# ENORMOUS INCREASE OF VOLCANIC CLOUDS IN THE STRATOSPHERE OVER FUKUOKA AFTER APRIL 1982

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## RESUMEN

Mediante el sistema lidar Yag se observaron sobre Fukuoka (33<sup>0</sup>N) aumentos muy grandes de los aerosoles estratosféricos causados por la erupción del volcán mexicano El Chichón, a principios de abril de 1982. Aquí se presenta la variación a largo plazo de los perfiles correspondientes a 15 meses. Las capas más densas de aerosol se localizaban a una altitud entre los 20 y los 30 km en los primeros seis meses y las capas más enrarecidas, por debajo de la altitud de alrededor de 20 km. La parte más densa descendió gradualmente después de octubre de 1982 a unos 20 km en enero de 1983, después de mezclarse con la parte inferior, y permaneció ahí desde entonces. La velocidad de descenso de la capa superior es aproximadamente de 1 km/mes o menos.

La profundidad óptica de las capas de aerosol en la longitud de onda de  $0.55 \,\mu$ m alcanzó su valor máximo de alrededor de 0.3 para el período comprendido entre diciembre de 1982 y febrero de 1983, y decreció entonces gradualmente a 0.05 en julio de 1983.

La difusión turbulenta en la parte superior se estima que fue extremadamente pequeña para el período de abril a octubre de 1982 en las direcciones vertical y meridional. Este hallazgo puede explicar congruentemente los resultados observados y apoya el enfoque originalmente propuesto por Matsuno (1980).

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## ABSTRACT

Very large-increases of stratospheric aerosols caused by the eruption of Mexican volcano El Chichón in early April 1982 were observed by a Yag lidar system over Fukuoka  $(33^{\circ}N)$  and the long term variation of the profiles for 15 months is presented. The densest aerosol layers were located in the altitude range from 20 to 30 km for the early six months and the lower rarer layers were located below the altitude about 20 km. The densest part gradually descended after October 1982 to about 20 km in January 1983, after merging with the lower part and remained there since that time. The descending velocity of the upper layer is about 1 km/month or less.

The optical depth of the aerosol layers at the wavelength 0.55  $\mu$ m attained maximum value about 0.3 for the period from December 1982 to February 1983 and then gradually decreased to 0.05 in July 1983.

The eddy diffusion in the upper part is estimated to be extremely small for the period from April through October 1982 in the vertical and meridional directions. This finding can explain observed results consistently and supports the view originally proposed by Matsuno (1980).

## INTRODUCTION

Very large increases of stratospheric aerosol particles caused by the eruption of Mexican volcano El Chichón in early April 1982 were observed by a Yag lidar system at the wavelengths  $F = 1.06 \,\mu\text{m}$  and  $S = 0.53 \,\mu\text{m}$  over Fukuoka (33.6°N, 130.2°E) since April 1982 (Hirono and Shibata, 1983). The profiles for 15 months are shown in Fig. 1. The increases of the stratospheric aerosols were so enormous that the optical depth of the aerosol layers at the wavelength 0.55  $\mu$ m attained maximum value about 0.3 for the period from December 1982 through February 1983,



Fig. 1. Profiles of lidar scattering ratio R(F) at the wavelength  $F = 1.06 \,\mu\text{m}$ . The value (R-1) is proportional to the aerosol mixing ratio. During volcanic quiet time  $(R-1) \sim 0.5$  at the altitude 20 km as seen on April 15, 1982.

(a) April - May 1982, (b) July - Dec. 1982, (c) Jan. - July 1983.

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slowly decreasing to 0.05 in July 1983 as inferred from the ordinate values of Fig. 2 multiplied by 100-130 sr. The conversion rate of backscattering into optical depth is explained in section 3.



Fig. 2. Variation of integrated aerosol backscattering  $B = \int \beta_A(F) dz$  from altitudes 13.5 to 28.5 km. Optical depth at the wavelength 0.55  $\mu$ m is obtained from B multiplied by 100 - 130 sr.

During the period from April to August 1982, the profiles were divided into two parts: upper denser part including very fine structures of the profiles above about 20 km and lower rarer part below as seen from Fig. 1 and shown by a sketch in Fig. 6. The intervening region at about 19–21 km was very clean until mid-June, the content of the remaining aerosols there being roughly proportional to the upper peak value at about 24 km.

According to the balloon observation (Hofmann and Rosen, 1983) the size distribution at 25 km in May 1982 at Laredo Texas (27.3°N) was bimodal and most mass of the aerosols was concentrated at the radius near 1  $\mu$ m. Revised interpretation of the two wavelengths lidar data can be reconciled with the results of direct observations.

The very slow meridional diffusion of the upper part until October (McCormick *et al.*, 1983a) and the existence of fine structures in the upper part and minimum aerosol concentration at 19-21 km until August should be examined. In the following we shall discuss the problem on the basis of the theory of Matsuno (1980).

#### THE EDDY DIFFUSION IN THE STRATOSPHERE

The distribution of the tracers in the stratosphere and its variations are strongly affected by the transport with eddy diffusion. The eddy transport is hardly attribut-

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able to random eddies, because it is often observed that countergradient diffusion takes place in the stratospheric atmosphere. Reed and German (1965) postulated a mixing length hypothesis. It is assumed that an airparcel moved a distance L (~100 km) on a straight line that makes a small angle  $\alpha_{\rm R}$  (downward to higher latitude) ~5x10<sup>-4</sup> with the horizontal, before mixing with the environmental air (Fig. 3 b). They succeeded to explain the meridional distribution of tracers by using this hypothesis. But this was not well founded on the principle of atmospheric dynamics.

 $d \chi/dt = (\chi - \chi)/\zeta$ 

 $\tau$  : MIXING TIME

L MIXING LENGTH





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Fig. 3. Projection of trajectories of airparcels onto the meridional plane which give rise to the eddy diffusion according to the authors:

(a) Matsuno (1980); in the presence of planetary waves within the westerly wind,
 (b) Reed and German (1976); mixing length hypothesis.

Matsuno (1980) on the other hand attempted to construct the diffusion theory on the basis of atmospheric dynamics. He formulated the planetary waves in the atmosphere on the beta plane. If the trajectory of the airparcel, affected by the planetary waves in the westerly wind, is projected on the meridional plane, it delineates an ellipse which has a major axis inclined at an angle  $\alpha_{\rm M}$  with the horizontal being almost equal to  $\alpha_{\rm R}$  used by Reed *et al.* (1965) as shown in Fig. 3 a. Further introducing the mixing time  $\tau$ 

$$d\chi/dt = (\chi - \chi)/\tau$$
 (1)

where  $\chi$  is a physical quantity carried with the airparcel and  $\overline{\chi}$  is the environmental value of  $\chi$ , he could formulate the eddy diffusivity on sound dynamical principle (see Appendix).

Danielsen (1981) showed that the mechanism of Matsuno can be a good basis of conventional two dimensional transport model, but the mixing time  $\tau$  is of the order of one day, based on documents obtained mainly below altitude 20 km. This time is much shorter than the 100 days estimated by Matsuno.

The planetary waves generated in the troposphere cannot propagate upward beyond the critical layer (C. L.) which divides the westerly and easterly wind regions. The altitude of C.L. over Fukuoka is shown in Fig. 4 against progress of time in months. Below the altitude 20 km, where planetary waves can exist, the dispersion of volcanic aerosols seemed to be rapid as suggested by conventional model simulations (Cadle *et al.*, 1976) and the 15 - 18 km layer reached West Germany (47°N) in May 1982 (Reiter *et al.*, 1983) and Brasil (23°S) in July 1982 (Clemesha *et al.*, 1983).



Fig. 4. Altitudes of critical layer over Fukuoka which divide easterly and westerly wind region. In the middle latitudes the altitude of C.L. is much the same.

During April and May 1982, the height of the C.L. was variable but located near the densest part of the upper layer. The meandering of the zonal wind was appreciable to displace the central part of El Chichón cloud to Fukuoka at times. The very large standard deviation of the upper peak scattering ratio  $R_p$  over Fukuoka in May as illustrated in Fig. 5 would be due to such an air motion and also to significant inhomogeneity of tracer mixing ratio.

In May the upper layer was detached in case of large meandering and transported over to Spain (40<sup>o</sup>N) (Ackerman *et al.*, 1983) and to Italy (D'Altorio *et al.*, 1983). From June to August the upper densest part was in the easterly wind region and C.L. was descending from 20 to 18 km, the occasional very low altitude in August

being caused by the local wind due to anticyclone. After June the zonal wind became nearly parallel to latitude circle. The central part of the cloud would return to about  $20^{\circ}$ N, producing the variation of R<sub>p</sub> shown in Fig. 5, and also the variation of the integrated aerosol backscattering shown in Fig. 2 which had minimum in August. The effect of settling of large ash particles until this time should be taken into account in addition to the above advection effects.





There seems to be no significant eddy diffusion in the higher region than 20 km, in the absence of planetary waves during summer. At such a situation the general circulation in the lower latitudes than  $33^{\circ}$  should be of the type originally advocated by Brewer-Dobson and calculated by Murgatroyd *et al.* (1961) and Vincent (1968). The calculated mean ascending velocity by these authors is of the order of magnitude of 0.03 cm/s (= 0.8 km/month) in summer at altitudes 21 - 24 km and around  $30^{\circ}$ N. This ascending motion would cancel the settling velocity of 0.5  $\mu$ m particles at the altitude 22 km. Smaller particles will ascend and larger particles including giant particles (r > 1  $\mu$ m) will descend but at reduced settling velocity. This mechanism might partly explain the existence of minimum aerosol concentration near 19 - 21 km in summer. On the other hand the absence of diffusion due to vanishing planetary waves can explain the observed results that there were almost no meridional diffusion in the upper densest part during this season (Labitzke *et al.*, 1983, McCormick *et al.*, 1983a).

The height of C.L. in the middle latitudes is not much different from that over Fukuoka beginning to ascend gradually from September. The mean vertical velocity obtained by Vincent (1968) begins to descend from October at  $35^{\circ}$ N at altitudes 21 - 24 km. This Eulerian mean velocity would include the influence of the planetary waves (Matsuno, 1983) and real tracer descending cannot be expected without careful examination of dynamical processes.

Very rough estimate of the mixing time  $\tau$  is obtained by  $\tau = L^2/D$  where L is the characteristic length of the horizontal inhomogeneity of the airparcel containing tracers and D is the coefficient of horizontal random diffusion. In cases of volcanic injection into the stratosphere, L should be much smaller than that for the ozone distribution. For the present case it is tentatively estimated that  $L \leq 300$  km.

In December 1979, one month after the eruption of the volcano Sierra Negra  $(0.83^{\circ}S, 91.17^{\circ}W) L \sim 300 \text{ km}$  is estimated by lidar observation (Fujiwara *et al.*, 1982). In November 1982, Thomas (1982) estimated that L = 200 - 400 km at Wales (52°N). If the value of D is estimated to be  $3 \times 10^5 \text{ m}^2$ /s according to Bauer (1974), then it follows that  $\tau \leq 3$  days which is sufficiently short to give effective meridional diffusion in the presence of planetary waves as shown in Appendix. The eddy diffusion will be increasingly effective since October towards winter. As shown in Fig. 6 the peak of the densest part of the upper layer was located at about 24.2  $\pm$  0.8 km until October, but since then gradually descended to 20 km until January 1983.



Fig. 6. Outline of distinct maxima and minima of R(F) with half width of (R-1) shown by vertical bars. Fine structures during early period and broadening after autumn are seen. Dashed lines show mean height of upper maxima above altitude 21 km and standard deviation until Oct. 1982.

• : greatest peak,  $\circ$  : smaller peak,  $\blacktriangle$  : minima.

## INTERPRETATION OF THE TWO-WAVELENGTH LIDAR SIGNAL RATIOS

In the previous paper (Hirono *et al.*, 1983) we assumed a truncated power law size distribution of aerosols and made an interpretation of the two-wavelength lidar signal ratios.

It is regret that in the absence of other informations, this method cannot discriminate more complicated size distributions which may be present for the disturbed state, under consideration of the very inhomogeneous distribution of ejecta.

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Hofmann and Rosen (1983) measured the aerosol size distribution in the stratosphere over Laredo, Texas (27.3<sup>o</sup>N) in May, August and October in 1982. Their results in the upper part than 21 km show bimodal distributions represented by the following expressions:

$$n(r) = \left\{ \sum N_i / (2\pi)^{1/2} \ln \sigma_1 r \right\} \cdot \exp[-(1/2) \left\{ \ln(r/r_i) / \ln \sigma_i \right\}^2 \right]$$
(2)  
(i = 0, 1)

At 24.5 - 25.5 km on May 18 - 19 1982 it follows that

$$N_0 = 150 \text{ cm}^{-3}, \sigma_0 = 2.8, r_0 = 0.02 \mu \text{m}, m_0 = 1 \mu \text{gm}^{-3}$$

for the 1st mode, and

$$N_1 = 4 \text{ cm}^{-3}, \sigma_1 = 1.77, r_1 = 0.72 \mu \text{m}, m_1 = 35 \mu \text{gm}^{-3}$$

for the 2nd mode.

Thus most part of the mass was involved in the 2nd mode. This size distribution quite resembles that calculated by Turco *et al.* (1983) to simulate formation processes one month after Mt. St. Helens eruption in 1980. This result suggests that in May the aerosol size distribution over Fukuoka would be shown by a bimodal one, which contains most part of mass concentration in the larger radius mode.

The backscattering coefficient  $\beta_A(\lambda)$  from aerosols of lidar with wavelength  $\lambda$  is represented by

$$\beta(\lambda) = (1/4\pi) \int \pi r^2 Q(x) n(r) dr$$
(4)

(3)

where  $x = 2\pi r/\lambda$ , Q = backscattering efficiency, n (r) = differential size distribution for radius r. Since approximately Q(x)  $\propto$  r, then the integrand of the right hand side of Equ.(4) is approximately proportional to the differential mass distribution m(r). Therefore the obtained lidar echo comes mainly from the second mode, which would be produced by condensational growth of the pre-existing particles as suggested by Hofmann and Posen (1983).

The aerosol backscattering coefficient at three wavelengths F, S, and 0.69  $\mu$ m are approximately equal for the size distribution expressed by (2) and (3) as follows:

$$\beta_{\rm A}({\rm F}) \sim \beta_{\rm A}({\rm S}) \sim \beta_{\rm A}(0.69) \sim 9.25 \,\mathrm{x} \, 10^{-7} \mathrm{m}^{-1} \,\mathrm{sr}^{-1}$$
 (5)

and hence their ratios are nearly equal to unity.

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The scattering ratios are given by R(F) = 290, R(S) = 19 and R(0.69) = 54.

On the other hand, the observed values at 25 km over Fukuoka on May 21 are as follows:

$$\beta_{A}(F) = 1.5 \times 10^{-7} \text{m}^{-1} \text{sr}^{-1}$$
,  $R(F) = 50$  and  
 $\beta_{A}(S)/\beta_{A}(F) = 3.2$ . (6)

The value of  $\beta_A(F)$  is about one sixth of that estimated on Texas and therefore it is inferred that the cloud over Fukuoka was near the edge of the main zonal stream, and contained lower concentrations of volcanic gases i.e.  $SO_2$ ,  $H_2SO_4$ , etc. than at the center. At such circumstances, homogeneous or ion nucleation would be few, but condensational growth of pre-existing particles would be still active.

If the pre-existing particles are represented by  $N_p = 5 \text{ cm}^{-3}$ ,  $r_p = 0.08 \,\mu\text{m}$  and  $\sigma_p = 1.6$  in Equ.(3), for which i = p, then the size distribution n(r') after the condensational growth is given by

$$\mathbf{r}' = \mathbf{r} + \Delta \mathbf{r} \quad (\mathbf{r}; \ \Delta \mathbf{r} > \mathbf{0}) \tag{7}$$

where  $\Delta r$  being almost independent of radii which are considered here. It follows that

$$\mathbf{n}(\mathbf{r}') = \mathbf{n}(\mathbf{r})_{\mathbf{i}=\mathbf{p}} \tag{8}$$

with equation (7).

The geometric mean radius  $r_m$  after the growth  $\Delta r$  is  $r_m = r_p + \Delta r$ , and calculated ratio  $\beta_A(S)/\beta_A(F)$  is shown by a curve s in Fig. 7. For reference, the following step-like functions of size distributions are also assumed:

$$n(r) = cr^{-2} \tag{9}$$

for the interval

$$r_m - b/2 < r < r_m + b/2$$
 (10)

with 
$$b = 0.1 \mu m$$
 and  $0.2 \mu m$   
and  $n(r) = 0$  outside interval shown by Eq.(10)

The ratios  $\beta_A(S)/\beta_A(F)$  are calculated for these size distributions and shown by the curves I and II for  $b = 0.1 \,\mu\text{m}$  and  $b = 0.2 \,\mu\text{m}$ , respectively in Fig. 7. The aerosol backscattering  $\beta_A(F)$  can be calculated against  $r_m$  for the distribution (8). We have  $r_m = 0.37 \,\mu\text{m}$  for the value of  $\beta_A(F)$  of (6), using the above relation between

the values. This  $r_m$  value gives rise to the ratio  $\beta_A(S)/\beta_A(F) = 2.8$  on the curve s of Fig. 7. This value agrees with observed value 3.2 if uncertainty of normalization  $\sim 10\%$  is taken into account.

The ratio of the extinction at  $\lambda = 0.55 \ \mu m \ E(0.55)$  to  $\beta_A(F)$  depends on the size distribution of aerosols. The vertical profiles of the size distributions by six channel detector of Hofmann *et al.* (1983) in August, October 1982 and those in January 1983 (Hofmann and Rosen, August 1983, Hamburg IUGG) are used together with our lidar profiles of  $\beta_A(F)$  and  $\beta_A(S)$  to calculate the ratio of the integrations between altitudes 13 and 28 km:



Fig. 7. Variation of the ratio  $\beta_A(S)/\beta_A(F)$  against  $r_m$  for the size distributions I and II of Equ. (9) with (10) at  $b = 0.1 \mu m$  and  $b = 0.2 \mu m$ , respectively; s for Equ.(8). See text for details.

 $b = \int E(0.55) dz / \int \beta_A(F) dz$  based on the Mie theory.

It is shown that  $b = 130.19 \pm 3.2$  sr which essentially agree with the value of Pinnick *et al.* (1980). According to the discussion in the present section, the ratio b in May 1982 is estimated approximately at 100 sr, which would be gradually changed to 130 sr until August due to the decrease of vast giant particle concentrations in the stratosphere.

## DISCUSSION

In the previous paper (Hirono *et al.*, 1984) it was shown that in the Mt. St. Helens 1980 event, the eddy diffusion at altitudes several km above 20 km was quite small i.e. about  $10^{-2}$  m<sup>2</sup>/s in summer and the vertical shear of the easterly wind transformed the aerosol distribution at the altitudes to horizontal layer with very thin vertical width about 1 km throughout the season.

As shown in section 2, for the El Chichón event, extremely small vertical eddy diffusion compatible with very fine structures in vertical profiles are seen in the easterly wind region above about 20 km in summer. At the same time, the meridional diffusion of the upper aerosol layer was almost absent until October. These findings strongly support the idea that the eddy diffusion is produced by the combined effects of planetary waves and the dissipation including the small scale turbulence, and the absence of the former minimizes the eddy diffusion.

According to the results of model calculation of Shibata (1983), the ion nucleation and homogeneous nucleation are effective to diminish the mean radii of aerosol particles and hence their settling velocities. The results are thought to be important to produce aerosol free region at 19 - 21 km. If the vertical diffusion is negligibly small, the volcanic gases  $SO_2$ ,  $H_2SO_4$  etc. gave rise to the growth of particles to supplement large particles lost by settling at the initial altitude of injection until October 1982.

The giant particles  $(r>1 \mu m)$  involving ash will fall into the troposphere without appreciable divergence of the vertical flux near the altitude 20 km, the settling velocity of which is greater than 1.6 km/month at 22 km. The majority of newly formed aerosol particles in the upper layer would have fairly smaller radii than about 0.5  $\mu$ m due to the abovementioned effects and hence remain in the region above 20 km also assisted by ascending motion in summer mentioned in section 2

In the lower region than about 20 km, in the presence of planetary waves, the meridional diffusion would be significantly larger to diminish aerosol concentrations. Along the critical surface dividing the easterly and westerly wind, the meridional diffusion is thought to be particularly effective (Fujiwara *et al.*, 1982). Thus a 19 - 21 km clean region would be produced.

The integrated aerosol backscattering  $B = \int \beta_A dz$  had maximum value around December 1982 to February 1983 as shown in Fig. 2 and gradually decreased. The optical depth at the wavelength  $0.55 \,\mu$ m is obtained after B is multiplied by 100-130 sr. In February 1983 optical depth is about 0.3 and in July 1983 about 0.05.

Column mass concentration M is obtained approximately by  $M = 33.3 \times B(g/m^2)$  after Pinnick *et al.* (1980). In March 1983 M is estimated at 0.04 g/m<sup>2</sup> over Fukuo-

ka. At this time M seems to be nearly equally distributed with latitude in Northern Hemisphere and hence the total stratospheric burden of the aerosols would be about 10 Tg ( $T = 10^{12}$ ). The stratospheric aerosols seem to be transported poleward by eddy diffusion, and in May maximum and minimum values of M are found in higher latitude than 50°N and about 30°N, respectively (McCormick, 1983b). Therefore, the total burden in the Northern Hemisphere would decrease more slowly than that over Fukuoka.

## CONCLUDING REMARKS

Enormous increases of stratospheric aerosols were observed by a Yag lidar system over Fukuoka and the long term variations for 15 months are presented. According to the theory of Matsuno (1980), the eddy diffusion in the stratosphere, effective for the global transport of tracers, is produced by the planetary waves, if the mixing time of the tracer is sufficiently short. Above C.L. (Critical Layer) at about 20 km over Fukuoka in summer, there is no planetary waves and hence the eddy diffusion would be extremely small. This hypothesis is consistent with quite slow meridional diffusion in the densest part of the volcanic cloud at about 24 km (30mb), until October 1982 (Labitzke et al., 1983; McCormick et al., 1983a). The ascending air motion estimated by Vincent (1968) will be true upward motion of tracer in this case and cancel some part of the settling velocity of large aerosol particles. In the region below C.L. there will be planetary waves and the meridional diffusion will be effective for transport of tracers, because the mixing time is estimated to be less than 3 days. The vertical diffusion of volcanic gases injected above C.L. will be quite small in summer. This effect combined with some ascending motion mentioned above and the increased meridional diffusion particularly along C.L. and below C.L. as mentioned above would produce minimum concentration near C.L. for about four months after volcanic injection. According to the model calculation results by Shibata (1983) the decrease of mean aerosol size due to the ion nucleation (Arnold, 1982) will be helpful to diminish the settling velocity of the particles and the above scenario would be promising.

After October the upper densest part was gradually immersed in westerly wind and hence suffered from eddy diffusion. The upper part fell down through C.L. due to both vertical diffusion of volcanic gases and to settling of large particles.

The ratio of two wavelength lidar return signals was measured at 25 km over Fukuoka on May 21 when the aerosol content was about one sixth of that at the same altitude over Texas (27.3°N) on May 19. In the volcanic cloud over Fukuoka, it is inferred that the condensational growth of pre-existing particles was active but the ion nucleation or homogeneous nucleation were few due to lower concentrations of volcanic gases. Then the half width of assumed lognormal size distribution is held constant at about  $0.1 \mu m$ , and the observed backscattering coefficient is simulated at geometrical mean radius  $r_m = 0.37 \mu m$  on the curve showing functional rela-

tion of both quantities. This size distribution gives  $\beta_A(S)/\beta_A(F) = 2.8$  which is approximately equal to observed value of 3.2 if uncertainty at normalization is considered. Much more wavelengths are necessary in lidar system to determine full size distribution without other kind of information.

The column mass concentration of aerosols is estimated at  $0.04 \text{g/m}^2$  in March 1983 over Fukuoka. In the Northern Hemisphere the total burden of aerosols is estimated at about 10 Tg in March 1983 which would decrease more slowly than that estimated from Fig. 2 over Fukuoka since poleward transport by eddy diffusion is significant (McCormick, 1983b).

## APPENDIX

According to Matsuno (1980), the parameterization of eddy transport of tracers by planetary waves is carried out as follows: The trajectories of airparcels in the atmosphere on the beta plane due to planetary waves in the westerly wind are denoted by X, Y, and Z which are measured in eastward, northward and upward directions, respectively. The trajectories projected on the meridional plane can be given by

$$Y(t;\phi) = a_y \left\{ \sin(\omega t + \phi) - \sin\phi \right\}$$
(A1)

$$Z(t;\boldsymbol{\phi}) = -a_{z} \left\{ \cos(\omega t + \boldsymbol{\phi}) - \cos\boldsymbol{\phi} \right\} - \left\{ b_{2} \sin(\omega t + \boldsymbol{\phi}) - \sin\boldsymbol{\phi} \right\}$$
(A2)

where  $a_y$  is the amplitude of the parcel oscillation in the northern direction and  $a_z$ ,  $b_z$  are those in the vertical upward direction and  $\phi$  is the parameter designating the initial position of the parcel. After careful dynamical consideration based on these equations, the elliptical trajectory shown in Fig. 3 is obtained.

Introducing the mixing time  $\tau$  defined by Equ. (1) the following flux vector is obtained:

$$-\overline{V\chi} = (\Phi_1 K_s + \Phi_2 K_A) \nabla \overline{\chi}$$
(A3)

where 
$$K_s = \begin{bmatrix} (\omega/2)a_y^2 & , & -(\omega/2)a_yb_z \\ \\ -(\omega/2)a_yb_z & , & (\omega/2)(a_z^2 + b_z^2) \end{bmatrix}$$

denotes symmetric tensor similar in form to Reed and German (1965) contributing to meridional diffusion and

$$\mathbf{K}_{\mathbf{A}} = \begin{bmatrix} 0 & , & -(\omega/2)\mathbf{a}_{\mathbf{y}}\mathbf{a}_{\mathbf{z}} \\ \\ \\ (\omega/2)\mathbf{a}_{\mathbf{y}}\mathbf{a}_{\mathbf{z}} & , & 0 \end{bmatrix}$$

denotes an anti-symmetric tensor which does not contribute to true diffusion. Introducing the following scalar weighting factors is the highlight of the problem:

$$\Phi_1 = \omega \tau / (1 + \omega^2 \tau^2)$$
,  $\Phi_2 = \omega^2 \tau^2 / (1 + \omega^2 \tau^2)$  (A4)

In winter the mean westerly wind u is about 20 m/s at the altitude 18 km at latitude 45<sup>o</sup>N. When a geostationary planetary wave with wavenumber two is present, an airparcel passes through one wave pattern in a time

$$T \sim 1.42 \times 10^7 \text{ m/}\overline{u} \sim 7.1 \times 10^5 \text{ s} = 7.27 \text{ days}$$
  
therefore  $\omega = 2\pi T^{-1} = 0.88 \times 10^{-5} \text{ s}^{-1}$  (A5)

The value of  $\Phi$  takes maximum 0.5 for  $\omega_r = 1$  and very small values for  $\omega \tau \ll 1$ and  $\omega \tau \gg 1$ . When  $\tau \sim 3$  days,  $\Phi_1 \sim 0.3$  and the meridional diffusion takes place at a significant rate which can produce observed global distribution of tracers like ozone. The values of  $\omega$  are less as compared with equation (A5) for other seasons than winter since  $\overline{u}$  takes less values. In summer above about 20 km altitude  $a_y \sim a_z \sim$  $b_z \sim 0$ , therefore all elements of  $K_s$  and  $K_A$  vanish, and the diffusion due to this mechanism is absent.

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(Accepted: February 2, 1984)