

Generalization of K Theory for Turbulent Diffusion. Part I: Spectral Turbulent Diffusivity Concept

RUWIM BERKOWICZ

Danish Meteorological Institute, Air Pollution Section, Lyngbyvej 100, DK-2100 Copenhagen, Denmark

LARS P. PRAHM

*Danish Air Pollution Laboratory, National Agency of Environmental Protection,
Risø National Laboratory, DK-4000 Roskilde, Denmark*

(Manuscript received 15 August 1978, in final form 4 December 1978)

ABSTRACT

The gradient transfer theory for turbulent diffusion is reformulated in order to obtain an improved method for applied dispersion studies. The basic innovation is that diffusivity of single Fourier components of the concentration field is treated separately, i.e., spectral turbulent diffusivity coefficients are introduced. The value of the diffusivity decreases with increasing wave vector k of the concentration spectrum. The rate of growth of an expanding cloud of material thus becomes dependent on the stage of growth. This is in qualitative agreement with the statistical dispersion theory. It is shown that the assumption of k -dependent diffusivity leads to a nonlocal flux-gradient relation. A new function, the turbulent diffusivity transfer function, is introduced. The turbulent diffusive flux depends on concentration gradients at all points in the space. The diffusion equation is written in terms of the turbulent diffusivity transfer function. The width of the turbulent diffusivity transfer function is shown to determine the validity of the traditional gradient transfer theory formulation. The turbulent dispersion can be considered as Gaussian when the size of the cloud is considerably larger than the size of the most energetic turbulent eddies. These eddies determine the width of the turbulent diffusivity transfer function.

In general, it is shown that the shape of the cloud is non-Gaussian and the width, computed in terms of spectral turbulent diffusivity coefficients, is smaller than in a Gaussian distribution. This deviation decreases with increasing size of the cloud.

The present theory reveals properties in agreement with experiments and Lagrangian statistical dispersion theory and has the advantage of being an Eulerian method which can be used for air pollution dispersion models treating multiple, interacting sources.

1. Introduction

The gradient transfer theory (K theory) (Schmidt, 1925) is much used in the field of air pollution studies. The theory is based on the analogy with molecular diffusion. One of the most important advantages of the gradient transfer theory is that it describes the diffusive transport in an Eulerian framework. In the atmosphere, diffusive transport is caused by turbulent eddies. The rate of diffusion of a cloud of material generally depends on the cloud dimensions and the intensity of turbulence. As the cloud enlarges, larger eddies come into play and, as the larger eddies represent more energy, the rate of diffusion increases. However, eddies larger than the cloud itself are relatively unimportant in its expansion. They represent meandering, not expansion. The turbulent diffusion process, in contradiction to molecular diffusion, is therefore scale-dependent. The gradient-transfer theory works well enough when the dimensions of the dispersed material are much larger than the size

of the turbulent eddies involved in the diffusivity process (e.g., in case of dispersion in a vertical direction from sources situated near the earth's surface). In general, however, and especially for horizontal diffusion from point sources, the gradient transfer theory with constant diffusivity yields erroneous results for dispersion close to a source where the size of the dispersed material is smaller than the most energetic turbulent eddies. Introduction of a diffusivity coefficient as a function of time of travel or distance from a source allows us, however, to obtain results which are physically justified; but the time-dependence makes it difficult to treat the diffusion equation in an Eulerian system because the diffusivity cannot be defined on a fixed-coordinate system if dispersion from multiple sources has to be treated simultaneously.

Turbulent diffusion has extensively been treated in terms of statistical theory. Since the pioneering works of Taylor (1921), the statistical theory has been applied with success to diffusion from both continuous

Generalization of K Theory for Turbulent Diffusion. Part I: Spectral Turbulent Diffusivity Concept

RUWIM BERKOWICZ

Danish Meteorological Institute, Air Pollution Section, Lyngbyvej 100, DK-2100 Copenhagen, Denmark

LARS P. PRAHM

*Danish Air Pollution Laboratory, National Agency of Environmental Protection,
Risø National Laboratory, DK-4000 Roskilde, Denmark*

(Manuscript received 15 August 1978, in final form 4 December 1978)

ABSTRACT

The gradient transfer theory for turbulent diffusion is reformulated in order to obtain an improved method for applied dispersion studies. The basic innovation is that diffusivity of single Fourier components of the concentration field is treated separately, i.e., spectral turbulent diffusivity coefficients are introduced. The value of the diffusivity decreases with increasing wave vector k of the concentration spectrum. The rate of growth of an expanding cloud of material thus becomes dependent on the stage of growth. This is in qualitative agreement with the statistical dispersion theory. It is shown that the assumption of k -dependent diffusivity leads to a nonlocal flux-gradient relation. A new function, the turbulent diffusivity transfer function, is introduced. The turbulent diffusive flux depends on concentration gradients at all points in the space. The diffusion equation is written in terms of the turbulent diffusivity transfer function. The width of the turbulent diffusivity transfer function is shown to determine the validity of the traditional gradient transfer theory formulation. The turbulent dispersion can be considered as Gaussian when the size of the cloud is considerably larger than the size of the most energetic turbulent eddies. These eddies determine the width of the turbulent diffusivity transfer function.

In general, it is shown that the shape of the cloud is non-Gaussian and the width, computed in terms of spectral turbulent diffusivity coefficients, is smaller than in a Gaussian distribution. This deviation decreases with increasing size of the cloud.

The present theory reveals properties in agreement with experiments and Lagrangian statistical dispersion theory and has the advantage of being an Eulerian method which can be used for air pollution dispersion models treating multiple, interacting sources.

1. Introduction

The gradient transfer theory (K theory) (Schmidt, 1925) is much used in the field of air pollution studies. The theory is based on the analogy with molecular diffusion. One of the most important advantages of the gradient transfer theory is that it describes the diffusive transport in an Eulerian framework. In the atmosphere, diffusive transport is caused by turbulent eddies. The rate of diffusion of a cloud of material generally depends on the cloud dimensions and the intensity of turbulence. As the cloud enlarges, larger eddies come into play and, as the larger eddies represent more energy, the rate of diffusion increases. However, eddies larger than the cloud itself are relatively unimportant in its expansion. They represent meandering, not expansion. The turbulent diffusion process, in contradiction to molecular diffusion, is therefore scale-dependent. The gradient-transfer theory works well enough when the dimensions of the dispersed material are much larger than the size

of the turbulent eddies involved in the diffusivity process (e.g., in case of dispersion in a vertical direction from sources situated near the earth's surface). In general, however, and especially for horizontal diffusion from point sources, the gradient transfer theory with constant diffusivity yields erroneous results for dispersion close to a source where the size of the dispersed material is smaller than the most energetic turbulent eddies. Introduction of a diffusivity coefficient as a function of time of travel or distance from a source allows us, however, to obtain results which are physically justified; but the time-dependence makes it difficult to treat the diffusion equation in an Eulerian system because the diffusivity cannot be defined on a fixed-coordinate system if dispersion from multiple sources has to be treated simultaneously.

Turbulent diffusion has extensively been treated in terms of statistical theory. Since the pioneering works of Taylor (1921), the statistical theory has been applied with success to diffusion from both continuous

and instantaneous sources (Batchelor, 1953). The statistical theories explain quite well the most important features of turbulent diffusion and the results of all other theories are usually compared with the results predicted by the statistical theory. The weakness of the statistical theory, however, is that it expresses the diffusion process in terms of Lagrangian parameters and is thus not suitable for multiple-source Eulerian air pollution models. Richardson (1926) has formulated a theory of cloud diffusion taking into account the dependence of the diffusivity on the effective size of the cloud. Richardson's model deals with dispersion of pairs of particles, i.e. with relative diffusion. The diffusion equation is formulated in terms of the so-called "distance-neighbor function." In practical air pollution studies, results in terms of concentration distribution are, however, more desirable. Most of the existing atmospheric diffusion models and theories are discussed in detail by Monin and Yaglom (1973) and Pasquill (1974).

Inspired by our previous numerical studies (Christensen and Prahm, 1976; Prahm and Christensen, 1977; Berkowicz and Prahm, 1978) and the need of an improved Eulerian model of turbulent diffusion, we propose here an alternative theory, a spectral turbulent diffusivity theory. This theory makes it possible to handle the diffusive transport in an Eulerian system, but with diffusivity being effectively dependent on the actual size of the concentration distribution. The theory is based on a spectral generalization of the K theory. Because of its simple mathematical formulation, the spectral turbulent diffusivity theory is applicable in Eulerian numerical models for simulation of atmospheric turbulent dispersion.

This research is based primarily on phenomenological experience but also on the basic physical properties of turbulent diffusion. The work is presented in two parts. This paper (Part I) contains a general formulation of the spectral diffusivity concept and deals with diffusion of a puff or a cloud of material. In Part II (Prahm *et al.*, 1979), the parameters of the theory are discussed in more detail, and the spectral turbulent diffusivity concept is applied on dispersion of a plume. In Part II, we also suggest methods for direct experimental testing of this new concept for diffusion studies.

2. Spectral turbulent diffusivity concept

We consider the case of one-dimensional diffusion of a concentration distribution $c(y,t)$, where y is the spatial coordinate and t the time. The diffusion equation, written in terms of gradient transfer theory, is

$$\frac{\partial}{\partial t}c(y,t) = \frac{\partial}{\partial y}K\frac{\partial}{\partial y}c(y,t). \tag{1}$$

If the diffusivity coefficient K is constant in space, it is easy to show that

$$c(y,t) = \sum_{k_n=-\infty}^{\infty} A(k_n,t)e^{ik_n y} \tag{2}$$

is the solution to (1), providing that

$$\frac{d}{dt}A(k_n,t) = -k_n^2 K A(k_n,t), \tag{3}$$

where the wave vectors $k_n = \pi n/L$, $n = \pm 0, 1, \dots, \infty$, and $2L$ is the length of the region to which the diffusion is limited. With boundary conditions

$$\frac{\partial}{\partial y}c(y,t) = 0 \text{ at } y = \pm L \tag{4}$$

and for an instantaneous point source of a strength Q_0 and located at $y=0$, the solution to (1) becomes

$$c(y,t) = \frac{Q_0}{2L} \left[1 + 2 \sum_{n=1}^{\infty} \cos k_n y \exp(-k_n^2 K t) \right]. \tag{5}$$

If $L \rightarrow \infty$, the summation in (5) can be replaced by integration with respect to k , by the transformation

$$\frac{1}{L} \sum_n \rightarrow \frac{1}{\pi} \int dk.$$

As a result, we obtain the well-known Gaussian shape of the concentration field

$$c(y,t) = \frac{Q_0}{\pi^{1/2} \sigma} \exp(-y^2/2\sigma^2), \tag{6}$$

where $\sigma^2 = 2Kt$ is the Gaussian standard deviation coefficient.

Examining Eq. (2), we can see that the concentration field is written as a Fourier series, or for $L \rightarrow \infty$, as a Fourier integral. We rewrite (2) in the form

$$c(y,t) = \sum c_{k_n}(y,t), \tag{2a}$$

where

$$c_{k_n}(y,t) = A(k_n,t) \exp(ik_n y), \tag{2b}$$

where $c_{k_n}(y,t)$ are Fourier components of the concentration field or concentration mode. Eq. (1) can be written for each single concentration mode as

$$\frac{\partial}{\partial t}c_{k_n} = K \frac{\partial^2}{\partial y^2}c_{k_n} \tag{7}$$

and the solution is

$$c_{k_n}(y,t) = c_{k_n}(y,0) \exp(-k_n^2 K t). \tag{8}$$

Each mode is thus attenuated with the attenuation rate determined by the wave vector k_n and diffusivity K , the latter being the same for all modes.

The resulting shape of the concentration field becomes Gaussian with the standard deviation coefficient $\sigma^2 = 2Kt$.

Our generalization of the diffusivity concept is based on the hypothesis that the diffusivity is dependent on the wave vector k_n of the concentration field. Contrary to molecular diffusivity, the turbulent diffusivity is known to be scale-dependent. An optimal way to take this scale-dependence into account is to treat each Fourier mode of the spatial concentration distribution separately. Thus, we define the spectral turbulent diffusivity coefficient $K(k_n)$ as a function of the concentration wave vector. By analogy to Eq. (7), we will assume the following diffusion equation for each Fourier mode:

$$\frac{\partial}{\partial t} c_{k_n} = K(k_n) \frac{\partial^2}{\partial y^2} c_{k_n}, \tag{9}$$

$$c_{k_n}(y,t) = c_{k_n}(y,0) \exp[-k_n^2 K(k_n)t]. \tag{10}$$

The spectral diffusivity coefficients $K(k_n)$ should not be confused with Fourier coefficients of a space-dependent diffusivity, as we still consider a homogeneous turbulent field.¹ Eq. (9) describes diffusion of a single wave component of the concentration field; however, the diffusion equation for the total concentration field cannot be written in a form similar to (9) when $K(k_n)$ is not constant. We introduce a new function, a turbulent diffusivity transfer function, which is defined as

$$D(y,y') = \frac{1}{2L} \sum_{k_n=-\infty}^{\infty} K(k_n) \exp[ik_n(y-y')]. \tag{11}$$

Also here the summation is replaced by integration when $L \rightarrow \infty$. It is now easy to show that the following equation describes the turbulent diffusion in terms of the spectral diffusivity coefficients

$$\frac{\partial}{\partial t} c(y,t) = \frac{\partial}{\partial y} \int_{-L}^L D(y,y') \frac{\partial}{\partial y'} c(y',t) dy'. \tag{12}$$

Substituting (11) for $D(y,y')$, and (2a) and (2b) for $c(y',t)$ and using the orthogonality relation

$$\int_{-L}^L \exp[i(k_n - k_m)y] dy = \begin{cases} 2L, & k_n = k_m \\ 0, & k_n \neq k_m \end{cases}$$

we obtain

$$\sum_{k_n} \frac{\partial}{\partial t} c_{k_n} = \sum_{k_n} K(k_n) \frac{\partial^2}{\partial y^2} c_{k_n}, \tag{13}$$

¹ It should not be surprising that Eq. (9) does not allow for the interaction among various Fourier modes. Such an interaction would violate the requirement of invariance of the diffusion equation toward a homogeneous translation in space.

which is equivalent to (9). Eq. (12) is a generalization of the gradient transfer theory for turbulent diffusion given by (1). The integral term on the right-hand side of (12), taken with the minus sign, is the turbulent diffusivity flux. In contradiction to the traditional gradient transfer theory formulation, the flux here depends not only on local values of concentration gradients, but also on values at remote points. This depicts the fact that turbulent eddies of different sizes and energy take part in the diffusive transport process. In case of a spectral diffusivity $K(k_n) = K_0$ for all k_n , it follows from (11) that $D(y,y') = K_0 \delta(y-y')$ and Eq. (12) reduces to (1). Eq. (1) can thus be considered as a special case of the more general formulation given by Eq. (12). The nonlocal character of the turbulent diffusion is the main feature of this type of diffusion. Batchelor and Townsend (1956) were the first to suggest that this nonlocalness may be best expressed by an integro-differential equation. In the case of the molecular diffusion, only the local fluctuations are responsible for the dispersion process. Because the rate of diffusion defined in (12) depends on the whole concentration field, it becomes clear why the diffusivity behaves as if it were dependent on the extension of the dispersed cloud of material. In (11), it is assumed that the spectral diffusivity coefficients are constant in space. This corresponds to homogeneous atmospheric turbulent diffusion, which is relevant, e.g., in the horizontal direction. The turbulent diffusivity transfer function $D(y,y')$ depends in this case on $y-y'$ only and not on y . In the vertical direction, however, the diffusivity is in general dependent on the height above the earth's surface and therefore can no longer be treated as a constant. The turbulent diffusivity transfer function is thus, in this case, not only a function of $z-z'$, but also a function of z and is defined as

$$D(z,z') = \frac{1}{L} \sum_{k_n} K(z,k_n) \exp[ik_n(z-z')]. \tag{14}$$

Because of the spatial dependence of the spectral diffusivity, interaction among various Fourier modes can be expected in the nonhomogeneous case. The appropriate diffusion equation in the Fourier space now is

$$\frac{\partial}{\partial t} A(k_n,t) = -k_n \sum_{k_m} \bar{K}(k_n - k_m, k_m) k_m A(k_m,t),$$

where $A(k_m,t)$ is defined in (2b) and $\bar{K}(\kappa,k_m)$ are Fourier coefficients of the now space-dependent spectral diffusivity $K(z,k_m)$, i.e.,

$$K(z,k_m) = \sum_{\kappa} \bar{K}(\kappa,k_m) e^{i\kappa z}.$$

The nonhomogeneous diffusion will be treated in detail in a forthcoming study and in the following we will

limit the discussion to the homogeneous case. Expression (12), with the turbulent diffusivity transfer function given by (14), can, however, be considered as a general diffusion equation but Eq. (10) can directly be used to compute the concentration field in the homogeneous case.

After the first draft of this manuscript was completed, our attention was led to a turbulent diffusivity theory based on the so-called direct interaction approximation (Kraichnan, 1959; Roberts, 1961). As the results of the theory happen to be in a very close relation to our theory, we find that a comparison is required and this comparison is given in the Appendix to this paper.

3. The spectral diffusivity coefficients

It is necessary to determine the spectral diffusivity coefficient $K(k)$ in order to use the theory formulated here. Turbulent eddies of dimensions comparable with the wavelength of a concentration wave will be most effective in dispersing this wave. The form of $K(k)$ can be deduced from studies on expansion of a puff or dispersion from a line source. As long as the cloud dimension is small (from 10 cm to 500 m), the standard deviation of its linear dimension can be expressed as (Pasquill, 1974)

$$\sigma = \alpha \epsilon^{1/3} l^{2/3}, \tag{15}$$

where α is a dimensionless constant of the order of unity and ϵ is the turbulent kinetic energy dissipation rate (Lumley and Panofsky, 1964). From the relation

$$K = -\frac{1}{2} \frac{d\sigma^2}{dt}, \tag{16}$$

it follows that

$$K = \frac{3}{2} \alpha^3 \epsilon^{1/3} l^{2/3}. \tag{17}$$

Where $l = \sigma$, the typical scale size is significant for the turbulent diffusion process. When the cloud dimension enlarges and becomes greater than the size of the most energetic eddies, the diffusivity coefficient tends to a constant value. Relating the typical turbulence scale to the vector k , i.e., $k = 2\pi/l$, we can make the following first estimate for $K(k)$:

$$K(k) = \frac{K_0}{1 + Bk^3}, \tag{18}$$

where K_0 is the diffusivity of the long-wave range of the concentration spectrum, and B is a constant determining the width of the spectral diffusivity function. One may expect that the value of K_0 is determined by the length l_m of the most energetic eddies, resulting in $K_0 \sim l_m^3$. In the short-wave range of the concentration spectrum, we have $K \sim l^3$; B must therefore be proportional to l_m^3 . Expression (18) should be considered as strongly approximated and

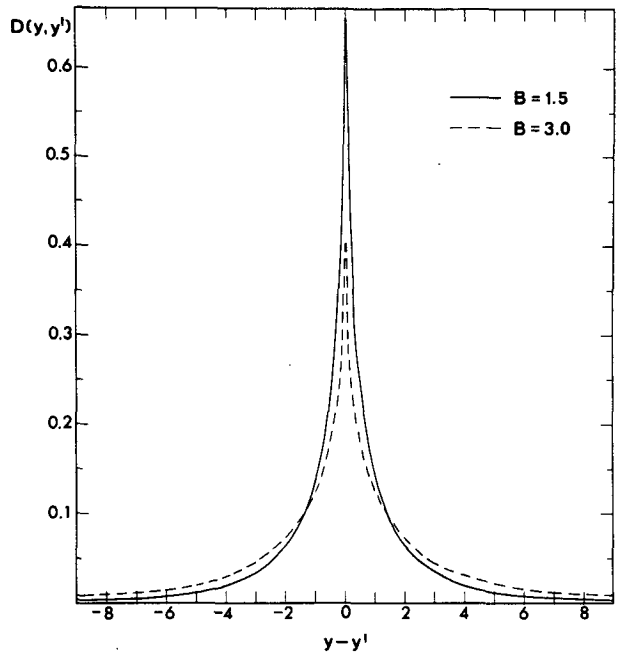


FIG. 1. The turbulent diffusivity transfer function. Eq. (18) is used for the spectral diffusivity coefficients. $D(y, y')$ is in units of $K_0/\Delta y$, where Δy is the length unit.

further study is necessary in order to determine the spectral turbulent diffusivity coefficient.

It follows from Eq. (10) that the amplitude of the shorter waves in the concentration spectrum decreases faster with time than the amplitude of the longer waves. This results in enlarging of the cloud with time. After sufficiently long time, only components with small k contribute to the concentration distribution and because $K(k) \rightarrow K_0$ as $k \rightarrow 0$, the further diffusion becomes nearly Gaussian. On the other hand, as long as the size of the cloud is small, Fourier components with large k values play an important role and the effective diffusivity is weaker than Gaussian, because $K(k)$ decreases as k increases. In this way, the diffusivity becomes effectively dependent on the size of the cloud, increasing with the growing cloud.

4. The turbulent diffusivity transfer function

We have computed the turbulent diffusivity transfer function using Eqs. (11) and (18) for the spectral diffusivity coefficients. In Fig. 1, $D(y, y')$ is plotted as function of $y - y'$ for two values of B (1.5 and 3.0). The length of the computational region is $2L = 32$ length units. Doubling of L has only little influence on the shape of $D(y, y')$. For larger values of B , the value chosen for L will be more important. As expected, the shape of the turbulent diffusivity transfer function becomes broader as B increases.

In the case of $B = 0$, $D(y, y')$ is a δ -like distribution with a zero width, and the corresponding diffusion

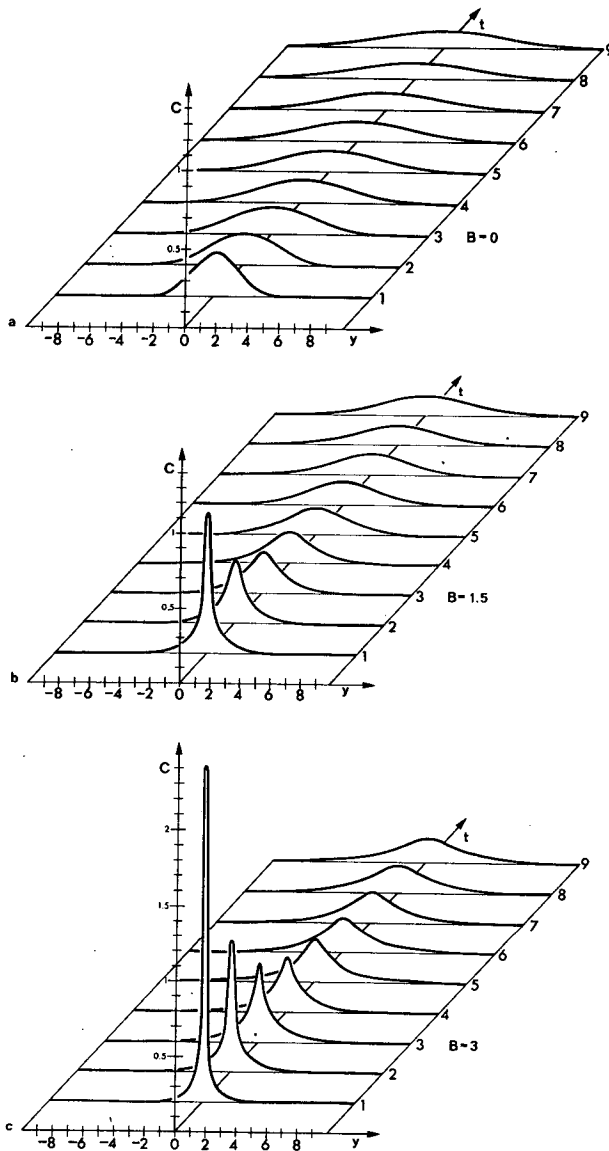


FIG. 2. Concentration profiles of an expanding one-dimensional cloud of material as a function of time: (a) $B=0$, (b) $B=1.5$, (c) $B=3.0$. Concentration is in units of $Q_0/\Delta y$, time in units of $(\Delta y)^2/K_0$ and (Δy) is the length unit.

becomes Gaussian. When the width of the expanding cloud is considerably larger than the width of $D(y, y')$, the turbulent diffusivity transfer function can be regarded as a δ -like distribution and the diffusion is again Gaussian. Because the width of $D(y, y')$ is determined by B , and $B \sim l_m^{\frac{2}{3}}$, the length l_m of the most energetic eddies can be used as a scale for determination of the validity of the traditional gradient transfer theory. In other words, the gradient transfer theory is applicable when the spread of the expanding material is much larger than the length of the most energetic eddies.

5. Cloud dispersion

In order to illustrate the theory of the spectral turbulent diffusivity, we present a computation of the dispersion of a one-dimensional cloud using (18) for the spectral diffusivity coefficients $K(k_n)$. The concentration field is then given by

$$c(y, t) = \frac{Q_0}{2L} \left[1 + 2 \sum_{n=1}^{\infty} \cos k_n y \exp[-k_n^2 K(k_n) t] \right], \quad (19)$$

where $k_n = \pi n/L$. In the case of $K(k_n) = K_0$, Eq. (19) becomes identical with (5). Summation in (19) is continued until the last term is only 0.01% of the concentration value. In Fig. 2, the concentration profiles are shown for three different values of the coefficient B : $B=0$ (Gaussian), $B=1.5$ and $B=3.0$. The units used in Fig. 2 are as follows: t -unit = $(\Delta y)^2/K_0$; c -unit = $Q_0/\Delta y$; and Δy is the length units in the y -direction. The unit for B is in this case $(\Delta y)^{\frac{2}{3}}$. We use $L = 16\Delta y$ in our computations. The choice of L is of minor importance as long as the spread of the cloud is smaller than L .

As one can see from Fig. 2, at the initial stage the spread of the cloud with $B=1.5$ and 3.0 is considerably smaller than the spread of the Gaussian ($B=0$) cloud. The shape of the cloud is also different from the Gaussian profile. At later stages this difference decreases.

In Fig. 3, the concentration at the center of the cloud is shown for $B=0, 0.5, 1.5$ and 3.0 . The rate of the change of the concentration with time increases with increasing value of B . Again, the difference decreases as the cloud grows. In Figs. 4, isopleths for concentrations equal to 10% of the maximum concentration are shown. For $B=0$, the isoline corresponds to a curve given by $y \sim t^{\frac{1}{2}}$. For $B=1.5$ and $B=3.0$, a considerable deviation from this relation can be seen for short times. Here the width of the cloud is smaller than in Gaussian dispersion. This behavior is in qualitative agreement with results predicted by the statistical theory and is due to the fact that the effective diffusivity increases as the cloud enlarges. In Part II, it is even shown that the spectral turbulent diffusivity theory reveals the well-known $\frac{3}{2}$ -power dependence of puff growth.

6. Conclusions

Introduction of spectral turbulent diffusivity coefficients, which are functions of the k vector of the concentration waves, results in a diffusivity which is effectively dependent on the actual extension of the dispersing material. On the basis of a spectral turbulent diffusivity concept, a new dispersion relation is defined, and the turbulent diffusivity transfer function is introduced. The theory reveals that turbulent fluxes of material are related not only to local concentration gradients, but also to values at remote points. The

traditional K theory formulation is shown to appear as a special case of the more general spectral turbulent diffusivity theory. Cloud dispersion treated by the latter theory reveals features in agreement with statistical theory. There is an essential difference, however, between the statistical theory and the spectral turbulent diffusivity theory. The first describes the diffusion in a Lagrangian reference system and the history of the dispersion is required. The spectral theory, however, treats the diffusion as depending only on the actual stage of dispersion and can handle the dispersion from an arbitrary initial concentration field with parameters defined on an Eulerian coordinate system.

The theory presented here should only be considered as a first attempt to formulate a dispersion theory based on a spectral turbulent diffusivity concept, but the results quoted here represent the essential features of such a theory. An improvement of the gradient transfer theory is of great importance if it leads to a concept which is easy to apply, even if this improvement does not lead to an exact theory for turbulent dispersion.

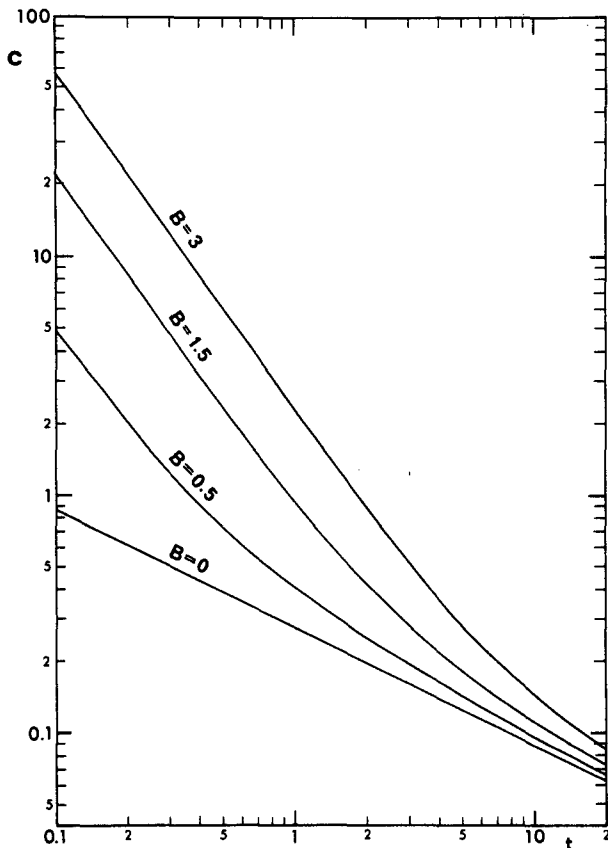


FIG. 3. Concentration at the center of a one-dimensional cloud as a function of time for $B=0, 0.5, 1.5$ and 3.0 . Units as in Fig. 2.

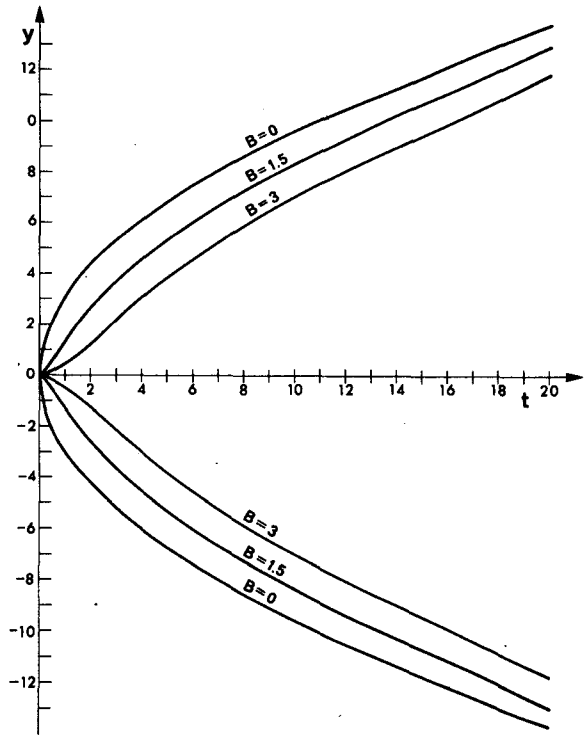


FIG. 4. Isopleths for concentrations equal to 0.1 of the maximum concentration. Units as in Fig. 2.

Acknowledgments. We would like to thank F. A. Gifford, NOAA, for encouragement during this research. Thanks also due to F. B. Smith, British Meteorological Office, for many critical comments and inspiring discussion and to an anonymous reviewer for leading our attention to the book by Leslie. We would like to extend our appreciation to Marie Bille who typed the manuscript. This study was activated as a part of the project Mesoscale Dispersion Modelling, coordinated by the Scandinavian Council for Applied Research. The project was supported by the Danish National Technical Scientific Research Council.

APPENDIX

Comparison between Robert's Theory and Spectral Diffusivity Theory

The spectral diffusivity concept presented in this paper is based on the assumption that each Fourier mode of the concentration distribution diffuses with a diffusivity coefficient characteristic for this mode. The physical justification of this non-traditional assumption is that the spectrum of the turbulent eddies effectively participating in the diffusivity process changes with the size of the diffusing cloud. A similar result was obtained from more basic relations by Roberts (1961) who formulated a theory of turbulent diffusion using the so-called direct interaction approximation (DIA). DIA was originally formulated by Kraichnan (1959) who focused on the theory of

homogeneous turbulence. An excellent review of the works connected with the DIA method and other related topics is given by Leslie (1973).

In this Appendix, a short comparison between the results from Roberts' theory and our spectral diffusivity theory is presented. In spite of strong similarity, the theory by Roberts appears to be more complex than ours, but we find that this complexity appears to be unnecessary in most of the practical applications for turbulent diffusion in the atmosphere. Roberts (1961) writes the diffusion equation for the probability distribution function $G(x,t)$, and for the one-dimensional case, the following Fourier transformed equation is presented:

$$\frac{\partial}{\partial t} G(k,t) = -2\pi k^2 \int_0^t dt' \left[\int \tilde{U}(k-k', t') \tilde{G}(k', t') dk' \right] \times \tilde{G}(k, t-t'), \quad (A1)$$

where $\tilde{U}(k,t)$ is a spectral component of the velocity correlation function for a time delay t . Using the relation

$$\tilde{G}(k,t) = \tilde{c}(k,t) / \int c(x,t) dx, \quad (A2)$$

where $\tilde{c}(k,t)$ is the Fourier component of the concentration distribution, Eq. (A1) can be written in the form

$$\frac{\partial}{\partial t} \tilde{c}(k,t) = -k^2 \int_0^t dt' \left[2\pi \int \tilde{U}(k-k', t') \tilde{G}(k', t') dk' \right] \times \tilde{c}(k, t-t'). \quad (A3)$$

Eq. (A3) is an Eulerian diffusion equation. Both $\tilde{U}(k,t)$ and $\tilde{G}(k,t)$ are universal functions depending only on the character of the turbulence and not on the initial and boundary conditions of the concentration distribution. $\tilde{G}(k,t)$ is the solution to (A1) with the initial conditions given by

$$\tilde{G}(k, t < 0) = 0 \quad \text{and} \quad \tilde{G}(k, 0) = (2\pi)^{-1}.$$

Note that, in the case of homogeneous turbulence, the diffusion equation must be invariant with respect to any uniform translation in space (Galilean transformation). It is easy to show that (A3) would not be invariant if $\tilde{G}(k,t)$ and $\tilde{c}(k,t)$ refer to the same boundary and initial conditions. In the paper by Roberts (1961), a relation similar to (A3) was not presented or discussed.

The term in square brackets in (A3) has dimensions of diffusivity \times time⁻¹ and is evidently a function of the k vector of the concentration distribution. The similarity between Eq. (A3) and the spectral diffusivity equation [(9)] of this paper is quite obvious. The substantial difference between Roberts' and our spectral formulation is the time nonlocality appearing in (A3) and absent in (9). According to the spectral diffusivity concept, the rate of change of a concen-

tration distribution is uniquely determined by the field of turbulence and the actual concentration distribution only. In the DIA formulation, however, the rate depends also on the history of the concentration distribution. It follows particularly from (A3) that the rate of expansion is always zero for $t=0$ independently on the initial concentration distribution. This is a somewhat dubious result as the shape of the concentration distribution should mainly determine the rate of diffusion. The time nonlocality might be important in the case of diffusion in a nonstationary turbulence field. When a turbulent diffusion model refers to a stationary turbulence field, only behavior on time scales longer than a relaxation time of the turbulence is of relevance. In this case, the time nonlocality cannot be of significant importance, and it is the authors' opinion that the spectral turbulent diffusivity model presented in this paper accounts for the essential physical properties of turbulent diffusion in stationary and homogeneous turbulent fields. It is worth noting that the theory by Richardson (1926) did not include any explicit time-dependency and one would wish an experimental indication of the necessity of the time nonlocality before accepting that such a complexity is required in applied studies of turbulent diffusion.

REFERENCES

- Batchelor, G. K., 1963: *The Theory of Homogeneous Turbulence*. Cambridge University Press, 197 pp.
- , and A. A. Townsend, 1956: Turbulent diffusion. *Surveys in Mechanics*, G. K. Batchelor and R. M. Davies, Eds., Cambridge University Press, 352-399.
- Berkowicz, R., and L. P. Prahm, 1978: Pseudospectral simulation of dry deposition from a point source. *Atmos. Environ.*, **12**, 379-387.
- Christensen, O., and L. P. Prahm, 1976: A pseudospectral model for dispersion of atmospheric pollutants. *J. Appl. Meteor.*, **15**, 1284-1294.
- Kraichnan, R. H., 1959: The structure of isotropic turbulence at very high Reynolds numbers. *J. Fluid. Mech.* **5**, 497-543.
- Leslie, D. C., 1973: *Developments in the Theory of Turbulence*. Clarendon Press, 368 pp.
- Lumley, J. L., and M. A. Panofsky, 1964: *The Structure of Atmospheric Turbulence*. Interscience, 239 pp.
- Monin, A. S., and A. M. Yaglom, 1973: *Statistical Fluid Mechanics of Turbulence*, Part 1. Trans-Joint Publication Research Service, Washington, 769 pp.
- Pasquill, F., 1974: *Atmospheric Diffusion*, 2nd ed. Wiley, 425 pp.
- Prahm, L. P., and O. Christensen, 1977: Long range transmission of pollutants simulated by the 2-D pseudospectral dispersion model. *J. Appl. Meteor.*, **16**, 896-910.
- , R. Berkowicz and O. Christensen, 1979: Generalization of K theory for turbulent diffusion. Part II. Spectral diffusivity model for plume dispersion. *J. Appl. Meteor.*, **18** (in press).
- Richardson, L. F., 1926: Atmospheric diffusion shown on a distance-neighbour graph. *Proc. Roy. Soc. London*, **A110**, 709.
- Roberts, P. H., 1961: Analytical theory of turbulent diffusion. *J. Fluid Mech.*, **11**, 257-283.
- Schmidt, W., 1925: Der Massenaustausch in freier Luft und Verwandte Erscheinungen. *Probleme der Kosmischen Physik*, Hamburg, Verlag Von Henri Grand, 176 pp.
- Taylor, G. I., 1921: Diffusion by continuous movements. *Proc. London Math. Soc.*, **20** (Ser. 2), 196-211.