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Some outstanding issues in the theory and practice of estimating diffusion from sources*

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In this survey I would like to begin by going over the theoretical principles which provide the framework within which we currently attempt generalizations and predictions about the diffusive spread of material released into the atmosphere. The atmosphere being as complex as it is, in practice these attempts often meet with difficulties, and these difficulties appear in their most acute form when we are concerned with medium-range or long-range dispersion, or with the spread of material from a considerable height above ground. With these difficulties in mind I propose therefore to focus the remainder of my remarks on two main topics - the understanding of the growth of horizontal spread with distance of travel - and the implications of the knowledge which has accumulated on the statistical properties of turbulence over the whole depth of the planetary boundary layer.

Principles for estimating diffusion from sources

There are three main approaches that are used in theoretically representing the spreading action of a turbulent atmosphere:

(a) The adoption of a 'simple diffusion' model in which the turbulent transfer of material across any plane is given by the product of the gradient of the material (normal to the plane) and an 'eddy diffusivity'.

(b) The use of statistical descriptions and laws for the velocities of typical material particles.

(c) The application of dimensional analysis (or similarity theory, as it is now usually called).

The diffusivity approach is basically still rather empirical, relying for its rationale largely on analogy with molecular transfer and kinetic theory,

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*Text for presentation at a symposium on turbulence and diffusion at the Sandia Corporation, Albuquerque, New Mexico, 5-7 December 1967.

with 'eddies' taking the place of molecules. This analogy is particularly useful in setting a scale relation between process and effect. Thus, just as molecular diffusion can be satisfactorily described only when it has been effective over dimensions large compared with the separation and mean free path of the molecules, so the analogous representation of transfer by eddies is obviously reasonable only if the dimensions occupied by the diffusing material are large compared with the dimensions and range of action of the eddies. This criterion is implicit in the statistical-theory result that after a long distance of travel (x) from a source, when the spread of particles (σ) becomes large, $\sigma \propto \sqrt{x}$, a variation which is also indicated by the classical diffusivity approach with constant eddy diffusivity. It is also relevant to note here that for the constant-stress layer in neutral conditions similarity theory predicts $\sigma \propto T$ for the vertical spread of particles at time T after leaving the surface, a result which also follows from taking $K \propto z$. This is an example of diffusive spread in which it might reasonably be argued that the effective eddies, being limited in size and vertical action by the presence of the boundary, are those which are smaller than, or at the most similar to the magnitude of vertical spread. It is therefore possible to accept certain situations as suitable for treatment by the diffusivity method. One important example which is certainly not in this category is the time-mean distribution from a continuous source, for here the distribution is largely controlled (especially at short range) by the scattering effect of eddies which are themselves larger than the cross-section of the plume stretching downwind from the source. When the 'K' approach is valid, however, it has the additional advantage that in principle the variation of diffusive power with position, and the effect of the fall and deposition of particles, can be allowed for. But before this advantage can be exploited to the full the distribution of K in absolute terms must be given.

The statistical theory is basically kinematical in nature, being concerned with the velocity fluctuations (of the particles). Given the statistics of these fluctuations it is no more than a formal (mathematical) step to convert these into statistics of particle displacement. The only obvious advantage of

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this approach is that if the flow is homogeneous and stationary in its properties, measurements referring to a typical particle or to a typical pair of particles provide a statistical estimate of the behaviour of all particles. In the sense that measurements of the behaviour of typical particles may sometimes be no more convenient than measuring the net effect of diffusion (i.e. the distribution of material concentration as a function of distance from a source) this advantage may be rather slight. A more significant gain follows if a satisfactory link is formed between the particle velocity (Lagrangian) statistics which determine the diffusion and the 'fixed-point' statistics which are conveniently attainable from the use of wind measuring instruments. Here the crucial feature has been shown to be the ratio of the integral time-scales characterising the velocity fluctuations in the two systems. As yet no completely satisfactory theoretical derivation of this ratio has appeared. The empirical evidence, though exhibiting the scatter characteristic of atmospheric turbulence data, nevertheless indicates some system in the variations of the ratio, to the extent of providing a useful working basis from which diffusion estimates may be made entirely in terms of measureable wind velocity fluctuations. In this method there is no limitation in scale or range except that imposed by departure from the assumed conditions of homogeneity and stationarity. Also the approach is potentially capable of providing the estimates of K which are required in exploiting the particular advantages of the diffusivity approach.

The similarity method was first used in the context of the expansion of clusters of particles. Here the essential argument is that over a certain range of cluster size the rate of growth must be controlled by eddies in the inertial sub-range and must therefore be determined by the rate of dissipation of turbulent kinetic energy. More recently the idea of 'Lagrangian' similarity has been developed, in the sense that the rate of vertical spread of particles starting at the ground is postulated to be dependent only on the aerodynamic drag of the ground and the turbulent heat flux. It is this treatment, for neutral flow (i.e. zero heat flux), which leads to the result quoted in the earlier discussion on the validity of the diffusivity concept. The great attraction of the treatment is that without any additional assumptions further dimensional analysis leads to

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the apparently successful prediction of the relative variation of concentration with distance from a continuous point source, valid over a range of some hundreds of kilometres for which the vertical spread remains within the height range of effectively constant stress (see Gifford 1962). There are however important shortcomings which remain to be resolved, one of which is that the variation of vertical spread with stability does not appear to be adequately predicted by the present form of the theory (Pasquill 1966). Another problem is that whereas the variation of concentration with distance from a point source is apparently correctly predicted, implying that irrespective of thermal stratification the effects of lateral spread are determined by those features which determine vertical spread, this implication has been shown not to be borne out by the variation of point-source concentration with stability (Klug 1967).

The growth of horizontal spread with distance of travel

The extrapolation of the continuing horizontal growth of a plume or cloud of particles beyond the distances at which it is directly measureable, or can be estimated with some confidence, is one of the most important issues facing those responsible for estimating the effects downwind of large individual sources. On the simplest basis both the diffusivity approach and statistical theory indicate that ultimately the growth of the horizontal dimensions should be proportional to the square-root of the distance. The fact that such a variation has not been demonstrated may at first sight be attributed to the effect, first conjectured by L.F. Richardson, that the effective diffusivity increases as the spread of the cloud increases, simply as a consequence of a progressive increase of the size-range of effective eddies. A closer look at the theoretical and observational evidence for this effect reveals some rather interesting features, especially as regards the widely accepted four-thirds power increase of diffusivity with the size of the diffusing cloud.

A variation of cloud spread (standard deviation σ) with time (T) in the form $\sigma \propto T^{3/2}$, and the implication that the effective K ($= \sigma^2/2T$) therefore depends on $\sigma^{4/3}$, follows from dimensional analysis when the effective range of eddies is assumed to be within the inertial sub-range. There is however considerable doubt about the validity of this argument on the scales up to 1000 km which were involved in Richardson's empirical demonstration of the law for K. The point may be

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examined further in terms of F.B. Smith's (1961) explicit evaluation of cluster growth in relation to the scale of turbulence. From this analysis (actually for three-dimensional spread in isotropic turbulence) it appears that accelerated spread ($d\sigma/dt$ increasing with σ) occurs only up to the stage when σ is roughly equal to the integral length scale l. At this stage ($d\sigma/dt$ constant) the corresponding K would be proportional to σ, and thereafter would increase less rapidly than σ. Observations of the variation of wind velocities on a synoptic scale give values of the integral time-scale of order 10 hrs, hence a corresponding length scale of the order of 400 km. On this basis the probability of a $K \propto \sigma^{4/3}$ law up to the scale covered by Richardson's analysis cannot be ruled out but the implication of such a law is by no means decisive.

It is interesting now to examine in more detail the data on balloon spread reported by Richardson and Proctor (1925), which in quality probably represent the best of the diffusion data used by Richardson in empirically deriving the law. In fact Richardson and Procter's analysis points to an even more rapid increase of K with σ i.e. $K \propto \sigma^{5/3}$. However, it should be emphasised that all the foregoing arguments about accelerated spread refer to the spread of particles travelling as a cluster, whereas it appears that the balloons in each group for which a spread was determined were actually released at intervals, over a total period which in many cases amounted to as much as 10 hours. This means that the results must contain a considerable element of continuous source behaviour, and in this respect $d\sigma/dT$ should decrease with T from the beginning. Finally, and probably most important, it is evident that most of the experiments provided estimates of σ at a single distance only. Direct comparisons of σ at two or more distances are provided in only 5 of the 19 experiments, and for the seven pairs of distances thereby involved only two show σ increasing more than in proportion to the distance.

In these circumstances the support thereby provided for the $\sigma^{4/3}$ law for K is very questionable and it seems likely that the original result is largely a consequence of bulking together the results of separate experiments showing a considerable scatter of σ at similar values of x. Since K is calculated as σ^2/T any scatter at given x or T will introduce a tendency towards $K \propto \sigma^2$.

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Thus, even if the true variation were, for example, $\sigma \propto T$, and hence $K \propto \sigma$, an ensemble of results such as those used by Richardson and Proctor will automatically lead to an exponent greater than unity in the K versus σ relation. In further support of this it may be noted that the long range diffusion data collected by Heffter (1965) have recently been shown by Machta to support $K \propto \sigma^{4/3}$ even though there does not appear to be any indication from individual experiments of a growth of σ more rapid than in proportion to time or distance.

Returning to the continuous-source aspect, for homogeneous turbulence with Lagrangian time-scale t_L the growth of spread is theoretically $\sigma \propto T$ for small T/t_L, tending asymptotically to $\sigma \propto T^{\frac{1}{2}}$ for large T/t_L. A fairly high intermediate value of the exponent of T, say $\geqslant 0.8$ may be expected to provide a reasonable average fit to the growth over the range $0<T<5t_L$. So if we take $t_L \simeq 10$ hr, as indicated by balloon and trajectory data on a large scale, the time limit for an exponent $\geqslant 0.8$ is roughly 50 hr and the corresponding distance limit about 2000 km. Thus in so far as the Richardson-Proctor data have a continuous-source element, an exponent in the range 0.8 to 1.0 might be expected. With data so scattered a decisive indication is not possible, but it is interesting that an exponent of 0.87 was obtained by Sutton (1932) from a re-analysis in which the values of σ were averaged in groups for specified ranges of distance.

So far we have considered only the direct effect of the horizontal component of turbulence, but there is also an indirect effect from the variation of mean velocity with height. The significance of this indirect effect was first pointed out by G.I. Taylor in connection with longitudinal dispersion of particles in pipe flow, and the idea was subsequently applied to atmospheric flow by Saffman (1962). In this case the analysis was carried out in terms of the 'simple diffusion model', but the essential features are brought out more clearly and completely in a 'statistical theory' treatment subsequently provided by F.B. Smith (1965).

Smith's analysis is concerned with the crosswind spread of particles released from a point in a field of turbulence, with mean wind speed constant with height but with a systematic (linear) variation of direction. In qualitative terms, one can see immediately that the particles passing through a given crosswind line, downwind of the point of release and at some specified level, will exhibit crosswind displacements which have two distinct contributions. The first

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will arise from the familiar effects of the crosswind component of turbulence, the second will be a consequence of the variations of horizontal velocity experienced by a particle as a result of variations in height along its trajectory. The full mathematical analysis leads to three terms for the crosswind spread at any level after time T:

$$\sigma_y^2 = \tfrac{1}{4}\sigma_w^2 u^2 \psi^2 T^4 \int_0^\infty F_L(n) \left(\frac{\text{Sin } r - r \text{ Cos } r}{r^2}\right)^2 dn \qquad \text{A}$$

$$+ \sigma_v^2 T^2 \int_0^\infty G_L(n) \left(\frac{\text{Sin } r}{r}\right)^2 dn \qquad \text{B}$$

$$- \rho^2 \sigma_v^2 T^2 \int_0^\infty F_L(n) \left(\frac{\text{Sin } r}{r}\right)^2 dn \qquad \text{C}$$

where $r = \pi n T$, ψ is rate of turning of wind in radians per unit height, σ_w, σ_v are the root-mean-square fluctuations of the vertical and crosswind components, $F_L(n)$, $G_L(n)$ are the corresponding normalized Lagrangian spectrum functions, and ρ is the correlation coefficient for the vertical and crosswind velocities, i.e. $\overline{w'v'}/\sigma_w \sigma_v$.

Term A, in which no effect of horizontal turbulence is involved, is the effect of the systematic wind shear. Essentially it represents the effect of the particle taking up instantaneously the mean horizontal velocity at each level. The weighting function applied to $F_L(n)$ is a narrow filter with a maximum value (0.19) at $\pi n T = 2$. For large T this filter transmits only very low frequencies, for which $F_L(n)$ is effectively constant and equal to $4t_L$. Accordingly, for large T, this term tends to $\frac{1}{6} u^2 \psi^2 \sigma_w^2 t_L T^3$.

Term B is the direct effect of the horizontal component of turbulence (no effect of wind shear involved), which for large T tends to $2\sigma_v^2 t_L T$.

Term C represents the effect of that part of the horizontal component which is correlated with the vertical component. It acts in opposition to term A, being a reflection of the extent to which the particle fails to adjust its horizontal velocity instantaneously to the mean velocity at its level. At large T its magnitude is $-2\rho\sigma_v^2 t_L T$ (with ρ normally < 1).

It is obvious that at sufficiently large T term A must become dominant. At small enough T on the other hand this term becomes very small and the resultant of terms B and C becomes dominant (except in the special case of $\rho = 1$)*. It is

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*and assuming the same t_L to apply to both F and G, which is unlikely.

especially interesting to note that the direct effect of the crosswind component is partially neutralized by the effect of correlation between w' and v'. The essential point is that some of the crosswind displacement due to the crosswind component occurs in conjunction with a systematic shearing of the plume of particles, upward moving particles tending to be displaced to one side and downward moving particles to the other.

As already mentioned these effects were first examined in terms of the simple diffusion model. The analytical solutions so obtained are valid only for large T, but they reveal certain additional features which should now be noted. Saffman's solutions actually give the alongwind spread resulting from change of wind speed with height. For diffusion in a bounded layer of depth h, and assuming K_z = constant and $u = 2U_z/h$ the result is

$$\sigma_x^2 = \frac{2\,U^2 h^2 T}{30\,K_z}$$

while for diffusion from a source on the ground with no upper boundary, assuming $u = \alpha z$

$$\sigma_x^2 = 0.036\alpha^2\,K_z T^3$$

A third solution for large T was obtained by F.B. Smith for completely unbounded diffusion, i.e. a source remote from the ground, for the case of wind direction turning with height. Transformed to the case of wind speed increasing with height, $u = \alpha z$, this becomes

$$\sigma_x^2 = \frac{1}{6}\alpha^2\,K_z T^3$$

noting that from statistical theory $K_z = \sigma_w^2 t_L$ at large T, and that α is equivalent to $u\phi$ in Smith's analysis, the last expression is identical with the large T limit of Smith's term A above.

According to the above results the effect on σ for a ground level source is less than half that for a source remote from the ground, for the same linear increase of wind with height. The result for diffusion over a finite depth (equivalent to the result originally obtained by G.I. Taylor for pipe flow) shows that the ultimate effect of shear increases much more slowly than when the diffusion is partially or completely unbounded in the vertical. Another

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interesting feature to note here is the entirely different effect of the vertical diffusivity according as the vertical diffusion is bounded or unbounded.

It should be emphasised that the results obtained so far from the simple diffusion model do not contain anything equivalent to Smith's term C. This is of course a direct consequence of the a priori neglected of correlation between w' and u'. In the present solution of the classical equation of diffusion this neglect is implied by writing

$$\overline{u'\chi'} = \overline{u'l} \frac{\partial\bar{\chi}}{\partial x} = K \frac{\partial\bar{\chi}}{\partial x}$$

(where $\bar{\chi}$, χ' are the mean and departure therefrom of material concentration). Strictly this should be generalized to represent the three-dimensional character of the mixing process, i.e.,

$$\overline{u'\chi'} = u'l_x \frac{\partial\bar{\chi}}{\partial x} + \overline{u'l_y} \frac{\partial\bar{\chi}}{\partial y} = + \overline{u'l_z} \frac{\partial\bar{\chi}}{\partial z}$$

and in the present context the important point is that the third term on the right-hand-side cannot be neglected if there is correlation between u' and w', for this would imply correlation between w' and l_z and hence between u' and l_z. An attempt to allow for this in extending Saffman's solution has in fact been made by Gee and Davies (1963), by making tractable assumptions about $\overline{u'l_z}$, with the result that the systematic shear term is reduced, qualitatively in keeping with the effect of Smith's term C.

Consider now the practical significance of the contribution of shear to the overall horizontal spread and its growth with distance. If the vertical spread is bounded both above and below the ultimate tendency is for the contribution from shear to increase with $T^{\frac{1}{2}}$. This is slower than we would expect from the direct action of the horizontal component of turbulence provided that the time scale of this turbulence is large enough, and it is certainly slower than the overall growth that is normally observed. If the vertical diffusion remains unbounded in at least one direction the tendency is for a variation with $T^{3/2}$ in the limit, and since the most rapid growth we can expect from the direct action of turbulence is with T to a power less than unity, this means ultimate dominance of the spread produced by shear.

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A rough assessment of the relative magnitudes of the two contributions may be made by using the expression for large T and neglecting the correlation effect. For wind turning with height at ψ radians/metre, the respective values of cross-wind dispersion for a ground level source, are as follows:

$$\sigma_s^2 = 0.036\, u^2 \psi^2 \sigma_w^2 \tau_w T^3$$

$$\sigma_T^2 = 2\, \sigma_v^2 \tau_v T$$

where τ_w, τ_v are the Lagrangian time-scales for the w and v components. The product $\sigma_w^2 \tau_w$ is unlikely to be as large as $\sigma_v^2 \tau_v$, and taking $\psi = 0.7 \times 10^{-3}$ radian/metre (i.e. a turning of 40° in 1000 m) and u = 10 m/sec (at 1000 m) this would mean that the contribution from turbulence (σ_T^2) would be greater than that from shear for 1000 sec of travel at least. Assigning to the speed of travel of the cloud a value of say one-half of the wind speed at the top of the layer, this corresponds to a distance of 5 km at least.

Using numerical solutions of the equation of diffusion (but still omitting the effect of correlation between the vertical and longitudinal components) Tyldesley and Wallington (1965) have calculated values of longitudinal spread in stable flow which appear to account for the whole of the spread observed at a distance of 5 km from a long crosswind line source. On the other hand, from an examination of data obtained by Högstrom (1964) on crosswind spread in stable conditions it can be argued that a turning of wind direction at the substantial rate of 14° per 100 metres would not dominate the spread from a ground-level source at distances up to 5 km. It is clear therefore that the precise relative significance of turbulence and shear remains to be evaluated. Also, even accepting the theoretical argument for ultimate dominance of the shear at long range, it has to be borne in mind that this conclusion requires that the vertical spread be maintained. In practice, in the very conditions which bring large shear the vertical spread is likely to be slowed up or even terminated by suppression of the vertical component of turbulence. On the other hand when diffusion over complete diurnal cycles is involved, with nocturnal suppression of vertical mixing, it is obvious that shear must exert an important influence. During the night the cloud of material will be sheared and distorted without necessarily

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affecting the spread at any given level, but with the onset of further vertical mixing this distortion will be converted to horizontal spread at all levels affected by the cloud.

Another feature in which the contribution of shear to horizontal (crosswind) spread may be of importance is the maximum concentration produced at ground level by an elevated source. Classical treatment, assuming a simple 'reflection' of the plume from the ground, with the basic crosswind and vertical distributions Gaussian in form, leads to the result that the maximum ground-level concentration is inversely proportioned to the square of the height of the source. This result assumes that the lateral and vertical spreads of the plume grow in the same proportion. However if their growth with distance is different, say

$$\sigma_y \propto x^m \text{ and } \sigma_z \propto x^n$$

then it follows (again assuming Gaussian distribution) that

$$x_{max} \propto 1/H^{1+m/n}$$

The tendency in stable conditions to maintain lateral spread more effectively than vertical spread, so leading to values of m/n greater than unity, will be augmented by the effect of shear. A value of 1.5 seems not unlikely, making the exponent on H 2.5. In stable conditions therefore, increase in source height is likely to have even greater benefit than is usually argued, partly as a result of the effect of shear.

Implications of the detailed turbulent structure of the planetary boundary layer

In relation to the various methods of analysing and estimating diffusion I would now like to consider some of the implications of the information currently available on the properties of turbulence over the depth of the planetary boundary layer. Of the various features which may be listed as important in this respect the following are probably uppermost - the Lagrangian/Eulerian time-scale ratio, which is one of the essential properties for prescribing diffusion and diffusivity in terms of 'fixed-point' measurements of turbulence - the variation with height and weather conditions of the intensity and scale of turbulence, essential in the same connection - the variation with height and weather conditions of the rate of dissipation of turbulent kinetic energy, a basic parameter in similarity predictions of the diffusion of clusters of particles.

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The Lagrangian/Eulerian scale ratio

In practice we are usually concerned with the ratio β of the integral time-scales evident in the turbulent velocity fluctuations exhibited by an ideal particle and by the measurements made at a fixed point. Without exception the various attempts to deduce a relation theoretically lead to a result of the form $\beta \propto 1/i$, where i is the intensity of turbulence (ratio of the r.m.s. velocity fluctuation to the mean velocity). The only approach which I propose to discuss here is that based on the properties of the spectra in the inertial sub-range of frequencies. This approach was first suggested by Corrsin (1963), and in essence the argument may be put as follows. It is assumed that both the 'fixed-point' and the Lagrangian spectra may be completely represented by the forms which are appropriate to the inertial sub-range. With subscripts E and L representing the 'fixed-point' and Lagrangian properties these forms are

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\begin{aligned}
S_E(n) &= C\,u^{2/3}\epsilon^{2/3}n^{-5/3} && n \geqslant n_E \\
&= 0 && n < n_E \\
S_L(n) &= B\epsilon\, n^{-2} && n \geqslant n_L \\
&= 0 && n < n_L
\end{aligned}
$$

where $\int_0^{\infty} S(n)dn = \sigma^2$ (the variance of the velocity component), n is frequency, u mean wind speed, ϵ rate of dissipation of turbulent kinetic energy per unit mass of air. C and B are universal constants to be specified. Integrating the two forms, eliminating ϵ and equating σ_E and σ_L, it follows that

$$n_E/n_L = \left(\frac{3}{2}\right)^{3/2} \frac{C}{B} \frac{u}{\sigma}$$

In the original argument upper limits of frequency are specified beyond which S(n) is effectively zero, but the terms involving these are ultimately neglected, and the present analysis follows an equivalent procedure by assuming the forms applicable to $n = \infty$. Regarding n_E and n_L as the inverse of characteristic time scales t_E and t_L and incorporating the definition of intensity of turbulence, $i = \sigma/u$,

$$\beta = {}^{t_L}/{t_E} = 1.84 \;\frac{C}{B}^{3/2} \;\frac{1}{i}$$

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This argument may be developed further, using forms for the complete spectra which are more realistic and which involve the integral time-scales as customarily defined. Probably the simplest extension of the above forms is

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\begin{aligned}
S_E(n) &= S_E(o) = 4\sigma_E^2 t_E & n<n_E \\
&= C_1 u^{2/3} \epsilon^{2/3} n^{-5/3} & n>n_E \\
S_L(n) &= S_L(o) = 4\sigma_L^2 t_L & n<n_L \\
&= B\, \epsilon n^{-2} & n>n_L
\end{aligned}
$$

the identity involving the time-scales being a general result for any stationary random function of time. By the same process as above this gives

$$\beta = 2.48 \frac{C}{B}^{3/2} \frac{1}{i}$$

An alternative approximation, which is certainly more represehtative of the observations of fixed point spectra, is

$$nS_E(n) = \sigma_E^2 N_E/(1 + \tfrac{3}{2} N_E)^{5/3}$$

$$nS_L(n) = \sigma_L^2 N_L/(1 + N_L)^2$$

in which $N = n/n_p$, n_p being the frequency at which $n\,S(n)$ is a maximum, and $1/4n_p$ is the integral time-scale. Equating the above forms to the inertial sub-range forms at large n, and then eliminating ϵ and equating σ_E and σ_L, we now find

$$\beta = 2.76 \frac{C}{B}^{3/2} \frac{1}{i}$$

Note that all the above approximation to the spectra give $B \propto \frac{C}{B}^{3/2} \frac{1}{i}$ and differ only in the numerical coefficient.

The magnitude of the universal constant C in the inertial sub-range form is now well established but no estimates have yet appeared for B. However, a direct estimate of $C^{3/2}/B$ would be provided by comparable measurements of the 'fixed-point' and Lagrangian spectra at appropriately high frequencies. The only example of such data at present available is due to Angell (1964) who has given a few cases of Lagrangian spectra at a height of about 750 m which are not inconsistent with the n^{-2} law over frequencies in the region of 1 c/sec. From the inertial sub-range laws

$$C^{3/2}/B = (nS_E(n))^{3/2}/unS_L(n)$$

and from average values of nS(n) over a frequency range 0.2 - 2c/sec in four

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separate runs the magnitudes of the above quantity were 0.15, 0.38, 0.019, 0.15. If we adopt the average of the three most consistent values, i.e. 0.23, the third of the foregoing expressions for β then reduce to

$$\beta = 0.6/i.$$

The experimental data on β show a good deal of scatter. The values provided by Angell's study, based on estimates of n_p in 24 comparisons of the 'fixed-point' and Lagrangian spectra, fall within the limits $0.15/i$ and $1.0/i$. Even greater scatter is shown in a more comprehensive plot (including Angell's results) recently prepared by Gifford (1967). Although it may be argued that certain aspects of diffusion are not sensitively dependent on the magnitude of β, the considerable scatter evident in the data is nevertheless rather disturbing and it would be desirable to uncover the reasons for it. It should be borne in mind that the time-scale t_E deduced from the fixed-point measurements themselves exhibits considerable scatter, which is presumably a reflection of the non-stationary, inhomogeneous and intermittent character of atmospheric turbulence and it does not seem reasonable to expect this scatter to be completely cancelled out in ratios of t_L/t_E. It is also interesting to refer back to the above implications of the forms of the spectra, and to note that the dependence of β on $1/i$ has emerged for spectral forms in which the common element is the $n^{-5/3}$ and n^{-2} laws, but in which widely different assumptions were made about the low-frequency parts of the spectra. If we experiment further in this matter by taking the same Lagrangian form but an arbitrary fixed-point form

$$S_E(n) = C^{1} u^{4-2r} \epsilon^{r-1} u^{-r} \qquad n \geq n_E$$

with $S(n) = S(o)$ for $n < n_E$, analysis on the same lines as before gives

$$1/\beta \propto i^{(4-2r)/(r-1)}$$

Now it is well known that the empirical magnitudes of the exponent r in the high-frequency spectra obtained in the atmosphere are rather varied. Values in the range 1.5 to 2 are quite common, and values outside this range are by no means unknown. The exponent on i in the relation above is obviously quite sensitive to such variations in r e.g.

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r =	1.5	1.67	2.0
$(4-2r)/(r-1)$ =	2	1	0

This argument can of course be criticised on the grounds that the 'fixed-point' and Lagrangian spectra are linked and that changes in the form of one are unlikely to occur without related changes in the form of the other. However it does suggest the possibility that the lack of a clear-cut dependence of β on $1/i$ could be associated with variability in the slope of the frequency spectra at high frequency, and it may be that further study of this connection would be rewarding.

The distribution and variation of turbulence in the planetary boundary layer

For the planetary boundary layer as a whole the details of the distribution of the relevant properties of turbulence - the intensity, scale and rate of dissipation - are not yet clearly defined, but there are several broad features which have emerged, particularly as far as the vertical component is concerned.

1. The familiar near-linear increase with height of the scale of the vertical component extends only to a height which depends on the thermal stratification. In stable conditions this height may be no more than a few metres, whereas in unstable conditions it may be several hundreds of metres. Thereafter the scale may change little with height, or, especially in stable conditions, it may even decrease quite sharply. In practice the evidence is usually in the form of estimates of the frequency n_p already referred to, which is inversely related to the time-scale t_E by a factor which depends on the shape of the spectrum. The considerable scatter evident in these parameters has already been mentioned.

2. Except when the wind is strong the r.m.s. vertical component (σ_w) tends to increase with height in unstable conditions and to decrease in stable conditions. At levels well above the ground (say >100 m) there is growing evidence that in strong winds the maximum magnitudes of σ_w are not greatly different from those in light winds, in contrast to the increase of σ_w with mean wind speed at lower levels. This implies that in strong winds there is a general tendency for a reduction of σ_w with height. A certain

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amount of confirmation is provided by a comparison of statistics recently obtained separately from instruments on a tower and on a captive balloon, but more direct and simultaneous comparisons would be desirable.

3. In the surface layer, when turbulence is produced mechanically, the rate of dissipation ϵ is expected to be proportional to u^3/z, and there is a fair amount of indirect observational evidence for such a variation. At greater heights, when the production or extraction of turbulent energy by buoyancy forces becomes more important this relation cannot be expected to hold. Wind speed is unlikely to be so important and the fall of ϵ with height is likely to be less rapid in unstable conditions and more rapid in stable conditions. As a result the general statistics on the variation of ϵ with height may be expected to show considerable scatter, and this is indeed the case. At 1000 m for example a recent Russian review (Zilitinkevic et al 1967) shows values of ϵ over a range 10^{-1} to 10^3 cm^2 sec^{-3}.

In the context of diffusion of windborne material the magnitude of σ_w is obviously directly important. For a continuous source it determines the initial rate of vertical spread of particles ($d\sigma_p/dt$). It also determines the significant rate of vertical spread of a cluster of particles or of a section of a plume. (According to F.B. Smith's treatment the maximum value of $d\sigma_p/dx$ is proportional to βi^2. On substituting the result given above for β ($\propto 0.6/i$) it follows that $d\sigma_p/dx$ is proportional to i and $d\sigma_p/dt$ to σ_w).

The relevance of the rate of dissipation ϵ may be seen in two ways. There is good observational evidence for $\epsilon \propto \sigma_w^3$ irrespective of stability conditions, which with Smith's result implies $d\sigma_p/dt \propto \epsilon^{1/3}$ for the significant stages of growth of a cluster. Then there is Batchelor's earlier prediction from similarity theory to the effect that when spread is dominated by the inertial subrange of turbulence $d\sigma_p/dt \propto \epsilon^{\frac{1}{2}}T^{\frac{1}{2}}$. So in a general way an estimate of the rate of spread of a puff or section of plume would be provided by observations of σ_w, or of the high-frequency contributions to σ_w, from which in turn estimates of ϵ may be attempted.

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The use of Smith's treatment or the similarity prediction is reasonable as long as the spread of material is not large compared with the scale of turbulence, and as long as there is no large change in the intensity and scale of turbulence over the vertical dimension occupied by the material. While these conditions may be satisfied in the early stages of spread of an elevated cluster or plume, they cannot be satisfied in the final and important stage of diffusion to ground levels. In this respect the most difficult case will be relatively stable conditions when both intensity and scale may be expected to be small and to have a complex variation with height. For such circumstances, in the lack of an extension of the statistical theory to turbulence which is not homogeneous in a vertical plane, the only practical recourse immediately available is the diffusivity approach. A combination of the two approaches would have merit as a reasonable compromise, the results of the first being accepted for σ_p below some fraction (say one-half) of the smallest scale of turbulence obtaining in the height range encompassed by σ_p, those of the second for σ_p above some multiple (say two) of the largest scale of turbulence so involved. Completion of the results for the intermediate range of σ_p could then be attempted by interpolation.

An experimental series of such calculations, using best estimates of the expected magnitudes of the turbulence and diffusivity characteristics would be enlightening. The degree of compatibility of the two approaches would be demonstrated and some impression would be obtained of the reliability of the interpolated results. In applying the scale criteria there is the further complication that the relevant scale of turbulence is that appropriate to variations along the vertical direction, whereas the data usually refer to variations along the direction of the mean wind (by implication from variations in time). It seems likely that the latter scale estimates will be overestimates of those actually required, in which case the criterion for acceptance of the diffusivity approach would be on the safe side.

In applying the diffusivity approach the outstanding requirement is that of expressing the diffusivity as a function of height and weather conditions, preferably in an explicit and independent way. With the accumulation of data

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on the statistical properties of turbulence a possible method for adoption outside the constant stress layer (within which estimates of K are already available) is the statistical theory result that $K = \sigma_w^2 t_L$. This is strictly valid only for homogeneous conditions but using it as a working assumption, and substituting the empirical relations

$$t_L = \beta t_E, \; \beta = 0.6/i, \; t_E = \frac{1}{2n_p} = \lambda_p/2u$$

the result is

$$K \simeq 0.3 \, \sigma_w \lambda_p$$

(where λ_p is the wavelength equivalent to the frequency at which n S(n) is a maximum). This expression for K is formally identical with that for K in the constant stress layer in neutral conditions, $K = ku_* z$, since $\sigma_w \propto u_*$ and $\lambda_p \propto z$. However, on substituting the latest estimates for the proportionality factors ($\sigma_w/u_* = 1.3$, $\lambda_p/z \simeq 3$) the constant stress layer K is actually equivalent to 0.1 $\sigma_w \lambda_p$. The reason for the lack of compatibility in the numerical coefficients is not evident, and this is an issue requiring elucidation if further progress is to be made on these lines. Of all the empirical factors involved above, that relating t_E and n_p is probably the most suspect. It is interesting to note that if the correlogram is of simple exponential form the quantity $1/n_p t_E$ is 2π, which would give a numerical coefficient 0.1 in the statistical theory result for K, identical with that in the 'constant stress layer' result. There is however a strong indication from spectra observed near the ground that $1/n_p t_E$ is substantially less than 2π. The value of 2 used above was thought to be the best estimate available. Clearly some further critical examination of these matters would be desirable.

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