

**Horizontal Diffusion in the Atmosphere:  
A Lagrangian-Dynamical Theory**

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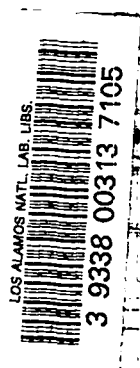
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# Horizontal Diffusion in the Atmosphere: A Lagrangian-Dynamical Theory

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HORIZONTAL DIFFUSION IN THE ATMOSPHERE:  
A LAGRANGIAN-DYNAMICAL THEORY

by

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ABSTRACT

A form of Langevin's equation is derived that is applicable to the atmospheric diffusion problem. The resulting equation for the particle displacement variance  $\sigma_y^2$  has limits at small and large diffusion times equal to asymptotic predictions of statistical diffusion theories but provides, in addition, estimates over the broad, middle range of diffusion, which is important in regional and larger-scale atmospheric applications. Predictions of the theory compare well with standard atmospheric diffusion data sets over a range of diffusion times, from seconds to days. When parameters of the theory are determined from short-range plume diffusion data, the theory predicts large-scale eddy diffusivity,  $K$ , in the known atmospheric range, a striking confirmation of the ability of this theory to describe atmospheric diffusion.

INTRODUCTION

Conventional wisdom is to the effect that there are three theories from which useful working models of atmospheric diffusion can be derived by analytical techniques. These are statistical theory, gradient-transfer or  $K$ -theory, and similarity theory; see, for example, such applications-oriented surveys as those by Pasquill (1975), Gifford (1975), Drake, et al. (1979), and Barr and Clements (1980). This paper attempts to focus attention on a fourth, alternative theory of diffusion and to demonstrate applications to horizontal atmospheric diffusion over a wide range of diffusion times. The theory goes by various names: the Brownian-motion analogy (Chadam, 1963; Lin and Reid, 1962); the Langevin model (Krasnoff and Peskin, 1971); the random-force method (Novikov, 1963). It is probably fair to say that all these studies were to a considerable extent motivated by the brief paper by Obukhov (1959), in which he proposed that the evolution of a diffusing air particle's motion in the atmosphere forms a Markov process and can be described by the Fokker-Planck equation in physical space.

In this report, a form of Langevin's equation will be derived directly from an expression for the diffusing air particle's motion, proposed by F. B. Smith (1968);

$$v(t + \tau) = R(\tau)v(t) + n(t) . \quad (1)$$

The velocity,  $v$ , of a small volume of air (a "particle") at a time  $\tau$  seconds after the present time,  $t$ , is given according to this simple hypothesis by the sum of a correlated part,  $R(\tau)v(t)$ , and a purely random, uncorrelated part,  $n(t)$ . For a stationary, homogeneous turbulent flow,  $R(\tau)$  can be shown to be the Lagrangian velocity autocorrelation function. The random increment to the wind speed,  $n(t)$ , satisfies the conditions that  $\bar{n} = 0$  and  $\overline{n\bar{v}} = 0$ ; the overbar indicates ensemble averaging. Smith derived a number of statistical-kinematical consequences from (1) for diffusion in stationary, homogeneous turbulence. In particular, he was able to show, by assuming an exponential form for  $R$ , that if the particle statistics are conditioned by the requirement that the initial velocity,  $v(0)$ , have a constant value over the ensemble, then the mean square particle displacement,  $\sigma_y^2 = \overline{y^2}$ , where  $y = dv/dt$ , is proportional to  $t^3$ . More recently, Reid (1978) and Hanna (1979) have made equation (1) the basis of Monte-Carlo computer simulations of atmospheric diffusion and have shown, again by assuming an exponential  $R$ , that asymptotic results of statistical diffusion theory are reproduced by these calculations. In addition, Hanna showed, by extensive direct comparisons with atmospheric turbulence data, that equation (1) is widely applicable in the atmospheric boundary-layer.

#### The Langevin equation for turbulent diffusion.

The turbulent wind speed  $v(t + \tau)$  can be expanded in series of powers of  $\tau$ ;

$$v(t + \tau) = v(t) + \tau dv/dt + (\text{higher order terms}). \quad (2)$$

Since for small enough  $\tau$ , say  $\tau = \tau_S$ , the higher order terms can be neglected, it follows directly from (1) that

$$dv(t)/dt + \{[1-R(\tau_s)]/\tau_s\} v(t) = \eta(t), \quad (3)$$

a form of Langevin's equation, where  $\eta(t) = n/\tau_s$ .

The exact magnitude of  $\tau_s$  will play no specific part in the following discussion. It is for our purpose most simply considered to be an averaging (running mean) time of the "true" microscopic particle velocity,  $v_i$ , which in general is the sum of macroscopic and molecular components; thus

$$v(t) = \tau_s^{-1} \int_t^{t+\tau_s} v_i(\tau) d\tau . \quad (4)$$

Considered from this point of view,  $\tau_s$  has to be large enough to smooth out molecular irregularities in  $v_i$ ; see, for example, Obukhov (1959) or Papoulis (1965, pp. 514-517). Thus it plays a role similar to that of the continuum hypothesis of fluid mechanics. As a practical matter, even the most sensitive anemometer measures  $v$ , not  $v_i$ . Dynamical, as well as thermodynamical, properties of the microstructure of  $v_i$  are naturally of fundamental physical concern; some aspects have been discussed by Novikov (1963). For present needs it is adequate to assume that the ratio  $(1 - R)/\tau$  approaches a definite limit as  $\tau$  approaches  $\tau_s$ . A similar assumption was introduced by G. I. Taylor (1921) in discussing diffusion by discontinuous movements.

Langevin's equation is usually presented in the form

$$dv/dt + \beta v(t) = \eta(t) \quad (5)$$

from which, with (2), it follows that  $\beta = [1 - R(\tau_s)]/\tau_s$ . Comparison of (5) with the Navier - Stokes equation (Lin and Reid, 1962) suggests that the particle motion is nearly free, but is influenced by a small, random, non-resistive force  $\eta$  which arises from the pressure terms, acting at large scales, and by a smaller resistive force  $-\beta v$ , associated with the viscous terms, which acts as a local drag force on the particle. If (5) is multiplied by  $v$  and averaged, the stationary condition,  $d\overline{v^2}/dt = 0$ , implies that  $\overline{\eta v} = \beta \overline{v^2}$ . This is interpreted to mean that  $\overline{\eta v}$  is the average rate of energy supply to the particle due to work by the random pressure forces. Novikov

(1963) states that its value should differ from the eddy-energy dissipation rate,  $\epsilon$ , by at most a factor of order unity. This is balanced by the term  $\beta v^2$ , which equals the viscous dissipation rate.

The system of stochastic differential equations, (5) together with

$$v = dy/dt, \quad (6)$$

and the boundary conditions,  $y = 0$  and  $v = v_0$  at  $t = 0$ , possesses solutions that are well known from studies of Brownian motion; see especially Chandrasekhar (1943) and Uhlenbeck and Ornstein (1930). Formulations in terms of the turbulent diffusion problem are mathematically equivalent to these. However, the papers by Lin and Reid (1962), Chadam (1963), Novikov (1963), and Krasnoff and Peskin (1971) bring out many important dynamical aspects that are specific to the turbulent diffusion application and should be consulted especially for these insights. The following discussion concentrates on the applicability of these solutions to atmospheric diffusion, for which purpose they have a number of interesting and potentially quite useful properties.

#### The mean-square particle displacement.

Equation (5) can be solved for  $v(t)$  by standard methods (variation of parameters), with the result that

$$v(t) = v_0 e^{-\beta t} + \int_0^t e^{-\beta(t-s)} \eta(s) ds = dy/dt. \quad (7)$$

Despite the complicated, stochastic nature of (5), due to the random-force term  $\eta$ , existence and uniqueness of solutions are guaranteed by appropriate theorems; see Yaglom (1962), for instance. Equation (7) can be integrated once again to get the particle displacement,  $y$ ;

$$y(t) = (v_0/\beta)(1-e^{-\beta t}) - \beta^{-1}e^{-\beta t} \int_0^t e^{\beta\xi} \eta(\xi) d\xi + \beta^{-1} \int_0^t \eta(\xi) d\xi. \quad (8)$$

If (8) is averaged, it is found that

$$\bar{y}(t) = (v_0/\beta)(1 - e^{-\beta t}); \quad (9)$$

and so in this model only for suitably large  $t$  does the mean particle's axis approach a constant value,  $\bar{y} = v_0/\beta$ .

The mean-square displacement,  $\overline{\sigma_y^2}$ , is found directly, by squaring and averaging (8); the result is

$$\sigma_y^2 = (2\overline{v^2}/\beta)t + (\overline{v_0^2}/\beta^2)(1 - e^{-\beta t})^2 + (\overline{v^2}/\beta^2)(-3 + 4e^{-\beta t} - e^{-2\beta t}). \quad (10)$$

The limiting value of this equation for large  $t$  is

$$\sigma_y^2 = (2\overline{v^2}/\beta)(t - 3/2\beta) = (2\overline{v^2}/\beta)t,$$

from which it can be concluded by analogy with results from K-theory that  $(\overline{v^2}/\beta) = K$ , the (large scale) eddy diffusivity. Thus we may also write (10) as

$$\sigma_y^2 = 2Kt + (v_0^2/\beta^2)(1 - e^{-\beta t})^2 + (K/\beta)(-3 + 4e^{-\beta t} - e^{-2\beta t}). \quad (10a)$$

Except that the parameters are defined in terms of the macroscopic turbulent velocity,  $v$ , these results are mathematically identical to the corresponding ones for Brownian motion.

The initial velocity,  $v_0$ , affects the dispersion near the source but not at large values of  $t$ . If, however, a further averaging is performed, over all possible values of  $v_0$ , equation (10a) becomes (since then  $v_0^2 = v^2$ )

$$\left\langle \sigma_y^2 \right\rangle_{v_0} = (2\overline{v^2}/\beta^2)(\beta t - 1 + e^{-\beta t}). \quad (11)$$

Taylor (1921) obtained this result for stationary, homogeneous turbulence, by assuming an exponential autocorrelation,  $R$ . Contrasting with this, in the present theory, the autocorrelation function,  $R$ , can be derived directly from equation (5). The result, when the initial velocity  $v_0$  has the specific value zero, is (Papoulis, 1965, eq. 15-17)



$$R(t, \tau) = \overline{v(t + \tau)v(t)}/v^2 = (1 - e^{-2\beta t})e^{-\beta \tau}. \quad (12)$$

This time-dependent function approaches the steady value  $R = e^{-\beta \tau}$  only after diffusion times equal to several times the Lagrangian time scale,  $t_L = \beta^{-1} = K/v^2$ , which will ordinarily equal on the order of  $10^2$  to  $10^3$  seconds in the atmosphere. Thus, the influence of a specified initial velocity (even one equal to zero) persists for considerable travel times of the particle. A simple exponential correlation thus defines the particle motion only after a displacement of the time origin (equal, according to (9), to  $v_0/\beta$ ) such that the velocity can be considered to be a stationary random process, i.e., when (corresponding to Taylor's statistical theory)  $v(t)$  has random values, unconditioned by  $v_0$ .

#### Limiting cases of the displacement variance.

Equation (12) can be written in a concise, nondimensional form by means of the following substitutions;  $a = K/\beta$ ,

$$b = \tau_0^2/\beta^2, \quad \Sigma_y^2 = \sigma_y^2/2a, \quad c = 1 - b/a, \quad \text{and} \quad T = \beta t.$$

Then equation (10a) becomes

$$\Sigma_y^2 = T - (1 - e^{-T}) - (c/2)(1 - e^{-T})^2. \quad (13)$$

The nondimensionalized dispersion,  $\Sigma_y^2$ , is thus seen to depend on a single parameter  $c = 1 - b/a = 1 - v_0^2/(\beta K) = 1 - v_0^2/v^2$ . On the average, this parameter varies between zero and unity depending on the value of  $b/a$ , which, in effect, is the ratio of the instantaneous turbulence level at the source to that of the entire flow.

For large values of  $T$ , equation (13) has the limit

$$\Sigma_y^2 = T \quad (14)$$

which is equivalent to

$$\sigma_y^2 = 2Kt.$$

Thus, it reproduces the well-known constant  $K$ , or Fickian diffusion solution, which is also the large-time limit of the statistical theory of diffusion, to which reference has already been made.

If the exponential terms in (13) are replaced by their power series, it develops that

$$\Sigma y^2 = (b/2a)T^2 + (1/3 - b/2a)T^3 + \text{h.o.t.} \quad (15)$$

Two distinct limiting cases for small values of  $T$  follow, depending on whether  $b/a$  is large or near zero. For large values of  $b/a$ , equation (15) reduces to

$$\Sigma y^2 = (b/2a)T^2, \quad (16)$$

or

$$\sigma_y^2 = v_0^2 t^2. \quad (16a)$$

For  $v_0^2$  averaged over all possible values, i.e.,  $\overline{v_0^2} = \overline{v^2}$ , (16a) provides the usual small-time limit of statistical theory. However, when the initial velocity,  $v_0$ , is specified to have a fixed value, (16a) equally indicates a small-time dispersion proportional to  $t^2$  but resulting, in this case, from an initial source effect. Thus (16a), with constant  $v_0$ , is related to the corresponding result from the theory of relative diffusion, for which the instantaneous cloud spreading is as  $t^2$  up to times large enough that the effect of the initial source configuration disappears.

When  $b/a$  is small, i.e., when  $c$  approaches unity, the small- $T$  limit becomes

$$\Sigma y^2 = T^3/3. \quad (17)$$

Since  $t_L = \beta^{-1} = K/\overline{v^2}$  is the Lagrangian time scale of the turbulent flow, this is equivalent to

$$\sigma_y^2 = (2/3)(\overline{v^2}/t_L)t^3. \quad (17a)$$

This result was derived by Smith (1968) as a consequence of his conditioned particle motion diffusion theory, but by assuming an exponential autocorrelation. Relative diffusion theory also produces a result of this form for the

instantaneous cloud spreading, applicable after initial source-size effects are negligible, but it involves an undetermined constant.

Consequently, equation (13) includes all the asymptotic results of both the time-averaged, Taylor form of diffusion theory and the relative, instantaneous-spreading form. Moreover, it is able to quantify some of these by providing explicit coefficients. Since equations (1) and (5) imply one another, equation (13) also reproduces results from the conditioned particle motion model, such as equation (17a), as well as various results of recent random-walk computer simulations based on equation (1) and an assumed exponential velocity correlation. However, the exponential form of  $R$  is, in the theory based on Langevin's equation, deduced from equations (1) and (5), rather than assumed; and the exponential form is shown to apply only when the initial velocity is averaged. That is to say, the exponential correlation applies only to the Taylor, or averaged, kind of diffusion. Instantaneous particle spreading depends at first on the influence of a specified initial velocity,  $v_0$ , and on the resulting, time-dependent autocorrelation. The applicability of the exponential Lagrangian correlation function, which has been assumed as a matter of expedience by most writers on atmospheric diffusion starting with Taylor (1921), has been clarified recently in the interesting papers by Neumann (1978) and Tennekes (1978). It is hoped that the above results may serve to further specify the role of  $R$  in the particle dispersion problem.

Equation (13) is illustrated in Fig. 1 for the extremes  $c = 0$  and  $c = 1$ , and for a typical intermediate value. All the preceding limiting forms can be seen. The  $T^{1/2}$  regime of equation (14), which is independent of  $c$ , is approached rapidly when  $T > 1$  and is reached in all the curves by  $T \sim 10$ . For  $c = 0$ , that is for  $v_0$  averaged over all possible values, (13) becomes

$$\Sigma_y^2 = T - (1 - e^{-T}), \quad (18)$$

which is the dimensionless, universal form of Taylor's result, equation (11), in which  $\Sigma_y$  varies at first linearly and later as  $T^{1/2}$ . At the other extreme, when  $v_0 = 0$  so that  $c = 1$ , the diffusion is of the instantaneous type but without an initial size effect. In this case,  $\Sigma_y$  varies as  $T$  for  $T \lesssim 10^{-1}$  and as  $T^{1/2}$  for  $T \gtrsim 10$ . For intermediate values of  $c$ , the behavior of the  $\Sigma_y$ -curve is complicated. Initial growth is as  $T$  because of the

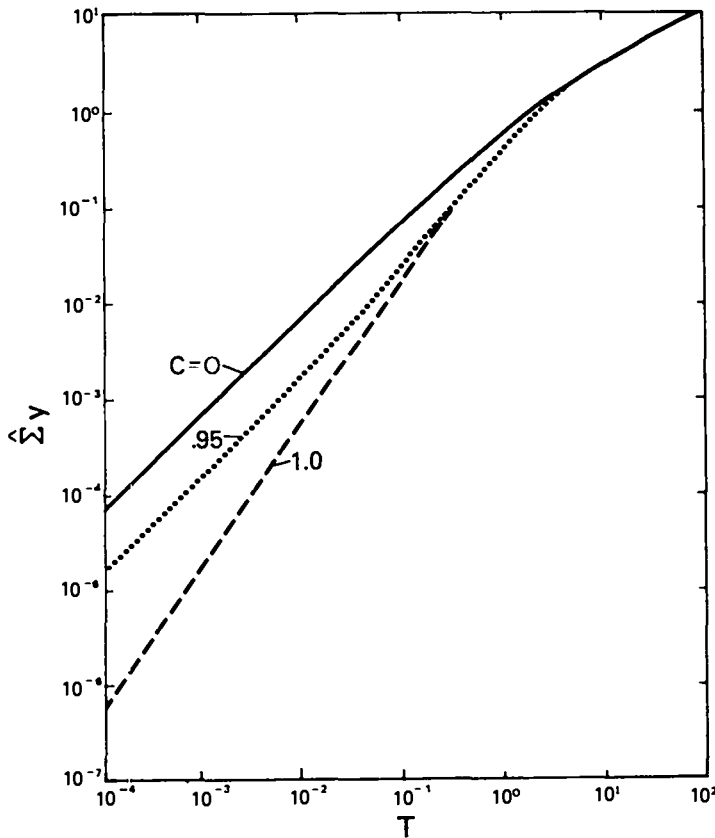


Fig. 1. Plot of nondimensionalized displacement standard deviation,  $\langle \Sigma_y \rangle$ , vs. nondimensionalized time,  $T$ , according to equation (13).

influence of  $v_0$ . This is followed by an accelerating diffusion range beginning when  $T$  becomes greater than about  $10^{-1}$  in this particular instance, and reaching the value  $T^{3/2}$  at about  $T \approx 0.3$ . The final  $T^{1/2}$  growth begins at around  $T = 5$ .

Comparisons with atmospheric diffusion data.

Horizontal diffusion:

Richardson (1926) was the first to draw attention to a remarkable property of horizontal diffusion in the atmosphere, namely that the phenomenon can be observed to occur at an accelerating rate. Particle clouds spread rapidly; the bigger the cloud, the faster

it spreads. Subsequent summaries of horizontal cloud spreading data in the atmosphere, by Heffter (1965), Hage (1964), Crawford (1966), and others (who mainly based their work on these studies) have amply confirmed this finding. This suggests that atmospheric diffusion results not solely from the comparatively small-scale, three-dimensional, inertial turbulence that is responsible for the well-known micrometeorological energy-spectrum maximum, but in addition, that it is augmented by the presence of much larger-scale turbulent wind-field heterogeneities. These must necessarily be quasi-horizontal and may be caused by large-scale surface inhomogeneities of various kinds. Additionally (or alternatively), there may be a reverse transfer of energy, toward lower frequencies (larger scales), from the micrometeorological maximum, as proposed by Gage (1979). It seems that the Langevin model, which provides for distinct small- and large-scale turbulence effects, might provide solutions to the atmospheric diffusion problem of the type that are observed.

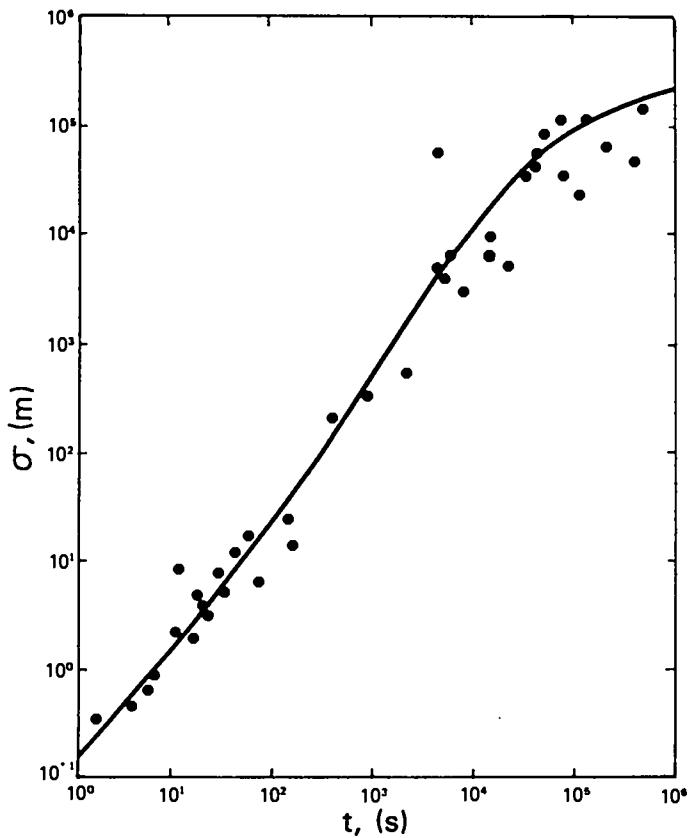


Fig. 2. Plot of Crawford's (1966) tropospheric diffusion data (Gifford, 1977) and  $\sigma_y$  from equation (10a).

In Fig. 2, the standard deviation of the instantaneous particle position displacement,  $\sigma_y$ , according to equation (10a), is compared with a subset of Crawford's summary of horizontal atmospheric diffusion data. This particular selection includes only tropospheric diffusion experiments; all stratospheric data have been removed, and the resulting data are more homogeneous and broadly representative of tropospheric diffusion conditions. For a description of, and references to, the several different data sets that are combined in Fig. 2, see Gifford (1977). The parameter values,  $K = 5 \times 10^4 \text{ m}^2 \text{ s}^{-1}$  and  $v_0 = .15 \text{ m s}^{-1}$ , were determined by comparing data extremes with equations (14a) and (16a); together with the value  $\beta = 10^{-4}$  they

provide the solid curve in the figure. The parameter  $c$  has the value  $c = 0.9955$  for these choices.

Figure 3, from Hage and Church (1967) illustrates an empirical curve of horizontal atmospheric diffusion over the entire atmospheric range that has been generally accepted as the best representation of the existing body of data. It was obtained by Hage et al. (1966) by analyzing the data compilations mentioned earlier, using standard curve-fitting techniques. The empirical equation (Hage et al., 1967) for this curve has the form (for  $\sigma_y$  in kilometers,  $t$  in seconds)

$$\log_{10} \sigma_y = -2.81524 + 0.228807 \log_{10} t + 0.274799 (\log_{10} t)^2 - 0.0241565 (\log_{10} t)^3 . \quad (19)$$

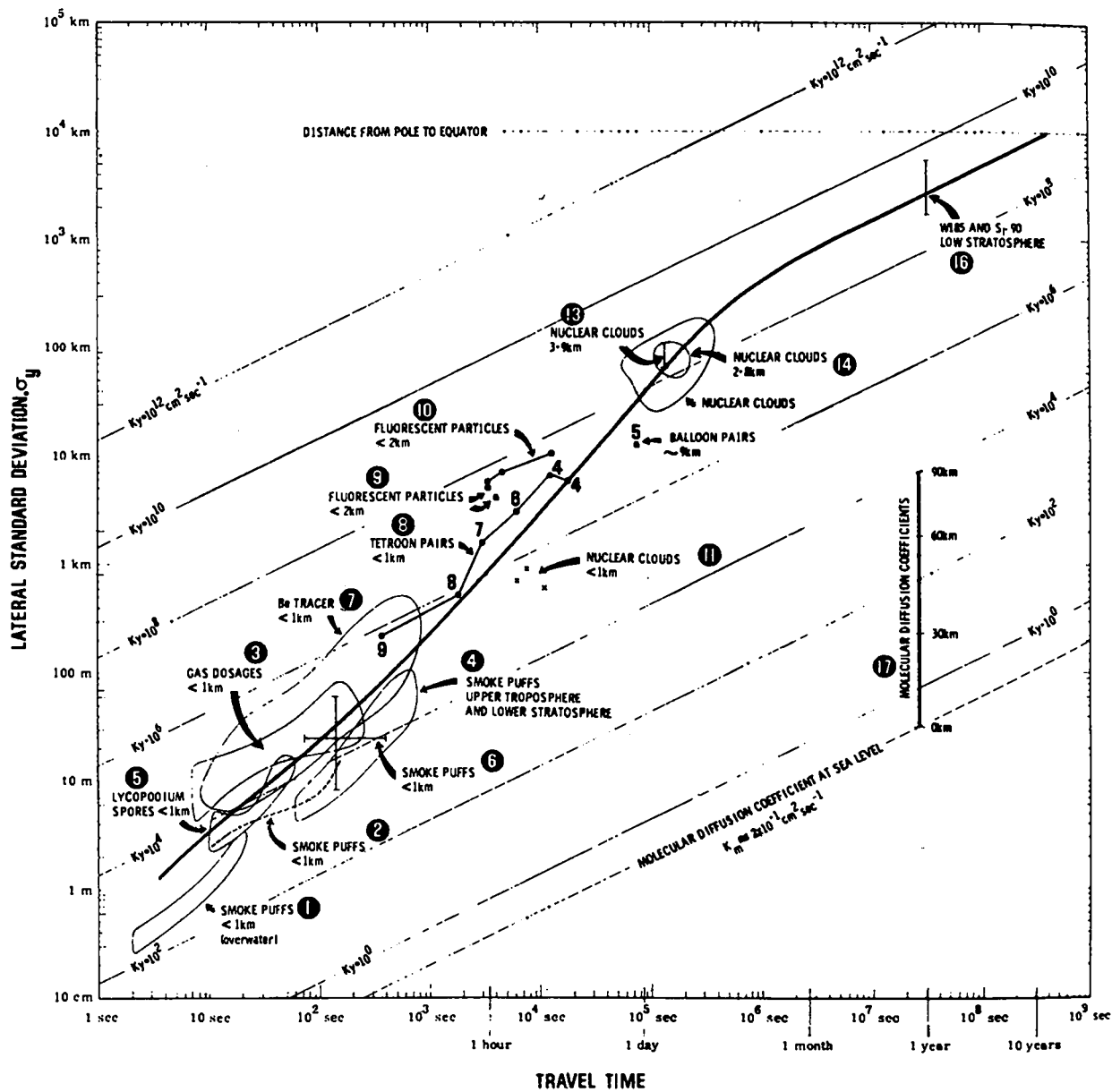


Fig. 3. Summary of data on horizontal atmospheric diffusion, from Hage and Church (1967). The solid curve, see Hage et al. (1967), illustrates equation (19).

If values of  $\sigma_y$  are computed from equation (19) for short, medium, and large dispersion times ( $t = 30, 10^5, \text{ and } 10^7$  seconds) and introduced into the asymptotic equations (14a), (16a), and (17a), the physical parameter values are found to be:  $K = 1.235 \times 10^5 \text{ m}^2\text{s}^{-1}$ ,  $v_0 = 0.253 \text{ ms}^{-1}$ , and  $t_L = \beta^{-1} = 2.13 \times 10^5 \text{ s}$ . When these values are introduced into equation (10a), the comparison with equation (19) displayed in Fig. 4 results.

The theoretical equation (10a), which contains three physical parameters, is seen in Fig. 4 to reproduce the four-parameter empirical curve, equation (19), quite faithfully. Although the small-scale parameter,  $v_0 = 0.253 \text{ ms}^{-1}$ , is not much different from the value used in the previous comparison, both large-scale parameters,  $K$  and  $t_L$ , are an order of magnitude larger. This reflects the fact that Fig. 3 and equation (19) include the

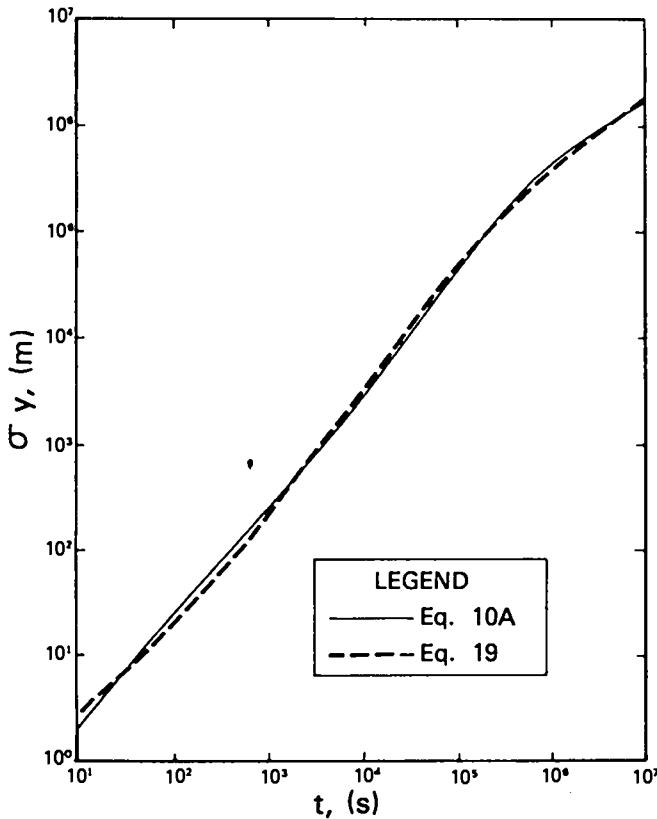


Fig. 4. Plots of theoretical curve, equation (10a), and empirical curve, equation (19), of horizontal diffusion.

stratospheric data points that were excluded from Fig. 2. The quantity  $v^2/t_L$  measures the turbulent energy dissipation. For these two cases, it has the values  $5 \text{ cm}^2\text{s}^{-3}$  for the tropospheric data of Fig. 2 and  $0.16 \text{ cm}^2\text{s}^{-3}$  for the data of Fig. 3, which includes the stratospheric points. Energy dissipation in the troposphere has been shown by various lines of reasoning to average about  $5 \text{ cm}^2\text{s}^{-3}$ , and the smaller value clearly indicates that stratospheric conditions dominate there. Thus the values of  $t_L$  and  $K$  associated with Fig. 2 appear to be more representative of tropospheric dispersion.

It can be concluded from Fig. 4 that the present theory results in a high-quality fit to data over a very wide range of atmospheric diffusion scales. Neither the asymptotic

predictions of other available theories, nor the often-quoted empirical power law,  $\sigma_y \propto t^{1.2}$ , provides this quality of overall agreement with the data. Note in particular that if typical short-range, averaged-diffusion  $\sigma_y$ -values are extrapolated to times corresponding to distances on the order of 100-200 km, the result will fall short of both observed values and the theoretical curve, by amounts ranging up to nearly an order of magnitude. The practice of extrapolating standard short-range values of  $\sigma_y$  to estimate large-scale values seems to be common among air-pollution modelers, see, for instance, Lange (1978) who treated horizontal diffusion in the well-known ARAC model this way. Modelers should be wary of the effect on pollution estimates of such brute-force extrapolations.

Averaged-to-instantaneous displacement variance ratio: The ratio of the averaged cloud dispersion, equation (18), to the instantaneous value, equation (13), is a quantity of interest. It is, in the first place, related to the ratio of instantaneous peak to time-averaged concentration values, a quantity of considerable importance in air pollution regulation. This ratio of the dispersions is

$$\left\langle \frac{\sigma_y^2}{v_0} \right\rangle / \sigma_y^2 = [T - (1 - e^{-T})] / [T - (1 - e^{-T}) - (c/2)(1 - e^{-T})^2]. \quad (20)$$

A plot for a range of  $c$ -values is shown in Fig. 5.

Measurements of both instantaneous and time-averaged horizontal spreading of smoke plumes have been made by several researchers, by combining instantaneous and time-lapse photography of a smoke plume. Experiments reported by Hilst (1957), Byzova, et al. (1970), and Nappo (1979) can be used to determine the variance ratio as a function of travel time,  $t$ . Since for each of these experiments the quantity  $\overline{v^2}$ , the mean square turbulent velocity, is either given or can be estimated from the averaged plume dispersion near the source,  $\beta = t_L^{-1}$  can be estimated from equation (17a), using the instantaneous plume variance values,  $\sigma_y^2$ . Then, from the resulting plots on Fig. 5, the parameter  $c$  can be estimated for each plume. Since  $c = 1 - b/a = \overline{v^2} t_L / K$ , a large-scale eddy diffusivity is implied for each experiment, even though the photographs upon which these determinations are based were necessarily representative of fairly short travel times, a small fraction of  $T$ .



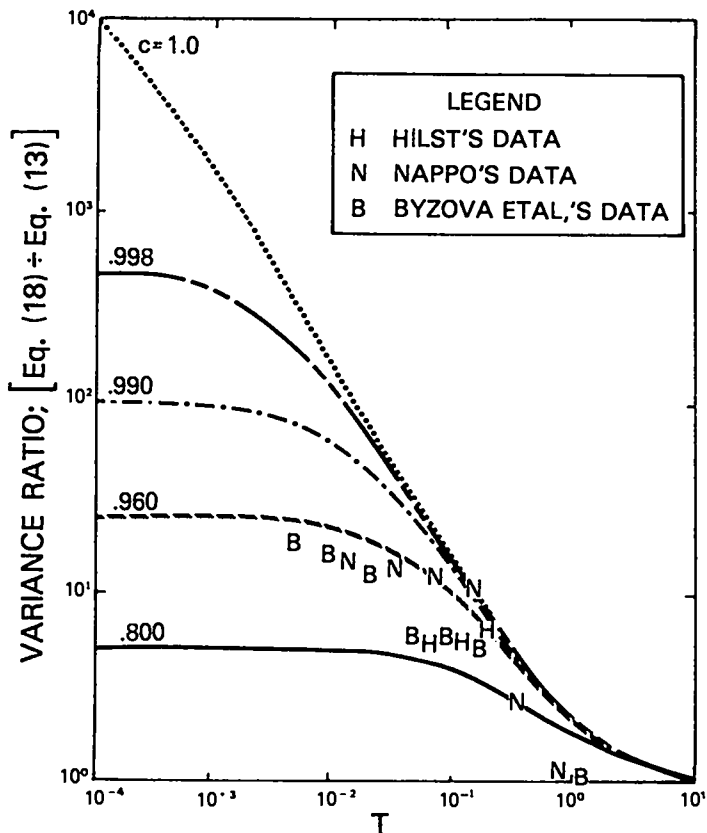


Fig. 5. Plot of the ratio of the displacement variance averaged on  $v_0$ , equation (18) to the un-averaged value, equation (13);  $T = \beta t$  is nondimensionalized time.

A summary of the various quantities entering the determinations of  $K$  is given in Table I, based on the ratios plotted in Fig. 5. The  $K$ -values deduced in Table I are certainly reasonable for large-scale atmospheric diffusion. They provide a convincing demonstration of the general correctness of the Langevin-equation model of atmospheric diffusion, in that the estimated  $K$ -values are based on diffusion data at only very small distances downwind. The  $c$ -values used were based on the smallest available values of  $T$  because for these the variance ratios are in theory nearly constant; approximately,  $T$  values were in the range  $5 \times 10^{-3} < T < 5 \times 10^{-2}$ , typical of plume spreading within a few hundred meters of the source, at most. Yet these values correctly imply the

TABLE I

Large-scale eddy diffusivity,  $K$ , determined from average-to-instantaneous displacement variance ratio,  $\langle \sigma_y^2 \rangle_{v_0} / \sigma_y^2$

Reference	$v^2, m^2/s$	$t_L, s$	$b/a$	$K, m^2 s^{-1}$
Hilst (1957)	: .56	550	.02	$1.5 \times 10^4$
Byzova et al. (1970)	: .18	4600	.04	$2.1 \times 10^4$
Nappo (1979)	: .70	1350	.03	$3.2 \times 10^4$

order of magnitude of the large-scale  $K$ , which controls diffusion only after many hours of downwind cloud travel.

#### SUMMARY AND CONCLUSIONS

A form of Langevin's equation has been derived from a simple, linear equation for the turbulent velocity. Solutions provide equations for the horizontal mean square particle displacement,  $\sigma_y^2$ , for the case for which the initial velocity is fixed and for the case of a random initial velocity. These correspond to the familiar instantaneous and time-averaged cloud variances of statistical diffusion theory; and all the limiting cases of that theory can be derived in terms of the physical parameters that appear in this form of the Langevin equation, namely the initial velocity, the Lagrangian time scale, and the large-scale eddy diffusivity. For reasonable choices of these parameters, determined primarily by the small- and large-time limits, the displacement equation demonstrates an excellent fit to atmospheric horizontal diffusion data, over diffusion times ranging from seconds to days. The fit is good not only at the upper and lower time limits, but also through the broad middle range of times, corresponding to distances from the source in the range of several hundred kilometers. It is pointed out that extrapolation to these distances of commonly used short-range values of  $\sigma_y$  can be in error by appreciable amounts. Finally, photographic observations of instantaneous and time-averaged smoke plumes are compared with the theoretical spreading curves. By this procedure, which involves observations of plumes to only a few hundred meters, all the parameters of the theory can be determined, including the large-scale eddy diffusivity,  $K$ . Three independent sets of plume photographs are analyzed in this way, and the large-scale  $K$ -values that are determined compare well with typical atmospheric values. The theory appears to predict horizontal atmospheric diffusion over a wide scale range quite well and provides, in particular, a basis for estimating diffusion at regional scales for modeling purposes, something that has not so far been achieved by other methods.

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