Diffusion of Gaussian puffs

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SUMMARY

A formula for the relative diffusion of a puff is proposed in which the rate of growth is related to common, one-dimensional velocity spectra obtainable, for instance, from direct measurements in the atmosphere. Often, an equation of this type is in demand for modelling of instant or short-term release of potentially harmful gases or smoke, for example, in connection with a puff diffusion model.

A simple expression like Taylor's equation for single-particle diffusion is not obtainable in general for relative diffusion. Therefore, the present approach is based on a kinematic-statistical model in which a Gaussian approximation has been applied to the relative displacement process of the fluid particles. This is along lines suggested previously, where the expansion of a Gaussian puff has been related to the kinematic energy spectrum in three-dimensional, isotropic turbulence. Here, however, we attempt to relate the growth rate directly to the one-dimensional velocity spectra (or their corresponding correlations) most commonly available in the atmosphere. In particular, this is of value for the surface layer, where diffusions in the horizontal and vertical directions are different (anisotropy) due to the presence of the ground.

Experimental evaluation is based on a series of smoke release experiments carried out in the surface layer over homogeneous terrain in Denmark. Solutions of the puff growth rate equation with the measured correlations are found to compare well with the experimental data of cross-wind relative diffusion over the limited scale of the experiments.

1. INTRODUCTION

One of the most important characteristics of turbulent flow is its ability to disperse particles at a rate many orders of magnitude greater than diffusion by molecular collisions. Two basically different aspects of this problem can be distinguished. One is the mean square displacement of a marked fluid particle with respect to its origin of release. The other is concerned with the relative displacement of two particles. The mean square particle separation is a critical parameter in the two-particle probability distribution function from which the mean square particle concentration may in principle be calculated for an arbitrary source configuration, as described by, e.g., Batchelor (1952). The problem of relative dispersion of marked fluid particles was investigated by Roberts (1961) using the Eulerian direct-interaction approximation. However, this representation is not invariant with respect to random Galilean transformations. The Lagrangian direct-interaction approximation described by Kraichnan (1966) remedies this shortcoming at the expense of a complication of the theoretical analysis. The analyses of, e.g., Smith (1959), Smith and Hay (1961) and Sawford (1982) bring out the underlying physical mechanism more clearly. The present work attempts a theoretical description of the expansion of a cloud of marked fluid particles released in a stationary, homogeneous turbulent flow which is related to these latter works (see also Mikkelsen 1982). The physical mechanism characterizing this problem can briefly be outlined as follows:

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Fluctuations (or eddies) having a scale size larger than the instantaneous size \( \sigma \) of the cloud will give rise to a random motion or meandering of the entire cloud ("a random Galilean displacement") while only scales smaller than or comparable with \( \sigma \) will contribute to an expansion. However, as the cloud expands, fluctuations characterized by a scale size which originally gave rise only to a bulk displacement of the cloud may at a later instant be smaller than \( \sigma = \sigma(t) \) and, consequently, now contribute instead to the expansion. In particular, it can be argued on intuitive grounds that as the cloud expands, a larger and larger fraction of the spectral energy will be available for the expansion, which consequently proceeds at an increasing rate. Obviously, the spectral index (i.e. the slope of the power spectrum on a log-log plot) is a significant parameter for the temporal expansion rate. Our analysis gives a satisfactory account of these physical arguments, and the results compare favourably with the measurements in, e.g., the Borris field experiment described by Mikkelsen (1983) and Mikkelsen and Eckman (1983). The analysis reproduces also some limiting cases known from the literature and suggests a numerical value for the universal constant in the Lagrangian structure function.

2. Theory

(a) The basic equations

Following Csanady (1973), we consider the release of a cloud of marked fluid at the position \( x = 0 \) and at a time \( t = 0 \) into a field of stationary and homogeneous turbulence. Let the observed concentration field at subsequent times of the experiment in each realization be \( C(x, t) \) and define

\[
Q = \int C(x, t) \, dx. \tag{1}
\]

The quantity \( Q \) is the total amount of matter released with the puff. The volume integral extends over all space. The first moment of the instantaneous normalized concentration field \( Q^{-1}C(x, t) \) yields the instantaneous position \( c(t) \) of the centre of mass of the cloud

\[
c(t) = Q^{-1} \int xC(x, t) \, dx. \tag{2}
\]

This quantity executes random movements as a function of time in a turbulent environment. The velocity of the centre-of-mass position vector, \( \mathbf{V}_{cm} = dc/dt \), follows from a differentiation of Eq. (2) and some simple manipulations (Csanady 1973), i.e.

\[
\mathbf{V}_{cm}(t) = Q^{-1} \int \mathbf{u}(x, t) C(x, t) \, dx \tag{3}
\]

where \( \mathbf{u}(x, t) \) represents the fluid velocity.

A coordinate system \( y \), attached to the puff's centre of mass \( c \), may now be defined by

\[
y = x - c. \tag{4}
\]

This 'relative' or 'moving' frame of reference is exposed to random accelerations by the turbulence and is as such a non-inertial reference frame. The ensemble average of the velocity of the centre-of-mass vector \( \mathbf{V}_{cm} \) may be determined from Eq. (3):

\[
\langle \mathbf{V}_{cm} \rangle = Q^{-1} \int (\langle \mathbf{u} \rangle \langle C \rangle + \langle \mathbf{u}'C' \rangle) \, dx. \tag{5}
\]
Primes denote fluctuations, i.e. departures from the ensemble mean in an individual realization. The mean product \( \langle u' C \rangle \) is identified as a local turbulent flux vector. Csanady (1973, p. 86) argues that for a homogeneous field and provided that the cloud when released is symmetrical about the origin, this flux must be antisymmetrical, so that its space integral is zero. Thus, for symmetrically released clouds, and for others at least approximately, the relation

\[
\langle V_{cm} \rangle = Q^{-1} \int \langle u \rangle \langle C(x, t) \rangle \, dx
\] (6)

replaces Eq. (5). In homogeneous and isotropic turbulence we have \( \langle V_{cm} \rangle = 0 \) while, obviously, the quantity \( V_{cm} \) defined in Eq. (3) refers to one realization of the flow and is nonzero in general. The quantity \( Q^{-1} \langle C(x, t) \rangle \) is the probability that marked particles will be found at position \( x \) at time \( t \).

In the following, we will consider diffusion along a single, but arbitrarily chosen, coordinate direction \( x \). Introducing the average as e.g. \( \langle x^2 \rangle = Q^{-1} \int x^2 \langle C(x, t) \rangle \, dx \) the relation

\[
\langle x^2(t) \rangle = \langle y^2(t) \rangle + \langle c^2(t) \rangle
\] (7)

is obtained from Eq. (4), i.e. absolute dispersion is the 'sum' of relative dispersion and meandering, in the sense that their variances are additive.

Introducing the two-particle displacement probability \( Q^{-2} \langle C(x, t) C(x', t) \rangle \) we find the mean square separation \( \langle l^2 \rangle \) between two diffusing particles as

\[
\langle l^2 \rangle = Q^{-2} \int \int (x' - x)^2 \langle C(x, t) C(x', t) \rangle \, dx' \, dx
\]

\[
= 2\langle x^2(t) \rangle - 2\langle c^2(t) \rangle.
\] (8)

By use of Eq. (7) this simply becomes

\[
\langle l^2 \rangle = 2\langle y^2(t) \rangle
\] (9)

which states that the mean square separation of two diffusing particles along the coordinate \( x \) is just twice their mean square separation from the centre of mass.

We introduce the velocity \( v = dy/dt \) of a marked particle with respect to the centre of mass. Referring to (4) we have

\[
v = u - V_{cm}
\] (10)

where \( u \) is the absolute velocity of the reference particle. With

\[
y(t) = \int_0^t v(\tau) \, d\tau
\] (11)

where the integration runs along a Lagrangian orbit we obtain the cloud's rate of growth

\[
\frac{d}{dt} \langle y^2 \rangle = 2 \left\langle \frac{dy}{dt} \right\rangle = 2 \int_0^t \langle v(\tau)v(\tau) \rangle \, d\tau.
\] (12)

The overbar in Eq. (12) denotes averaging over all the marked particles in the cloud. It is convenient to consider the cloud as composed of a very large number \( N \) of individual but identical particles. In this case the cloud averaging is understood as the operation \( N^{-1} \sum_{n=1}^N \). Introducing \( \sigma = (\langle y^2 \rangle)^{1/2} \) we may rewrite Eq. (12) as
\[
\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \langle \overline{v(t) \nu(t - \tau)} \rangle \, d\tau \\
= \langle \overline{\nu^2(t)} \rangle \int_0^t r(t, \tau) \, d\tau = \langle \overline{\nu^2(t)} \rangle t_e(t).
\]
(13)

The relative velocity auto-covariance has been introduced as
\[
r(t, \tau) = \frac{\langle \overline{v(t)\nu(t - \tau)} \rangle}{\langle \overline{\nu^2(t)} \rangle}.
\]
This quantity is written explicitly as a function of \(t\) and \(\tau\) to emphasize that relative diffusion is not a stationary process. Consequently, the relative time scale \(t_e(t)\) derived from \(r(t, \tau)\) depends on the time \(t\) after release. Intuitively \(t_e(t)\) is expected to be increasing with \(t\), since as the cloud grows larger and larger, eddies contribute to the expansion with correspondingly increasing time scales (or 'memory').

Using Eq. (10), and realizing that \(\langle V_{cm}(t)u(t - \tau) \rangle = \langle V_{cm}(t)V_{cm}(t - \tau) \rangle\) Eq. (13) can be written as
\[
\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \{\langle u(t)u(t - \tau) \rangle - \langle V_{cm}(t)V_{cm}(t - \tau) \rangle\} \, d\tau \\
= \int_0^t \{R_{abs}(\tau) - R_{cm}(t, \tau)\} \, d\tau.
\]
(14)

Here, the relations
\[
R_{abs}(\tau) = \langle \overline{u(t)u(t - \tau)} \rangle \quad \text{and} \quad R_{cm}(t, \tau) = \langle \overline{V_{cm}(t)V_{cm}(t - \tau)} \rangle
\]
have been introduced, where \(R_{abs}(\tau)\) is the Lagrangian auto-covariance function associated with absolute or single-particle diffusion. \(R_{cm}(t, \tau)\) is the corresponding auto-covariance function for the puff’s centre-of-mass motion, a non-stationary function, which in general depends on both the lag, \(\tau\), and the absolute time \(t\).

An alternative form of Eq. (14) was obtained by Batchelor (1950). The mean square separation of an arbitrary pair of particles belonging to the puff is
\[
d(\ell_0^2(t))/dt = 2 \int_0^t \langle \overline{(u_i(t) - u_j(t))(u_i(t - \tau) - u_j(t - \tau))} \rangle \, d\tau
\]
(16)

where \(u_i(t)\) is the Lagrangian velocity of a marked fluid particle belonging to the puff. As pointed out by e.g. Sawford (1982), Eq. (16) contains two types of velocity products. One is of the form \(u_i(t)u_i(t - \tau)\) and refers to the same particle at two different times. We recognize this as the Lagrangian auto-covariance, \(R_{abs}(\tau)\). The second, of the form \(u_i(t)u_j(t - \tau)\), is a two-particle Lagrangian covariance involving one particle at time \(t\) and a second at time \(t - \tau\).

For the case of stationary and homogeneous turbulence we have for a single pair
\[
d(\ell_0^2(t))/dt = 4 \int_0^t \{R_{abs}(\tau) - \langle u_i(t)u_i(t - \tau) \rangle\} \, d\tau.
\]
(17)

Formally we average this over all \(N(N - 1)\) particle pairs in the puff to obtain the cloud average. By denoting the pair-average defined as
\[
(N(N - 1))^{-1} \sum_{i=1}^{N} \sum_{j \neq i}
\]
by a double overbar, we have for the pair-averaged quantity

$$\bar{d}\langle \bar{u}_j(t) \rangle / dt = 4 \int_0^t \{ R_{\text{abs}}(\tau) - \langle u_i(t)u_j(t-\tau) \rangle \} d\tau. \quad (18)$$

By use of Eq. (9) this can be cast into a form which compares with the previous result in Eq. (14)

$$\frac{1}{2}d\bar{a}^2 / dt = \int_0^t \{ R_{\text{abs}}(\tau) - \langle u_i(t)u_j(t-\tau) \rangle \} d\tau. \quad (19)$$

From here we readily identify

$$R_{\text{cm}}(t, \tau) = \langle u_i(t)u_j(t-\tau) \rangle. \quad (20)$$

This leaves us with two principal different relationships for the centre-of-mass covariance function:

(1) $R_{\text{cm}}(t, \tau) = \langle V_{\text{cm}}(t)V_{\text{cm}}(t-t-\tau) \rangle$ from Eq. (15), which in turn, through Eq. (3), relates to the Eulerian properties of the turbulence.

(2) Equation (20) above, which relates $R_{\text{cm}}(t, \tau)$ to the two-particle Lagrangian covariance of marked fluid particles, or hypothetical fluid elements, belonging to the puff.

For the discussion to follow, we shall finally consider the averaging of particle pairs involved in Eq. (20). Sawford (1982) emphasizes that the different particles in the puff are identifiable only because of the influence their initial positions have on their subsequent trajectories. The averaging in Eq. (20) therefore reduces to averaging over the distribution of initial separations

$$\langle u_i(t)u_j(t-\tau) \rangle = \int_{-\infty}^{\infty} \langle u_i(t)u_j(t-\tau) \rangle P(\xi_{i,0}) d\xi_{i,0}. \quad (21)$$

Here, $P(\xi_{i,0})$ denotes $Q^{-1} \int_{-\infty}^{\infty} \langle C(x,0)C(x+\xi_{i,0},0) \rangle dx$, the initial distance-neighbor distribution introduced earlier in this section. If it is assumed that the initial puff release is a Gaussian $G_{\sigma(0)}(x)$, having the standard deviation $\sigma(0)$, then $P(\xi_{i,0})$ is a Gaussian too, with standard deviation equal to $\sqrt{2}\sigma(0)$.

At this point we note that the two-particle Lagrangian covariance function in its most general form, shows functional dependence on the initial separation $\xi_{i,0}$, in addition to the two times $(t, \tau)$, apparent from Eq. (17):

$$\langle u_i(t)u_j(t-\tau) \rangle = f(\xi_{i,0}, t, \tau). \quad (22)$$

(b) Previous approximations for $R_{\text{cm}}(t, \tau)$

Various approximate forms of the two-particle covariance function $\langle u_i(t)u_j(t-\tau) \rangle$, on which $R_{\text{cm}}(t, \tau)$ in Eq. (20) depends, have earlier been proposed in the literature. Recently, Sawford (1982) compared approximations suggested by G. I. Taylor (in an unpublished note (1935) referred to by Batchelor (1952)), Brier (1950) and Smith and Hay (1961), in order to assess the effect of these approximations on predicting the mean square separation of a cluster of particles. The present theory, to be outlined in section (c) below, bears resemblance to these previous approximations, in particular to the work by Smith and Hay. Therefore, we include a brief review of this earlier work in order to emphasize later some fundamental differences in the present theory, and for the comparison of results.

Presented here in a one-dimensional version, Smith and Hay in effect assumed

$$\langle u_i(t)u_j(t-\tau) \rangle = R_E(\xi_{ij} + Ut/\beta) \quad (23)$$
where the Lagrangian time lag \( \tau \) has been incorporated into the argument of the Eulerian space correlation function \( R_E(\xi_y) = \langle u(x,t)u(x+\xi_y,t) \rangle \). \( U \) is the mean wind speed.

The centre-of-mass covariance function is subsequently calculated as

\[
R_{cm}(t, \tau) = \int_{-\infty}^{\infty} R_E(\xi_y + Ut/\beta)G_{\epsilon 2,\sigma(\epsilon)}(\xi_y) d\xi_y
\]

(24)

where \( G_{\epsilon 2,\sigma(\epsilon)} \) is the distance-neighbour distribution function of the particle pair separations. These equations, (23) and (24), were adopted by Smith and Hay based on the following set of approximations:

(1) The Lagrangian separation of particles in time has been incorporated into the argument of the Eulerian space correlation function \( R_E \), by use of the Hay–Pasquill (Hay and Pasquill 1959) and the Taylor (Pasquill and Smith 1983, p. 21 and p. 42) hypotheses.

(2) The distance-neighbour distribution function is assumed to be Gaussian and is evaluated at the fixed time \( t \) only.

**Point (1).** These are the more fundamental assumptions involved in the theory by Smith and Hay. Implicitly assumed by adopting this approach for \( \langle u_i(t)u_j(t-\tau) \rangle \) is that any memory or bias that the two-particle correlation function would carry due to the initial separation \( \xi_y \) of the particles, is lost. This is in general true, however, only after such long travel times that the corresponding trajectory-correlation \( \langle x_i(t)x_j(t-\tau) \rangle \) effectively becomes independent of \( \xi_y \). At short to intermediate travel times, we are in principle dealing with conditional sampling, and the corresponding covariance, calculated from the sub-ensemble of particle pairs holding the fixed separations \( \xi_y \) after a travel time \( t \): \( \langle u_i(t)u_j(t-\tau) \rangle \big| \xi_y \), accordingly, would depend on the separation at time \( t \) in addition to the initial separation (compare Eq. (22)):

\[
\langle u_i(t)u_j(t-\tau) \rangle \big| \xi_y = g(\xi_y, \xi_y^0, t, \tau).
\]

(25)

Generally, we would expect the function \( g \) to differ from the unconditional sampled covariance \( R_E(\xi_y + Ut/\beta) \) adopted in Eq. (23).

**Point (2).** This form of the distance-neighbour distribution function neglects the actual change in pair-separation during the period \( t - \tau \) to \( t \). Sawford’s analysis, however, supports this approximation originally proposed by Smith and Hay. Time lags near zero dominate the puff growth due to the weighting by \( R_E(\xi_y + Ut/\beta) \), and the effect can safely be ignored. Consequently, the distance-neighbour function in Eq. (24) has been based on values of the puff size at the single time \( t \) only.

(c) **The present analysis**

In section (a) we pointed to the existence of two different relationships for the centre-of-mass covariance function. One, Eq. (20), relates \( R_{cm}(t, \tau) \) to the Lagrangian covariance of two marked fluid particles. The approach was reviewed in section (b) above. The other, Eq. (15), relates \( R_{cm}(t, \tau) \) to the Eulerian properties of the flow (through Eq. (3)). Using this latter approach here, we shall now proceed from Eq. (14) in order to relate the puff growth rate \( \frac{d\sigma^2}{dt} \) to some more fundamental properties of the turbulence.

The basic problem in evaluating Eq. (14) is caused by the expression \( R_{cm}(t, \tau) \), which accounts for the intrinsic non-stationary property of relative expansion. The analysis of the first term, \( R_{obs}(\tau) \), is well known from investigations of absolute diffusion, as given by, e.g., Taylor (1921), and Lumley and Panofsky (1964). Introducing Eq. (3) we rewrite \( R_{cm}(t, \tau) \) from Eq. (15) as
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\[ R_{cm}(t, \tau) = Q^{-2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle u(x', t)u(x'', t - \tau)C(x', t)C(x'', t - \tau) \rangle \, dx' \, dx''. \]  

(26)

This expression is more conveniently expressed in the frame of reference moving with the centre-of-mass velocity defined in Eq. (4), i.e.

\[ R_{cm}(t, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle u(y' + c, t)u(y'' + c, t - \tau)C(y' + c, t)C(y'' + c, t - \tau) \rangle \, dy' \, dy''. \]  

(27)

where \( x = y + c \) with \( c = c(t) \) defined in Eq. (2).

In order to proceed from this rather general relation the following simplification of the random turbulent flow will be introduced:

Assumption 1: The macroscopic turbulent field is based on a micro-structure consisting of a very large but numerable number of individual fluid particles. Of these particles some will be 'marked' in order to identify the cloud, but otherwise the particles are identical. In essence: we are dealing with a passive contaminant.

The connection with measurable, macroscopic fluid properties, such as a fixed point (Eulerian) velocity \( u(x, t) \) and the instantaneous concentration \( C(x, t) \), will be established by counting the collective contribution from the sample of fluid particles that at time \( t \) occupy the interval between \( x \) and \( x + dx \), where \( dx \) is a small increment. This increment, \( dx \), is assumed to be small relative to the smallest structure in the turbulence (the Kolmogorov microscale), but still much larger than the linear extension of a single fluid particle. Uncertainties due to a finite number, \( M \), of fluid particles in the sample interval \( dx \) will accordingly be ignored. In this way, the actual choice of the sample interval \( dx \) will not enter into the calculations. The fluid velocity \( u(x, t) \) associated with the point \( x \) is consequently given by the sum \( (1/M) \Sigma u_i(t) \), where \( u_i(t) \) is the (Lagrangian) velocity of the individual fluid particles, marked or unmarked, occupying the small interval between \( x \) and \( x + dx \) at time \( t \). However, the corresponding instantaneous concentration \( C(x, t) \) relates only to the number of *marked* fluid particles in this interval. Based on these two macroscopic quantities the cloud's centre-of-mass velocity obtained as in Eq. (3) is

\[ V_{cm}(t) = Q^{-1} \int_{-\infty}^{\infty} u(x, t)C(x, t) \, dx. \]  

(28)

Assumption 2: When a test cloud of initial concentration \( C(x, 0) \) is released, its centre-of-mass coordinate \( c(t) = \int_{-\infty}^{\infty} xc(x, t) \, dx \) defines the origin of a moving coordinate system. Relative to this randomly moving coordinate system, the stochastic displacements of fluid particles will be assumed to obey identical and uncorrelated Gaussian statistics.

Assumption 2 is the most restrictive in the present approach, and later it will be evident that Gaussian-shaped puffs result as a consequence of this.

Consider assumption 2 in greater detail: Fig. 1 shows the Lagrangian trajectory \( y_i(t) \) of a marked fluid particle \( i \), that at the previous time \( t - \tau \) was in position \( y_i(t - \tau) \). In the moving frame, the displacement \( \Delta y_i = y_i(t) - y_i(t - \tau) \) constitutes a stochastic process, having the assumed Gaussian density distribution function \( G_{\Delta y_i} \) as indicated. The quantity \( \langle \Delta y_i^2 \rangle \) equals the contribution of this particular particle to the growth of the cloud in the period \( t - \tau \) to \( t \). The growth of the entire cloud is consequently related to the collective motion of all the marked fluid particles over this interval.

Not only the marked, but *all* the displacement distribution functions \( G_{\Delta y_i} \) of the fluid
particles are assumed to be identical, and any pair of particles is assumed to disperse without trajectory correlation in the moving frame. This is mathematically contained in the following expression

$$
\langle \Delta y_i \Delta y_j \rangle = \begin{cases} 
\sigma^2(t) - \sigma^2(t - \tau) & \text{for } i = j \\
0 & \text{for } i \neq j.
\end{cases}
$$

(29)

Here, $\sigma(t)$ represents the standard deviation of the test cloud at travel time $t$ after release. A physical interpretation of this is that the length scale of particle motion in the cloud's coordinate system

$$
l(t) = \frac{\langle \nu^2(t) \rangle^{-1}}{\langle \nu(y, t) \nu(y + \xi, t) \rangle} d\xi
$$

is assumed to be small relative to the instantaneous cloud size $\sigma(t)$. Both $l(t)$ and $\sigma(t)$ are increasing functions with time, and therefore the inequality $l(t) \ll \sigma(t)$, which could satisfy Eq. (29), expresses a (rather strong!) limitation that must be imposed on an initially small Gaussian cloud in order to evolve in an uncorrelated and Gaussian manner. In atmospheric turbulence, $l(t)$ is usually of a size comparable with the cloud itself ($l(t) \sim \sigma$). Consequently, the particle motion inside the cloud (as seen from the cloud's moving frame of reference) will to some extent always be correlated, and a 'true' Gaussian cloud is accordingly never seen. Nevertheless, it is worth emphasizing that the condition in Eq. (29) is realizable, i.e. a physical system can be imagined where (29) is strictly valid.
We may then use this with some confidence as an \textit{approximation} for atmospheric turbulence.

The derivation of an equation for the puff growth $\sigma(t)$ now follows directly as a consequence of assumptions 1 and 2, which in turn are encompassed in Eqs. (28) and (29), respectively.

Based on the approximation that the moving frame dispersions of any two particles are uncorrelated, the instantaneous concentration of the test cloud at time $t$ can be calculated as a superposition of all the marked fluid constituting the cloud at a previous time $t - \tau$:

\[ \hat{C}(y, t) = \int_{-\infty}^{\infty} G_{\Delta y}(y - y_0) \hat{C}(y_0, t - \tau) dy_0. \]  

(30)

For later convenience, we have here introduced the notation $\hat{C}(y, t) = C(y + c, t)$.

From this it follows that an initially Gaussian cloud, with standard deviation $\sigma(0)$, will preserve its Gaussian form at all subsequent times $t > 0$, see Fig. 2. Equation (30) therefore takes the form

\[ G_{\sigma(t)}(y) = \int_{-\infty}^{\infty} G_{\Delta y}(y - y_0) G_{\sigma(t - \tau)}(y_0) dy_0 \]  

(31)

where $G_{\sigma(t)}(y)$ is a Gaussian with standard deviation $\sigma(t)$. It also follows that the spread $\langle \Delta y^2 \rangle$, associated with the individual fluid particles transition\* from time $t - \tau$ to time $t$,

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{gaussian_puff.png}
\caption{The motion of a Gaussian cloud $G$, in the fixed frame of reference $x$ as a function of time $t$. The centre-of-mass coordinate of the cloud $c$ defines the origin of the moving frame $y$, relative to which the dispersion of the cloud in terms of the standard deviation $\sigma$ is defined. Also shown are the two fixed points in the moving frame, $y'$ and $y''$, on which the covariance function $\langle u(y' + c, t)u(y'' + c, t - \tau) \rangle$ depends.}
\centering
\footnotesize\textsuperscript{*} The distribution function $G_{\Delta y}$, together with its spread $\langle \Delta y^2(t, \tau) \rangle$ can easily be shown to satisfy the Chapman-Kolmogorov equation for a transition probability (see e.g. van Kampen 1981). Consequently, the diffusion process in question classifies as a (non-stationary) Markov process.
\end{figure}
obeys the relationship given in Eq. (29)

$$\langle \Delta y_1^2(t, \tau) \rangle = \sigma^2(t) - \sigma^2(t - \tau).$$

(32)

We note that the transition-spread $\langle \Delta y_1^2 \rangle$ does not have functional dependence on values of $\sigma$ prior to $t - \tau$. In particular, it does not depend on the initial puff size or particle separation; cf. the discussion in sections (a) and (b). In other words, 'memory' in the relative diffusion process is limited in history and goes back only to the state at time $t - \tau$. This is plausible because the relative diffusion process involves only the high-pass filtered energy (eddies smaller than $1/\sigma$). Compared with the unfiltered eddies of the full turbulence, these in-cloud eddies have significantly shorter 'turn-around' time, or memory.

Replacing the instantaneous concentrations in Eq. (27) by the corresponding Gaussians, we obtain the simpler form

$$R_{en}(t, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle u(y' + c, t)u(y'' + c, t - \tau) \rangle G_{\sigma(t)}(y') G_{\sigma(t - \tau)}(y'') dy' dy''.$$  (33)

From here we proceed by calculating the two-point, two-time (Eulerian) velocity covariance, which in the moving frame of reference can be written as $\langle \hat{\mathbf{u}}(y', t)\hat{\mathbf{u}}(y'', t - \tau) \rangle$, where the moving frame fluid velocity $\hat{\mathbf{u}}$ has been related to the fixed frame fluid velocity $u$ by the relation $\hat{\mathbf{u}}(y, t) = u(y + c, t)$. Previously, we considered the turbulent field as composed of a very large number of fluid particles, some of which were marked in order to identify the cloud. Suppose that the $i$th of these particles at time $t$ occupies the interval between $y'$ and $y' + dy'$ associated with the point $y'$. The Lagrangian velocity of this fluid particle, $\bar{u}(y(t))$, then contributes to the Eulerian macroscopic velocity $\bar{u}(y', t)$ through the sum $(1/M)\Sigma_i \bar{u}(y_i(t))$ at this point and time. Equivalently, if another fluid particle $(j)$ at time $t - \tau$ is inside the interval $y''$ and $y'' + dy''$, it contributes to the Eulerian fluid velocity here through $(1/M)\Sigma_i \bar{u}(y_j(t - \tau))$.

![Figure 3](image_url)

Figure 3. The trajectory of an arbitrary fluid particle $j$, which at time $t - \tau$ is in the position $y_j(t - \tau)$ and another particle, $i$, which at the same time holds a position displaced a distance $\xi_y$ relative to $j$. 
Figure 3 shows the trajectories of an arbitrary chosen pair of fluid particles, \(i\) and \(j\). Obviously, the separation of these particles is the same as measured in both the fixed and moving frame:

\[
\xi_{ij} = x_i(t - \tau) - x_j(t - \tau) \\
= y_i(t - \tau) + c(t - \tau) - \{y_j(t - \tau) + c(t - \tau)\} \\
= y_i(t - \tau) - y_j(t - \tau). \tag{34}
\]

By fixing the variable \(\xi_{ij}\) at a constant value, we consider the joint probability distribution of the remaining two variables \(y_i(t)\) and \(y_j(t - \tau)\). This is called the conditional probability of \(y_i(t), y_j(t - \tau)\); it is conditional on \(\xi_{ij}\) having a prescribed value. We denote this probability \(S(y_i(t), y_j(t - \tau) | \xi_{ij})\). Suppose we know the joint probability \(S(y', y'' | \xi_{ij})\) for finding the fluid particle \(i\) in the interval \(dy'\) associated with \(y'\) at time \(t\) and the fluid particle \(j\) in the interval \(dy''\) associated with \(y''\) at time \(t - \tau\), under the condition that the separation of the two particles, at time \(t - \tau\), is given by the fixed distance \(\xi_{ij} = y_i(t - \tau) - y_j(t - \tau)\). Then, the contribution from this particular particle pair, \(i\) and \(j\), to the total covariance \(\langle \hat{u}(y', t)\hat{u}(y'', t - \tau) \rangle = \langle (1/M) \Sigma_i \hat{u}(y_i(t)) (1/M) \Sigma_i \hat{u}(y_i(t - \tau)) \rangle\), can be calculated as \(S(y', y'' | \xi_{ij}) \langle \hat{u}(y_i(t))\hat{u}(y_j(t - \tau)) \rangle\), where the ensemble-averaged covariance function of the velocity of the pair

\[
\langle \hat{u}(y_i(t))\hat{u}(y_j(t - \tau)) \rangle = \langle \hat{u}(y_i(t))\hat{u}(y_j(t - \tau) - \xi_{ij}) \rangle \tag{35}
\]

is also subject to the condition that the particle pair separation at time \(t - \tau\) equals the fixed quantity \(\xi_{ij}\). The fixed point covariance function \(\langle \hat{u}(y', t)\hat{u}(y'', t - \tau) \rangle\) can in this way be obtained by summing the pair contributions from all possible values of the fixed separation \(\xi_{ij}\) in the fluid. This leads to an integral over \(\xi_{ij}\), where we explicitly make use of assumptions 1, that changes in the turbulence characteristics are all on scales larger than the small increment \(d\xi_{ij}\), and 2, that any two particles are uncorrelated in the moving frame:

\[
\langle \hat{u}(y', t)\hat{u}(y'', t - \tau) \rangle = \int_{-\infty}^{\infty} S(y', y'' | \xi_{ij}) \langle \hat{u}(y_i(t))\hat{u}(y_j(t - \tau) - \xi_{ij}) \rangle d\xi_{ij}. \tag{36}
\]

It is now convenient to change the frame of reference for the two-particle covariance function on the right-hand side of Eq. (36) back to the fixed coordinate system. We do this by use of the relation \(\hat{u}(y_i) = u(y_i + c) = u(x_i)\). The two-particle covariance becomes

\[
\langle \hat{u}(x_i(t))\hat{u}(x_j(t - \tau) - \xi_{ij}) \rangle = \langle u(x_i(t))u(x_j(t - \tau) - \xi_{ij}) \rangle \tag{37}
\]

where the condition \(x_i(t - \tau) = x_j(t - \tau) + \xi_{ij}\) is now imposed on the separation of the two particles. As Fig. 4(a) shows, Eq. (37) expresses the correlation between the velocity of a fluid particle \(i\) at time \(t\) in the position \(x_i\), and the velocity at time \(t - \tau\) of fluid particle \(j\) that is displaced by a distance \(\xi_{ij}\) relative to \(x_i(t - \tau)\).

Alternatively, by referring the fixed distance \(\xi_{ij}\) separating the two particles to time \(t\) as shown in Fig. 4(b), rather than to time \(t - \tau\), the covariance function between the two particles reads \(\langle u(x_i(t - \tau))u(x_i(t) + \xi_{ij}) \rangle\), where now \(x_i(t) = x_j(t) + \xi_{ij}\). In a stationary and homogeneous turbulent field these alternative definitions are identical, since the situation in Fig. 4(b) follows immediately from a time reversal of the situation in Fig. 4(a). Moreover, these covariance functions will be independent of both absolute position \(x\) of the fluid particles and of the absolute time \(t\). This leaves a function of the time lag \(\tau\) and separation \(\xi_{ij}\) alone, which we define as

\[
R_{abs}(\xi_{ij}, \tau) = \langle u(x_i(t))u(x_i(t - \tau) - \xi_{ij}) \rangle \tag{38}
\]

\[
= \langle u(x_i(t - \tau))u(x_i(t) + \xi_{ij}) \rangle.
\]
Though similar, the present unconditionally sampled covariance function $R_{ab}(\xi_{ij}; \tau)$ differs in significant ways from the two-particle, two-time Lagrangian covariance $\langle u_i(t)u_j(t-\tau) \rangle$ introduced by Batchelor (1952). His covariance applies to the marked fluid particles only and, as discussed in section (a), will in general depend on the particle separation at zero time $\xi_{ij}^0$, in addition to the variables $(t, \tau)$, see Eq. (22).

Our covariance in Eq. (38) is based on an unconditional ensemble of both marked and unmarked fluid. This leaves a function, which in homogeneous and stationary turbulence depends only on (1) the time lag $\tau$, and (2) the instantaneous particle

![Diagram](image)

**Figure 4.** The two-particle covariance function defined in Eq. (38). (a) Referring the fixed particle separation $\xi_0$ to time $t - \tau$: $\langle u_i(t)u_j(t-\tau) - \xi_0(t-\tau) \rangle$. (b) Referring $\xi_0$ to time $t$: $\langle u_i(t-\tau)u_j(t) + \xi_0(t) \rangle$. In homogeneous and stationary turbulence, these two definitions are identical.
separation \(\xi_{ij}\). Also, from a modelling point of view, \(R(\xi_{ij}, \tau)\) is definitely the more tractable of the two.

Setting \(\xi_{ij} = 0\) reduces our two-particle covariance function to the Lagrangian autocovariance function of a single particle: \(R_{abs}(0, \tau) = \langle u_i(t) u_i(t - \tau) \rangle\). On the other hand, by setting \(\tau = 0\), a pure Eulerian space-covariance results (where the separation \(\xi_{ij}\) is along the direction of the velocity component \(u_i\)). With both \(\xi_{ij}\) and \(\tau\) set equal to zero, it yields the total energy \(\langle u^2 \rangle\) of the turbulence. \(R_{abs}(\xi_{ij}, \tau)\) therefore also defines, with \(\xi_{ij} = 0\), the Lagrangian integral time scale appropriate for a single particle through

\[
\tau_L = \langle u^2 \rangle^{-1} \int_0^\infty R_{abs}(0, \tau) d\tau. \tag{39}
\]

Also, a (fixed point) Eulerian integral length scale for the turbulence is given by

\[
\ell_E = \langle u^2 \rangle^{-1} \int_0^\infty R_{abs}(\xi_{ij}, 0) d\xi_{ij}. \tag{40}
\]

It remains to investigate the joint probability distribution \(S(y', y''|\xi_{ij})\) in Eq. (36) for finding the \(i\)th fluid particle inside the interval \(y', y' + dy'\) at time \(t\), and the \(j\)th fluid particle in the interval \(y'', y'' + dy''\) at time \(t - \tau\), given that \(\xi_{ij} = y_j(t - \tau) - y_i(t - \tau)\) is the separation of the pair. This is accomplished by means of the previously introduced Gaussian transition probability \(G_{\Delta y_i}\), for a fluid particle. For the (arbitrary) fluid particle \(j\) (in the interval \(y'', y'' + dy'')\) we identify a corresponding particle \(i\) by keeping \(\xi_{ij}\) fixed at a constant value. The probability that the \(i\)th particle in the interval \(t - \tau\) to \(t\) will reach the interval \(y', y' + dy'\) is here given by

\[
S(y', y''|\xi_{ij}) = G_{\Delta y_i}[y' - y_i(t - \tau)]
= G_{\Delta y_i}[y' - y_i(t - \tau) - \xi_{ij}]
= G_{\Delta y_i}(y' - y'' - \xi_{ij}) \tag{41}
\]

where the spread \(\langle \Delta y_i^2 \rangle\) associated with the transition probability \(G_{\Delta y_i}\), is given by \(\sigma^2(t) - \sigma^2(t - \tau)\), cf. Eq. (32). By substituting this, together with the two-particle covariance function in Eq. (38), the following expression is finally obtained for the fixed point Eulerian velocity covariance in Eq. (36):

\[
\langle \bar{u}(y', t) \bar{u}(y'', t - \tau) \rangle = \int_{-\infty}^{\infty} G_{\Delta y_i}(y' - y'' - \xi_{ij}) R_{abs}(\xi_{ij}, \tau) d\xi_{ij}. \tag{42}
\]

It is now possible to calculate the centre-of-mass covariance function in Eq. (27) explicitly. The following integral results

\[
R_{cm}(t, \tau) = \iint R_{abs}(\xi, \tau) G_{\Delta y_i}(y' - y'' - \xi) G_{\sigma(t)}(y') G_{\sigma(t - \tau)}(y'') dy' dy'' d\xi. \tag{43}
\]

By keeping \(\xi = \xi_{ij}\) fixed, the remaining two integrals are simply (double) convolutions of two Gaussian distribution functions. The result of this is another Gaussian with standard deviation equal to the square root of the sum of the individual variances: \([\sigma^2(t) - \sigma^2(t - \tau)] + \sigma^2(t) + \sigma^2(t - \tau)]^{1/2} = \sqrt{2/\sigma(t)}\). This leads to the following result for the centre-of-mass covariance function expressed in terms of the two-particle covariance function \(R_{abs}(\xi, \tau)\) defined in Eq. (38)

\[
R_{cm}(t, \tau) = \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi} \sigma(t)} \exp \left\{ -\frac{1}{4} \frac{\xi^2}{\sigma(t)} \right\} d\xi. \tag{44}
\]
Our expression has the following properties:
(a) When an initially small puff is released, $\sigma$ is much smaller than $l_E$ and in this case $R_{cm}(t, \tau) = R_{abs}(0, \tau)$. This implies that the centre-of-mass covariance function and, consequently, the centre-of-mass spread, equals that of a single particle in this limit.
(b) In the other limit, when $\sigma$ has grown to a size much greater than the length scale $l_E$, $R_{cm}(t, \tau)$ becomes small compared with $R_{abs}(\tau)$. This implies that the centre-of-mass dispersion ($c^2$) becomes negligible in this far-field limit, and that the relative diffusion $\sigma^2$ is entirely dominated by single-particle diffusion $\langle x^2 \rangle$.

With the centre-of-mass covariance function Eq. (44) inserted into Eq. (14), a first-order differential equation for the growth of a Gaussian puff results:

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_0^t \left\{ R_{abs}(0, \tau) - \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi \cdot \sigma(t)}} \exp\left(-\frac{1}{4 \frac{\xi^2}{\sigma^2(t)}}\right) d\xi \right\} d\tau.$$  \hspace{1cm} (45)

In Eq. (13) the cloud dispersion was defined in terms of a mean square relative velocity $\langle v^2(t) \rangle$ and a relative Lagrangian time scale $t_r(t)$:

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \overline{\langle v^2(t) \rangle} t_r(t).$$  \hspace{1cm} (46)

Setting $\tau = 0$ in Eq. (44), and by use of Eq. (10), the mean square relative velocity can here be identified as

$$\overline{\langle v^2(t) \rangle} = R_{abs}(0, 0) - \int_{-\infty}^{\infty} R_{abs}(\xi, 0) \frac{1}{2\sqrt{\pi \cdot \sigma(t)}} \exp\left(-\frac{1}{4 \frac{\xi^2}{\sigma^2(t)}}\right) d\xi.$$  \hspace{1cm} (47)

Equivalently, the relative correlation function $r(t, \tau)$ as defined in Eq. (13) here explicitly becomes

$$r(t, \tau) = \overline{\langle v^2(t) \rangle}^{-1} \left\{ R_{abs}(0, \tau) - \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \frac{1}{2\sqrt{\pi \cdot \sigma(t)}} \exp\left(-\frac{1}{4 \frac{\xi^2}{\sigma^2(t)}}\right) d\xi \right\}. \hspace{1cm} (48)$$

Given this correlation function, the relative time scale $t_r(t)$ is easily obtained from an integration of $r(t, \tau)$ with respect to $\tau$: $t_r = \int_0^t r(t, \tau) d\tau$ (cf. Eq. (13)).

(d) Spectral formulation of the relative diffusion model

It is convenient to introduce a spectral representation for the two-particle covariance $R_{abs}(\xi, \tau)$ defined in Eq. (38) for stationary and homogeneous turbulence. We can define the spectrum $S(k, \omega)$, where $k$ is the wavenumber and $\omega$ the frequency, by the Fourier transform

$$S(k, \omega) = (2\pi)^{-2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R_{abs}(\xi, \tau) \exp\{-i(k\xi + \omega\tau)\} d\xi d\tau$$  \hspace{1cm} (49)

and the covariance $R_{abs}(\xi, \tau)$ is correspondingly given by

$$R_{abs}(\xi, \tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \exp[i(k\xi + \omega\tau)] dk d\omega. \hspace{1cm} (50)$$

By inserting this in Eq. (45) for the growth rate of the cloud, followed by integrations over the lags $\xi$ and $\tau$, we arrive, without loss of generality, at

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} S(k, \omega) \sin(\omega t) / \omega t [1 - \exp(-k^2 \sigma^2(t))] \, d\omega \, dk.$$  \hspace{1cm} (51)
As before, we will also consider here the limiting cases: (a) In the limit where the cloud size $\sigma$ is large compared with the length scale $l_T$ of the turbulence (cf. Eq. 40), Eq. (51) reduces to the well-known G. I. Taylor's formula for single-particle diffusion:

$$\sigma^2(t) = t^2 \int_{-\infty}^{\infty} S_L(\omega) \{ \sin^2(\frac{1}{2} \omega t) / (\frac{1}{4} \omega t)^2 \} \, d\omega$$  \hspace{1cm} (52)

where the Lagrangian spectrum $S_L(\omega)$ is simply related to $S(k, \omega)$ by

$$\langle u^2 \rangle S_L(\omega) = \int_{-\infty}^{\infty} S(k, \omega) \, dk.$$  \hspace{1cm} (53)

Not surprisingly, we find that the different behaviour of the spread of a cloud, when compared with that of a single particle, is closely related to the spatial correlation of the turbulence. (b) In the limit where the time $t$ is also large compared with the time scale $t_s$, Eq. (51) reduces to the usual form $\sigma^2 = 2 \langle u^2 \rangle t_s t$, appropriate for single-particle dispersion in the far-field limit.

(e) Simplified solution of the relative diffusion equation

In his unpublished note in 1935 G. I. Taylor suggested that a two-particle covariance function could be estimated as the product of a two-point (Eulerian) space-correlation: $\rho_E(\xi) = \langle u(x, t) u(x + \xi, t) \rangle / \langle u^2 \rangle$ and a single-particle (Lagrangian) autocorrelation: $\rho_L(\tau) = \langle u(x(t)) u(x(t - \tau)) \rangle / \langle u^2 \rangle$, see for instance Batchelor (1952) or Sawford (1982) for a detailed discussion. With this approximation, the two-particle covariance function in Eq. (38) becomes

$$R_{ab}(\xi, \tau) = \langle u^2 \rangle \rho_E(\xi) \rho_L(\tau).$$  \hspace{1cm} (54)

In his intercomparison Sawford (1982) found that this type of approximation was the most appropriate. However, already Batchelor (1952) had pointed out that this approximation cannot in general be valid, except perhaps in the limit when $\tau$ is small compared with the Lagrangian integral scale $t_s$.

Based on Eq. (54) the spectrum in Eq. (49) reduces to

$$S(k, \omega) = \langle u^2 \rangle S_E(k) S_L(\omega)$$  \hspace{1cm} (55)

where the wavenumber spectrum $S_E(k)$ and the frequency spectrum $S_L(\omega)$ are the spectra corresponding to the Eulerian ($\rho_E(\xi)$) and the Lagrangian ($\rho_L(\xi)$) correlations, respectively, defined as

$$S_E(k) = (2\pi)^{-1} \int_{-\infty}^{\infty} \rho_E(\xi) \exp(-ik\xi) \, d\xi$$  \hspace{1cm} (56a)

and

$$S_L(\omega) = (2\pi)^{-1} \int_{-\infty}^{\infty} \rho_L(\tau) \exp(-i\omega\tau) \, d\tau.$$  \hspace{1cm} (56b)

Consequently, with the approximation proposed in Eq. (54) the puff growth rate formula, Eq. (45), reduces to

$$\frac{1}{2} d\sigma^2 / dt = \langle u^2 \rangle \int_{0}^{t} \rho_L(\tau) \, d\tau \int_{-\infty}^{\infty} S_E(k) \{1 - \exp(-k^2 \sigma^2)\} \, dk.$$  \hspace{1cm} (57)
This equation is of the form
\[ \frac{d\sigma^2}{dt} = \overline{(v^2(t))} t_e(t) \]  
(58)
from which we can identify the mean square relative velocity
\[ \overline{(v^2(t))} = \langle u^2 \rangle \int_{-\infty}^{\infty} S_E(k) \{1 - \exp(-k^2 \sigma^2)\} \, dk \]  
(59)
and the relative Lagrangian time scale function
\[ t_e(t) = \int_0^t \rho_L(\tau) \, d\tau. \]  
(60)
If Taylor’s formula for single-particle diffusion is written as
\[ \frac{d\sigma^2}{dt} = \langle u^2 \rangle \int_0^t \rho_L(\tau) \, d\tau = \langle u^2 \rangle t_s(t) \]
(see e.g. Pasquill and Smith (1983)), we find that the relative time scale \( t_e(t) \) now becomes identical to the time scale \( t_s(t) \) appropriate for single-particle diffusion. This results as a consequence of the ‘factorization’ of \( R_{abs} \) in Eq. (54). The mean square relative velocity, however, is still, as already found in Eq. (47), exclusively related to the Eulerian properties of the turbulence.

Before proceeding by solving the set of equations (58), (59) and (60), it remains to investigate whether the now very simple formula for the relative time scale, Eq. (60), yields results that are consistent with our expectations for turbulent dispersion. In the limit for ‘large’ times \( t \gg t_L \), the relative time scale \( t_e(t) \) becomes equal to \( t_L \) as it properly should in this limit where the particles in effect are separated so far from each other that they move independently, and also have forgotten their initial velocity. According to Eq. (60), on the other hand, the relative time scale for small diffusion times becomes
\[ t_e(t) = t \quad \text{for} \quad t \ll t_L \]  
(61)
since for small time lags, \( \rho_L(\tau) \approx 1 \). The same limiting value, \( t_e(t) \approx t \), is also obtainable from the spectral version of the more general growth rate formula, Eq. (51), without ‘factorization’ of \( R_{abs} \): for small values of \( t \), the function \( \sin(\omega t)/(\omega t) \) in Eq. (51) remains close to unity for all values of \( \omega \) where \( S(k, \omega) \) contributes to the integral (see Fig. 5).

To first order we therefore have the approximation
\[ \int_{-\infty}^{\infty} S(k, \omega) \{\sin(\omega t)/(\omega t)\} \, d\omega = \int_{-\infty}^{\infty} S(k, \omega) \, d\omega = \langle u^2 \rangle S_E(k). \]  
(62)
Therefore Eq. (51) becomes
\[ \frac{d\sigma^2}{dt} = t \langle u^2 \rangle \int_{-\infty}^{\infty} S_E(k) \{1 - \exp(-k^2 \sigma^2)\} \, dk = t \overline{(v^2(t))} \]  
(63)
from which we retrieve that \( t_e(t) \approx t \) in this small time limit.

Any difference between the relative time scales with and without the assumed ‘factorization’ of \( R_{abs}(S, \tau) \) is therefore to be expected only for values of \( t \approx t_L \).

(f) **Comparison with the analysis of Smith and Hay**

At this point we can compare our approach with that of Smith and Hay (1961) as outlined in section (b). First, we point to some fundamental differences in the theoretical
Figure 5. Iso-contour plot of a hypothetical spectrum $S(k, \omega)$. Its maximum value is at $(k, \omega) = (0, 0)$ from where the function monotonically decreases through the levels I, II and III. The cut-off frequency associated with the low-pass filter $\sin(\omega t)/\omega t$ is schematically drawn as the vertical line at $\omega = \tau^{-1}$. Correspondingly, the high-pass filter $\{1 - \exp(-k^2\sigma^2)\}$ cuts away wavenumbers that are smaller than $\sigma^{-1}$. The shaded area therefore represents the part of the spectrum $S(k, \omega)$ that essentially contributes to the integral over $k$ and $\omega$ in Eq. (51).

derivations. Next, we compare the resulting growth rate equations and finally, we investigate the corresponding solutions for $\sigma(t)$.

In retrospect, our theoretical approach was based on the hypothesis of vanishing trajectory correlations between fluid particles in the moving frame, cf. Eq. (29). Our justification thereof is that the high-pass filtered energy, contributing to the expansion of the puff, is limited in eddy size to the smaller eddies only $(k > 1/\sigma)$, which in turn have limited length and time scales. As a consequence, the transition probability function for diffusion in the relative frame $G_{\Delta\nu}$ could be described by a non-stationary Markov process. This property allowed us to relate the centre-of-mass covariance $R_{cm}(t, \tau)$ to the two-particle covariance function for unconditional sampled fluid particles $R_{sab}(\xi, \tau)$ in Eq. (44).

Now compare our expression Eq. (44) with Eq. (24) of Smith and Hay. Despite significant differences in derivation, these results differ only by the covariance functions in the integral. Here, Smith and Hay's approach relates to the covariance $R_{E}(\xi + Ut/\beta)$, whereas we arrive at $R_{sab}(\xi, \tau)$. The fixed form of the space and time lag argument $(\xi + Ut/\beta)$ reflects the 'frozen turbulence' hypothesis adopted in Eq. (23), whereas at this point we deal with space and time as independent variables. In our opinion this is the most significant difference between the two theories.

To bring out any differences in the predicted puff spread, we proceed as in section (e) and estimate $R_{sab}(\xi, \tau)$ by the Taylor approximation, Eq. (54). This additional approximation brings our covariance in Eq. (44) to a form identical to the one Sawford obtained by use of the G. I. Taylor approach, cf. Sawford (1982), his Eq. (7):

$$R_{cm}(t, \tau) = \langle u^2 \rangle \rho_{L}(\tau) \int_{-\infty}^{\infty} \rho_{E}(\xi) G_{\sqrt{2}, \sigma(t)}(\xi) d\xi.$$  

(64)
Consequently, Sawford’s study applies to our Eq. (57) as well. In his conclusion, Sawford, p. 204, comments on the existence of a retarded growth region in the Smith–Hay solution which is neither predicted by Batchelor’s similarity theory nor supported by observations. This gives rise to differences in the predictions, between our approach and the Taylor approach solutions on the one hand, and the Smith–Hay solution on the other. This difference is most significant at intermediate travel times ($0.1 < Ut/\beta L_E < 10$), and increases with decreasing initial puff size. However, since the simplified equation (57) also invokes the Taylor approximation, our finding cannot be used as an independent assessment of this difference.

In summary, the growth rate equation (45) in section (c) and the corresponding spectral form, Eq. (51) in section (d), seem to have fewer constraints than the corresponding Smith–Hay approximation. At this point of derivation, our approach does not invoke either the Taylor frozen-turbulence hypothesis nor the Hay–Pasquill $\beta$ hypothesis. However, these general equations are practical only for calculating the spread in the near-field limit for very small diffusion times, cf. Eq. (63), and in the trivial far-field limit for very large times. To obtain feasible solutions for the intermediate diffusion times as well, we also adopt the Taylor approximation, thereby separating space and time variables in the product of an Eulerian and a Lagrangian covariance function. This can give rise to significant differences in predictions based on our Eq. (57) and the Smith–Hay approach. Although the limited observations compiled by Sawford seem to indicate that the Taylor approximation is more appropriate, this conclusion cannot yet be regarded as decisive.

(g) *Puff diffusion related to Eulerian power law spectra*

In search for analytical solutions in the near-field limit, we now proceed from our result in Eq. (63). In the following the Eulerian wavenumber spectrum is assumed to be given by a power law

$$ S_E(k) = \delta k^p $$

where $\delta$ is a constant with dimensions $m^{1+\gamma}$. It should be emphasized that any power law representation is of relevance only over limited ranges of wavenumbers. For instance, at very small wavenumbers ($k \sim 0$), realistic spectra tend to be flat ($p = 0$) and their asymptotic amplitude is $S_E(k) \sim l_E/\pi$ where $l_E$ is the Eulerian integral length scale. Also, spectra with spectral index $p \geq -1$ are meaningful only in connection with a high-frequency cut-off that limits their total energy.

Further, we consider only puff diffusion times $t$ that are small relative to the Lagrangian time scale $t_L$, estimated as $l_E/(u^2)^{1/2} = \pi S_E(k \sim 0)/(u^2)^{1/2}$. Consequently, we have $t_L \sim t$, and the puff growth can be calculated from Eq. (63). For values of the spectral index within the interval $-3 < p < -1$, we find

$$ \langle v^2(t) \rangle = \frac{-2(u^2)\delta/(p + 1))\Gamma(\frac{1}{p + 3})\sigma^{-(p + 1)}}{\langle v^2(t) \rangle} $$

where $\Gamma$ denotes the gamma function.

For values of $p \leq -3$ we approximate the high-pass filter $\{1 - \exp(-\frac{1}{2}k^2\sigma^2)\}$ by a Heaviside step-function and get

$$ \langle v^2(t) \rangle = \frac{-2(u^2)\delta/(p + 1))\sigma^{-(p + 1)}}{\langle v^2(t) \rangle} \quad \text{for} \quad p \leq -3. $$

The puff growth rate equation (63) is now easily solved to give

(i) for $-3 < p < -1$ \quad $\sigma(t) = (ct^2 + \sigma_0^{1/\gamma})^q$

(ii) for $p = -3$ \quad $\sigma(t) = \sigma_0 \exp(\frac{1}{2}\delta (u^2) t^2)$

(iii) for $p < -3$ \quad $\sigma(t) = (ct^2 + \sigma_0^{1/\gamma})^q$. 

\[ \{ \]
Here, \( q = 1/(3 + p) \), \( \varepsilon = -\langle u^3 \rangle \delta (3 + p)/(1 + p) \) and \( c = \varepsilon \Gamma[\xi/(p + 3)] \). \( \sigma_0 \) is the initial puff size, i.e. \( \sigma(t = 0) \). The solution (iii) requires in addition that \( t < t_{\max} \), \( t_{\max} \) being \( (\sigma_0^{1/4}/|\varepsilon|)^{1/2} \). The limit \( t = t_{\max} \), however, is never approached with a physically meaningful spectrum, which tends to be flat \( (p \sim 0) \) at small wavenumbers.

In the initial phase of spread where \( \sigma_0 \) is an important scaling parameter, the behaviour of \( \sigma(t) \) can be deduced from Eq. (63) by substituting Eq. (66) (or Eq. (67)) for \( \langle u^3(0) \rangle \) with \( \sigma(0) = \sigma_0 \). Hence, for diffusion times \( t \ll (\sigma_0^2/\langle u^3(0) \rangle)^{1/2} \), an expansion of the initial spread to second order gives
\[
\sigma^2(t) = \sigma_0^2 + \langle u^3(0) \rangle t^2.
\]
(69)

This result is in accordance with the similarity scaling suggested by Batchelor (1952) for the near-field limit.

Next, consider the time interval described by Batchelor as "intermediate", i.e. when viscosity and the initial puff size are no longer dominant parameters, but before the integral time scale \( t_L \) becomes an important scaling parameter. The solution for the spectral index in the interval \(-3 < p < -1\) follows from Eq. (68):
\[
\sigma(t) = c t^{(3 + p)/4}.
\]
(70)

For instance, by setting \( p = -5/3 \) the cloud is found to grow as
\[
\sigma(t) = [2\langle u^3 \rangle \Gamma(2/3)\delta]^{3/4} t^{3/2}
\]
(71)

which also is in agreement with the inertial subrange '3/2 power law' scale-argument suggested by Batchelor (1952). In order to evaluate the coefficient, we make use of the spectrum suggested for homogeneous and isotropic turbulence described by, e.g., Tennekes and Lumley (1972):
\[
\langle u^2 \rangle S_E(k) = \alpha e^{2/3} k^{-5/3}.
\]

The Eulerian correlation function \( \rho_E(\xi) \), also leading to \( S_E(k) \) in Eq. (56a), is defined in terms of velocities that are parallel to the line of separation \( \xi \). The constant \( \alpha \) is consequently chosen to be \((9/55) \times 1.5\) for the longitudinal, one-dimensional spectrum in question. Setting \( \delta = \alpha e^{2/3}/\langle u^2 \rangle \), Eq. (71) now yields
\[
\sigma^2(t) = 0.534\varepsilon t^3
\]
(72)

which applies to 'intermediate' diffusion times within the inertial subrange. In addition to this (already well-known) proportionality between \( \sigma^2 \) and \( \varepsilon t^3 \), our model also suggests a value for the constant of proportionality \((0.534)\). For this inertial subrange several other authors have derived corresponding formulae. Based on the Langevin equation, both Smith (1968) and Gifford (1982) calculated the following equation for three-dimensional, isotropic turbulence:
\[
\sigma^2(t) = \frac{4}{3} \varepsilon t^3.
\]
(73)

The two authors used very different interpretations of the Langevin equation, but Sawford (1984) has recently pointed out the similarity between these for three-dimensional inertial subrange turbulence. From his analysis it also becomes clear that the numerical coefficient in Eq. (73), \( 2/3 \), results as a consequence of the exponential autocorrelation function inherent in the Langevin equation. With \( \rho_L = \exp(-\tau/t_L) \), the universal constant \( c_0 \) in the Lagrangian structure function \( D(\tau) = c_0 \varepsilon \tau \) equals 2. Since \( \sigma^2(t) = \frac{4}{3} c_0 \varepsilon t^3 \), our model calculates \( c_0 \) to be 1.60. Another interesting case is obtained by setting \( p = -3 \) which corresponds not only to the enstrophy cascade subrange in global-scale two-dimensional turbulence (Gage 1979), but also to observations obtained in very stable stratified surface
layer turbulence (Olesen et al. 1984). For these cases, Eq. (68) predicts an exponential puff growth. Similar results were obtained by Lin (1972) based on dimensional analysis for relative diffusion within the enstrophy cascade subrange.

3. Experimental evaluation

(a) Instantaneous diffusion observations in the atmospheric surface layer

The experimental evaluation of the puff growth formula is based on a recent series of experiments carried out over homogeneous terrain in Denmark. Combined with aerial photographs of surface-released smoke plumes, wind data were obtained from a horizontal array of tower-mounted sonic anemometers. These data provided information about the spatial and temporal variability of the dispersing wind field, including estimates of the correlations $\rho_E(\xi)$ and $\rho_L(\tau)$.

Appropriate for direct use with measured correlations the growth rate Eqs. (58)–(60) were written in the form

$$\frac{1}{2} \frac{d\sigma^2}{dt} = \langle \bar{v}^2(t) \rangle t_c(t)$$

(74a)

where

$$t_c(t) = \int_0^t \rho_L(\tau) d\tau$$

(74b)

and

$$\langle \bar{v}^2(t) \rangle = \langle u^2 \rangle \left[ 1 - \frac{1}{2\sqrt{\pi} \sigma} \int_{-\infty}^{\infty} \rho_E(\xi) \exp \left( -\frac{1}{4} \frac{\xi^2}{\sigma^2} \right) d\xi \right].$$

(74c)

Equation (74c) is merely a non-spectral form of Eq. (59), as can be seen from Eqs. (45) and (54). If preferred, Eq. (74c) can be interchanged with Eq. (59) for use with measured spectra.

The experimental site was a 1 km $\times$ 1 km homogeneous area in the middle of the Borris Moors, a part of the Danish army exercise area in Jutland. Four 10 m-high masts were placed in a north–south array, and a three-axis sonic anemometer/thermometer (Kaijo Denki type DAT300) was mounted on top of each mast. By suitable positioning of the masts, it was possible to obtain estimates of the cross-covariances of all three velocity components for six, approximately logarithmically equidistant, lateral displacements in the range from 4.5 to 30 m. Smoke pots (Hexit) were placed in a stack on the base line between the masts and, when ignited, the smoke pots produced a white visible plume that could be observed from a small aircraft as far as ~1 km down-wind under light wind conditions. The visible contour of the white smoke plume was registered on photographs taken from an airplane circling above the test site. Cross-wind arrays of white contrast plates were placed at down-wind distances of 31.25, 62.5, 125, 250 and 500 m. Figure 6 gives an example of a smoke plume photograph taken from an altitude of 1000 feet. A series of approximately 20 such pictures could be obtained during each 40 min smoke release.

By comparing the outline of the plume with the marker plates, the visible contour of the smoke plume was determined as a function of the distance from the source point. By subsequent use of an opacity method (Gifford 1980) the corresponding lateral standard
deviation, $\sigma_r$, of the instantaneous plume was inferred. This method was used to obtain ensembles of the instantaneous spread (standard deviations) as a function of down-wind distance. A more detailed description of the experimental set-up and the subsequent processing of both the smoke and sonic data are given by Mikkelsen (1983) and Mikkelsen and Eckman (1983).

Figure 6. Photograph of the instantaneous smoke plume taken from $\sim$1000 ft above the source point. The white contrast plates constitute a network of lines normal to the $x$ axis at the following downwind distances: 31.25, 62.50, 125, 250 and 500 m. Their lateral spacing is 5 m in the first three rows, 10 m in the 250 m row, and 20 m in the 500 m row.
(b) Results

On 28 August 1980, between 1520 and 1600 h local time, 22 plume photographs were taken at approximately two-minute intervals from an elevation of 1000 feet. The sky was clear and the mean wind, $U$, was later determined to be 4.72 m s$^{-1}$ over this period, indicating slightly unstable conditions. (A Monin–Obukhov length, $L = -150$ m was later determined from the sonic anemometers.)

Figure 7 shows the entire ensemble of obtained instantaneous standard deviations, $\sigma_x$, from the experiment BOREX80, run 6, as a function of the downwind distance. By averaging the observed instantaneous $\sigma_x$ values at fixed positions, the ensemble spread $\langle \sigma_x \rangle$ was obtained as a function of downwind distance, $x$. Figure 8 shows this averaged standard deviation $\langle \sigma_x \rangle$, together with the centre-of-mass dispersion $\langle y_{cm}^2 \rangle^{1/2}$ of the plume. The latter is measured with respect to a coordinate axis aligned with the mean wind direction over the 40 min period. Also shown in Fig. 8 is the total lateral dispersion: $\Sigma^2 = \langle y_{cm}^2 \rangle + \langle \sigma_x \rangle^2$ corresponding to Taylor's (1921) theory for single-particle diffusion $\frac{d\Sigma^2}{dt} = \langle u^2 \rangle t_\alpha(t)$ where $t_\alpha = \int_0^\infty \rho_L(\tau) \, d\tau$. The average standard deviation of the instantaneous lateral spread, $\langle \sigma_x \rangle$, is found to a first approximation to grow linearly with the downwind distance. Within the surface layer, this observation is to be expected based on the scaling argument by Chatin (1968, p. 351): $\langle \sigma_x \rangle = \beta u_* t$ where $\beta$ is a constant (of order unity).

The measured lateral velocity spectrum indicated an Eulerian integral time scale of the order of $\sim 100$ s. Assuming for the unstable flow considered that the Lagrangian time scale $t_L$ is also $\approx 100$ s, the following simple expression for the absolute time scale

$$t_a(t) = t_L (1 + t_L / t)^{-1}$$

(75)
was used for calculation of $\Sigma(t)$. Figure 8 shows that this models the single-particle diffusion data very well. Therefore Eq. (75) was also used as a model for $t_1(t)$ in Eq. (74b), cf. the discussion in connection with Eq. (60).

The six measured cross-correlations of the lateral velocity component were well approximated by the function $\rho_\ell(\xi) = 1.05 - 0.20 \log_{10}(\xi)$, appropriate for values of the cross-wind separation $\xi$ in the interval from 4.5 to 30 m. Also the lateral velocity variance, $\langle u^2 \rangle$, was measured to be 0.98 m$^2$s$^{-2}$.

In this way, by specifying $\langle u^2 \rangle$, $\rho_\ell(\xi)$ and $t_0$ in accordance with data obtained during the experiment, our puff formula, Eq. (74), could be integrated to obtain $\sigma$ as a function of the diffusion time $t$. By subsequent use of the relation $x = Ut$, the calculated puff size $\sigma$ can be compared with the experimentally determined values of the ensemble mean plume width $\langle \sigma \rangle$ in Fig. 8.

The specifications of $\langle u^2 \rangle$ and $\rho_\ell(\xi)$ were, from a sensitivity analysis, found to be the more critical input parameters, whereas changes in the specification of $t_0$ showed relatively little influence on the result. Use of $\rho_\ell(\xi)$ from a single mast (possible only with an additional assumption about frozen and isotropic turbulence) consequently led to a significantly poorer agreement of $\sigma(t)$ with the data in Fig. 8 (cf. Mikkelsen 1983).

For comparison, Fig. 8 also shows $\sigma$ calculated from the formula originally derived by Smith and Hay (1961) and later calibrated by Pasquill (see Pasquill and Smith 1983): $\sigma = 0.22ix$, where $i$ denotes the turbulence intensity based on the total energy of the

![Figure 8. Ensemble-averaged standard deviation ($\sigma$) of the instantaneous observed spread in Fig. 7 versus down-wind distance $x$. The bars through the data points represent the standard deviation of the ensemble of instantaneously observed spreads at a fixed down-wind distance. Dispersion associated with the movement of the centreline of the instantaneous plume $(\langle y^2 \rangle)^{1/2}$ has been indicated by \$\Delta\$. The total lateral plume dispersion $\Sigma$ is indicated by $\bullet$. The upper solid line is calculated from Taylor's (1921) single-particle diffusion theory: $t_{\Sigma} \Sigma^2/dt = \langle u^2 \rangle t_0(i)$. The lower solid lines show integrations of the puff-growth equation (74) with two instantaneous initial values: $\sigma(0) = 0.5$ m (upper branch) and $\sigma(0) = 0.25$ m (lower branch). Also shown (dashed line) is the puff formula of Smith and Hay (1961): $\sigma = 0.22ix$.](image-url)
turbulence. For operational and quick calculations, this formula has been used successfully in the Risø small-scale puff diffusion model (Mikkelsen et al. 1984). In this study this formula estimates the relative diffusion a little too high, although it has the linear character implied by the data. The value of $\beta_i$ given in Pasquill and Smith (1983) is tentative and subject to future modification. Moreover, in a strong anisotropic surface layer, it is not surprising that horizontal spread (as seen by the aircraft) is not altogether well described by the total (three-dimensional) intensity of turbulence.

A similar experiment carried out under more neutral atmospheric conditions (BOREX81, run 1B) has also been compared with calculations based on the puff growth equation (74) (see Mikkelsen and Eckman 1983). In both experiments the solutions were found to compare well with the experimental data on the limited scale considered.

4. CONCLUSION

We have proposed a statistical model for the turbulent diffusion of an instantaneously released puff in which a Gaussian approximation has been applied to the relative displacement process of the fluid particles. As a test of this assumption our analysis shows that the puff expansion follows the 3/2-law when the turbulent flow field is characterized by a Kolmogorov subrange.

With an Eulerian spectral index of $-3$, an exponential growth is predicted for the puff, representing, for instance, relative lateral diffusion in a very stably stratified atmosphere. This particular power law for the spectrum was also investigated in detail by Pécseli et al. (1983) and Misguich et al. (1985) in connection with studies of dispersion of puffs in plasma turbulence.

For the model to apply under more general conditions we made use of an additional assumption regarding a 'factorization' of the two-particle covariance function, $R_{ab}(\xi, \tau)$, into a product of a two-point Eulerian and a single-particle Lagrangian correlation.

Experimental evaluation of the model took place in the surface layer over homogeneous terrain in Denmark. Combined with aerial photographs of surface-released smoke plumes, wind data were obtained from a horizontal array of tower-mounted sonic anemometers. Integration of the puff growth rate equation gave good agreement with the smoke diffusion data over the limited scale considered.

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