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The Random-Force Method Applied to Calculating Short-Range Atmospheric Diffusion

Los Alamos
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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F. A. Gifford*

*Consultant. 109 Gorgas Lane, Oak Ridge, TN 37830.

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by

F. A. Gifford

ABSTRACT

The random-force equation, a simplified, stochastic, Lagrangian equation for the lateral component of the motion of a diffusing air particle, is applied to the analysis of short-range, short-time-averaged, horizontal atmospheric diffusion data. The velocity autocovariance and the velocity variance of the diffusing particle are derived and found to vary with the diffusion time, t. The normalized velocity autocorrelation function formed from these quantities is found to be a stationary exponential function of lag time. The corresponding energy spectrum of the particle's motion has the form of (frequency)$^{-2}$, over a broad high-frequency range. All these particle statistics, as well as the root mean square displacement of the particle about its mean position, i.e., the instantaneous plume spreading, $\sigma_y(t)$, depend on three atmospheric flow parameters: the total turbulent energy, $\overline{v^2}$; the Lagrangian integral time scale, t_L; and the initial velocity of the particle at the source point, v_o. By averaging the equation for σ_y, and the variance function, $\sigma_v^2(t)$, over a time equal to that of the standard, low-level, short-range atmospheric diffusion experiments, an analytical expression is deduced for the "plume shape-factor," f_1, in the equation $\sigma_y = \sigma_v t f_1$, which has often been used to correlate the results of such experiments. This is compared with the f_1-curve determined empirically from the large collection of low-level time-averaged atmospheric diffusion experiments made by Draxler. By comparing the theoretical and experimental f_1-curves it is shown that the Lagrangian time scale, t_L, is on the order of 10^4 s and that the averaging times of these diffusion experiments, 10 minutes to an hour, was too short to support their interpretation in terms of fully time-averaged diffusion. The shape of these time-averaged plumes is shown to have been intermediate between the instantaneous and fully time-averaged limits but in this instance rather nearer the former. From this it is inferred that large-scale, quasi-horizontal atmospheric turbulent motions influence the shape and concentration patterns of diffusing plumes in the planetary boundary layer, even when these are averaged over time periods of up to an hour. The controlling role of tropospheric, turbulent, kinetic-energy transfer in this process is briefly discussed.

I. INTRODUCTION

The random-force, or Langevin-equation method of calculating particle diffusion has been shown (Gifford 1981) to compare well with available data on horizontal diffusion in the atmosphere over a wide range of scales. This method assumes that an air "particle" obeys the following equation of motion:

$$dv/dt + \beta v = \eta(t) \; ; \tag{1}$$

v is the horizontal (lateral) component of the particle velocity, $\beta \equiv t_L^{-1}$, where t_L is a Lagrangian turbulence time-scale and η is a random acceleration that is assumed to have a flat spectrum and zero mean. In this simplified equation of motion, the term βv represents the local drag force on the particle, and the random acceleration η represents the effects on the particle motion that arise from large-scale pressure forces.

The solution to this one-dimensional, linear, stochastic differential equation is

$$v(t) = v_o \exp(-\beta t) + \exp(-\beta t)\int_o^t \exp(\beta\xi)\eta(\xi)d\xi \quad . \tag{2}$$

Since $v \equiv dy/dt$, a second integration provides the lateral particle displacement, y, from the fixed time or mean-wind axis. This can be squared and averaged over an ensemble of particles to find the mean-square displacement, σ_y^2. The good agreement of the resulting expression with observations of the instantaneous cross-flow spreading of puffs and plumes in the atmosphere, over a range of time scales from 10 to 10^7 s, suggests that the stochastic process represented by Eq. (1) is broadly applicable to atmospheric diffusion problems. In this report, some further useful statistical quantities are derived from Eq. (1), including the (one-dimensional) time-dependent velocity variance, correlation, spectrum, and the time-averaged displacement variance. These results are then compared with the extensive empirical analysis of time-averaged atmospheric horizontal diffusion data that was performed by Draxler (1976).

So that "he may run who reads it," a fairly detailed summary of the following results is included at the end of this report. References to the very limited amount of literature on the theory of atmospheric diffusion application of Eq. (1) can be found in Gifford (1981), where the relation of the random-force equation to Smith's (1968) linear-velocity hypothesis and to several recent Monte Carlo studies of atmospheric diffusion was also pointed out.

II. PROPERTIES OF THE RANDOM-FORCE MODEL

The particle-attached, or Lagrangian correlation for the y-, or cross-flow component of the motion can be formed directly from Eq. (2) as follows:

$$\begin{aligned}\overline{v(t)v(t+\tau)} = {} & v_o^2 \exp(-\beta t) \exp(-\beta(t+\tau)) \\ & + \exp(-\beta(2t+\tau)) \int_o^t \int_o^{t+\tau} \exp\beta(\xi+\zeta)\overline{\eta(\xi)\eta(\zeta)}d\xi d\zeta \\ & + \text{ensemble averages of products involving single integrals} \; .\end{aligned} \tag{3}$$

The overbar indicates ensemble averaging. Single-integral averages all equal zero because $\overline{\eta} \equiv 0$. To evaluate the double-integral term, the new variable $w = \xi - \zeta$ is introduced, and this term becomes

$$\exp[-\beta(2t+\tau)] \int_o^t \exp(2\beta\xi)d\xi \int_o^\tau A(w)\exp(\beta w)dw \; .$$

The acceleration correlation A depends only on the lag-time w because stationarity is assumed. It is also assumed that this correlation drops to zero very rapidly, in a time much less than $\beta^{-1} = t_L$. This proposition can be argued in various ways, for instance, on the basis that A was assumed to have a white-noise spectrum. But in the final analysis, it is an assumption that has to be judged by the quality of agreement of the results with data, which so far appears to be good. If A drops rapidly enough, the second integral can be treated as a constant, $\int_0^{\tau} A(w)e^{\beta w}dw = c_1$, and the (cross-flow component of) Lagrangian velocity covariance is found to be

$$\overline{v(t)v(t+\tau)} = e^{-\beta\tau}\{v_o^2\ e^{-2\beta t} + (c_1/2\beta)[1 - \exp(-2\beta t)]\}\ ; \tag{4}$$

note that it is time dependent.

The velocity variance is given by Eq. (4) with $\tau = 0$;

$$\sigma_v^2(t) = v_o^2 \exp(-2\beta t) + (c_1/2\beta)\ [1 - \exp(-2\beta t)]\ . \tag{5}$$

Thus, the velocity variance experienced by a diffusing particle is also a function of time, even in the assumed stationary turbulent flow, and becomes equal to the flow variance, $\overline{v^2} = c_1/2\beta$, only after a sufficiently long diffusion time, t, on the order of several times t_L.

The constant c_1 equals the rate of turbulent kinetic energy transferred to the particle by the action of large-scale, quasi-horizontal, random atmospheric motions; cf.Tennekes (1978), Tennekes and Lumley (1972, pp. 20-21). This must, in turn, be proportional to the average rate of energy dissipation, ε, in the boundary layer, and so

$$c_1 = B\varepsilon = 2\overline{v^2}t_L^{-1}\ ,\ B \approx 1\ . \tag{6}$$

In this way Eq. (1) provides the necessary direct linkage between the small-scale, dissipative, atmospheric motions and the quantities that characterize the largest, integral, or outer turbulence scales. Tennekes (1978) obtained the same result [cf. his Eq. (12)] by evaluating particle dispersion in the inertial sub-range and assuming an exponential autocorrelation. Here Eq. (6) follows directly from Eq. (1) for the stochastic process assumed to govern the entire range of atmospheric diffusive motions. Thus it appears that the random-force model, Eq. (1), is capable of describing one of the most fundamental aspects of turbulent kinetic-energy transfer in the lower atmosphere, i.e., its flow toward smaller scales from the large-scale motions and dissipation in the boundary layer.

By forming the quotient of Eqs. (4) and (5), it is found that $\overline{v(t)v(t+\tau)}/\sigma_v^2(t) = R(\tau) = \exp(-\beta\tau)$ and so the Lagrangian particle autocorrelation function, R, is exponential for the assumed stochastic process provided that the correlogram, $\overline{v(t)v(t+\tau)}$, is normalized with the time-dependent variance, $\sigma_v^2(t)$. If Eqs. (4) and (5) are averaged over all possible v_o values, then $\overline{v_o^2} = \overline{v^2} = c_1/2\beta$, since all turbulence fluctuations are in this case accounted for. In this case also, it is seen that $\overline{v(t)v(t+\tau)}/\overline{v^2} = R(\tau) = \exp(-\beta\tau)$. Thus both for a specified initial v_o and for v_o averaged over all possible values, an exponential Lagrangian autocorrelation, R, is determined. This reflects the nature of the assumed stochastic process, Eq. (1). The difference, which is quite significant for diffusion-modeling applications, is that in the fully averaged case, which corresponds to the averaged, Taylor-type of diffusion that is ordinarily assumed in most modeling applications, the mean-squared particle velocity is constant and equal to the mean-squared flow velocity, $\overline{v^2}$. In contrast, for the unaveraged v_o, which corresponds to the relative, instantaneous, or "puff"-type of diffusion, the variance of the diffusing particles' velocities, $\sigma_v^2(t)$, is time-dependent, as shown by Eq. (5). The resulting mean-square displacement relative to an instantaneous puff or plume centroid exhibits accelerating diffusion behavior, $\sigma_y^2 \propto t^3$, as is well known.

A. The Energy Spectrum

The kinetic-energy spectrum of the horizontal particle motion, $\overline{v^2}F(n)$, corresponding to an exponential Lagrangian velocity autocorrelation is given by

$$\overline{v^2}F(n) = 4\overline{v^2}\int_o^\infty R(t)\cos 2\pi n t\,dt = 4\overline{v^2}t_L\left[1 + (2\pi t_L n)^2\right]^{-1} , \qquad (7)$$

where n is frequency in Hz, and the customary meteorological normalization, $\int_o^\infty F(n)dn = 1$, has been applied. Thus the random-force model has an energy spectrum that behaves as $(\overline{v^2}/\pi^2 t_L)n^{-2}$ over a broad range of frequencies, in agreement with the well-known equilibrium-theory result that $\overline{v^2}F(n) \propto n^{-2}$ [cf.

Tennekes and Lumley (1972)]. Just as in the case of the correlation function, however, the Lagrangian spectrum derived from Eq. (1) is expressed in terms of the large-scale parameters that control atmospheric diffusion, namely the mean-square turbulence velocity, $\overline{v^2}$, and the Lagrangian integral scale, t_L.

B. Scale Analysis of Atmospheric Diffusion

The two large-scale parameters, $\overline{v^2}$ and t_L, define an (effective) eddy-viscosity of the atmosphere, $K = \overline{v^2} t_L$, and a characteristic length, L; $L \sim (\overline{v^2} t_L^2)^{1/2} \sim (K t_L)^{1/2} \sim \varepsilon^{-1/4} \; K^{3/4}$ (by Eq. 6). The last of these relations is just a form of the Richardson-Obukhov "4/3 law" of diffusion. The length, L, characterizes the smallest size-range of the large-scale, horizontal motions that is capable of maintaining the energy-transfer rate, ε, to the small, three-dimensional boundary-layer eddies, given the large-scale flow "viscosity," K, as was pointed out by Monin (1972). By comparing the mean-squared particle-displacement formula that follows, by integration and averaging, from Eq. (2) with a wide range of tropospheric contaminant-cloud diffusion measurements, Gifford (1981) found that, typically, $K \sim 5 \times 10^4$ m^2 s^{-1} and $t_L \sim 10^4$ s. From these values it follows that $L \sim 20$ km, which is approximately at the location of the so-called "spectral gap," or mesometeorological minimum, of the atmospheric energy spectrum.

Scales of motion greater than L, up to the synoptic, are quasi-horizontal and two-dimensional. In this range the largest "eddies" lose significant energy to the next smaller ones in a time $t_L \sim L^2/K \sim \varepsilon^{-1/3} L^{2/3}$ (e.g., Monin 1972 and Golitsyn 1973), which is the relaxation time of the diffusion process. By substituting the above values of K and t_L, it is found from Eq. (6) that $\varepsilon \sim 5$ cm^2 s^{-3}. This evaluation of ε agrees well with other estimates of energy dissipation in the planetary boundary layer that have been made by various authors, who used quite different lines of reasoning.

At scales smaller than L, in the range of typical three-dimensional planetary boundary-layer turbulence, energy is derived from the larger, two-dimensional eddies both indirectly from synoptic-scale motions and directly from instability of the vertical-shear layer created above the earth's surface by the frictional drag of these large-scale motions. The latter source operates, however, at the comparatively slow rate of viscous decay, i.e., ν/t_L^2, because it is controlled by the atmosphere's (kinematic) molecular viscosity, ν. This is much smaller than the rate, $\varepsilon \sim K/t_L^2$, of energy transfer, which depends on the apparent viscosity, K, of the atmospheric turbulence. Consequently the bulk of the net energy that drives small-scale, three-dimensional, boundary-layer turbulence is supplied by quasi-horizontal, diffusive atmospheric motions whose scale is greater than L. These are maintained by the large synoptic-scale motions from which they derive their energy. The exact nature and properties of this scale of motions, intermediate between the lower end of the synoptic scale and the small-scale, three-dimensional turbulence of the planetary boundary layer, as Golitsyn (1973) points out, have been very little studied. In particular the mechanisms of kinetic energy transfer that are involved are essentially unknown. But whatever these may prove to be, they necessarily must involve a net transfer of turbulent kinetic energy to the boundary layer. The properties of diffusion in the boundary layer are in this way always being directly influenced to some degree by the larger scale (i.e., > L), quasi-horizontal, two-dimensional motions. In attempting to estimate or model atmospheric diffusion, even (as will be demonstrated below) over fairly short times and distances, it is for this reason essential to account fully for the influence on plume or cloud spreading of the large-scale atmospheric-turbulence parameters $\overline{v^2}$ and t_L. Tennekes (1978) also stresses this point.

C. The Role of v_o in Particle Dispersion

Draxler (1976) analyzed short-range, horizontal atmospheric diffusion data for which displacement variances, σ_y^2, were formed from air-concentration observations that had been averaged over short times, on the order of the travel-time, t, from source to sampling point. Tracer gases and particles were released over time periods intermediate between the fully averaged and unaveraged cases, and this suggests that the role of the initial source velocity, v_o, in the process should be considered in more detail.

Clearly, the initial particle velocity, v_o, in Eq. (2) should be that of the air motion at the source at the initial time of the diffusion process.

Only in this case will $v(t)$ be a stationary random function. This requirement (Yaglom 1962) dictates the following choice of v_o:

$$v_o = \int_{-\infty}^{0} e^{\beta\xi}\, \eta(\xi)d\xi \ , \tag{8}$$

so that, from Eq. (2),

$$v(t) = \int_{-\infty}^{t} e^{-\beta(t-\xi)}\eta(\xi)d\xi \quad . \tag{9}$$

Equation (9) defines a unique stationary solution of Eq. (1), whose autocorrelation is exponential. The initial particle velocity, v_o, is a random "constant," having the value at any instant of the air velocity at the source point. Consequently, any particular source configuration defines an averaging of v_o over corresponding space-time points. For averaged-type diffusion from a point source, v_o is averaged over all possible values at the fixed source point and $\overline{v_o^2}$ equals the value for the entire flow, $\overline{v^2}$. For the case of instantaneous dispersion from a source of finite width, the v_o-averaging is over the source-width dimension. Similarly, dispersion from a point source that operates (or is sampled) only for a certain time interval can be related to a finite-time average of v_o over that interval. Lee and Stone (1982) have analyzed these various source configurations in detail for the random-force model, relating the space- and time-extension of sources to the corresponding Eulerian space- and time-correlation scales. This requires generalizing certain of the present single-point results to a large number of source points. Although the single-point model will be pursued here to keep the discussion as simple as possible, and because it seems to be adequate for present purposes, their paper should be consulted for the detailed discussion it provides of the effects of source configurations on dispersion.

D. Horizontal Cloud Spreading and Meandering

A single integration of Eq. (2) provides the horizontal particle displacement, y, from the time or mean-wind axis;

$$y(t) = (v_o/\beta)[1 - \exp(-\beta t)] - \beta^{-1} \exp(-\beta t) \int_o^t \exp(\beta\xi)d\xi + \beta^{-1} \int_o^t \eta(\xi)d\xi \quad .(10)$$

The (nondimensional) horizontal displacement variance follows from Eq. (10) by squaring and averaging over an ensemble of particles all released with the initial velocity v_o;

$$\Sigma_y^2(T) = [T - (1 - \exp(-T)) - (1/2)(1 - v_o^2/\overline{v^2})(1 - \exp(-T))^2], \tag{11}$$

where $T = t/t_L$ and $\Sigma_y^2 = \sigma_y^2/2\overline{v^2}t_L^2$. From Eq. (10), the average displacement, $\overline{y}$, of a cloud's centroid is

$$\overline{y} = v_o t_L [1 - \exp(-T)] \ , \tag{10a}$$

and so Eq. (11) can be written in the equivalent form

$$\Sigma_y^2(T) = \Sigma_R^2(T) + Y^2(T) \ . \tag{11a}$$

That is, the (mean-squared) cloud spreading with respect to the (fixed) time or mean-wind axis equals the sum of the instantaneous cloud spreading about its centroid and the (square of) the displacement of that centroid. The terms on the right-hand side are the spreading about the centroid, or relative diffusion,

$$\Sigma_R^2(T) \equiv \sigma_{yR}^2/2\overline{v^2}t_L^2 = T - (1 - \exp(-T)) - (1/2)(1 - \exp(-T))^2 \ ; \tag{11b}$$

and the centroid displacement, or meandering,

$$Y^2(T) \equiv \overline{y}^2/2\overline{v^2}t_L^2 = (v_o^2/2\overline{v^2})(1 - \exp(-T))^2 \ . \tag{11c}$$

(Since the present analysis has been based, for simplicity of presentation, on a single particle, the mean of the square of its displacement equals the square of its mean displacement, for constant v_o.)

When a further averaging of Eq. (11) is performed over all possible v_o, the final term drops out since then $\overline{v_o^2}/\overline{v^2} = 1$; and the equation for averaged diffusion from a point source is

$$\overline{\Sigma_y^2} \equiv \overline{\sigma_y^2}\ 2\overline{v^2}t_L^2 = [T - (1 - e^{-T})] \ , \tag{12}$$

a result first obtained by Taylor (1921). In principle, it should be possible to interchange such (ensemble) averaging with an average of Eq. (10) performed over a long period of time, such that for it $\overline{v_o^2}$ would in fact approach $\overline{v^2}$. In practice (Culkowski 1975; Csanady 1973; and Ferrara and Cagnetti 1980), this can require several days for atmospheric boundary-layer turbulence. Thus, the atmospheric diffusion data to be discussed in the next section will correspond to time averages of Eq. (10) with respect to v_o^2, such that $\langle v_o^2 \rangle$ is considerably less than $\overline{v^2}$, where the notation $\langle\rangle$ indicates averaging over a time period t_a that is generally small compared to such long averaging times.

III. SHORT-RANGE ATMOSPHERIC DIFFUSION DATA

Models of atmospheric dilution that are used in a wide variety of practical air-pollution and environmental-impact estimations are based on dispersion parameters derived from a number of field observations of time-averaged plume-concentration patterns made during the 1950s and 1960s under a variety of experimental conditions. Draxler (1976) summarized these as empirical curves of σ_y and σ_z, the horizontal and vertical plume standard deviations of the averaged plume-concentration distributions. He presented the results of these extensive data comparisons for the case of horizontal spreading, in the form of a similarity equation originally proposed by Pasquill (1971),

$$\sigma_y = \sigma_v\ t\ f_1(t/t_L) \ ; \tag{13}$$

σ_y, m, is the standard deviation of the plume-concentration distribution after t seconds of downwind travel; σ_v, m s^{-1}, is the standard deviation of the y-component of the horizontal wind measured at the source; f_1 is a universal dimensionless function.

Draxler determined the following equation for f_1 based, essentially, on all the available short-range plume diffusion data:

$$f_1 = [1 + 0.90\ (t/t_i)^{0.5}]^{-1}\ . \tag{14}$$

The quantity t_i equals the t-value for which f_1 equals 0.5. Equation (14) was found to apply to σ_y in all conditions of stability and for short-term-averaged concentrations from ground-level and elevated sources. (It also applies to σ_z, but vertical cloud spreading is not considered here.) It can be written in terms of the Lagrangian integral time-scale, t_L, by defining a quantity α such that

$$f_1 = [1 + 0.90\ \alpha\ (t/t_L)^{0.5}]^{-1}\ . \tag{15}$$

The form of Eq. (14) was chosen so that the limiting values for small and large diffusion times, t, agree with the Taylor, i.e., time-averaged expression for σ_y,

$$\sigma_y^2(t) = 2\overline{v^2} \int_o^t \int_o^\tau R(\xi) d\xi d\tau\ . \tag{16}$$

Release times for the various tracers used in these experiments ranged from 10 minutes up to an hour, depending to some degree on the distance to the farthest concentration-sampling arc, which was 5 km or less in the majority of cases. The horizontal wind standard deviation, σ_v, and the ground-level concentration patterns were as a rule sampled over a like time, and so usually represent averages over periods from several tens of minutes to an hour.

Draxler's results are of very great interest, not only because they comprise a practically useful method of diffusion estimation, but also for the following theoretical reason. Since Taylor's form of σ_y for averaged diffusion, Eq. (16), was assumed to hold for the experiments, the Lagrangian autocorrelation function $R(\tau)$ could be found from Eq. (14) by combining it with Eq. (13) and differentiating twice. Draxler showed that the R-function determined in this way dropped rapidly at first but had a long positive "tail" and could not be described by an exponential form for any reasonable choice of t_L. This fact about atmospheric diffusion has been somewhat puzzling since he first demonstrated it.

An equation for the averaged cloud width as measured at a fixed sampling arc over an experimental time period t_a follows from averaging and rearranging Eq. (11a) to get $\langle\sigma^2_{yR}\rangle = \langle\sigma^2_y\rangle - \langle\bar{y}$&2. Performing the averaging on Eq. (11) for σ^2_{yR} and Eq. (10b) for $\bar{y}$, it is found that

$$\langle\sigma^2_{yR}(T)\rangle = 2\overline{v^2}\ t_L\ [T - (1 - \exp(-T)) - (\langle c\rangle/2)(1 - \exp(-T)^2)]\ , \tag{17}$$

where $\langle c\rangle = [1 - (\langle v^2_o\rangle - \langle v_o\rangle^2)/\overline{v^2}]$. Note that, for a vanishingly small averaging time, $\langle c\rangle = 1$ and Eq. (17) reduces to Eq. (11b) for the instantaneous spreading. When t_a is very large, $\langle c\rangle = 0$, since $\langle v_o\rangle = 0$ and $\langle v^2_o\rangle = \overline{v^2}$ then; and Eq. (17) reaches the Taylor limit, Eq. (12). The velocity variance experienced by the diffusing particle in the same circumstances, $\langle\sigma^2_{vR}\rangle$, can similarly be formed from Eq. (5) and the (ensemble) average of Eq. (2), since $\langle\sigma^2_{vR}\rangle = \langle\sigma_v{}^2\rangle - \langle\overline{v}^2\rangle$; the result is that

$$\langle\sigma^2_{vR}\rangle = (\langle v^2_o\rangle - \langle v_o\rangle^2)\exp(-2T) + \overline{v^2}\ [1 - \exp(-2T)]\ . \tag{18}$$

Equation (11b), for the relative cloud diffusion, contains a range for which $\sigma^2_{yR} \propto t^3$. This accelerating diffusion range begins to approach the large-time asymptotic state, $\sigma^2_{yR} \rightarrow 2\ kt$, for T-values of about 2 or 3. For the fully averaged Taylor diffusion of Eq. (12), $\overline{\sigma^2_y}$ increases at first as t^2 and then later as 2 kt also, joining the Eq. (11b) curve. In the intermediate case, for diffusion averaged over t_a, the mean-square cloud spreading is at first as t^2, but below the fully averaged spreading case of Eq. (12), to a degree depending on how different $\langle c\rangle$ is from zero. Accelerating diffusion begins at some value of T that is also controlled by $\langle c\rangle$, i.e., by t_a; and the final stage of diffusion is reached, as with the other two curves, at some large value of T. Figure 1 illustrates the behavior of cloud spreading as determined by Eqs. (11b), (12), and (17) for, respectively, the instantaneous diffusion about the centroid, the fully averaged Taylor limit, and the diffusion averaged over an experimental time period, t_a, corresponding in the example shown to the value $\langle c\rangle = 0.95$.

Equations (17) and (18) define the averaged cloud spreading and the corresponding wind variance experienced by a particle, over an experimental averaging time t_a, and so are the appropriate quantities to compare with Eq. (13). When the substitutions are made it is found that

$$f_1(T) = \frac{2^{1/2}}{T}\left\{\frac{T-(1-e^{-T})-(\langle c\rangle/2)(1-e^{-T})^2}{1-\langle c\rangle e^{-2T}}\right\}^{1/2}, \tag{19}$$

where the symbol $\langle\rangle$ indicates as always a time averaging over t_a. The present theory in this way provides an analytical formulation of the function f_1, which can be compared with Draxler's empirical formula, Eq. (14). Figure 2 illustrates Eq. (19) for a range of values of the parameter $\langle c\rangle$ that includes the Taylor average-diffusion limit ($\langle c\rangle = 0$) as well as the limit ($\langle c\rangle = 1$) of instantaneous, or relative, diffusion. Since Draxler's curve is based on concentrations averaged over periods of from tens of minutes to an hour, it can be expected that $\langle c\rangle$ will be found to have some intermediate value.

From Fig. 2, it can be seen that when $T > 5$ the curve f_1, according to Eq. (19), is approaching its asymptotic behavior for large values of T and is, consequently, little influenced by $\langle c\rangle$. The value $f_1 = 0.5$, upon which Draxler based his time-scale t_i, corresponds closely to $T \simeq 6.4$ for any value of $\langle c\rangle$. With these values it follows from Eq. (15) that $\alpha = 0.44$. To specify $\langle c\rangle$ it seems most reasonable to require agreement between Eqs. (15) and (19) for some smaller value of T, say $T = 1$. When this is done, it is found that $\langle c\rangle = 0.68$,

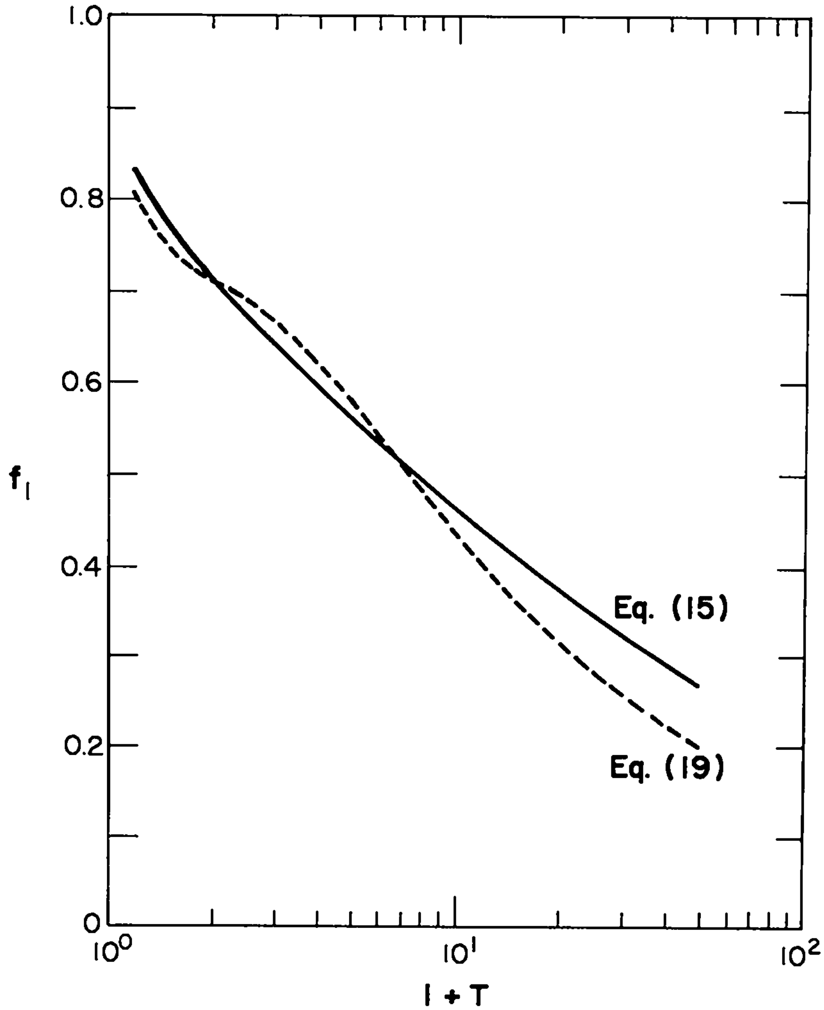

Fig. 1. Instantaneous, horizontal puff, or plume element, spreading relative to the centroid, Σ_R, Eq. (11b); fully averaged (over all possible initial velocities) plume dispersion relative to the (fixed) time or mean-wind axis, $\overline{\Sigma_y^2}$, Eq. (12); and plume dispersion averaged over an experimental time period t_a, $\langle\Sigma_y^2\rangle$, Eq. (17) with $\langle c\rangle = 0.95$.

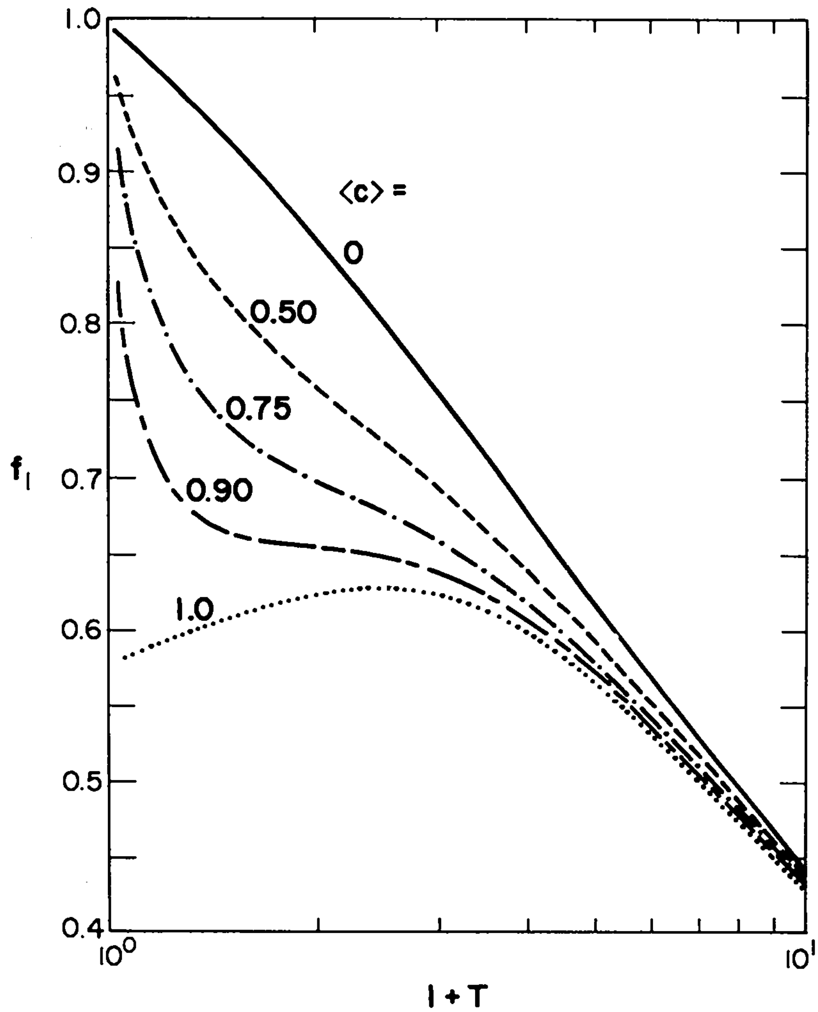

Fig. 2. Plot of theoretical curves of the plume "shape-factor," $f_1(T)$, as a function of nondimensionalized travel time, $T = t/t_L$, for various values of the parameter $\langle c \rangle$, based on Eq. (19).

which implies that $\langle V_o^2 \rangle \equiv \langle v_o^2 \rangle - \langle v \rangle^2$ has attained 32% of the total $\overline{v^2}$ for the time averaging of these data. Equations (15) and (18), with $\alpha = 0.44$ and $\langle c \rangle = 0.68$, are plotted in Fig. 3 for a range of T-values adequate to cover the experimental data. In Fig. 4, Eq. (19) has been expressed in terms of t/t_i, for the same parameter values, and superimposed on Draxler's Fig. 1, permitting a direct comparison with the diffusion data as well as with Draxler's curve, Eq. (15). Equation (19) appears to fit the data equally as well as Eq. (15). It may in fact fit slightly better, since Draxler's f_1-curve seems a bit high for $t/t_i > 2$, particularly if a few outliers are ignored.

IV. THE LAGRANGIAN TIME SCALE

Equation (14) does not involve t_L explicitly, and so Draxler estimated t_L directly from the diffusion data. Basing the determination on the near-source observations, he found that $t_L = t_i/6.36$. Since t_i equalled about 300 s, from the asymptotic behavior of Eq. (14) for large t, he concluded that $t_L \sim 50$ s for diffusion from ground-level sources. But Draxler could not explain the behavior of Eq. (15), which he derived assuming the applicability of Eq. (16) for fully averaged diffusion (i.e., $\sigma_v^2 = \overline{v^2}$), in terms of a single exponential autocorrelation and, hence, a single time scale, t_L. He concluded that the implied Lagrangian autocorrelation, according to Eq. (16), "approaches zero much too soon to properly describe diffusion at large distances." When he fit the longer range portion of the data, he found a considerably larger apparent Lagrangian time scale, $t_L = t_i/1.64$. In the present analysis, which explicitly accounts for the averaging time of the diffusion data through the parameter $\langle c \rangle$, this problem does not occur. Equation (19) is derived from Eq. (1), which defines a random process that has a single, exponential correlation, given by Eqs. (4) and (5), and a single Lagrangian time scale, t_L, which analysis of tropospheric instantaneous puff- and plume-spreading data has indicated to equal several hours (Gifford 1981).

Just as in the case of Draxler's empirical Eq. (14), for f_1, the time scale t_L is not explicitly defined by Eq. (19), even after the single disposable parameter, $\langle c \rangle$, has been determined. Some further information on the energy content of the large-scale diffusive motions has to be supplied in order to specify t_L; one way to derive an estimate is as follows. The value

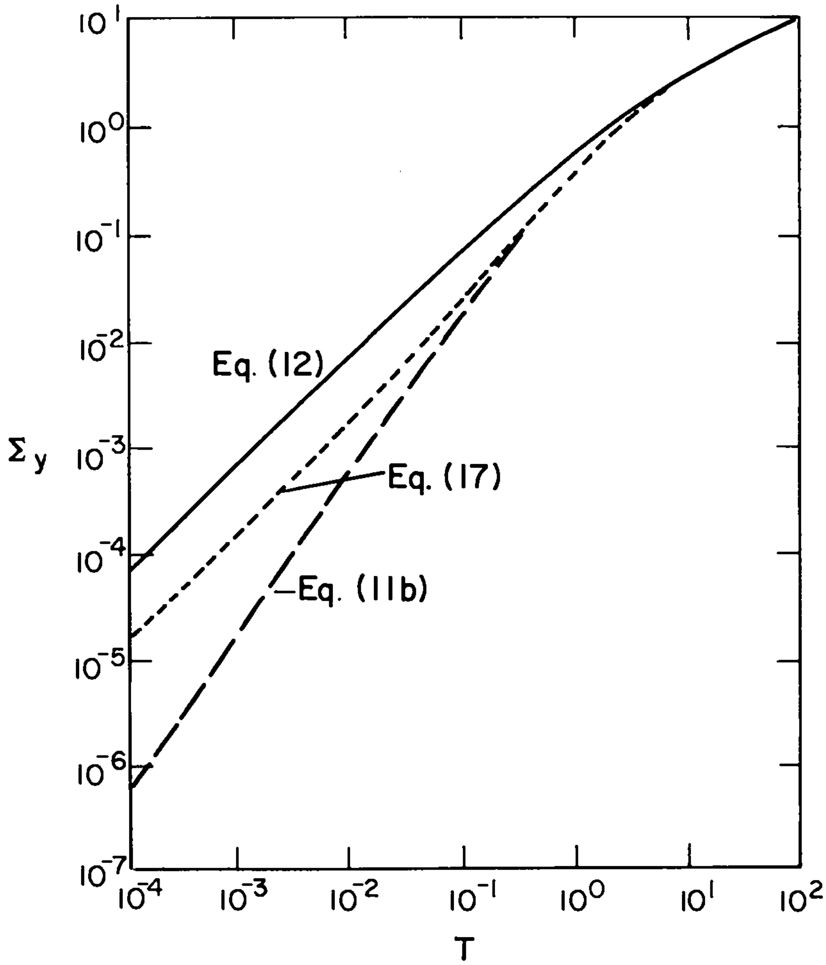

Fig. 3. Draxler's (1976) empirical $f_1(T)$-curve, Eq. (15), and the theoretical $f_1(T)$-curve, Eq. (19), with $\langle c\rangle = 0.68$ chosen to give the best matching.

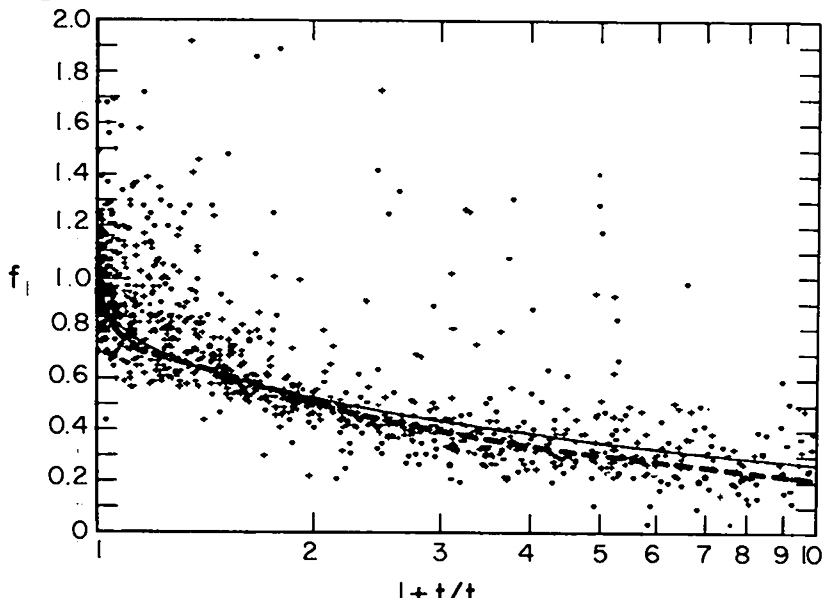

Fig. 4. Draxler's (1976) ground-source plume diffusion data (plus signs) plotted as a function of travel time, t; for $t = t_i$, $f_1 = 0.5$. The solid line is Draxler's empirical f_1-curve, Eq. (15); the dashed line is the theoretical f_1-curve, Eq. (19).

$\langle c \rangle = 0.68$ was found to produce the best match between the present theory, Eq. (19), and Draxler's empirical curve, Eq. (15). This means, since $\langle c \rangle \equiv 1 - \langle V_o^2 \rangle / \overline{v^2}$, that $\langle V_o^2 \rangle = 0.32\ \overline{v^2} = 0.32\ K/t_L = 0.16\ \varepsilon t_L$, based on various relationships that were previously introduced. Although Draxler's data are widely scattered over a broad range of stability conditions, many of the points are clumped near neutral stability (Draxler 1976, cf. Fig. 3). Consequently Eq. (15) can probably be taken to approximate near-neutral conditions. In such conditions the horizontal wind direction standard deviation, σ_θ, equals about 10^o, and the mean transport wind, $u = x/t$ (where x is downwind distance from the source), is about 5 ms^{-1}. It follows that $\langle V_o^2 \rangle^{1/2} \sim u \tan 10^o = 0.9\ ms^{-1}$. Making use of the fact that $\varepsilon \sim 5\ cm^2s^{-3}$ on the average in the boundary layer, it is found that $t_L \sim 10^4$ s.

This rough estimate of t_L, a quantity about which virtually nothing is known from direct observations, should not be taken as definitive for several reasons. The value assumed for ε may be too high, possibly by a factor of 2, since a significant fraction of the total tropospheric eddy-energy dissipation occurs at high altitudes, in the clear-air turbulence associated with jet streams. Also seasonal and other kinds of variations in the tropospheric turbulence level will affect t_L. The present estimate, $t_L \sim 10^4$ s, agrees with the estimate previously derived from the tropospheric instantaneous diffusion data (Gifford 1981). It should however be taken only as an order-of-magnitude indication that t_L, according to the random-force theory, is <u>much</u> larger than t_L estimated on the basis that fully averaged diffusion (Eq. 16) applies to the short-range diffusion data that were averaged over a few tens of minutes. Diffusing clouds and plumes, even when averaged over such time periods, are observed to have shapes (f_1-curves) that are influenced to some degree by large-scale diffusive motions, shapes that cannot be explained unless these are accounted for.

Draxler (1976) was well aware of this implication of his data analysis, i.e., that observed plume shapes do not reproduce the theoretical shape implied by analysis based on Eq. (16) for fully averaged Taylor-type diffusion, at intermediate and large diffusion times. He proposed to explain this on the basis that the observed accelerating diffusion is caused by vertical shear of the mean planetary boundary-layer wind. Diffusion in various sheared flows, including an Ekman boundary layer, has been the subject of several theoretical studies, which are well summarized in the book by Monin and Yaglom (1971) and in Csanady's (1973) thoughtful monograph. The main shear effect is to elongate a diffusing puff, or a plume element, in the direction of the mean flow. This has little effect on the along-wind structure of plumes, as Csanady pointed out. But the cross-wind shear occurring in the outer part of a skewed Ekman-layer also leads to enhancement of the effective lateral diffusion, and must be included in any boundary-layer K-model of plume diffusion.

Apart from the limited degree to which the Ekman-layer assumption of constant vertical diffusivity represents actual atmospheric boundary layers, the main problem with this interpretation, from the point of view of the present theory, is that boundary-layer shear is an <u>effect</u>, not a <u>cause</u>, of diffusive atmospheric motions. Accelerating diffusion, in the random-force model, is controlled by the same large-scale, quasi-horizontal motions that create and maintain the boundary-layer shear. Boundary-layer shear is in effect parameterized in the random-force model along, it should be noted, with a number of other dispersive flow phenomena of possible importance, such as certain wave instabilities, irregular terrain effects like flow separations and vortex shedding, and gravity flows.

Whether to regard the accelerating, horizontal cloud spreading as having been produced entirely by an imposed, constant, boundary-layer shear, or by the presence in the atmosphere of a steady transfer of energy from large to small scales, is to some degree a matter of choice; but it must be remembered (cf., for example, Monin et al., 1974) that the phenomena are entirely distinct from the modeling point of view and produce quite different physical effects. The accelerating diffusion described by Eq. (11b) disappears when the diffusion is averaged over a sufficiently long time. When t_a is large enough, and it should, for this purpose, equal at least several times t_L, the parameter $\langle c \rangle$ goes to zero as has been shown, and Eq. (17) reduces to

$$f_1(T) = 2^{1/2}T^{-1}[T - (1 - e^{-T})]^{1/2} \quad . \tag{21}$$

If Eq. (21) is introduced into Eq. (13), Eq. (12) for the averaged-plume diffusion is recovered. The accelerating diffusion is no longer present, even though the wind field, including any shear, remains the same. Consequently, accelerating diffusion is clearly associated, in the random-force model, with the influence of the averaging time of diffusion, t_a. This effect does not occur in the case of sheared boundary-layer K-theories, since the mean motion in these is regarded as fixed; consequently the cloud elongation and distortion produced by shear are not changed by time averaging. In contrast to this, the accelerating regime of lateral diffusion predicted by the random-force theory will disappear for a sufficiently large averaging time.

V. SUMMARY AND CONCLUSIONS

In the random-force method, the horizontal velocity and position displacement of a diffusing air particle are assumed to obey a simple Lagrangian equation of motion, Eq. (1). In this equation, the particle acceleration is the result of a local drag force, proportional to the particle's velocity, and a random acceleration, representing large-scale pressure forces; the latter is assumed to have a white-noise spectrum. Equation (1) is solved to give the following properties of a dispersing particle: the Lagrangian velocity, Eq. (2); the autocovariance, Eq. (3); the velocity variance, Eq. (5); the power, or variance, spectrum, Eq. (7); and the lateral displacement variance, σ_y^2, Eq. (11). These properties of the particle motion are shown to depend on three parameters of the flow: the initial value of the lateral wind component at the source, v_o; the lateral component of the total flow variance, $\overline{v^2}$; and the Lagrangian integral time-scale, t_L. All these particle properties depend, in addition, on the dispersion time, t, and become identical with the corresponding (stationary) properties of the tropospheric flow only after dispersion times equal to several times t_L. The normalized Lagrangian autocorrelation function, defined as the quotient of Eqs. (4) and (5), is found to be exponential and stationary; $R(\tau) = \exp(-\tau/t_L)$. The form of the corresponding variance (energy) spectrum, Eq. (7), agrees with that of the inertial-range frequency spectrum over a range of frequencies that is considerably wider than that which can be attributed to the action of three-dimensional boundary-layer turbulence alone.

Values of the large-scale flow parameters, $\overline{v^2}$ and t_L, can be combined to estimate the tropospheric eddy-diffusivity K, the eddy-energy dissipation rate ε, and a length scale L, which essentially separate the large, quasi-horizontal, two-dimensional dispersive motions of sub-synoptic scales from the smaller, three-dimensional, boundary-layer turbulence. The values $K = 5 \times 10^4$ $m^2\ s^{-1}$ and $t_L = 10^4$ s were estimated previously, by means of an extensive comparison of Eq. (11) with tropospheric, instantaneous cloud-spreading data extending over a wide range of diffusion times, from 10 to 10^7 s. From these values it is found that $L \sim 20$ km and $\varepsilon \sim 5\ cm^2 s^{-3}$, in agreement with accepted estimates of these quantities. Thus the random-force model correctly provides for the effects of two important properties of the tropospheric eddy-energy balance; namely, energy transfer from large-scale, two-dimensional motions to the three-dimensional, boundary-layer turbulence, and eddy-energy dissipation by the boundary-layer turbulence of the correct magnitude. The value found for the large-scale eddy-diffusivity, K, from the tropospheric cloud-spreading data is also in reasonably good agreement with other large-scale diffusion results.

These data comparisons are extended, in the present report, to the various canonical series of short-range (a few kilometers), short-time-averaged (a few tens of minutes) horizontal diffusion data that were summarized by Draxler (1976). Following Pasquill (1971), Draxler assumed a similarity form for the horizontal dispersion, Eq. (13), and fit the diffusion data to this to determine a universal, empirical plume-shape function, f_1, Eqs. (14) and (15). An analytical expression for this function f_1 can be derived by averaging Eqs. (2), (5), (10a), (11), and (11a) of the present theory over a time period equivalent to that of the diffusion data; Eq. (19) is the result. This equation depends on a single disposable parameter, $\langle c \rangle$. By choosing the value of $\langle c \rangle$ so as to provide the best match between Eq. (19) and Draxler's empirical curve, it is found that, over the averaging period of the experimental data, approximately a third of the total flow variance has occurred. It appears that these short-time averaged diffusion experiments, which have usually been applied as if they represented fully averaged, Taylor-type diffusion, in fact retain much of the character of the instantaneous cloud spreading described by Eq. (11b), since they correspond to the curve $\langle c \rangle = 0.68$ of Fig. 2, a plot of Eq. (19). Draxler was unable to interpret Eq. (15), for the empirical plume-shape function, f_1, in terms of a single Lagrangian time-scale, because he assumed that Eq. (16), the fully averaged Taylor-equation for σ_y, applied.

From the value $\langle c \rangle = 0.68$ it is estimated that the Lagrangian integral time scale implied by these boundary-layer, short-time averaged plume data is about 10^4 s. The fact that this agrees with the t_L-value previously determined from the instantaneous, tropospheric puff-spreading data must be regarded as somewhat fortuitous. Nevertheless it is a strong indication of the random-force model's essential correctness, inasmuch as the value 10^4 s considerably exceeds the travel, or diffusion time of these experiments. This value of the Lagrangian time scale, $t_L \sim 10^4$ s, is about two orders of magnitude larger than values previously inferred from boundary-layer diffusion experiments or balloon trajectory data, which are typically conducted over periods ranging up to an hour. In fact t_L computed from any particular experiment has always shown a disconcerting tendency to be approximately equal to the time-duration of that experiment. From the present results it is easy to see why this must be so.

If an "apparent" Lagrangian time scale for an experiment is defined as $t_{aL} \equiv \int_0^{t_a} R(\tau)d\tau$, as is usually assumed in estimates from data, then the present results imply that

$$t_{aL} = \int_0^{t_a} \exp(-\tau/t_L)d\tau = t_L \left[1 - \exp(-t_a/t_L)\right] \; ; \tag{21}$$

t_a is, as before, the averaging time, or duration, of an experiment. By expanding the exponent it is seen that, for small enough values of the experimental duration t_a, the apparent Lagrangian scale t_{aL} just equals t_a, as has so often been found. For experimental periods t_a of up to an hour, t_{aL} differs from t_a by less than 20%, if $t_L = 10^4$ s.

The general picture of tropospheric diffusion that is conveyed by these comparisons of the random-force model with the standard diffusion-data sets has some significant points of difference from more conventional interpretations. The major one is that the transfer of turbulent kinetic energy by quasi-horizontal atmospheric motions of lengths between the lower end of the synoptic scale and about 20 km has two important effects on the shape of diffusing clouds. The model predicts, and the instantaneous, long-range cloud- and plume-spreading data confirm, the presence of an extensive region of accelerating horizontal diffusion in the troposphere. This phenomenon evidently occurs up to time scales much larger (several times 10^4 s) than those of buoyant, boundary-layer convective motions, which are typically on the order of a few hundred seconds, and must necessarily be governed by larger scale, random motions. The exact nature and origin of these motions, it should be stressed, is not, at present, at all well understood. Terrain inhomogeneities, which exist over a broad range of sizes, land-use patterns, and so on, presumably exert an influence. The actual vehicle for the energy transfer could involve gravity-wave propagation, in modes that as yet have not been studied in relation to the diffusion problem. But that a flow of turbulent kinetic energy at large scales, from the lower (high-frequency) end of the scale of synoptic motions down to the dissipative, three-dimensional, boundary-layer turbulence scales, exists in some form is certain; and the diffusion data indicate that diffusive motions are involved.

A second point, the principal subject of this report, is that the large-scale, random motions of the lower troposphere also affect the shapes and concentration distributions of small-scale diffusing clouds and plumes in the boundary layer, even when the latter are averaged over times of up to a few tens of minutes. Explanation of the shapes and concentration distributions of all the basic plume-diffusion data sets by means of fully time-averaged, Taylor diffusion theory was shown by Draxler (1976) to be awkward at best, and impossible in terms of a single Lagrangian time-scale of the order of the experimental averaging time. The resolution of this problem in terms of the random-force theory of horizontal diffusion indicates that the observed plume shapes, which is to say the universal function f_1, formed from these extensive short-range, short-time-averaged diffusion trials, imply a large Lagrangian time scale. The experimental averaging periods of these data, a few tens of minutes, were large enough to include only about a third of the total turbulence of the flow. Consequently the observed plume shapes, intermediate between the shape of an instantaneous plume and a fully averaged plume, still reflected the influence of the former.

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