Gaussian 2-D LS model of Flesch and Wilson (1992) for dispersion in a canopy, which we discussed in Section 3.3.2, and which the authors showed to be *inferior* to an LS model based on a Gaussian pdf (even though the canopy turbulence was strongly non-Gaussian), failed for precisely the reason that the *arbitrary* pdf built to conformance with the specified flow moments was a "bad" choice.

5. Applications

From the foregoing it will be clear that for some applications the LS model is uniquely suited: alternative models are either inapplicable or inefficient (e.g., dispersion problems involving the near field of sources; or complex, recirculating windflows). We give two examples.

5.1. Transport and Dispersion on the Mesoscale

Recently several groups have coupled 3-D LS models with a mesoscale wind model, to address air quality problems – CSIRO in Australia (Physick *et al.*, 1993; Manins, 1995); and Colorado State University (Pielke *et al.*, 1992; Uliasz, 1993; McNider *et al.*, 1988). Strengths of combining a mesoscale wind field model and an LS model in this way include the ability to handle complex terrain, and to readily display the transport of pollutants using 2- or 3-D still and animated displays.

These models typically are applied over a domain of 50–500 km, with a resolution for the mean wind field of 1–10 km. Particle velocity consists of two components; that due to the resolved mean wind field (from the mesoscale model), and the turbulent fluctuation from the LS model. The mesoscale model provides parameters needed by the LS model, such as boundary-layer depth, and the surface fluxes of momentum and heat, and from these, turbulence variables (e.g., $\sigma_{u,v,w}$, T_L or ε, etc.) are diagnosed.

To reduce computation time, it may be necessary to simplify the LS model. The CSIRO model assumes (local) homogeneity in all three directions (permitting use of a long timestep), but under convective conditions includes the effect of skewness in W. Hurley and Physick (1993) showed that neglecting inhomogeneity in W near the ground (and inversion) is a reasonable approximation, but that skewness cannot be neglected. Hurley (1994) also proposed a technique ("PART-PUFF") in which horizontal dispersion is treated analytically, essentially modelling the vertical dispersion of puffs with a Gaussian distribution in the horizontal.

5.2. Source-Receptor Applications

"Backward" LS models calculate an ensemble of trajectories that are distinguished by each passing through a specified observation point, and are especially suitable for quantification of "source-receptor relationships." Consider, for example, the dimensionless ratio $UC/Q(= n)$, relating the strength (Q) of an extensive source (chemical spill; field emitting pesticide residue; etc.) to the mean concentration (C) at a nearby point (U is a reference windspeed). If n is known (e.g., from an LS model), Q can be inferred from the simpler measurements U, C. This approach was introduced for circular sources by Wilson *et al.* (1982).

In a horizontally-uniform atmosphere, this source-receptor relationship can be conveniently determined using a backward LS model. We firstly generate, for a given observation height h and atmospheric state $(u_*, L, \ldots)$, a catalog of "touch-down points": the locations (x_{0i}, y_{0i}) where the (backward) trajectories from $z = h$ reflect off ground. We store these points, and the associated (vertical) touchdown velocities (W_{0i}); i labels touchdowns (not particles). Then the flux-concentration relationship is (Flesch and Wilson, 1995):

$$n = \frac{UC}{Q} = \frac{1}{N} \sum_i L(x_{0i}, y_{0i}) \left(\frac{2U}{W_{0i}} \right) \tag{25}$$

where $L(x_{0i}, y_{0i})$ is an indicator function, with value unity if (x_{0i}, y_{0i}) lies within the (eventual) source region, zero otherwise; N is the number of backward trajectories in the archive; and i runs over all touchdowns (i may exceed N). The beauty of this approach is that the source need not be known or specified in advance; it may be of any shape, and in any position and orientation with respect to the mean wind vector and the observation point. A similar (and equally flexible) method (Flesch, 1996) determines the "footprint," i.e., the ground area contributing to the *vertical* flux at a given point.

6. Conclusion

Although continuing investigations of fundamental aspects of the single-particle LS model remain necessary (e.g., to resolve the non-uniqueness problem for models in multiple dimensions), most of the groundwork has been done. The modern 1-particle LS model is soundly-based in scientific principle, and provides an excellent description of absolute dispersion in the ABL, as judged by agreement with field observations. It is a simple and natural model of the process, and elegantly employs the available Eulerian information on a flow. In contrast Eulerian models, no matter the level of closure, are flawed through the need for assumptions on the joint concentration-velocity field, a crucial disadvantage for the description of the near field of a source (travel time not large with respect to turbulence timescale). For "far field" calculations, a simpler zeroth-order (i.e., "random displacement") Lagrangian model is legitimate, and Eulerian closure may be acceptable (an LS model can be used to provide guidance for Eulerian closure: e.g., van Dop *et al.*, 1985; Pope, 1994).

Our confidence in the single-tracer LS model stems from unambiguous criteria identifying those particular models (among all that intuition may throw up) that accord with established ideas regarding turbulence. Interesting challenges remain before we can have equal confidence in treating the simultaneous motion of multiple tracer particles (necessary to describe concentration fluctuation statistics), the motion of heavy particles and buoyant gases, or, cases in which the source itself significantly perturbs the flow (e.g., a buoyant industrial stack, or, a helicopter spraying pellets of insecticide). Such types of models, purporting validity of use in the atmosphere, already exist, but all are to some degree heurstic. In modelling some of these processes (e.g., the correlation in the driving fluid velocity along the path of a heavy particle) it seems doubtful that criteria might exist that will entirely dismiss need for guesswork! But then, the extent of the implications of the well-mixed criterion was unsuspected a decade ago, so an optimist may hope for further surprising discoveries.

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