A Simple Relative Dispersion Model for Concentration Fluctuations in Contaminant Clouds

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ABSTRACT

The relative dispersion process for clouds of contaminant in generic atmospheric flow is considered. The properties of the separation distance for pairs of particles are simplified by implicitly averaging over the spatial domain of the dispersing cloud. Representative statistics and simplified sets of measurements for characterizing two-particle dispersion in complex flows are identified. A Lagrangian stochastic model of relative dispersion equivalent to processes in homogeneous and isotropic turbulence at high Reynolds numbers is derived. The model uses a new formulation for parameterizing the acceleration of separation, satisfies the criterion of conserving a well-mixed distribution of particle separations, and accounts explicitly for non-Gaussian statistics of the turbulence velocity differences. The results are in very good agreement with similarity theory in the inertial range and are consistent with uncorrelated velocities at length scales larger than the turbulence integral scale. The model is applied to the estimation of fluctuating concentration fields, which is relevant for representing the relative dispersion part of popular meandering plume and puff approaches. The dependence of mean-square concentration and concentration fluctuations on the source size is eliminated via a new scaling law for the time, which in fact determines a universal behavior for the concentration field. Simple formulas are derived that are consistent with previous theories, and they are successfully tested against numerical simulations.

1. Introduction

Clouds of noxious chemicals are significant environmental hazards, and their behavior needs to be predicted for many public safety purposes. Such contaminant clouds are characterized by many factors, foremost being the highly variable fluctuating concentrations related to the highly variable shape and distribution of the cloud. Numerous models have been formulated, using a variety of approaches, to respond to the growing need for accurate estimates of the concentration fluctuations [see, e.g., Hanna (1984) for a review of concentration fluctuation models for continuous plumes and Lewis and Chatwin (1997), Thomson (1996), and Brown and Sawford (2000) for detailed models]. Such clouds or plumes are also commonly described by the relative dispersion properties, which control the instantaneous size of the cloud as opposed to its translation by the mean wind (Luhar et al. 2000).

To predict the mean-square concentration and relative dispersion, statistical modeling of the simultaneous trajectories of two particles is required [see, e.g., the recent review by Sawford (2001)]. The statistical approach for the description of concentration fluctuations parallels the evolution of the stochastic Lagrangian theory for the prediction of mean concentration fields.

A complete description of the trajectories of pairs of particles must include the formulation of the equations for positions in terms of either the six Cartesian coordinates of the two particles (Thomson 1990) or the three Cartesian coordinates of the pair center of mass plus the three components of the separation vector between the two particles. These formulations have six spatial coordinates and a corresponding number of velocity variables, so the mathematical description of the system is formidable. A simpler description just considers the magnitude of the separation vector, the so-called scalar separation, independent of the other variables.

The first model for scalar separation was proposed by Durbin (1982), who derived it from his original three-dimensional model (Durbin 1980). However, Thomson's (1990) analysis has shown that Durbin's model does not satisfy the well-mixed condition. Durbin's model also does not give inertial-range scaling for small-time La-

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grangian velocity increments (Borgas and Sawford 1991). Kurbanmuradov and Sabelfeld (1995) more recently gave an example of a scalar separation model that satisfies the well-mixed condition. However, because this model cannot be formulated in a closed form (i.e., the acceleration and the coefficient of the random term can only be obtained numerically), it is impractical for some applications and its computational efficiency is poor. In addition, the tails of the Eulerian probability density function (pdf) of the velocity differences fall off as a Gaussian distribution and do not reflect inertial-range behavior (Anselmet et al. 1984). The recent model of Reynolds (1999) is also an interesting preliminary model of relative dispersion, but it is not suited well as an applied model, nor does it formally indicate how averages of orientation of the separation vector lead to simple applications of scalar separation models in atmospheric flow.

In this paper, we present a stochastic model for the separation between particles of a passive contaminant released into a homogeneous, isotropic, and stationary turbulent flow at high Reynolds numbers. When the turbulence is isotropic, the orientation of the particle pairs is equally distributed in all directions. Although this is not the case in anisotropic turbulence (typical of the atmosphere), a model solely for the magnitude of the separation distance effectively averages over all orientations of the separation vector, and the properties for this distance are essentially equivalent to those in isotropic turbulence. Therefore, we model only the *magnitude* of separation, that is, the scalar separation, instead of the three components of the separation vector; the orientation of the pairs will be accounted for by implicit averaging.

In homogeneous turbulence, the approach used by Durbin (1980) proves convenient with its two systems of equations—one for the motion of the center of mass and one for the evolution of the separation. These two systems are coupled in a straightforward manner, and the relative dispersion can be solved independent of the center-of-mass dispersion. In addition, if the turbulence is isotropic, the orientation in space of each pair of particles is unimportant, and the three equations for the evolution of the separation vector reduce to a single equation for the magnitude of separation (Durbin 1982). For anisotropic atmospheric flow, provided that only the separation magnitude is considered, the same simplification to quasi-one-dimensional processes is available and is discussed in section 2. This averaged statistic is a robust indicator of typical separations within a cloud regardless of detailed atmospheric conditions and can be expected to be a measure of local dilution or levels of internal fluctuations in the cloud.

To formulate our model in section 4, we use a direct new formulation for the conditional acceleration that satisfies the well-mixed condition as well as consistency of the model with mean, variance, skewness, and kurtosis of the Eulerian distribution of velocity differences. The simple model that results from this approach has the advantage of very efficient computational times, which enables it to be used in applied problems, for example, within complex atmospheric flow models.

The dispersion model is applied to continuous (plume) and instantaneous (puff) releases from point sources. Because the scale of the fluctuations at the source is known to have an important effect on the second-order statistics of concentration, five different source sizes are considered with the focus on parameterizing this size dependence. This approach helps with analyzing scaling properties for near-source decay of internal fluctuations. The results presented in section 5 include simulations of mean-square concentration field $\overline{c^2}$, mean-square relative separation $\overline{r^2}$, mean concentration $\overline{c}$, standard deviation of concentration σ_c, and intensity of concentration fluctuations $\sigma_c/\overline{c}$. Also, a scaling law is defined that eliminates the dependence of these statistics on the source size and leads to analytical formulas describing their time decay. All of the simulation results are compared with those obtained from the application of the similarity theory in the inertial range.

Before discussing the model development and fluctuation calculations, we first elaborate (in section 2) on the simplification for relative dispersion provided by quasi-one-dimensional models. This identifies the scalar separation for pairs of particles as key and, furthermore, identifies the key Eulerian properties necessary for modeling.

2. Application of relative diffusion in the atmosphere

Practical atmospheric dispersion work with approximations provided by Gaussian plume or puff models, such as Industrial Source Complex (ISC3) and ''CALPUFF'' (Environmental Protection Agency 1995a–c), can describe bulk mean properties reasonably well with only a basic understanding of the turbulence in the atmosphere. To make progress on describing fluctuations of concentrations in atmospheric plumes in a similar manner is, at first sight, beyond the scope of current analytical techniques. This is mainly due to the complexity of the poorly known two-point flow statistics. For example, the *nine* two-point velocity correlations,

$$\overline{v_i(\mathbf{x}_1)v_j(\mathbf{x}_2)} \qquad (i, j = 1, 2, 3), \tag{1}$$

where the bar represents ensemble averaging and the subscripts i and j indicate vector components, are all formal inputs into two-particle Lagrangian stochastic models (Borgas and Sawford 1994a), but essentially none are well known for realistic flows and for general sampling positions $\mathbf{x}_1$ and $\mathbf{x}_2$. Thus the two-particle stochastic problem seems likely to be beyond detailed rational description for practical atmospheric flows. However, when the essential piece of information is the magnitude of the separation $r = |\mathbf{r}|$, which is a simple

scalar property (where $\mathbf{r}$ is the separation vector between two particles), much simpler statistics are required. To model generic properties of r, a quasi-one-dimensional model is required: say a Lagrangian stochastic equation for the rate of change, or separation velocity, $u = dr/dt$, where t is time. The appropriate Eulerian statistics for u are, however, novel. They are essentially averages over space and all orientations of the separation vector. Because u is defined as

$$u = [v_i(\mathbf{x}_1 + \mathbf{r}) - v_i(\mathbf{x}_1)]r_i/r, \tag{2}$$

where the convention of summation over repeated indices is adopted, instead of (1) we have a statistic of the form

$$\left\langle \overline{\left\{[v_i(\mathbf{x}_1 + \mathbf{r}) - v_i(\mathbf{x}_1)]\frac{r_i}{r}\right\}^2} \right\rangle = \frac{1}{4\pi}\int_{|\mathbf{r}|=r} \overline{\left\{[v_i(\mathbf{x}_1 + \mathbf{r}) - v_i(\mathbf{x}_1)]\frac{r_i}{r}\right\}^2}\, d\Omega_r, \tag{3}$$

where the integral is over all angles of the separation vector $\mathbf{r}$ (solid angles Ω_r). In addition, inhomogeneity can be accounted for by spatially averaging further over, say, $\mathbf{x}_1$. The detailed information required in (3) is not generally explicitly available, but because the important processes occur at small scale (e.g., while plumes remain smaller than the entire depth of the boundary layer), it is possible to use local inertial-range forms and to average over spatial variations of energy dissipation rate, mean velocity, and turbulent kinetic energy. The resulting averages for the fixed separation properties of $\langle\overline{u}\rangle$, $\langle\overline{u^2}\rangle$, $\langle\overline{u^3}\rangle$, and $\langle\overline{u^4}\rangle$ represent bulk separation-rate properties representative of the relevant atmospheric flow domain. These are simple functions of the separation r and, moreover, have simple inertial-subrange representation for small scales (with a bulk-averaged energy dissipation rate).

Because the plume structure is usefully thought of as a bulk meander with internal structure (Luhar et al. 2000; Yee and Wilson 2000; de Haan and Rotach 1998a,b; Rizza et al. 2000), the bulk representative separation behavior is the critical property, and detailed three-dimensional two-particle modeling is essentially redundant. Thus, the fundamentally useful model for applied concentration-fluctuation work is a stochastic model for separation r and separation rate u, consistent with the bulk-averaged moments. A remarkable thing is that this problem is mathematically identical to the problem in isotropic, homogeneous turbulence. The difference is in the detail of the spatial averages of the two-point Eulerian moments, and this difference affects the final parameters, with the local spatial structure (ignoring intermittency) being

$$\langle\overline{u}\rangle = 0, \qquad \langle\overline{u^2}\rangle \sim C_K\langle\varepsilon^{2/3}\rangle r^{2/3},$$
$$\langle\overline{u^3}\rangle \sim -(4/5)\langle\varepsilon\rangle r, \qquad \langle\overline{u^4}\rangle \sim C_4\langle\varepsilon^{4/3}\rangle r^{4/3}, \tag{4}$$

where the bar indicates ensemble averaging, and the angled brackets indicate space averaging. For example, $\langle\varepsilon\rangle$ is the local mean energy dissipation rate $\varepsilon(\mathbf{x})$ at position $\mathbf{x}$ in the boundary layer averaged over all positions $\mathbf{x}$ in the domain of interest (represented by angled brackets), that is, over the important part of the plume dispersal. The other constants in (4) are considered later, but it is already possible to develop a generic atmospheric model for relative dispersion, which is done in section 4.

3. Basic equations for second-order concentration statistics

In this section, we recall the general definition of mean-square concentration $\overline{c^2}$ according to the statistical theory of two-particle dispersion and derive an equation for $\overline{c^2}$ near plume and puff centers in terms of scalar separations in a frame of reference moving with the center of mass of the contaminant cloud. A relative dispersion model predicts, at instants in time, the distribution of separations for pairs of particles that were released from known sources at a given prior time. Once the joint pdf of the positions of two particles has been calculated, it is used to calculate the two-point covariance of concentration with the following expression (see, e.g., Batchelor 1952; Thomson 1990):

$$\overline{c(\mathbf{x}', t)c(\mathbf{x}'', t)} = \int_{t_o<t}\int p(\mathbf{x}', \mathbf{x}''; t\,|\,\mathbf{x}_o', \mathbf{x}_o''; t_o) \times \overline{S(\mathbf{x}_o', t_o)S(\mathbf{x}_o'', t_o)}\, d^3\mathbf{x}_o'\, d^3\mathbf{x}_o''\, dt_o, \tag{5}$$

where the vectors $\mathbf{x}'$ and $\mathbf{x}''$ represent the respective positions of particles labeled 1 and 2, the subscript o indicates conditions at the initial time t_o, $p(\mathbf{x}', \mathbf{x}''; t\,|\,\mathbf{x}_o', \mathbf{x}_o''; t_o)$ is the joint pdf of the positions of the two particles, and $S(\mathbf{x}_o, t_o)$ is the source strength distribution, that is, the distribution of the amount of material released per unit volume per unit time. The absolute second moment of the concentration, $\overline{c^2}(\mathbf{x}, t)$, can be obtained as the limit of (5) as $\mathbf{x}'$ and $\mathbf{x}''$ tend to the same position $\mathbf{x}$, at least to within some small separation of a scale much smaller than cloud dimensions (Durbin 1980).

For internal structure well within an almost homogeneous cloud, $\overline{c^2}(\mathbf{x}, t)$ can be approximated by evaluating it in a frame of reference moving with the mean advection velocity of a plume or puff and assuming that the fluctuations are almost homogeneous, at least away from the edge of the cloud. Another approach is to integrate $\overline{c^2}(\mathbf{x}, t)$ over the cloud cross section, so that a net mean-square concentration $\overline{c^2}(t)$ characterizes the plume or puff (Brown and Sawford 2000). From these simplifications, we can use $p(0; t\,|\,\mathbf{r}_o; t_o)$ proportional to $p(\mathbf{x}, \mathbf{x}; t\,|\,\mathbf{x}_o', \mathbf{x}_o''; t_o)$, and $\overline{c^2}(t)$ for $\overline{c^2}(\mathbf{x}, t)$, where $\mathbf{r}_o = \mathbf{x}_o' - \mathbf{x}_o''$. After this transformation, the time t after

release corresponds to the distance $x = Ut$ downwind of the source, where U is the advection velocity.

Next, we use the backward dispersion relationship $p(0; t \mid \mathbf{r}_o; t_o) = p(\mathbf{r}_o; t_o \mid 0; t)$ for incompressible flows (Thomson 1990), where $p(\mathbf{r}_o; t_o \mid 0; t)$ is the probability that two particles that are separated by the vector $\mathbf{r}$ (≈ 0) at the time t were separated by the small band of vectors $\mathbf{r}_o$ to $\mathbf{r}_o + d^3\mathbf{r}_o$ at the time t_o.

If, in addition, we assume instantaneous release, the source strength can be represented as $S(\mathbf{x}_o, t_o) = c(\mathbf{x}_o, t_o)\delta(t_o)$, where δ is the Dirac delta function, and $c(\mathbf{x}_o, t_o)$ is the source concentration distribution. Note that this representation is still a valid model for a plume, that is, a *continuous* release from a compact source, inasmuch as it is sufficient to represent $c(\mathbf{x}_o, t_o)$ as a virtual instantaneous *line* source, which starts from the actual source and extends in the along-wind direction (Thomson 1990).

Last, after averaging over all angles of the initial separation vector $\mathbf{r}_o$, we obtain the following expression:

$$\overline{c^2}(t) = \int_0^\infty p(r_o; t_o \mid 0; t) q(r_o)\, dr_o, \tag{6}$$

where $r = |\mathbf{r}|$, $p(r_o; t_o \mid 0; t) = 4\pi r_o^2 p(\mathbf{r}_o; t_o \mid 0; t)$ and $q(r_o) = \overline{c(x_o, t_o)c(x_o + r_o, t_o)}$. In the next section, we derive a stochastic model for scalar separations that will be used to calculate $p(r_o; t_o \mid 0; t)$, and hence $\overline{c^2}(t)$ through (6).

4. Model formulation

The scalar separation $r(t)$ between pairs of particles is modeled by a system of two stochastic differential equations, one for r itself, the other for its rate of change $u = dr/dt$. The scalar u is the longitudinal component of the three-dimensional velocity difference vector between two particles, namely

$$u = (v_i' - v_i'')r_i/r, \tag{7}$$

where the subscript i indicates a vector component, the vectors $\mathbf{v}'$ and $\mathbf{v}''$ represent the respective velocities of particles labeled 1 and 2, and the convention of summation over repeated indices is adopted. Again, for anisotropic flows we can formally define the angle-averaged longitudinal velocity u in (7), which removes the explicit role for orientation of the specific choices of $\mathbf{r}$.

For simplicity, we explicitly assume incompressible flows and isotropic turbulence in the high-Reynolds-number limit for all separation scales. Because the relative acceleration of two particles at high Reynolds number is practically uncorrelated in time (Monin and Yaglom 1975, p. 370; Borgas and Sawford 1991, 1994b), we can assume that the joint evolution of r and u is a Markov process and that the motion of independent pairs of fluid particles can be modeled by the following stochastic differential equations:

$$\begin{aligned} du(t) &= a(u, r)dt + (2C_o\varepsilon)^{1/2}dW \\ dr(t) &= u(t)dt, \end{aligned} \tag{8}$$

where dW are the random increments of a Wiener process with zero mean and variance dt, and the drift term $a(u, r)$, which will be determined below from a Fokker–Planck equation, incorporates Eulerian velocity structure functions and ε. The coefficient of the random increments, $(2C_o\varepsilon)^{1/2}$, ensures the consistency of (8) with the Lagrangian velocity structure function of the second order in the inertial range, which is written for the velocity of separation u as

$$\overline{[u(t + dt) - u(t)]^2} = 2C_o\varepsilon dt \qquad (dt \gg \tau_\eta), \tag{9}$$

where C_o is a universal constant and τ_η is the Kolmogorov microtimescale [$\tau_\eta = (\nu/\varepsilon)^{1/2}$, where ν is the kinematic viscosity]. To date there is no consensus on the value of C_o. For example, Anand and Pope (1985) estimate $C_o \approx 2$ based on a comparison with wind tunnel experiments on the thermal wake downstream of a heated wire in grid turbulence; Hanna (1981) obtains values of C_o ranging from about 2 to about 6, with an average $C_o \approx 4$, from spectral analysis of the velocity of neutral balloons released in the atmospheric boundary layer; Sawford's (1991) theoretical analysis supports values of C_o equal to or larger than 6, and recent direct numerical simulation calculations of Lagrangian statistics for homogeneous turbulence in uniform shear flow are consistent with a value of C_o equal to 6 (Sawford and Yeung 2001). In our simulations, we use $C_o = 6$.

The model is derived first by assuming a functional form for the relative acceleration $a(u, r)$ and then by imposing the well-mixed criterion as described by Thomson (1987), modified by Kurbanmuradov and Sabelfeld (1995) for the case of scalar separation models. We propose the following simple functional-form closure for $a(u, r)$:

$$a(u, r) = \alpha(r) + \beta(r)u + \gamma(r)u^2, \tag{10}$$

where the coefficients α, β, and γ are unknown functions of r. This form, a quadratic function of u, is of the same type as the one used by Franzese et al. (1999) in the context of one-particle models for the convective atmospheric boundary layer.

In accordance with the well-mixed criterion, below we will determine $a(u, r)$ by requiring consistency with the Fokker–Planck equation associated with the stochastic process (8).

The Eulerian pdf of the relative velocity $p_E(u; t \mid r)$, which is obtained by sampling all particle pairs at a given separation r, can be defined as the ensemble average of $p(u, r; t \mid u_o, r_o; t_o)$ over the set of initial conditions for three-dimensional pairs of trajectories (and then using the Jacobians of the transformation to scalar separation pdfs)

$$p_E(u;\, t|r) = \int \frac{r_o^2}{r^2} p(u,\, r;\, t|u_o,\, r_o;\, t_o) p_E(u_o|r_o,\, t_o)\, du_o\, dr_o. \quad (11)$$

By means of (11) and assuming statistically stationary conditions, the Fokker–Planck equation associated with (8) transforms into the following equation of evolution for $p_E(u;\, t \mid r)$ (Kurbanmuradov and Sabelfeld 1995):

$$\frac{u}{r^2}\frac{\partial(r^2 p_E)}{\partial r} = -\frac{\partial(a p_E)}{\partial u} + C_o \varepsilon \frac{\partial^2 p_E}{\partial u^2}, \quad (12)$$

where $p_E \equiv p_E(u \mid r)$. Because of the simple polynomial form of (10), the following hierarchy of equations can be obtained by multiplying (12) successively by powers of u and then integrating over u:

$$\alpha(r)\overline{u^{n-1}} + \beta(r)\overline{u^n} + \gamma(r)\overline{u^{n+1}} = \Gamma_{n+1}/n - (n-1)C_o\varepsilon\overline{u^{n-2}} \qquad n = 1, \ldots, N, \quad (13)$$

where

$$\Gamma_n = \frac{1}{r^2}\frac{\partial(r^2\overline{u^n})}{\partial r}. \quad (14)$$

System (13) involves the turbulence structure functions $\overline{u^n}$ and their derivatives with respect to r, as well as the coefficients α, β, and γ that appear in the drift term (10). The unknowns α, β, and γ are then obtained by solving (13) for $n = 1$, 2, and 3:

$$\gamma(r) = \frac{\Gamma_4/(3\overline{u^2}^2) - \overline{u^3}(\Gamma_3 - 2C_o\varepsilon)/(2\overline{u^2}^3) - \Gamma_2/\overline{u^2}}{\mu_4 - \mu_3^2 - 1}$$

$$\beta(r) = \frac{\Gamma_3 - 2\overline{u^3}\gamma(r) - 2C_o\varepsilon}{2\overline{u^2}}$$

$$\alpha(r) = \Gamma_2 - \overline{u^2}\gamma(r), \quad (15)$$

where μ_3 is the skewness and μ_4 is the kurtosis of $p_E(u;\, t \mid r)$ (i.e., $\mu_3 = \overline{u^3}/\overline{u^2}^{3/2}$ and $\mu_4 = \overline{u^4}/\overline{u^2}^2$). Therefore, the acceleration (10) is completely defined if the velocity structure functions up to the fourth order and ε are known.

This model represents a very efficient and simple parameterization of the problem and is superior to arbitrary assumptions about the functional form for p_E, which is the standard method for parameterizing Lagrangian stochastic models. In fact, (13) shows that the full set of moments is prescribed once the coefficients are set in (15). Thus, the closure prescribes a form for $p_E(u;\, t \mid r)$ that can be shown to be realistic. Furthermore, we know from Borgas and Yeung (1998) that quadratic form representations are a good fit to direct numerical simulation data for conditional accelerations.

Turbulence velocity structure functions and Eulerian probability of velocity differences

The velocity structure functions that are input to the model through (15) are derived from the formulas given in Borgas and Yeung (1998). These formulas were originally devised to fit results from direct numerical simulations of the turbulence at several different Reynolds numbers. The structure functions are for the increment u, which in the Eulerian sense becomes the standard longitudinal velocity increment

$$u = v_1(x_1 + r, x_2, x_3) - v_1(x_1, x_2, x_3), \quad (16)$$

with the indices labeling standard Cartesian axes in three-dimensional space. In the context of complex atmospheric flows, these structure functions are interpreted as composite averages over longitudinal increments in all directions and possibly over various positions in the boundary layer. Nevertheless, the character remains the same, with small-scale inertial-range properties blending into large-scale energy-containing properties.

In the limit as the Reynolds number tends to infinity the following parameterizations for the structure functions of second, third and fourth order are used:

$$\overline{u^2}(r) = 2(\varepsilon r)^{2/3}\left[\frac{1}{A_2 + (r/L)^2}\right]^{1/3}, \quad (17)$$

$$\overline{u^3}(r) = -\frac{4}{5}\varepsilon r\left[\frac{1}{1 + (r/L)^2}\right]^4, \quad \text{and} \quad (18)$$

$$\overline{u^4}(r) = 12(\varepsilon r)^{4/3}\left[\frac{1}{A_4 + (r/L)^2}\right]^{2/3}, \quad (19)$$

where L is a characteristic length scale for the energy-containing eddies that is defined as $L = \sigma_v^3/\varepsilon$, where σ_v is the standard deviation of the turbulent velocity fluctuations, and the constants A_2 and A_4 were determined by requiring that the structure functions (17) and (19) be consistent with inertial-range scaling. One obtains $A_2 = (2/C_K)^3$ and $A_4 = A_2(3/\mu_{4I})^{3/2}$, where C_K is the constant in the classical Kolmogorov formula for the second-order velocity structure function in the inertial range, namely $\overline{u^2} = C_K(\varepsilon r)^{2/3}$ (Kolmogorov 1941b), and μ_{4I} is the value assumed by the kurtosis μ_4 in the inertial range. Following the approach used by Borgas and Yeung (1998), we assume $C_K = 2.13$ and $\mu_{4I} = 3.4$.

Expression (17) is similar to that introduced by Durbin (1980) and since then widely used in relative dispersion modeling (see, e.g., Sawford and Hunt 1986; Thomson 1990; Borgas and Sawford 1994a). This expression reduces to $\overline{u^2} = C_K(\varepsilon r)^{2/3}$ in the inertial-range limit, namely for $\eta \ll r \ll L$, where $\eta = (\nu^3/\varepsilon)^{1/4}$ is the Kolmogorov microscale. The third-order structure function (18) similarly reduces, at inertial-range scales, to the exact relationship $\overline{u^3} = -(4/5)\varepsilon r$ (Kolmogorov 1941a).

Expressions (17), (18), and (19) are also consistent with the statistics of the Gaussian turbulence at large separations. As r/L tends to infinity, $\overline{u^2}$ tends to $2(\varepsilon L)^{2/3} \equiv 2\sigma_v^2$ (thus ensuring a vanishing correlation between turbulent velocities at points separated by a length $r \gg$

L), $\overline{u^3}$ vanishes, and $\overline{u^4}$ tends to $3\overline{u^2}^2$, consistent with the assumption that the pdf of the turbulent velocities is Gaussian (Townsend 1947). In complex atmospheric flows, such as convective turbulence, one-point statistics are not Gaussian (in the vertical), so large separation statistics will not tend to pure Gaussian behavior in the large time limit. However, these effects are not examined in this paper because we focus on small-scale relative dispersion properties.

Our model is now completely defined, in very simple terms, and because we did not assume any form for $p_E(u \mid r)$, but only used its second, third, and fourth moments, the drift term a is an algebraic function of u, allowing efficient computations.

5. Model results

We now use the model to estimate values of $\overline{c^2}$, $\overline{c}$, σ_c, and $\sigma_c/\overline{c}$ for the case of instantaneous and continuous releases from individual sources. Then, a simple scaling law that determines self-similar results independent of the source size is derived. Also, we derive analytical formulas for $\overline{c^2}$, $\overline{c}$, and σ_c far from the source through an application of the earlier scaling law combined with similarity theory relations and compare these formulas with our simulations.

a. Mean-square internal fluctuation field

The mean-square concentration field $\overline{c^2}(t)$ is calculated according to (6), which is reproduced here for convenience:

$$\overline{c^2}(t) = \int_0^\infty p(r_o;\, t_o \mid 0;\, t) q(r_o)\, dr_o. \tag{20}$$

Equation (20) shows that $\overline{c^2}(t)$ can be simply calculated as $\overline{c^2}(t) = \overline{q}(r_o)$, where r_o is a random variable whose pdf is $p(r_o;\, t_o \mid 0;\, t)$, with initial condition $r = 0$.

Therefore, the numerical evaluation of $\overline{c^2}(t)$ can be performed efficiently by simulating the backward trajectories of the released particles that have zero final separation and covariance at the source equal to $q(r_o)$, where r_o is a random variable.

To simulate backward trajectories, the dispersion model has to be written in a reverse form. However, the reverse formulation of our stationary model is equivalent to the forward formulation, in the sense that the same stochastic differential equations governing the motion of pairs of particles [namely (8), (10), and (15)] can be used to simulate backward trajectories, if the forward variables are replaced by backward variables. In other words, the equations are formally the same provided that we interpret the time as running in the opposite direction to real time and interpret the velocity as representing a reverse separation velocity (Durbin 1982; Thomson 1987, 1990).

All simulations presented in this paper were made by releasing 10^5 particle pairs and using a variable time step $dt(r) = 10^{-3}\overline{u^2}(r)/(2C_o\varepsilon)$. The function $q(r_o) = c_o^2 \exp[-r_o^2/(2\sigma_o^2)]$ was assumed for the source term, where σ_o is a characteristic length scale of the source and c_o is the concentration of the cloud center of mass at the source. By definition, $c_o \sim Q/(U\sigma_o^2)$ for a plume, where Q is the amount of contaminant released per unit time and U is the mean wind velocity at the source, and $c_o \sim M/\sigma_o^3$ for a puff, where M is the amount of contaminant released. Because the function $q(r_o)/c_o^2$ is the same for both a plume and a puff, the calculated $\overline{c^2}$, normalized over c_o^2, will also be the same for both plume and puff releases. Figure 1 shows $\overline{c^2}/c_o^2$, as predicted by the model, as a function of t/T, where $T = \sigma_v^2/\varepsilon$ is the Eulerian integral timescale of the turbulence. The simulations were performed for values of the source size σ_o ranging over four orders of magnitude, namely, from $\sigma_o = 10^{-4}L$ to L. As time increases, so the cloud size increases, and hence fewer particle pairs converge on the same position; thus, by definition, $\overline{c^2}$ decreases. The decay of $\overline{c^2}$ with time shown in Fig. 1 depends on the source size, with smaller sources decaying faster. The dependence of the statistics of a fluctuating concentration field on the source size is a well-established phenomenon, which was predicted by theoretical analyses (Chatwin and Sullivan 1979), observed in wind tunnel experiments for the case of continuous releases in a turbulent boundary layer (Fackrell and Robins 1982), and numerically simulated by Lagrangian stochastic models (Durbin 1982; Thomson 1990).

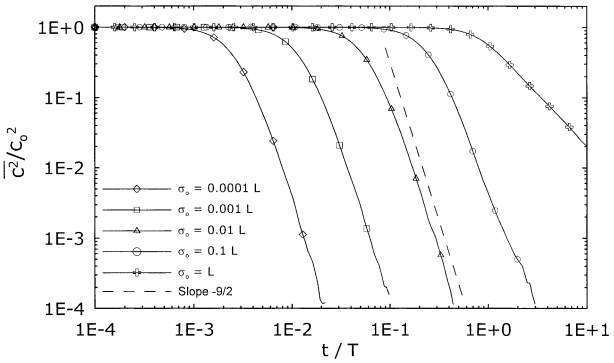

FIG. 1. Mean-square concentration $\overline{c^2}$ predicted by the model, normalized over the square concentration at the source c_o^2, plotted as a function of t/T for five values of the source size σ_o ranging from 10^{-4} L to L. The dashed line has a slope equal to $-9/2$.

Some time after the release, when the cloud size has grown much larger than the source size but is still smaller than L, the pdf of the particle separation, $p(\mathbf{r}, t \mid \mathbf{r}_o, t_o)$, becomes independent of $\mathbf{r}_o$ and scales according to the similarity theory as $p(\mathbf{r}, t) \propto (\varepsilon t^3)^{-3/2}$ (Monin and Yaglom 1975, p. 542). According to (20), we expect the scaling $\overline{c^2}(t) \propto (\varepsilon t^3)^{-3/2}$ to occur. Figure 1 shows that, after an initial transient, the power-law time decay matches the dashed line (proportional to $t^{-9/2}$) for sources smaller than L (e.g., $\sigma_o \leq 0.1\ L$). This indicates good

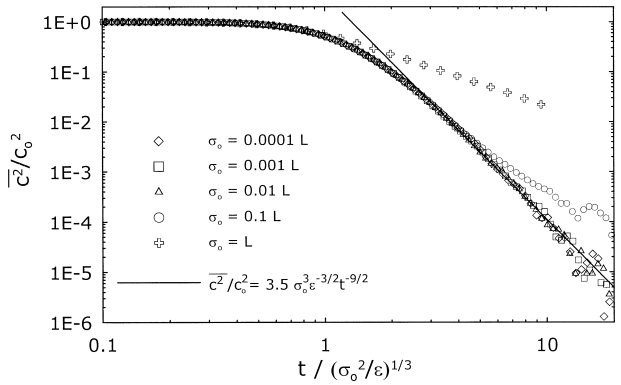

FIG. 2. Nondimensional mean-square concentration $\overline{c^2}/c_o^2$ as a function of the nondimensional variable $t/(\sigma_o^2/\varepsilon)^{1/3}$, predicted by the model for five values of σ_o. The solid line represents (22), i.e., $\overline{c^2}/c_o^2 = 3.5\sigma_o^3\varepsilon^{-3/2}t^{-9/2}$.

agreement between our model results and the predictions of the similarity theory.

1) DEPENDENCE OF THE MEAN-SQUARE CONCENTRATION ON THE SOURCE SCALE

In the following, we show how a universal behavior can be obtained for $\overline{c^2}$ in the inertial range for plumes and puffs released from different-sized sources.

The eddies that most efficiently disperse a cloud have a characteristic length scale d of the same order of magnitude as the cloud size. Therefore the initial timescale of the relative dispersion of a cloud is determined by the timescale of the eddies of the same size as the source. Because the timescale of an eddy is given by $(d^2/\varepsilon)^{1/3}$, a cloud released from a generic source σ_o disperses with an initial timescale equal to $(\sigma_o^2/\varepsilon)^{1/3}$. As a consequence, we expect that clouds released from different source sizes will have similar $\overline{c^2}$ at times t after release proportional to their respective initial timescales. Moreover, all statistics of the concentration field must depend on the initial concentration c_o.

Therefore, we conclude that $\overline{c^2}/c_o^2$ as a function of the nondimensional time $\tau = t/(\sigma_o^2/\varepsilon)^{1/3}$ is independent of the source size σ_o, and we can formally write

$$\overline{c^2} = c_o^2 f_1(\tau), \tag{21}$$

where the function $f_1(\tau)$ is a universal function of the sole variable τ. As mentioned earlier, this behavior is expected to occur as long as both σ_o and the cloud size $\overline{r^2}^{1/2}$ belong in the inertial range, that is, as long as the relationship $\overline{r^2} \propto \varepsilon t^3$ holds.

Figure 2 shows the simulated $\overline{c^2}/c_o^2$ plotted as a function of τ. This figure was obtained by appropriately scaling the data shown in Fig. 1. The data in Fig. 2 for $\sigma_o = L$ are seen to decay more slowly than releases from smaller sources because the universal behavior in (21) is only valid if the cloud size does not exceed L. We also note that the points obtained for the simulation with $\sigma_o = 0.1L$ deviate from the universal behavior when τ is larger than about 5, because $\tau \equiv t/(\sigma_o^2/\varepsilon)^{1/3} = 5$ corresponds for this case to $t \approx T$ and hence $\overline{r^2}^{1/2} = O(L)$. These results show that once a cloud has reached a size comparable with L its mean-square concentration starts decaying at a slower rate.

2) A UNIVERSAL TIME DECAY LAW FOR THE MEAN-SQUARE CONCENTRATION

A single formula describing the decay of $\overline{c^2}$ in plumes and puffs released from different source sizes can be derived by combining the similarity theory scaling $\overline{c^2}(t) \propto t^{-9/2}$ with (21). Using $f_1(\tau) = A[t/(\sigma_o^2/\varepsilon)^{1/3}]^{-9/2}$, where A is a constant, gives

$$\overline{c^2}(t) = Ac_o^2\sigma_o^3\varepsilon^{-3/2}t^{-9/2}. \tag{22}$$

The constant A was determined from the simulation results to be approximately equal to 3.5. From (22), $\overline{c^2}$ is estimated to be of order $Q^2/(U^2\sigma_o\overline{r^2}^{3/2})$ for a plume and $M^2/(\sigma_o^3\overline{r^2}^{3/2})$ for a puff. Note that the estimated magnitude of $\overline{c^2}$ for a puff is consistent with that obtained from a different approach by Chatwin and Sullivan (1979). It follows that $\overline{c^2}$ is much more sensitive to the source size for a puff ($\overline{c^2} \propto \sigma_o^{-3}$) than for a plume ($\overline{c^2} \propto \sigma_o^{-1}$).

The function $\overline{c^2}(t)/c_o^2 = 3.5\sigma_o^3\varepsilon^{-3/2}t^{-9/2}$ is plotted in Fig. 2 as a function of the nondimensional time τ along with our numerical simulations, showing very good agreement.

b. *Concentration fluctuations*

The standard deviation of concentration σ_c is defined as

$$\sigma_c(t) = [\overline{c^2}(t) - \overline{c}^2(t)]^{1/2}. \tag{23}$$

The mean concentration at the cloud center of mass was evaluated as $\overline{c}(t) = c_o\sigma_o^2/\overline{r^2}(t)$ for a plume and as $\overline{c}(t) = c_o\sigma_o^3/\overline{r^2}(t)^{3/2}$ for a puff, where $\overline{r^2}(t)$ is the mean-square separation between particles whose initial separation was equal to σ_o. Figure 3 shows our model results for $\overline{r^2}/L^2$ as a function of the nondimensional time t/T. The classical inertial-range formula for point sources,

$$\overline{r^2}(t) = C_r\varepsilon t^3, \tag{24}$$

where C_r is the Richardson–Obukhov constant, is also plotted as the solid line in Fig. 3 for $C_r = 1.96$, which is the value obtained from our model. Monin and Yaglom (1975, p. 567) report several estimates of C_r obtained by different authors on the basis of observations from diffusion experiments in the atmosphere. Such estimates of C_r range over three orders of magnitude, namely from $\approx$0.008 to $\approx$8, possibly because of the difficulties in the simultaneous measurements of ε and $\overline{r^2}$. Ott and Mann (2000) more recently obtained $C_r \approx 0.5$ from experiments in oscillating grid turbulence. Thus, the value obtained by our model falls within the observed values reported in the literature. However, the

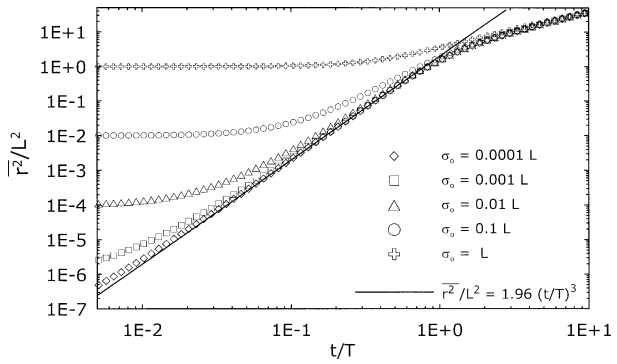

FIG. 3. Mean-square scalar separation $\overline{r^2}$ normalized over L^2, as a function of the nondimensional time after release t/T, predicted by the model for five values of σ_o. The solid line represents (24) after normalization for $C_r = 1.96$, i.e., $\overline{r^2}/L^2 = 1.96(t/T)^3$.

model-simulated C_r is strongly influenced by the assumed velocity-structure constant C_o in (9), whose value is still not well established. For example, a sensitivity test of our model to C_o provided $C_r \approx 5$ for $C_o = 4$ and $C_r \approx 1$ for $C_o = 8$.

Using the same scaling arguments as for $\overline{c^2}$, we find that $\overline{c}$, σ_c, and the intensity of fluctuations $\sigma_c/\overline{c}$ are independent of the source size when they are written as functions of the nondimensional time $\tau = t/(\sigma_o^2/\varepsilon)^{1/3}$; namely, we can write

$$\overline{c} = c_o f_2(\tau), \tag{25}$$

$$\sigma_c = c_o f_3(\tau), \quad \text{and} \tag{26}$$

$$\sigma_c/\overline{c} = f_4(\tau), \tag{27}$$

where the universal functions f_2, f_3, and $f_4 \equiv f_3/f_2$ depend only on τ.

Figures 4a,b show the model-simulated $\overline{c}$ for plume and puff releases, respectively. As for the plot of $\overline{c^2}$ shown in Fig. 2, it is clear that in each case (plume or puff) the data collapse to a single curve as long as the cloud sizes do not exceed L. As expected from the definition of $\overline{c}$, Figs. 4a,b show that the time decay of $\overline{c}$ is much faster for a puff than for a plume. In the inertial range these decays can be determined analytically, yielding the following formulas:

$$\overline{c}(t) = C_r^{-1} c_o \sigma_o^2 \varepsilon^{-1} t^{-3} \quad \text{for a plume,} \quad \text{and} \tag{28}$$

$$\overline{c}(t) = C_r^{-3/2} c_o \sigma_o^3 \varepsilon^{-3/2} t^{-9/2} \quad \text{for a puff.} \tag{29}$$

Formulas (28) and (29) are plotted in Figs. 4a and 4b, respectively, where they fit well the inertial-range decays of $\overline{c}(t)$.

Figure 5 shows the simulated σ_c/c_o as a function of τ for plumes released from five different-sized sources. This figure also represents puff data reasonably well, because σ_c/c_o is practically equivalent at both small and large times.

An interesting implication of (26) (also seen in Fig. 5) is that the maximum σ_c does not depend on the source

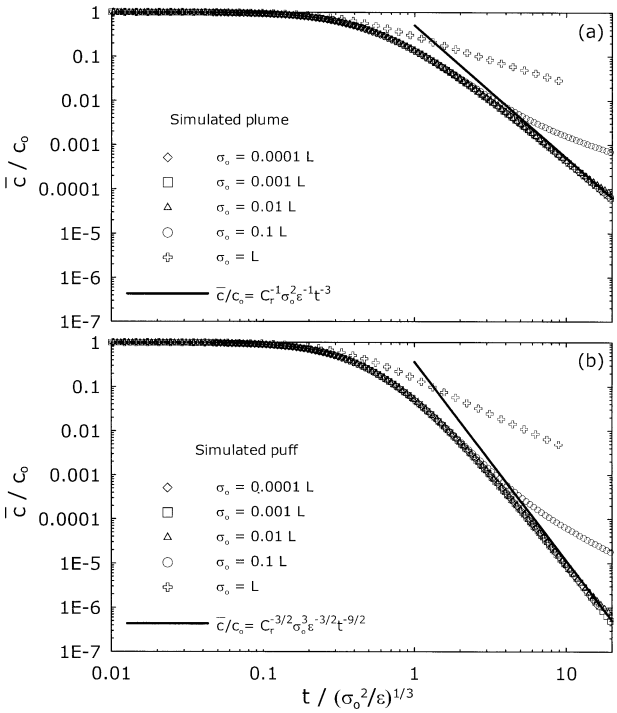

FIG. 4. Nondimensional mean concentration $\overline{c}/c_o$ as a function of $t/(\sigma_o^2/\varepsilon)^{1/3}$, predicted by the model for five values of σ_o: results for (a) simulated plumes and (b) simulated puffs. The solid lines plotted in (a) and (b) represent (28) and (29), i.e., $\overline{c}/c_o = C_r^{-1}\sigma_o^2\varepsilon^{-1}t^{-3}$ and $\overline{c}/c_o = C_r^{-3/2}\sigma_o^3\varepsilon^{-3/2}t^{-9/2}$, respectively.

size and that the maximum occurs at the same dimensionless time τ. Thus, from (26), we have the following result:

$$(\sigma_c)_{\max} = B_1 c_o, \tag{30}$$

which is reached after a time

$$t_{\max} = B_2(\sigma_o^2/\varepsilon)^{1/3} \tag{31}$$

from the release. The values of the constants B_1 and B_2

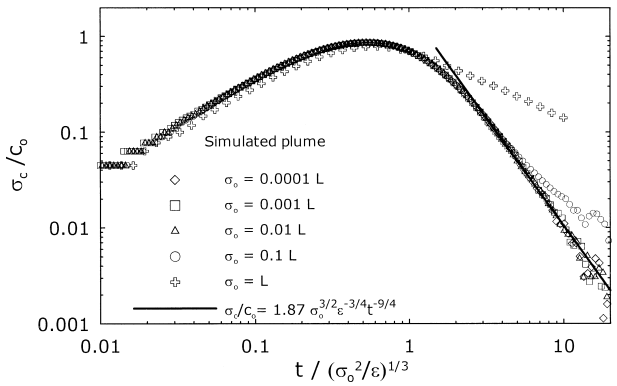

FIG. 5. Nondimensional standard deviation of concentration σ_c/c_o as a function of $t(\sigma_o^2/\varepsilon)^{1/3}$ for simulated plumes, predicted by the model for five values of σ_o. The solid line represents (32), i.e., $\sigma_c/c_o = 1.87\sigma_o^{3/2}\varepsilon^{-3/4}t^{-9/4}$.

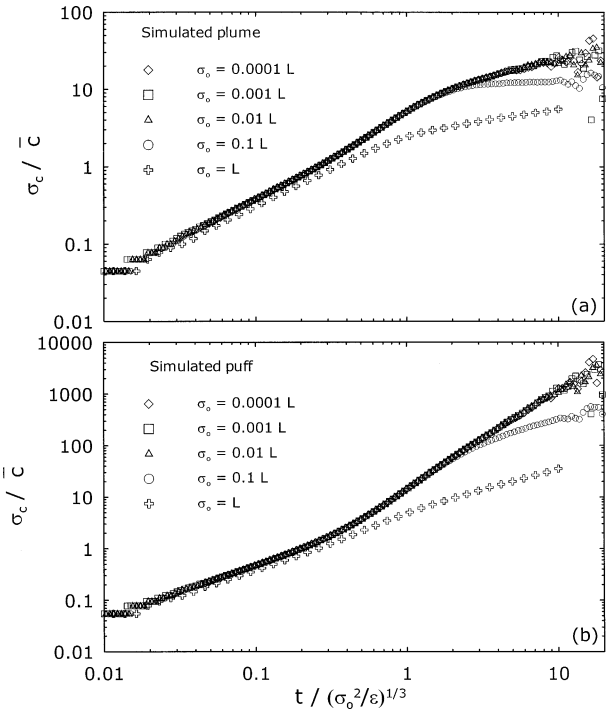

FIG. 6. Intensity of concentration fluctuations $\sigma_c/\overline{c}$ as a function of $t/(\sigma_o^2/\varepsilon)^{1/3}$, predicted by the model for five values of σ_o: results for (a) simulated plumes and (b) simulated puffs.

estimated by our model are $B_1 = 0.83$ and $B_2 = 0.53$ for plumes and $B_1 = 0.91$ and $B_2 = 0.48$ for puffs.

Because the square of the mean concentration, $\overline{c}^2(t)$, is proportional to $\overline{r^2(t)}^{-2}$ for a plume and to $\overline{r^2(t)}^{-3}$ for a puff, it decays as $\overline{c}^2(t) \propto t^{-6}$ for a plume and as $\overline{c}^2(t) \propto t^{-9}$ for a puff. Therefore, for large-enough times, $\overline{c}^2(t)$ is negligible when compared with $\overline{c^2}(t)$ (which scales as $t^{-9/2}$ for both plume and puff releases), and $\sigma_c^2(t)$ is equivalent to $\overline{c^2}(t)$. Thus, at this stage of the cloud growth ($\tau \gtrsim 3$), by virtue of (26), we can write the formula

$$\sigma_c(t) = \overline{c^2(t)}^{1/2} = \sqrt{A} c_o \sigma_o^{3/2} \varepsilon^{-3/4} t^{-9/4}, \qquad (32)$$

which is valid for both plumes and puffs, where the constant $\sqrt{A}$ is equal to 1.87. Formula (32) is plotted in Fig. 5, where it fits well with the simulations for sources smaller than the integral length scale.

Figures 6a,b show the intensity of concentration fluctuations $\sigma_c/\overline{c}$ for simulated plumes and puffs, respectively. At large times, $\sigma_c/\overline{c}$ for puffs is about two orders of magnitude larger than for plumes. This effect is due to the decay of $\overline{c}$, which is much faster for a puff than for a plume, as described earlier.

c. *Simple practical considerations for atmospheric scaling*

The universal scaling results are useful for practical estimates of near-source plume behavior. In fact, source sizes on the order of 1–10 m in, say, the convective boundary layer, where the energy-containing eddies fill the depth of the layer, with $L \sim 1$ km, give ratios from $\sigma_o/L \sim 10^{-3}$ to $\sigma_o/L \sim 10^{-2}$, with the small values clearly demonstrating the potential role for inertial-range processes. Indeed, many industrial chimney stacks release plumes with scales in the inertial subrange. Comparisons between emissions and mixing of plumes from different-size stack diameters may be made by collapsing the fluctuation fields with the source-size scaling used above. That is, the downstream development for a larger source, say a 10-m-diameter stack, will take a relatively longer time to develop than for a 1-m-diameter stack, typically $(10/1)^{2/3} = 4.6$ times as long for the scaled fluctuations to decay by the same amount. Absolute measures are also available by using parameterized functions in (25), (26), and (27) and providing data for c_o, but no explicit examples will be given here. In more stable surface-layer processes, the turbulence scales are much smaller, but applications will also often involve smaller-scale sources.

6. Conclusions

Relative dispersion in atmospheric flow can be described simply in terms of the magnitude of the separation of generic pairs of particles averaged over the plume domain, which is relevant for predicting and parameterizing internal concentration-fluctuation decay, particularly in meandering plumes (Luhar et al. 2000). The generic relative dispersion problem is equivalent in form with simpler homogeneous isotropic turbulence, but various constant factors require measurements of two-point velocity statistics, or at least energy dissipation statistics as functions of position in the mean-plume envelope, for inertial-range modeling of internal fluctuations. In this paper, a stochastic model of relative dispersion in isotropic and homogeneous turbulence has been derived using a new formulation for the acceleration of separation with a focus on simple robust formulation with efficient numerical solutions. The model is consistent with a well-mixed distribution of particle pairs and with the non-Gaussian turbulence statistics for the Eulerian velocity differences. In the future it is to be hoped that measurements of turbulence properties in the atmosphere can be used for the development of models of scalar dissipation in flows with significant atmospheric stability effects.

To demonstrate some simple internal fluctuation characteristics, the model has been applied to simulate plume and puff releases from single sources with sizes ranging over four orders of magnitude, namely, from $10^{-4}L$ to L, using parameterized structure functions as input. Such structure functions are consistent with the similarity theory in the inertial range and with the hypothesis of zero spatial autocorrelation of the Eulerian velocity at length scales exceeding the integral scale L. As a consequence,

the model correctly predicts the slower dispersion of a cloud that grows larger than L.

The simulations of mean-square concentration $\overline{c^2}$, mean concentration $\overline{c}$, standard deviation of concentration σ_c, and intensity of concentration fluctuations $\sigma_c/\overline{c}$ show the strong dependence of the results on the source size, consistent with observations, theoretical predictions, and numerical simulations in the literature. The results show a very good agreement with the predictions of the similarity theory. For instance, the model reproduces the prescribed $-9/2$ exponent in the time decay law for $\overline{c^2}(t)$.

We have proposed a scaling law that determines a universal behavior of $\overline{c^2}$, $\overline{c}$, and hence σ_c and $\sigma_c/\overline{c}$ for all sources and cloud sizes within the inertial range. The combined application of this scaling law along with the similarity theory has led to general analytical formulas for $\overline{c^2}$, $\overline{c}$, and σ_c, as well as to a simple formula for the maximum σ_c.

Future work with the current model clearly involves coupling with meandering plume and puff models, embedded within prognostic meteorological models. One-particle mean field modeling of atmospheric plumes embedded within more general Eulerian boundary layer models is practical and useful (Luhar and Sawford 1995; Anfossi et al. 1998; Nasstrom et al. 2000; Tinarelli et al. 2000; Kurbanmuradov and Sabelfeld 2000; Stein et al. 2000), and adapting simple two-particle relative dispersion models, at least the quasi-one-dimensional scalar separation models, to the same ends seems to be feasible. White-noise representation of fluid particle accelerations is certainly a reasonable approximation for atmospheric flows, despite known difficulties for such representation for low-Reynolds-number flows (Heppe 1998). Also, for reasons of complexity and numerical efficiency, highly sophisticated models of relative dispersion such as kinematic simulation (Flohr and Vassilicos 2000) are unlikely to be practical options for modeling plume dispersion within routine meteorological prognostic models (Hurley and Luhar 2000). The current results suggest that Lagrangian stochastic models of relative dispersion (suitably averaged) are efficient and simple enough for atmospheric applications; possible improvements within this class of models may be needed to reduce the mean-square dispersion in the inertial subrange, but this is not yet certain. Future development involves the extension to nonstationary conditions, expected for most atmospheric flows. It is also hoped that measurements of atmospheric flows yield the simplified two-point statistics that drive the current model and that the combination of all these efforts leads to operational prediction of concentration fluctuations in atmospheric releases for routine air quality and hazard response work.

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