Study on Horizontal Relative Diffusion in the Troposphere and Lower Stratosphere①

Zheng Yi (郑 毅)

State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029

(Received August 18, 1998; revised September 9, 1999)

ABSTRACT

The behaviour of relative diffusion theory and Gifford's random-force theory for long-range atmospheric diffusion is examined. When a puff scale is smaller than the Lagrangian length scale, $\sqrt{2KT_L}$, an accelerative relative diffusion region exists, i.e., $\sigma_y \propto t^{3/2}$. While the puff diffusion enters a two-dimensional turbulence region, in which the diffusion scale is larger than 500 km, or time scale is larger than 1 day, divergence and convergence are main cause of horizontal diffusion. Between the two above-mentioned regimes, diffusion deviation is given by $\sigma_y = \sqrt{2KT}$. The large-scale horizontal relative diffusion parameters were obtained by analyzing the data of radioactive cloud width collected in air nuclear tests.

Key words: Tropospheric and lower stratospheric diffusion, Relative diffusion, Large scale turbulence, Nuclear explosion clouds

1. Introduction

Relative diffusion problem, or instantaneous puff diffusion problem, was studied in the 1920's by Richardson (see Pasquill and Smith, 1983) who proposed a famous law that the diffusion parameter is proportional to diffusion scale to the power of 4 / 3. The property of relative diffusion is that the diffusion parameter is associated with the scale of the puff. For the diffusion of a continuous source, all scales of turbulence act upon the plume, while for the diffusion of an instantaneous source only the turbulence near the scale of the puff is important. When the diffusion time is large enough, puff diffusion changes to plume diffusion. From the viewpoint of Lagrangian reference, a continuous source is equivalent to a series of instantaneous sources. Therefore, it is more essential to study relative diffusion than continuous diffusion.

Nuclear explosion clouds in a test in the air are formed in a very short time in comparison to their diffusion time. They are considered to be instantaneous sources that can be studied by the theory of relative diffusion. On the contrary, air nuclear tests can be considered as atmospheric field studies in which nuclear radioactive debris is tracers. The diffusion data can be used to validate the relative diffusion theory. Randerson (1972) and Walton (1973) analyzed a cloud dataset in an air nuclear test in the US, but the agreement with the exact diffusion rules was not satisfactory due to the short of data.

The aim of the paper is to derive a rule for the horizontal relative diffusion in the

①This work is sponsored by the National Natural Science Foundation of China under Grant No. 49505064.

troposphere and the lower stratosphere by analyzing the nuclear clouds data of an air explosion test. The theoretical frame under which the present study is carried out includes the stationary and homogeneous turbulent relative diffusion theory and Gifford's random–force theory of long–range relative diffusion.

2. Relative diffusion theory in stationary and homogeneous turbulence

Batchelor (1950; 1952) established a theoretical basis of relative diffusion in homogeneous turbulence. His main idea was to use two particles to demonstrate the separation of clusters. He used the dimensional argument method and the theory of small–scale structure of turbulence to derive the following formulae:

$$\overline{y^2} = y_0^2 + c_1 T^2 (\varepsilon y_0)^{2/3} \qquad \text{(for a small } T\text{)}, \tag{1}$$

$$\overline{y^2} = c_2 \varepsilon T^3 \qquad \text{(for an intermediate } T\text{)}, \tag{2}$$

$$\overline{y^2} = 2\overline{Y^2} \qquad \text{(for a large } T\text{)}, \tag{3}$$

where y is the separation of the pair of particles at time T after initial separation y_0, c_1 and c_2 are two constants with the order of magnitude of unity, ε is the turbulent dissipation rate, $\overline{Y^2}$ is the mean square displacement of particles released continuously from a fixed position (a property of dispersion of a single particle). At very small values of T, the velocities of the two particles have not had time to change appreciably. Equation (2) brings out accelerative nature of the relative diffusion, which occurs as long as the separations involved are small compared with the scale of turbulence. This is associated with the $-5/3$ law of the atmospheric turbulent kinetic energy (TKE) spectrum. As T is large enough, the separation of two particles is so large that their velocities are not correlative. The mean square separation of a pair of particles approaches $\overline{Y^2}$, a parameter associated with dispersion of a single particle. Thus, Taylor's diffusion theory of continuous point source can be used to present instantaneous ones.

Gifford (1982a; 1984) used the Lagrangian movement equation which is the same as the Langevin's equation of Brownian motion. He concluded that the 'random–force' theory could be used in the meso– and large–scale relative diffusion. The Langevin's equation is written as:

$$\frac{dv}{dt} + Bv = a(t), \tag{4}$$

where v is the horizontal velocity component of the particle, B^{-1} is the integral or outer time scale of the turbulence process, and $a(t)$ is a random acceleration that is assumed to have a flat spectrum and zero mean. Integration of (4) under the condition of stationary and homogeneous turbulence yields:

$$\sigma_y^2 = \sigma_0^2 + 2KT_L\left[\frac{T}{T_L} - (1 - e^{-T/T_L}) - \frac{c}{2}(1 - e^{-T/T_L})^2\right], \tag{5}$$

where σ_y^2 is the standard deviation of particle displacement, $T_L (= 1/B)$, is the Lagrangian time scale of large scale turbulence, K is the effective atmospheric eddy diffusivity, T is the lapse of time since release, $c = 1 - v_0^2 / \overline{v^2}$, v_0 is the initial speed of particles, and $\overline{v^2}$ is the standard deviation of velocities. The Lagrangian time scale of the atmospheric turbulence T_L is equal to $1/f_z$ ($\approx 10^4$s in the mid–latitudes), in which f_z is the vertical component of the

Coriolis parameter, as Gifford concluded that the characteristic time scale of large–scale atmospheric diffusion is dominated by the rotation of the earth. Gifford (1982b; 1984) and Barr and Gifford (1987) compared some data of tropospheric relative diffusion with Eq. (5) and found the comparison quite satisfactory, but K and T_L can only be obtained by fitting data to Eq. (5).

For large values of T, Eq. (5) becomes

$$\sigma_y^2 = 2KT, \tag{6}$$

and this is coincident with Taylor's results. It shows that the correlation between the two particles is small. For small values of T, Eq. (5) becomes

$$\sigma_y^2 = \sigma_0^2 + v_0^2 T^2. \tag{7}$$

For intermediate values of T, Equation (5) becomes

$$\sigma_y^2 = \frac{2}{3}\frac{\overline{v^2}}{T_L}T^3, \tag{8}$$

which is consistence with Batchelor's results. Batchelor's similarity theory and Gifford's random–force theory have different concepts, but their symbols share the same physical meanings. We thus have $\sigma_0^2 = y_0^2$, $v_0^2 = (\varepsilon y_0)^{2/3}$. Comparing (7) and (8) with (1) and (2), respectively, we obtain

$$\varepsilon = \frac{2}{3}\frac{\overline{v^2}}{T_L}, \tag{9}$$

and

$$c = 1 - c_3\left(\frac{\sigma_0}{L}\right)^{2/3}, \tag{10}$$

where $c_3 = 2/3^{2/3} \approx 1$, $L = \sqrt{2KT_L}$ is the Langrangian length scale of large–scale turbulence, σ_0 is the initial standard deviation of particle displacement. As $\sigma_0 \approx L$, $c \approx 0$, Eq. (5) becomes the Taylor's equation. As $\sigma << L$, $c \approx 1$, Eq. (8) can be derived. This indicates that only when the initial size of the source is far less than L, the puff has accelerative diffusion nature; otherwise, accelerative diffusion does not occur, i.e. instantaneous source diffusion becomes continuous source diffusion. For typical values of $K \approx 5 \times 10^4\ \mathrm{m}^2/\mathrm{s}$, $T_L \approx 10^4\ \mathrm{s}$, and $L \approx 30$ km, Gifford (1984) proposed that the length L characterizes the smallest size range of large–scale, quasi–horizontal motions of the atmosphere that is capable of maintaining the energy–transfer rate to the small, 3–D PBL eddies, given the large–scale atmospheric "viscosity", K.

3. Relative diffusion theory on two–dimensional turbulence

Generally speaking, large–scale atmospheric motions are two–dimensional or quasi two–dimensional. Kraichnan (1967) proposed that there is an inertial region of two–dimensional turbulence in which the TKE spectrum is $E(k) \propto \eta^{2/3} k^{-3}$ and the cascade rate of mean–square vorticity, η, is constant. Charney (1971) provided similar results according to the geostrophic turbulence theory. Kao et al. (1968) calculated 200, 500 and 800 hPa relative velocities spectrum and found that the high frequency portion of the power spectra of both zonal and meridional components of the relative velocities was proportional to k^{-3}. Lilly (1989) proposed that the energy spectrum function exhibits an approximately −3 slope at low wavenumbers and a −5 / 3 slope at high wavenumbers. Lin (1972) deduced that the diffusion parameter is exponential extension with time according to the two−dimensional homogeneous turbulence theory. He derived the following result:

$$\overline{y^2} = \overline{y_0^2} \cdot e^{2c\eta^{1/3}T}, \tag{11}$$

where c is a positive constant with the order of unity, and η is the cascade rate of enstrophy (half−squared vorticity). The specialities of two−dimensional homogenous turbulence are energy spectrum that obeys the k^{-3} law. In addition, enstrophy cascade rate is constant and it easily appears as downscale transport, viz a negative η (Lilly, 1989). The quantity $\overline{y^2}$ changes less with time in this situation. From energy spectrum of Lilly (1989), there is a turning point from low wavelength −5 / 3 law to high wavelength −3 law. In other words, the atmospheric motion changes from three−dimensional to two−dimensional or quasi−two−dimensional as a puff extends to a large area. The traditional diffusion theory of small−scale turbulence is invalid in the two−dimensional regime. Consequently, a puff cannot increase with $t^{1/2}$ unlimitedly. On the contrary, the increase is sometimes negative, and this will be validated at following paragraphs.

During the long−range transport and diffusion, the main effects of wind fields are transportation, distortion and circumrotation (some scientists use the terms of dilution, deformation or shredding etc.). These processes cannot be explained by classical homogeneous diffusion theories. The author speculates that the horizontal change of the puff is mainly associated with the divergence of the wind fields. The rate of change of horizontal area for an air parcel is equal to the horizontal divergence and convergence (Wallace and Hobbs 1977). This can be written as:

$$\frac{1}{A}\frac{dA}{dt} = \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = div\vec{V}_H, \tag{12}$$

where A is the horizontal area of the air parcel, and u and v are the x− and y−component of velocity. By setting $A = \overline{A} + A'$, $u = \overline{u} + u'$ and $v = \overline{v} + v'$, (12) becomes:

$$\frac{f(\overline{A} + A'}{dt} = (\frac{\partial(\overline{u} + u')}{\partial x} + \frac{\partial(\overline{v} + v')}{\partial y})(\overline{A} + A'). \tag{13}$$

After taking ensemble averaging of (13), we have

$$\frac{\partial \overline{A}}{\partial t} + \overline{u}\frac{\partial \overline{A}}{\partial x} + \overline{v}\frac{\partial \overline{A}}{\partial y} + \overline{w}\frac{\partial \overline{A}}{\partial z} + \overline{u'\frac{\partial A'}{\partial x}} + \overline{v'\frac{\partial A'}{\partial y}} + \overline{w'\frac{\partial A'}{\partial z}}$$
$$= (\frac{\partial \overline{u}}{\partial x} + \frac{\partial \overline{v}}{\partial y})\overline{A} + \overline{A'\frac{\partial u'}{\partial x}} + \overline{A'\frac{\partial v'}{\partial y}}. \tag{14}$$

Simplification of (14) yields:

$$\frac{d\overline{A}}{dt} - \beta \cdot \overline{A} = \xi(t), \tag{15}$$

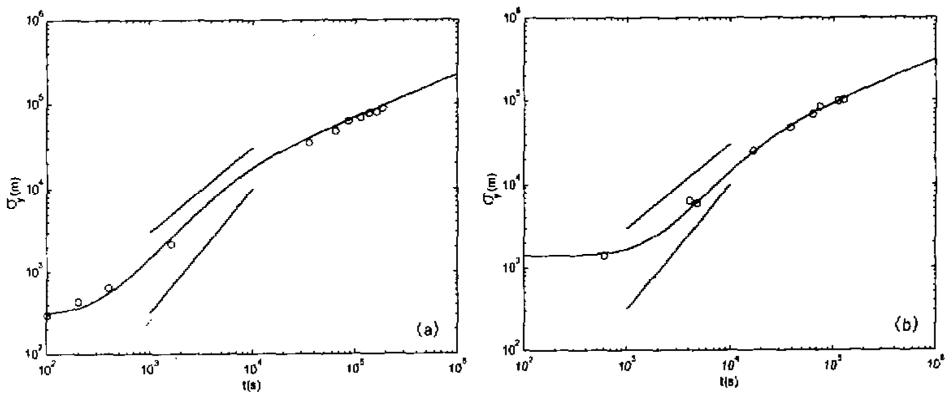
where $\beta = \frac{\partial \overline{u}}{\partial x} + \frac{\partial \overline{v}}{\partial y}$, is the horizontal divergence of the mean wind field, $\xi(t)$ includes all turbulent terms that can be considered as random functions. It is noted that Eq.(15) looks like Eq.(4). Supposing that the horizontal divergence does not change with time, we have

$$A = A_0 e^{\beta T} + \int_0^T e^{\beta(T-s)} \xi(s) ds, \tag{16}$$

where T is time, A_0 is the horizontal area of the puff at $T=0$. The first term at the right–hand side of the equation is caused by divergence and the second term by turbulence. Because divergence can be positive or negative, Eq. (16) can cause the horizontal area of the puff to be larger or smaller. The quantity has the same property with B in Eq. (4). It is a reciprocal of sluggish time. In Eq. (4), B is reciprocal of the Coriolis parameter, which implies that the rotation of the earth acts on the diffusion. In Eq. (15), β is divergence, which means that the distortion of flow fields acts on the diffusion. In the troposphere and stratosphere, β is of the order of $10^{-5}s^{-1}$; accordingly, $1/\beta$ is about 1 day. As a result, divergence becomes important after one day of release.

4. Dispersion parameters of air nuclear test clouds and atmospheric parameters analysis

Air nuclear test clouds occupy a very huge volume and they can enter the troposphere and the stratosphere by the buoyancy force. The fine radioactive debris or particles transport a long distance by the general circulation and therefore may stay in the atmosphere for a few hours to years. During nuclear tests in China, optical instruments were used to determine the width of nuclear clouds near the test site. An airplane equipped with sensitive radiation monitoring instruments flew back and forth to measure the trajectories and widths of nuclear debris clouds. We use air radioactive background values plus its three times standard deviation as the threshold to define the boundary of the radioactive debris clouds. We use widths along the direction perpendicular to the trajectory line as the widths of the cloud. Observation time ranged from a few minutes to about 50 hours. We apply the Gifford's theory to analyze 20 nuclear explosion clouds data sets. It is assumed that the horizontal radioactive concentration of cloud presents Gaussian distribution, with a standard deviation $\sigma_y = W / 4.3$, where W is the width of clouds. We obtain T_L and K by fitting curves of datasets (See Fig. 1). Solid lines are curves of Eq.(5). The parameters such as K and T_L are listed in Table 1.



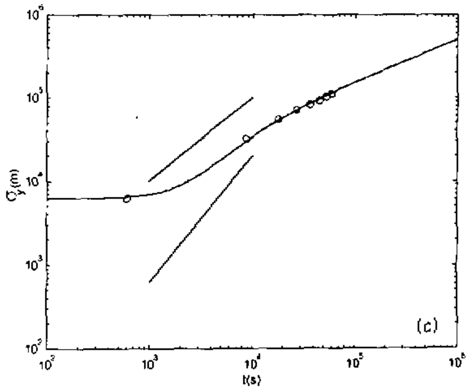


Fig. 1. Diffusion parameters of nuclear test clouds, their change against time and Eq.(5). (a) Test 10, (b) Test 2 and (c) Test 7.

Table 1. Horizontal diffusion parameters for 20 air nuclear tests

Explosion	Z (km)	K($10^4 m^2/s$)	T_L(s)	$\sqrt{v^2}$ (m/s)	ε (10^{-4} m^2/s^3)	L (km)
1	9	6	10000	2.45	6	34.6
2	7	5	100000	2.24	5	31.6
3	9	10	3000	2.77	111	24.5
4	6	3	10000	1.73	3	24.5
5	9	5	10000	2.24	5	31.6
6	12	12	7000	4.14	24.5	41.0
7	12	12	5000	4.90	48	34.6
8	14	12	5000	4.90	48	34.6
9	14	10	5000	4.47	40	31.6
10	7.5	2.5	3000	1.58	2.5	22.4
11	14	7	10000	2.65	7	37.4
12	14	10	2000	7.07	250	20.0
13	14	6	5000	3.46	24	24.5
14	3	2	7000	1.69	4.1	16.7
15	16	15	2000	8.66	375	24.5
16	9	6.5	7000	3.05	13.2	30.2
17	9	4	9500	2.05	4.4	27.6
18	5.5	2.8	7000	2.00	5.7	19.8
19	3	2.4	10000	1.55	2.4	21.9
20	14	8.5	3000	5.32	94.4	22.6

From Fig. 1a, we can find the accelerative nature of relative diffusion, viz $\sigma_y \propto t^\alpha$, $\alpha > 1$. In Fig. 1b, $\alpha \approx 1$. In Fig. 1c, $\alpha < 1$.

In Fig. 2, measurement of σ_y changes less as time goes longer, and this is different from the general understanding on the concept of the diffusion. In fact, the tropospheric diffusion is controlled by the distortion and divergence of the wind fields. Divergence causes clouds to expand and convergence causes clouds to shrink. Since Eq. (5) is derived from the small–scale homogeneous turbulence, it fails to explain the results in Fig. 2.

Horizontal diffusion parameters for 20 air nuclear tests are given in Table 1:
In the table Z is the altitude of airplane measurement, $\overline{v^2} = K / T_L$, $\varepsilon = K / T_L^2$, $L = \sqrt{2KT_L}$. It is noticed that Z ranges from 3 to 16 km (in the troposphere and the lower stratosphere); the large–scale effective eddy–diffusivity, K, ranges from 2 to $15 \times 10^4 m^2 / s$; the Langrangian time scale, T_L, ranges from 2000 to 10000 s; the Langrangian length scale L ranges from 16.7 to 41 km, which means that there is an accelerative diffusion region as the initial size of a puff is within L.

When the diffusion time is large enough, we can see an interesting phenomenon from Fig. 3. After 50 hours of dispersion, the horizontal diffusion deviations approach 100 km, no matter how large the initial cloud sizes are. At this time, the width of clouds is about 450 km that is the turning point of atmospheric motion from three–dimension to two–dimension. Therefore, the small puff diffusion is accelerative but this is not true for the diffusion at large scales.

Figure 4 is drawn from effective eddy–diffusivity K in Table 1 and Barr's et al. (1987) 12 fitting data. The figure shows that K changes with altitude. We can obtain an empirical equation using the regression method:

$$K = 1.42 \times 10^4 \exp(z / H), \quad (17)$$

where $H \approx 7 km$, is the scale height of the atmosphere and the correlation coefficient between the height and eddy–diffusivity K (converting to linear model) is 0.82. According to the Prandtl's theory, Eq. (17) can be explained that as the altitude increases, the air density decreases and the mixing length increases, and therefore K increases with the altitude. Equation (17) can be considered as an estimation of the effective eddy–diffusion parameter.

We use the cloud width data of a test to demonstrate the relationship between the diffusion and the divergence. In test 9, the nuclear clouds were transported and diffused to the downwind locations under the WNW wind and moved out of the frontier of China above the Yangtze River mouth.

The effective eddy–diffusivity K can be calculated by using

$$K = \frac{1}{2}\frac{d\sigma_y^2}{dt} = \frac{\Delta\sigma_y^2}{\Delta t}, \quad (18)$$

and the results together with the divergence for 14 km along the trajectory are listed in Table 2. In the table, T is the time after explosion, and divergence is calculated from the NCAR reanalysis① data which include wind at 17 pressure levels, geopotential height, air temperature, relative humidity, $2.5° \times 2.5°$ latitude–longitude global grid with 144×73 points for output every 24 hours. From the results, cloud diffusion is faster as divergence being positive and slower as divergence being 0 or negative. From the weather map, we can see that the debris cloud locates behind a high altitude trough, behind which there exists convergence. At low altitudes, however, there exists divergence. There is a convergence region above

①http: / / www.scd.ucar.edu / dss / pub / reanalysis / index.html

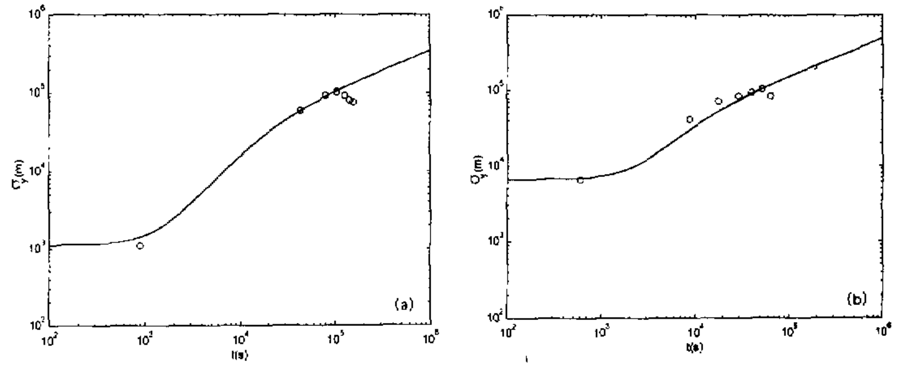


Fig. 2. Air nuclear test diffusion parameters change with time.

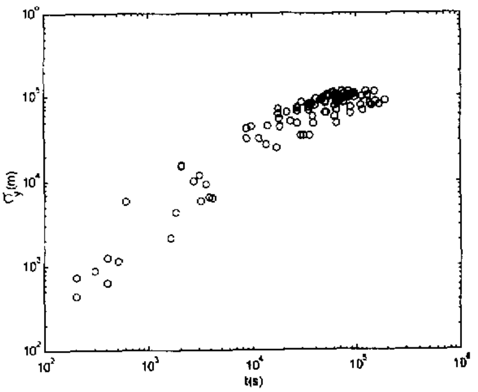


Fig. 3. Nuclear clouds sizes change against time for all tests.

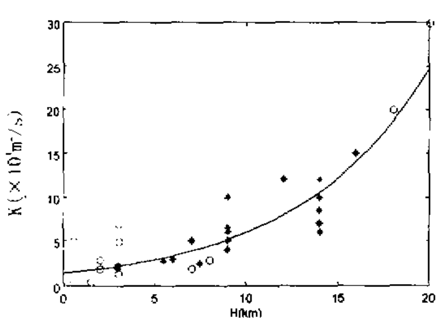


Fig. 4. Effective eddy–diffusivity K changes against altitude of release (* nuclear tests data, o Barr et al. (1987) data).

the Yangtze River valley, and this is the reason that the cloud width changed slowly in this region.

Table 2. Relationship between nuclear clouds effective eddy–diffusivity K and divergence

Longitude (degrees)	95	100	105	110	115	120
σ_y (km)	44.2	65.1	81.4	88.4	95.3	97.7
T(h)	2.7	5.9	9.5	13.8	18.5	23.5
Divergence ($10^6 s^{-1}$)	6	3	3	0	0	−10
$K(10^4 m^2 / s)$		1.1	9.3	3.8	3.7	1.3

5. Conclusions

From the above discussions, we can conclude that the horizontal relative diffusion in the troposphere and the stratosphere undergoes multiple phases according to the TKE spectrum $k^{-3/5}$ and k^{-3} laws. We can simply categorize the following three phases:

1) Accelerative diffusion phase: when the puff size is smaller than the Langragian length scale $\sqrt{2KT_L}$, we have $\sigma_y \propto T^{3/2}$, which is revealed by the diffusion theory for the three-dimensional inertial subrange.

2) Taylor's diffusion phase: at this phase, $\sigma_y = \sqrt{2KT}$, which is the Taylor's diffusion formula for a continuous point source to present an instantaneous source.

3) Divergence and convergence phase: when the puff enters the two-dimensional turbulence region (generally horizontal puff size greater than 500 km, or time scale greater than 1 day), the dominant factor to affect the puff's horizontal diffusion is divergence and convergence. Puff can grow or shrink, depending on the puff location in the weather system.

The horizontal relative diffusion processes are described as follows. The puff expands by the three-dimensional turbulence firstly. As it expands to a certain size, it enters a two-dimensional turbulence region. Then, the two-dimensional turbulence shreds the puff into smaller puffs which continue to diffuse by the three-dimensional turbulence. Finally, the puff is diluted. In fact, the atmospheric motions in the troposphere and the stratosphere are neither stationary nor homogeneous. Air nuclear tests show that the clouds were shredded into pieces in the beginning of the release. The large-scale turbulence is only effective for the deformation of puff. Therefore, it is not suitable to use parameter to demonstrate the diffusion of the puff.

The author would like to thank Prof. Chen Jiayi' Department of Geophysics of Peking University and Dr. Cai Xiaoming School of Geography and Environmental Sciences of Birmingham University for their helpful discussions.

REFERENCES

Barr, S., and F. A. Gifford (1987): The random force theory applied to regional scale tropospheric diffusion. *Atmos. Environ.*, **21**, 1737–1741.

Batchelor, G.K.(1950): The application of the similarity theory of turbulence to atmospheric diffusion. *Quart. J. R. Meteor. Soc.*, **76**, 133–146.

Batchelor, G.K.(1952): Diffusion in a field of homogeneous turbulence, II. The relative motion of particles. *Proc. Camb. Phil. Soc.*, **48**, 345–362.

Charney, J.G.(1971): Geostrophic Turbulence. *J. Atmos. Sci.*, **28**, 1087–1095.

Gifford, F. A. (1982a): Horizontal diffusion in the atmosphere: a Langrangian-dynamical theory. *Atmos. Environ.*, **16**, 505–512.

Gifford, F. A. (1982b): Long-range plume dispersion: comparisons of the Mt. Isa data with theoretical and empirical formulas. *Atmos. Environ.*, **16**, 883–886.

Gifford, F. A. (1984): The random force theory: application to meso- and large-scale atmospheric diffusion. *Bound.-Layer Meteor.*, **30**, 159–175.

Kao, S. K., and A. A. Al-Gain (1968): Large-Scale Dispersion of Clusters of Particles in the Atmosphere. *J. Atmos. Sci.*, **25**, 214–221.

Kraichnan, R. H. (1967): Inertial range in two-dimensional turbulence. *Physics of Fluids*. **10**, 1417–1423.

Lilly, D.K.(1989): Two-dimensional turbulence generated by energy sources at two scales. *J. Atmos. Sci.*, **46**, 2026–2030.

Lin, Jung–Tai (1972): Relative dispersion in the enstrophy–cascading inertial range of homogeneous two–dimensional turbulence. *J. Atmos. Sci.*, **29**, 394–396.

Pasquill, F., and F. B. Smith, (1983): *Atmospheric Diffusion* (3rd ed.). Ellis Horwood Limited, 149–158, 437pp.

Randerson, D. (1972): Temporal changes in horizontal diffusion parameters of a single nuclear debris cloud. *J. Appl. Meteor.*, **11**, 670–673.

Wallace, J. M. and Hobbs P. V. (1977): *Atmospheric Science: An Introduction Survey*, Academic Press, New York, 396–399, 467pp.

Walton, J. J. (1973): Scale–dependent diffusion. *J. Appl. Meteor.*, **12**, 547–549.