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The Observed Rate of Tropospheric Diffusion

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THE OBSERVED RATE OF TROPOSPHERIC DIFFUSION

by

F. A. Gifford

ABSTRACT

Recent Australian plume-width measurements to 1000 km downwind from an isolated stack, together with existing relative diffusion data from a wide variety of tropospheric relative diffusion sources, are analyzed to determine the value of p in the equation for the cloud-width standard deviation, $\sigma_y \propto t^p$, where t is travel time downwind. The data suggest that there are three regimes of quasi-instantaneous cloud diffusion: 1) an irregular but, on the average, approximately linear growth region extending from 0- to 2-h travel time; 2) a regime of accelerating diffusion (i.e. $p > 1$) extending from 2 to about 15 h; and 3) an approach to a parabolic ($p \approx 1/2$) stage of diffusion at about 40 h.

I. INTRODUCTION

The problem of atmospheric diffusion to great horizontal distances from a source is an old one. Like many other significant aspects of atmospheric dynamics, it was first studied by Richardson (1926). More recently, meteorologists have concentrated on short-range diffusion, and the long-range problem has usually been dealt with theoretically as an asymptotic or degenerate case (e.g., Fick's Law). The most useful operational specification of long-range diffusion has been purely empirical (Heffter, 1965). The trouble seems to be that, although we now know much about diffusion at short (0- to ~10-km) distances, none of the theories that are reasonably successful in that range extrapolates very well to greater distances. Something else, besides the vigorous and highly variable boundary-layer turbulence that drives

the short-range atmospheric diffusion, seems to be going on. From a few observational clues that have been published, for instance the interesting study by Weber (1980) of the diffusion of a krypton-85 plume at 100 km, the net result is more rapid diffusion to intermediate distances by a factor of greater than twice what would be expected based on short-range diffusion rates. Short-range diffusion theories must in general be corrected (Pasquill, 1974) when applied at distances beyond about 5 km. The correction is often ascribed to the influence of wind-direction shear in the upper part of the planetary boundary layer (PBL); but this assigns a new name to the phenomenon without really describing its mechanics. What controls this shear, and how does it produce diffusion? How is it related to large-scale atmospheric dynamics? Are there other significant dispersive motions of the atmosphere that operate at mesoscales? Such questions can be answered only speculatively, based on the short-range orientation of most present thinking.

II. MECHANISMS OF LONG-RANGE DIFFUSION

Richardson and Proctor (1926) analyzed serial balloon releases and other evidence, such as spreading clouds of volcanic ash, and concluded that the (r.m.s.) horizontal spreading, σ_y , of particles in the atmosphere to distances of at least 100 km could be described by the astonishing formula $\sigma_y^2 \propto t^3$. Since the apparent or eddy viscosity, K , of the atmosphere is defined by $d\sigma_y^2/dt = 2K$, Richardson derived from this his famous law of diffusion.

$$K = c\varepsilon^{1/3}\sigma_y^{4/3}, \quad (1)$$

where ε is the eddy-energy transfer rate. It appeared, contrary to expectation (indeed to common sense, which suggests that, at its most rapid, diffusion should be along straight lines and thus $\sigma_y \propto t$), that the particle spreading rate actually increases with time. The bigger the cloud, the faster it spreads. Nowadays this result is understood in the context of Obukhov's (1941) equilibrium-range similarity theory, along with the $k^{-5/3}$ spectrum, to apply to a turbulent flow in which the scales of eddy-energy production and dissipation are widely separated. Turbulent-flow properties like K and $E(k)$, the energy spectrum, then depend only on the eddy-energy transfer rate in the so-called inertial range in between.

By providing this theoretical explanation of Richardson's law, Obukhov also seemed to be limiting the region of atmospheric scales to which it could apply. It is difficult to accept its application to diffusion at horizontal distances much greater than the scales of the large, daytime, convective PBL turbulence, say 5 or 10 km, the region of the spectral gap. The downscale turbulent-energy cascade process is necessarily a three-dimensional one and does not involve a mechanism that would extend PBL diffusion into the range of more nearly two-dimensional motions above PBL scales. How then is Richardson's result to be interpreted?

Pasquill (1974) pointed out that Richardson's balloon-spreading data were actually influenced by the presence of fairly large intervals between releases and, consequently, do not strictly represent instantaneous spreading. Gifford (1977) looked at tropospheric puff- and plume-spreading data from many sources, including the collections by Heffter (1965) and Hage and Church (1967) as well as more recent data, and could conclude only that the various individual data sets of which the composite curves of σ_y consisted were "not inconsistent" with accelerating diffusion at large distances. This problem of lack of definitive diffusion data at mesoscales continues to plague diffusion researchers, as evidenced by a comment recently received from Pasquill (personal communication, 1982). Examining the 10 individual relative-diffusion data groups from which Fig. 1 of Gifford (1977) is formed, Pasquill points out that accelerating diffusion is clearly indicated only at the small travel times (10-20 s) of Frenkiel and Katz's data and at the large scales (10-20 h) of Crawford's data. Other, intermediate data groups appear to follow slower, more nearly linear diffusion patterns which, in some cases, may have been obscured by source-size effects. And yet the accelerating-diffusion trend of the composited data of Fig. 1 appears to be quite definite. Two interpretations are possible.

(A) If purely diffusive spreading, i.e., that which separates particles on an instantaneous basis, is limited to the effects of PBL turbulence, say to horizontal distances of 5 km or so, then (instantaneous) diffusion at larger scales will be at a rate no greater than linear ($\sigma_y \propto t^p$, $1/2 \lesssim p \lesssim 1$). Large values of p , indicating accelerating diffusion, can occur at these larger scales only in connection with the effect of time averaging, as a result of puff or plume displacements by large-scale motions that do not themselves diffuse the instantaneous cloud. Estimation, and numerical modeling, of diffusion at large distances reduces, under alternative (A), to the superposition of large-scale displacements on a short-range formulation of σ_y that depends strictly on PBL turbulent diffusion rates. F. B. Smith (1983),

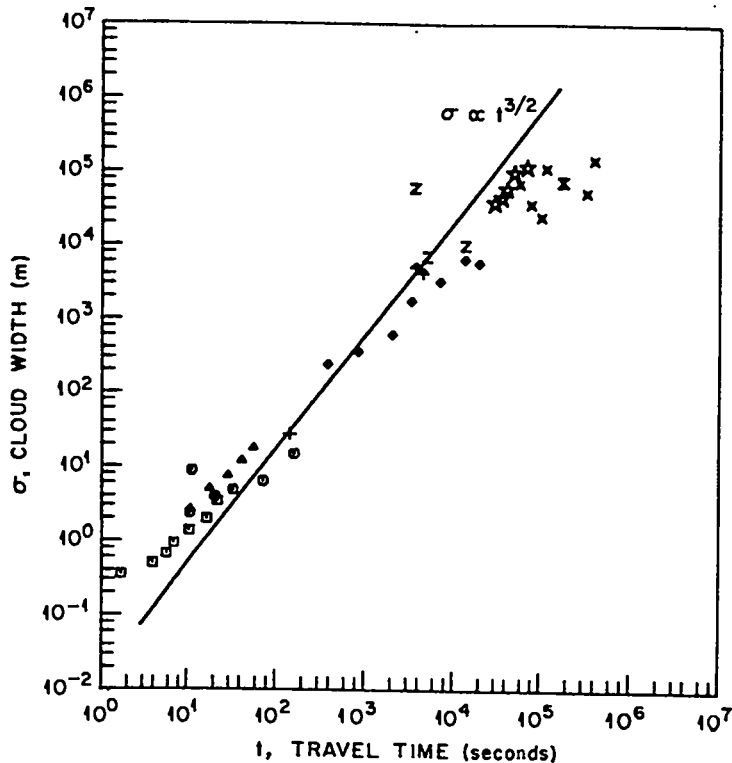


Fig. 1.

Observations of widths (horizontal standard deviation) of diffusing tracer clouds in the troposphere as a function of downwind travel time; for references to individual experiments and further details, see Gifford (1977).

commenting on the random-force model of diffusion proposed by Gifford (1982), seems to favor this hypothesis. In a similar vein, a reviewer of the National Science Foundation travel-grant application that led to the present study cautioned the writer not to attempt to apply the accelerating diffusion model beyond PBL scales. Most present-day operational models of long-range plume spreading are also based on this concept.

(B) But what if, on the contrary, downscale energy transfer from the small-scale end of the synoptic range of atmospheric motions does occur? Well-established global kinetic-energy production and dissipation rates suggest that, in some form, it must. Kinetic energy, produced at synoptic scales as a result of baroclinic instability, flows partly toward the general circulation and partly toward higher wave numbers. This synoptic-scale production rate must on the average just be balanced by an equal turbulent kinetic-energy dissipation at molecular scales of motion, so as to maintain the prevailing equilibrium state of the atmosphere. It is well established that in the range of two-dimensional, quasi-geostrophic motions extending from synoptic scales to smaller scales, up to about $k=20 \div$ earth's radius, a kinetic-energy cascade

cannot occur (Charney, 1971). In the broad spectral region between $kr \approx 20$ and the mesoscale minimum at $kr \approx 2000$, atmospheric motions are at best poorly understood, due mainly to the insufficient density of conventional data gathering networks; and this is just the range that is involved in the long-range diffusion problem. A classical, Richardson (i.e., three-dimensional) cascade of kinetic energy toward smaller scales would produce a $k^{-5/3}$ region of the spectrum there, as would the reverse, strictly two-dimensional cascade proposed by Gage (1979); or, which is more likely, elements of both processes may occur.

Present ignorance about motions in the range from $kr \sim 20$ to PBL scales does not imply the absence of diffusive effects in that range. Many atmospheric phenomena known to occur at these scales could conceivably diffuse particles. The first detailed energy spectra for the mesoscale region have just recently been obtained by Larsen et al. (1982). These (frequency) spectra behave generally as (frequency) $^{-5/3}$ between 2 and 50 h, but they also contain many fine-scale details, suggesting the presence of a number of active scales of turbulent motions. In this situation, the nature of (instantaneous) puff or plume spreading should be noticeably different from that of hypothesis (A). Examination of plume widths to distances beyond 5 or 10 km should show instantaneous spreading rates such that p reaches the value $3/2$, and these accelerating-diffusion rates may occur to considerable distances, on the order of hundreds of kilometers. As a result, numerical diffusion models will have to be handled differently to account for this enhanced diffusion and provide the correct spreading rates and plume shapes.

III. THE EVIDENCE OF RECENT AUSTRALIAN PLUME MEASUREMENTS

Fortunately for the progress of the discussion, a series of plume-width measurements over a wide range of downwind distances has been described recently by Carras and Williams (1981). These authors report plume widths measured during the (Southern Hemisphere) winters of 1977 and 1979, using airborne particle and gas recorders, to distances of up to 1000 km and travel times of up to 43 h from the Mt. Isa, Queensland (Australia), smelter. The extraordinary plume-travel times and distances achieved by the Commonwealth Scientific and Industrial Research Organization (CSIRO) group in these measurements extend present empirical data on plume spreading essentially by an order of magnitude in distance. This was possible because of the virtual absence of background concentrations and the comparative regularity of the winter, easterly wind-flow patterns in that region. The Mt. Isa data include

plume widths measured between travel times of 0.9 h and 43 h. These are plotted in Fig. 2 as $\sigma_y(t)$, along with a set of near-field plume widths obtained by the CSIRO scientists, using the same measurement techniques and equipment, at the Kalgoorlie smelter in Western Australia during 1980. Observed plume widths, W , have been converted to σ_y by assuming that $\sigma_y = W/4.28$.

The first point to be noticed is that the data are highly correlated; the linear correlation coefficient for these 101 data points equals 0.97. A third degree polynomial regression on the logarithms of $\sigma_y(\text{km})$ and $t(\text{h})$ provides the following interpolation formula:

$$\ln \sigma_y = 0.34 + 0.995 \ln t + 0.33 (\ln t)^2 - 0.009 (\ln t)^3. \quad (2)$$

Equation (2) illustrates why Heffter's (1965) straightforward proposal, $\sigma_y(\text{m}) = t(\text{s})/2$, has proved so durable. In present units this becomes $\sigma_y(\text{km}) = 1.8t(\text{h})$. If the small, higher order terms in Eq. (2) are ignored, the result is the formula $\sigma_y = 1.4t^{0.995}$, which differs from Heffter's only by the

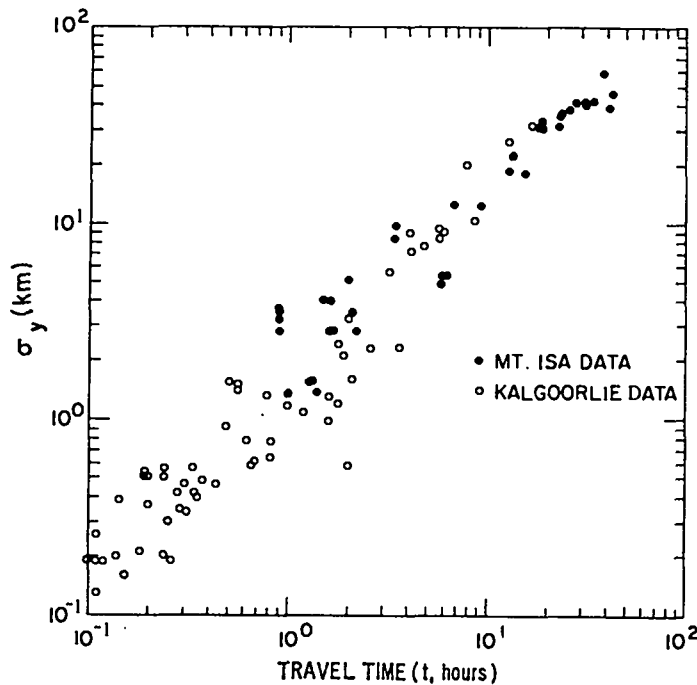


Fig. 2.

Values of horizontal plume standard deviations, $\sigma_y(\text{km})$ for the Mt. Isa (Carras and Williams, 1981) and Kalgoorlie smelter plumes as a function of downwind travel time, t , hours. Slopes of 1, 3/2, and 1/2 are indicated.

ratio of the coefficients, a factor of approximately 1.3. It follows that if a range of accelerating diffusion exists with powers of t greater than $p = 1$, it is embedded in a strong linear trend of the data.

The diffusion rates of these same data are shown in more detail in Figs. 3 and 4, which were prepared in the following way. The (101) data points of Fig. 1 were ordered in time and divided into contiguous subsets of 15 points each. This 15-point interval was chosen, after a certain amount of experimentation with smaller and larger data segments, to provide a reasonable amount of smoothing without destroying too much of the time resolution needed to study the variation of p . Then a standard linear regression analysis was performed on the logarithms of σ_y and t for each subset of 15 points, starting with point 1; and this process was repeated starting at points 4, 7, 10, and 13 in a kind of moving-average procedure. Figure 3 is a plot of averaged values of σ_y for each of the resulting 35 data segments. Figure 4 is a plot of the power-law exponent, p , calculated by the regression analysis as the coefficient of $\ln t$ for the same 35 data segments.

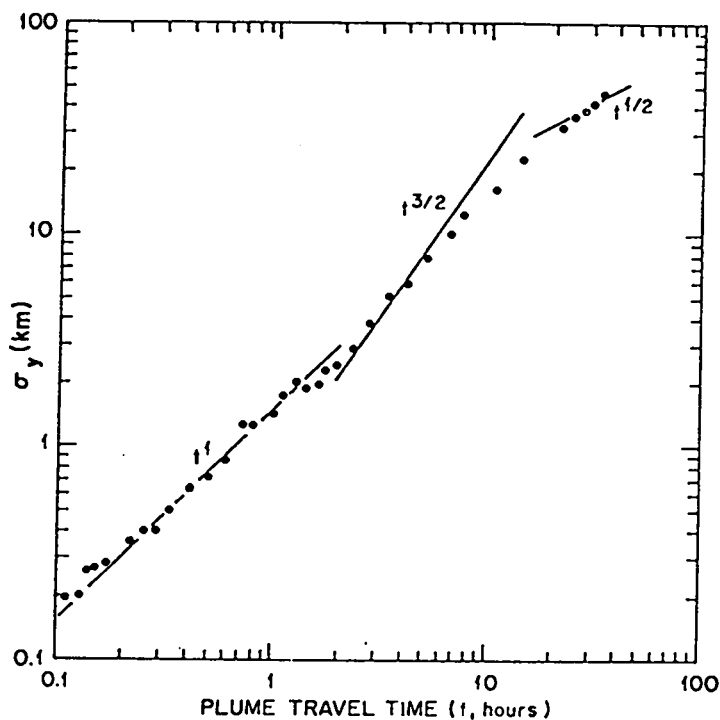


Fig. 3.

Plot of averaged values of σ_y (km) determined by least squares fits to the equation, $\ln \sigma_y = b + p \ln t$, for the data of Fig. 2; the averaged value in each segment is indicated. (See text.)

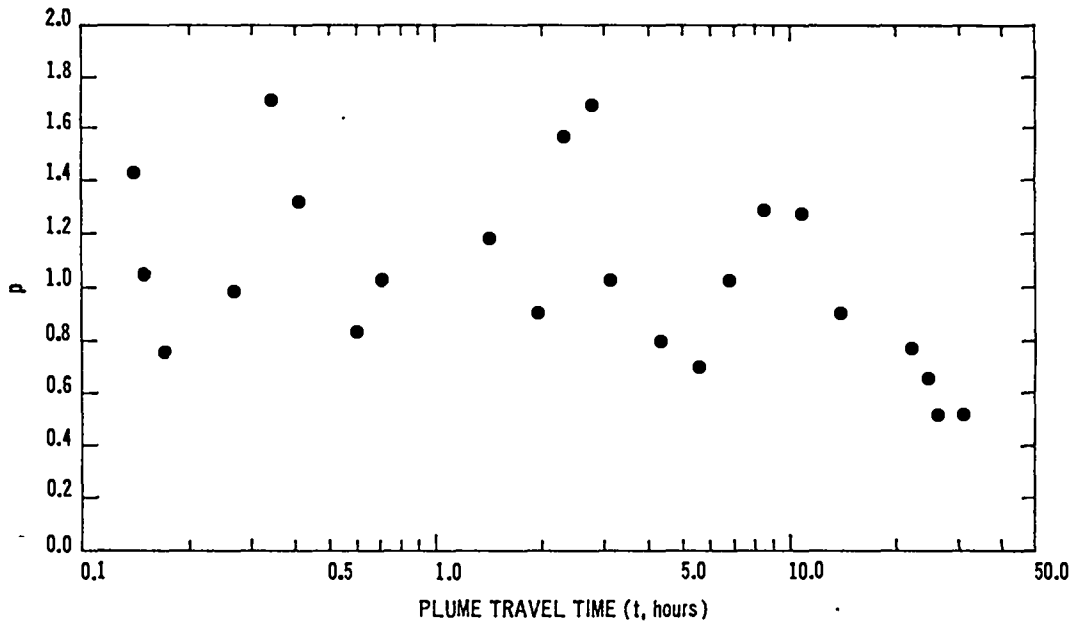


Fig. 4.

Cloud diffusion-rate power-law exponent, p , vs travel time, t , hours, for the same data segments as in Fig. 3.

The generally smooth increase of the averaged σ_y -values with t exhibited in Fig. 3 indicates that a 15-point average is adequate to define that variable. The same can not be said however for the corresponding values of p . Some calculated p -values were of doubtful significance due to the scattered nature of the data in their segments which, in a few cases, yielded very large positive or negative values of p or values corresponding to near-zero correlations between $\ln \sigma_y$ and $\ln t$. These segments appeared to differ qualitatively from the others in that their far greater scatter resulted in physically unreasonable values of p . To test the possibility that such segments should be excluded from the group used to determine the behavior of p as a function of t , the quotient of the variance attributable to $\ln t$ and the residual variance, the F -statistic, was calculated for each data segment. When F is large, the regression has accounted for a large part of the variation of the σ_y with $\ln t$; for small values, the residual variance is large and the data are very scattered. A critical value of the F -distribution can be identified such that segments with smaller values have variance properties that differ from those with higher F 's to any desired level of significance. At the 5% significance level, the critical value for these data segments is close to $F = 3$. This means that there is little possibility (< 5%) that segments having smaller F -values will have been identified incorrectly as having variances

qualitatively different from those of the main group. Segments with $F < 3$ were accordingly omitted from Fig. 4, which has 10 fewer points as a result.

IV. THE EVIDENCE OF EARLIER RELATIVE DIFFUSION EXPERIMENTS *

Some of the relative diffusion data sets discussed by Gifford (1977), augmented by several more recent data sets, can be analyzed in a similar way to determine values of p for individual experiments or, in some cases, for subsets of data from an experiment. The results are summarized in Table I and Fig. 5. Relevant new measurements are: those by Gifford (1980) who analyzed Randerson's (1977) Skylab photograph of a Gulf of Mexico smoke plume; the radar measurements by Moninger and Kropfli (1982) of a "chaff" plume; and Nappo's (1981) photographic analysis of a smoke plume at Idaho Falls. Several of the earlier sets consisted of too few data points and so have been omitted from this compilation. These include the data of Crozier and Seely (1955), Braham, Seely and Crozier (1952), Edinger (1955), and Roberts (1923). For the remainder of these data, Table I lists for each group of points used to calculate a p -value: the average diffusion time, t , hours, and the range of times over which p was determined; the averaged value of the spreading rate, p ; the linear correlation coefficient, r , and the number, n , of data points used; and the source reference of the data. These values of p are plotted as a function of t in Fig. 5. The data sources, sample sizes, and experimental methodologies are much too diverse to support a variance analysis like that carried out for the previous data set; and the scatter of the p -values in Fig. 5 is accordingly quite large. An attempt has been made in Fig. 5 to account for at least some of the large scatter of the p -values, compared with those of Fig. 4, at diffusion times less than a few hours by indicating whether the source involved a quasi-point release or a release having an effectively large initial volume. There is some indication of the expected effect; i.e., generally larger p -values occur near the source for the near-point releases.

*Analysis of the historical relative diffusion data, summarized in Section IV, was done under contract with Sandia National Laboratories for the U.S. Nuclear Regulatory Commission, Division of Risk Analysis.

TABLE I
INSTANTANEOUS DIFFUSION RATES OF TROPOSPHERIC PUFFS AND PLUMES

<u>t, h (range, h)</u>	<u>p</u>	<u>r</u>	<u>n</u>	<u>Source Type^a</u>	<u>Reference (data points used)</u>
0.9 (.24-5.5)	0.9	.98	7	1	Pack and Angell (1963)
11.1 (7.7-17.9)	1.5	.96	5	1	Crawford (1966)
0.13 (.06-.31)	1.2	1.0	11	2	Byzova, et al. (1970)(last 11)
0.42 (.12-.97)	0.7	.94	7	1	Kao and Wendell (1968)
0.14 (.03-.42)	1.0	1.0	6	1	Hanna (1975) (first 6)
1.0 (.63-1.5)	1.0	.95	5	1	" (last 5)
4.0 (.46-12.0)	0.8	.90	6	2	Peterson (1968) (outbound)
4.9 (3.3-6.8)	2.0	.95	6	2	" (inbound)
0.43 (1.0-9.5)	0.8	1.0	9	1	Angell, et al. (1971)(first 9)
16.6 (10.5-25.1)	0.9	1.0	9	1	" " (last 9)
11.8 (7.6-18.6)	1.0	1.0	9	1	" " (middle 9)
0.8 (.19-1.7)	1.4	.85	9	2	Gifford (1980) (first 9)
3.1 (1.9-4.8)	2.2	.84	10	2	" (middle 10)
4.4 (1.9-8.3)	1.0	.71	19	1	" (last 19)
0.43 (.15-1.0)	0.7	.90	6	1	Randerson (1972) (first 6)
4.5 (2.1-8.0)	1.1	1.0	6	1	" (second 6)
14.5 (9.0-23.6)	0.7	0.90	6	1	" (third 6)
52.4 (27.1-83.9)	0.6	.95	16	1	" (last 16)
0.14 (.03-.25)	1.5	>0.90	50	2	Nappo (1981)
0.9 (.24-1.69)	0.7	1.0	22	1	Moninger and Kropfli (1982)(first 22)
2.1 (1.77-2.6)	1.2	0.90	12	1	" " (next 12)
2.8 (2.63-3.0)	2.8	0.90	6	1	" " (last 6)

^aSources of type 1 had large initial volumes or separations; sources of type 2 approximated point releases.

V. CONCLUSIONS

The diffusion rates shown in Figs. 3-5 support the existence of three broad regions of instantaneous puff and plume diffusion: (1) a roughly linear region extending from 0.1 h to 1 or 2 h; (2) a region of accelerating diffusion, possibly with two separate maxima, extending from 2 h to about 15 h; and (3) a region of decreasing diffusion rates, with slopes approaching

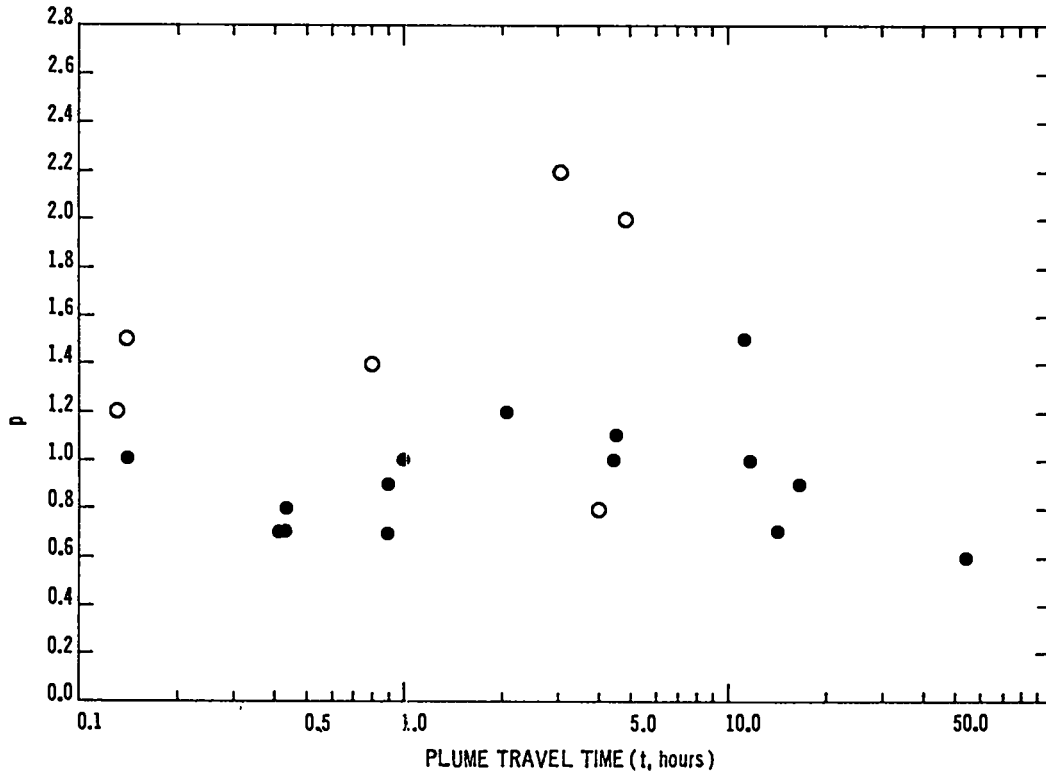


Fig. 5.

Cloud diffusion-rate power-law exponent, p , vs travel time, t , hours, for Table I data. Open circles approximated point releases; the remaining points involved large initial volumes or separations.

$p \approx 1/2$, beyond that point. In very general terms, these regions can be interpreted in the following way. In region (1) the instantaneous cloud spreading is dominated by various source-size and buoyant plume-rise effects, the latter involving entrainment of ambient air into a rising plume at a rate governed by the small-scale turbulence generated in the shear zone at its edges. Larger scale, convective PBL turbulence, which was quite active for much of these data, is important over most of this time range primarily in displacing the cloud. This affects time-averaged, but not instantaneous, cloud widths. In region (2) the dispersion is controlled by purely atmospheric processes and characterized by the presence of accelerating diffusion, possibly in two ranges whose peaks are at about 3 h and 10 h. Whether these maxima reflect distinct and possibly different diffusion and energy-transfer processes is unknown. It is tempting to associate them with spectral features and, in fact, the meridional component of the spectra by Larsen et al. (1982) exhibit (among several others) maxima at, roughly, 6 h and 20 h. But whatever the ultimate interpretation of such interesting details of diffusion rates may be, there appears to be no doubt about the existence of accelerating diffusion

between 2 h and 15 h, i.e., between the outer limit of the scale of typical PBL turbulence and a scale approaching that of the high wave-number end of the synoptic region.

Clearly, much more turbulence and diffusion information should be gathered at mesoscales in many places and meteorological situations. But these observations of instantaneous puff- and plume-spreading rates seem to support the second hypothesis, alternative (B). Because the general spreading behavior of the comparatively homogeneous Mt. Isa-Kalgoorlie plume-data set, Fig. 2, as well as the detailed spreading rates, Fig. 4, are similar to those of earlier, composite relative-diffusion plots, Figs. 1 and 5, it seems that the hypothesis of downscale energy transfer in some form between a scale of about 2000 km and the PBL scale should be taken fairly seriously now. Many, virtually all details of mechanisms remain to be clarified, particularly the role of PBL wind-direction shear in the diffusion process. This must involve the diurnal cycle of PBL stability changes, as Pasquill (1974) has pointed out. The average magnitude of PBL wind-direction shear is governed by the global requirement of a net, poleward meridional heat flux (Golitsyn, 1973; Monin, 1972), so at least in this very general way the largest scale of atmospheric motions is coupled to small-scale PBL diffusion processes. An interesting question raised by these data is whether the transition to a parabolic ($p = 1/2$) stage of tropospheric diffusion has occurred by the most distant of the Mt. Isa points, at 1000 km. The Australian group plans to attempt further observations of the width of the Mt. Isa plume, to distances of 2000 km or more. A successful outcome could resolve this particular question and add significantly to our present understanding of both long-range diffusion and mesoscale dynamics.

VI. ACKNOWLEDGMENTS

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