

Dynamical Component of Seasonal and Year-to-Year Changes in Antarctic and Global Ozone

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Using a zonally averaged model in which all transport fields are fixed by input temperature data, we study dynamical aspects of the Antarctic ozone hole problem as a response to the observed year-to-year temperature change in August, September, October, and November. For the period studied, from 1979 to 1985, October column ozone density averaged over longitude is observed to decrease by 125 DU at 85°S. Our model produces 100 DU of decline from temperature change alone. At 60°S the model ozone decreases from 375 DU in 1979 to 325 DU in 1985, while the observed zonal mean column ozone declined from 410 DU in 1979 to 325 DU in 1985. The quasi-biennial signal in the year-to-year variations in column ozone is well-produced by the model with low (high) ozone correlating with westerly (easterly) phase of the tropical quasi-biennial oscillation (QBO). It is estimated that our small underprediction of 25 DU of polar ozone decline for the period 1979-1985 is uncertain by a factor of 2 and that if the effect on temperature cooling by reduced ozone heating in later years is taken into account, our model underprediction may increase to 50 DU. The underprediction of the October mean for 1985 by the model appears to be caused by the underprediction of the seasonal decline in September in the present model without heterogeneous chemistry. However, the seasonal declines during the earlier years are well simulated. With dynamics accounting for between 60 to 80% of the Antarctic October mean, zonal mean ozone decline for the period 1979-1985, we infer, subject to uncertainties in the model and data inputs, that dynamics should play a more important role in the phenomenon than merely providing a special condition for heterogeneous chemical destruction of ozone. However, chemical depletion of ozone may have become more important after 1984. Because heterogeneous chemistry is not incorporated in the present model, the model may have overestimated the magnitude of upwelling after 1984. It is possible that the reduced springtime radiative heating due to chemically reduced ozone amount may have suppressed vertical motion in September and early October in the later years.

1. INTRODUCTION

Recent observations reveal that the Antarctic ozone hole phenomenon [Chubachi, 1984; Farman *et al.*, 1985; Stolarski *et al.*, 1986] has both a seasonal cycle and a year-to-year trend. When the Sun returns to the Antarctic lower stratosphere after the polar night, column ozone levels have been observed to decline to reach a minimum value sometime in October, before recovering in November as a result of the final warming. Since at least 1979, the October minimum in column ozone has been found to be generally lower each year until 1986.

Many investigators have concentrated on a purely chemical hypothesis that relates the recent "secular" trend to the effect of man-made chlorofluorocarbons, whose release into the atmosphere has increased steadily since the early 1970s [Farman *et al.*, 1985; Solomon *et al.*, 1986; McElroy *et al.*, 1986a] (see also the recent review by Solomon [1988]). The role of dynamics is relegated to the secondary role of maintaining an isolated region inside the polar vortex for special chemical reactions to take place without disruption by transports from outside the polar vortex. The special chemical reactions currently considered all require the presence of

polar stratospheric clouds (PSC) [Molina and Molina, 1987; Crutzen and Arnold, 1986; McElroy *et al.*, 1986b, 1988]. The PSCs are largely absent outside the cold Antarctic polar vortex.

However, there are some aspects of the Antarctic ozone hole phenomenon that can best be explained by dynamical redistributions. These include the large-scale nature of the problem, with year-to-year decline of ozone occurring both inside and outside of the polar vortex [Stolarski *et al.*, 1986]. There is even some preliminary indication that the year-to-year changes in the Antarctic ozone and temperature may be related to the equatorial quasi-biennial oscillation (QBO) [Stolarski *et al.*, 1986; Bojkov, 1986, 1987; Garcia and Solomon, 1987]. Such global patterns of temperature and ozone changes are not likely to be explained by the special chemical reactions postulated to occur on surfaces of PSCs present only inside the polar vortex. The nonmonotonic nature of the ozone decline (e.g., the ozone column minimum in 1986 was higher than in 1985 and that in 1984 was higher than that in 1983) suggests that a downward trend, if it exists, is superimposed upon large year-to-year variability. Whether or not one can infer an underlying long-term downward trend in ozone due to chemistry will depend on one's ability to understand the dynamical component of the variability.

A successful theory, be it chemical, dynamical, or a combination of the two, should address the following observed aspects of the phenomenon:

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1. The year-to-year decline of the October ozone minimum over Antarctica is accompanied by a decline of the mid-latitude ozone maximum [Stolarski *et al.*, 1986]. The decline over mid-latitudes is about half that over the pole from 1979 to 1985 in magnitude per unit area (but over a larger area) [Schoeberl *et al.*, 1986].

2. Such year-to-year declines in both minimum and maximum ozone appear to be correlated with a cooling of the lower stratosphere in middle and high latitudes (outside as well as inside the polar vortex) [Mahlman and Fels, 1986; Newman and Schoeberl, 1986].

3. The interannual October mean column ozone changes over Antarctica appear to follow a quasi-biennial oscillation in phase with the tropical oscillation of the east-west wind [Garcia and Solomon, 1987], with low (high) Antarctic ozone occurring in the westerly (easterly) phase of the tropical QBO for the period analyzed, 1979–1986.

4. The seasonal spring deepening of the minimum column ozone appears to be accompanied by a simultaneous intensification of the surrounding maximum in mid-latitudes, for some years (1979–1982, Stolarski and Schoeberl, [1986]). The area-weighted sum of column ozone in the two regions appears to have no seasonal depletion from September to November [Stolarski and Schoeberl, 1986] in these years.

5. There is spatial correlation between the total ozone pattern and the distribution of temperature in the lower stratosphere [Stolarski *et al.*, 1986; Newman and Schoeberl, 1986; Sekiguchi, 1986; Chubachi, 1986], with low column ozone over Antarctica correlated with low temperatures and the surrounding maximum column ozone correlated with warm temperatures in the mid-latitudes [Stolarski *et al.*, 1986].

6. There is recovery in November, when the hole is “filled” in.

Chemical theories proposed so far mostly concentrated on the depletion in the minimum region (the “hole”) and attributed the accompanying change in mid-latitudes to other mechanisms (e.g., dynamics). The correlation of the ozone minimum with cold temperatures is attributed to the formation of PSCs at low Antarctic temperatures and the surfaces that PSCs provide for heterogeneous chemical reactions. The correlation of the ozone maximum with warm temperatures is not addressed. The temperature decrease is sometimes attributed to the reduced ozone heating caused by chemically depleted ozone amounts [Solomon, 1987].

This last interpretation is currently the subject of much debate. The radiative calculations of Rosenfield and Schoeberl [1986], using the observed ozone amounts from 1980 and 1984, suggest a reduction of about 0.1°K/day in the ozone heating in the lower stratosphere due to changes in ozone concentration in October. When multiplied by the radiative relaxation time of about 40 days for this part of the atmosphere, this yields a cooling of about 4°K in November. This is consistent with the finding of Shine [1986], who found a 5°K change in his seasonally varying case. The actual temperature change for the period 1979–1985, according to Newman and Schoeberl [1986], is much larger, (about 12°K in the lower stratosphere) and occurs earlier (in October). However, there is currently also some debate concerning the magnitude of the temperature change observed from different data bases. Nevertheless, radiatively induced temperature change should be distinct from dynamical (eddy) induced temperature change; the former is not accompa-

nied by changes in large-scale wave forcing, while the latter is. The observational evidence [Nagatani and Miller, 1987] appears to favor the latter [Mahlman and Fels, 1986], but uncertainty remains.

We present here numerical model results on the zonally averaged ozone distribution for the years 1979–1985, attempting to address aspects 1–6 of the phenomenon, mentioned earlier. Furthermore, we also attempt to address the following:

7. The observed latitudinal and seasonal distribution of column ozone over the globe, including the equatorial minimum, the spring maximum over the Arctic, and the fall minimum at high latitudes in both hemispheres.

8. The contrast between the northern and southern hemispheres; in particular, the occurrence of polar maximum, instead of minimum, ozone in spring in the northern hemisphere.

It is not our intention to suggest that dynamics is the sole cause of the Antarctic ozone hole phenomenon, only that it is an important component. Ground and balloon measurements at McMurdo Station during the austral spring of 1986 suggest that the chlorine chemistry of the Antarctic lower stratosphere is highly anomalous [P. M. Solomon *et al.*, 1987; S. Solomon *et al.*, 1987; Hofmann *et al.*, 1987], and that the local depletion at certain thin layers in the lower stratosphere [Hofmann *et al.*, 1987] probably is more severe than can be accounted for by advection of air from elsewhere. It is likely that the relative importance of chemistry vs. dynamics is different for different years, with chemistry playing an important role in later years when chlorine content is higher and dynamics being responsible for the creation of the ozone spring minimum in the climatology of earlier years.

Our purpose here is to quantitatively delineate the dynamical components of the change over Antarctica and other parts of the world. By design, we have not introduced any special chemistry into the model, nor any change in the chemistry between years. The role played by chemical changes in the atmosphere is instead inferred by comparing the model results without these changes with the observational data.

2. BACKGROUND

The basic dynamical mechanism that can produce variations in column ozone density is that of vertical motions [Reed, 1950; Tung *et al.*, 1986; Tung, 1986a] and the accompanying equator-to-pole transport of ozone [Mahlman and Fels, 1986]. Column ozone density is lowered in the presence of upward motion in the lower stratosphere, which brings in ozone-poor air from the lower atmosphere and displaces ozone-rich air in the stratosphere to other latitudes. Conversely, downward motion of air transports ozone-rich air from the photochemical production region and increases the column density of ozone.

The existence of mean vertical motion is dependent on the temperature structure of the atmosphere; the temperature structure of the atmosphere is in turn maintained away from radiative equilibrium by irreversible wave transports of heat (more precisely, Eliassen-Palm flux divergence) [World Meteorological Organization (WMO), 1986; Newman *et al.*, 1986; Plumb and Mahlman, 1987]. From a diagnostic point of view, one expects that (at steady state) upward mean

motion should exist in regions where the atmospheric mean temperature is colder than the local radiative equilibrium value, such as in the tropical lower stratosphere, where the excess radiative heating causes air to rise; and downward mean motion should be expected in regions where the atmospheric mean temperature is warmer than the local radiative equilibrium value, such as in the high latitudes in the northern hemisphere [Tung, 1986a]. This is the main reason why the observed climatology shows that column ozone density attains its minimum in the equatorial region, despite the fact that ozone is predominantly produced by photochemistry there. This is also why the maximum ozone column density is usually found over the pole (in the northern hemisphere), despite a lack of ozone sources there.

The ozone-temperature correlation mentioned previously also follows, with low (high) column ozone density correlated with cold (warm) temperatures in the lower stratosphere.

The contrast in the observed temperature structure between the northern and southern hemispheres is due to the relative lack of wave transport in the southern hemisphere. The winter temperature in the lower stratosphere inside the circumpolar vortex over Antarctica becomes closer to radiative equilibrium and consequently is more sensitive to radiative and/or dynamical perturbations than in the northern hemisphere [Tung et al., 1986; Mahlman and Fels, 1986]. In other words, while the radiative dynamical balance of the northern hemisphere is such that downward mass transport is almost always the case in the high-latitude lower stratosphere, the downward branch of the diabatic circulation in the southern hemisphere does not reach over the pole, but is instead located over subpolar latitudes. The circulation over Antarctica is near radiative equilibrium and may be either upward or downward in spring, depending on sensitive balances between the radiative input as the Sun returns and the adjustment of the atmospheric temperature to this and other radiative and dynamical perturbations. In recent years, the atmosphere over Antarctica in early springs is probably even closer to radiative equilibrium than in earlier years because of the lower amount of ozone observed, which gives rise to a smaller change in radiative heating as the Sun returns.

The observed latitudinal and seasonal structure of column ozone density is shown in Figure 1, taken from Bowman and

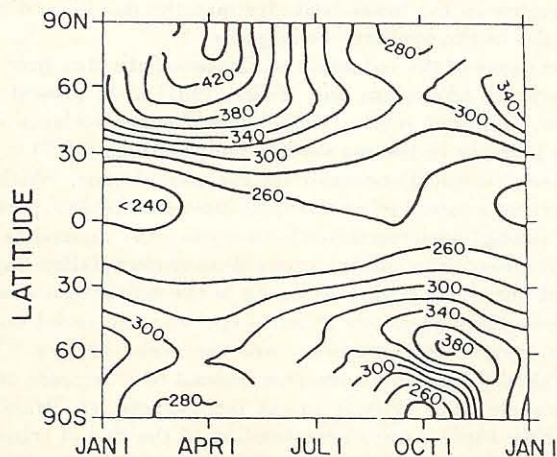


Fig. 1. The observed distribution, as a function of time and latitude, of the zonal-mean column ozone density (in Dobson units) averaged from TOMS data for the period from September 1978 to October 1982, taken from Bowman and Krueger [1985].

Krueger [1985]. The existence of the equatorial minimum is consistent with the cold tropical lower stratosphere, as discussed earlier. Similarly, the existence of high-latitude maxima is consistent with stratospheric conditions there. The seasonal variation of the high-latitude column density (in the northern hemisphere) is controlled by the varying intensity of the two-cell diabatic circulation pattern in the lower stratosphere (the downward branch over the Arctic being stronger during the winter than during the summer), and by the sign of the one-cell diabatic circulation which exists above 20–30 km, with rising motion in the summer hemisphere and downward motion in the winter hemisphere (see Figures 2a and 2b) [Dunkerton, 1978; Ko et al., 1985a, b; Stordal et al., 1985; Rosenfield and Schoeberl, 1986].

The seasonal reversal in the upper circulation occurs in spring and fall. Before the spring reversal, both the lower and upper circulations are downward over the (north) pole, thus leading to a build up of column ozone. This accounts for the spring maximum over the (north) pole [Ko et al., 1985b]. After spring, poleward transport of ozone is diminished by the reversal of the one-cell circulation above and the weakening of the winter circulation below. This situation continues until fall, leading to a seasonal minimum in fall of the high-latitude column density for ozone [Ko et al., 1985b]. The magnitude of the observed ozone seasonal cycle is a 40% decline from the spring maximum to the fall minimum.

The fall minimum in the southern hemisphere is similar to its counterpart in the northern hemisphere. As fall becomes winter, however, the contrast between the two hemispheres clearly manifests itself. The minimum ozone value attained in the fall over Antarctica is maintained, more or less, through the winter, enclosed inside the strong circumpolar vortex and isolated (to some extent) from the poleward transport from the equatorial region. In the northern hemisphere the winter polar vortex is often drastically disrupted by powerful wave events during mid-winter sudden warming episodes, when high-ozone air is transported into the Arctic lower stratosphere. In the southern hemisphere the poleward branch of the wave-driven diabatic circulation in the lower stratosphere terminates over the subpolar latitudes, creating the ozone maximum just outside the circumpolar vortex.

What happens in the spring over the middle and high latitudes in the southern hemisphere is the subject of much debate. However, circumstantial evidence indicates that if the same mechanism that explains the seasonal behavior of column ozone over other parts of the globe is to apply here, then the parallel intensification of the mid-latitude maximum and the deepening of the Antarctic minimum in spring in the climatology shown in Figure 1 is consistent with the development of a spring reverse circulation from pole to mid-latitudes [Tung et al., 1986; Stolarski and Schoeberl, 1986] for the years (1979–1982) that the climatology is based on (see Figure 2c). The possibility exists, however, that in recent years the seasonal minimum over Antarctica has further decreased in value as a result of chlorine chemistry and that the vertical motion in the spring may be suppressed by the lack of radiative heating due to the chemically depleted ozone amount.

The October monthly mean, zonal-mean column ozone over the pole has been found to decline from 280 Dobson units (DU) in 1979 to 170 DU in 1985 [Schoeberl et al., 1986].

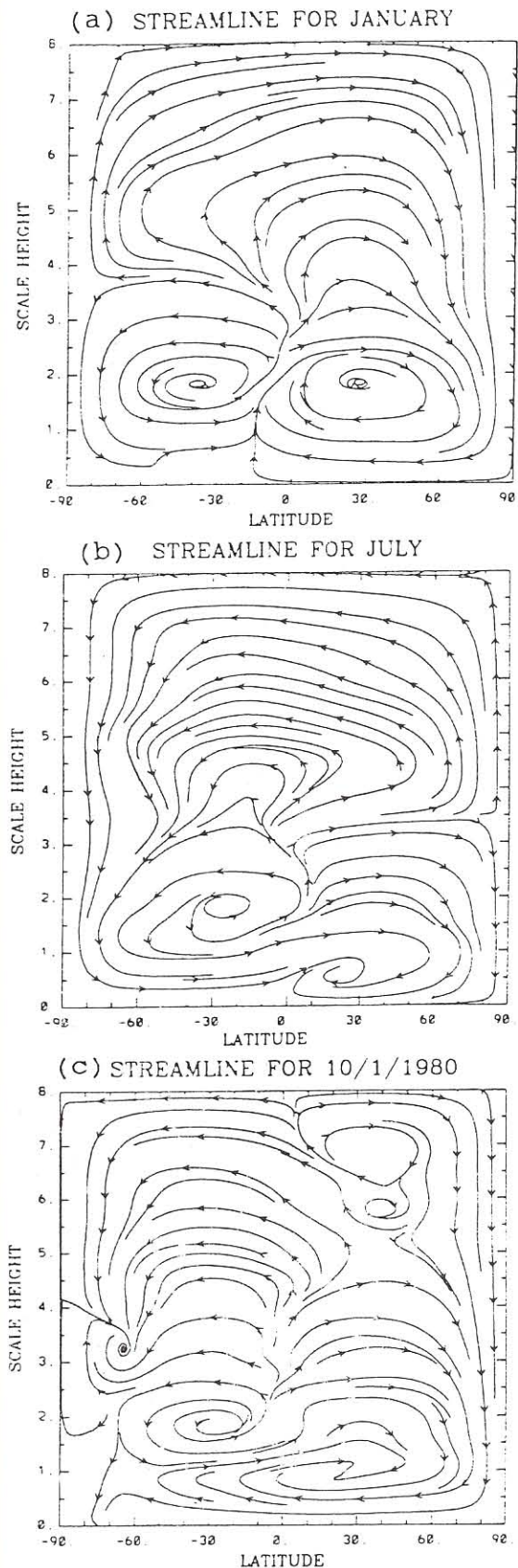


Fig. 2. Zonal-mean diabatic circulations for different seasons. Shown are model-generated diabatic streamlines based on Geller and Wu's [1987] 4-year temperature climatology for (a) January, (b) July, and (c) October 1980, using NMC data for the month for that year. The streamlines are calculated from (\bar{V}, \bar{W}) as if the latter were nondivergent.

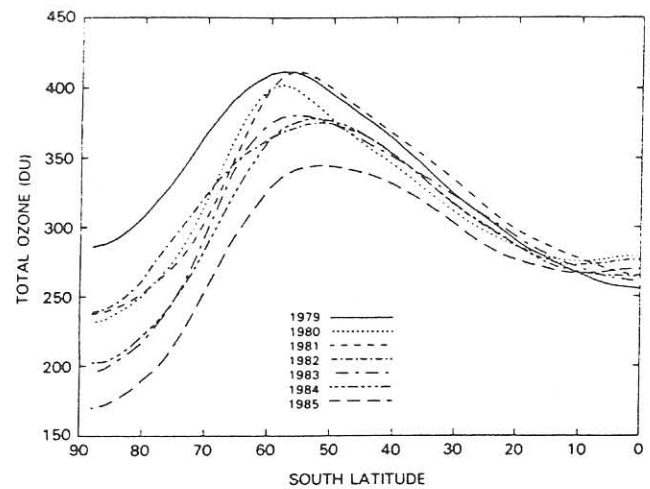


Fig. 3. The observed zonal-mean, monthly mean column ozone density as a function of latitude, for seven Octobers, taken from Stolarski and Schoeberl [1986].

(A later reprocessed data set [Fleig *et al.*, 1986] gave 300 DU for 1979 and 175 DU for 1985 (M. R. Schoeberl, personal communication)). As shown in Figure 3, taken from Schoeberl *et al.*, [1986], the decreases occurred not just over the pole, but also over the mid-latitudes, with some compensating increases over the tropics, although the correlation between the tropical increases and polar decreases was rather erratic from year to year. The year-to-year decline over the pole is by no means uniform but appears to exhibit a seesaw, up-down pattern, consistent with the phase of the tropical quasi-biennial oscillation [Garcia and Solomon, 1987].

Over the same period, significant cooling of the lower stratosphere in the mid- to high-latitude regions in October has been observed [Newman and Schoeberl, 1986; Mahlman and Fels, 1986].

Mahlman and Fels [1986] suggested that the temperature decrease in the lower stratosphere must have been caused by a substantial reduction of the wintertime planetary/cyclone scale wave activity in the troposphere since 1979. Accompanying such a diminution in wave activity is the reduction of the transport of ozone out of the photochemical production region in the lower latitudes into the middle and high latitudes in the southern hemisphere.

The cause of the reduction of Eliassen-Palm flux from the troposphere [Nagatani and Miller, 1987] is at present not known. Folland *et al.*, [1984] showed some evidence of a slight increase in the sea surface temperatures (SST) in the southern (tropical) oceans over the past decade. Without suggesting a causal relationship, Austin *et al.*, [1988] pointed to a "strong" anticorrelation between the SST anomalies and the October mean column ozone density over Halley Bay. If indeed there has been a warming of the southern oceans in the lower latitudes, one should expect an enhanced north-south temperature gradient over the ocean surface. This then should lead to an increase instead of a decrease of cyclone scale wave activity in the troposphere [cf. Mahlman and Fels, 1986] if our understanding of the role of transient eddy heat flux in controlling the north-south temperature gradient in the atmosphere is correct [Stone and Miller, 1980]. Nevertheless, as far as column ozone density is concerned, what matters is the large-scale wave activity in the lower stratosphere. A reduced penetration of large-scale

waves into the lower stratosphere would still lead to the cooling suggested by *Mahlman and Fels* [1986].

Given such large uncertainties concerning the long-term variations of wave driving (namely, Eliassen-Palm flux divergence) in the lower stratosphere, it is at present not feasible to trace the ultimate cause of the temperature change that has been observed for the past several years. Nevertheless, it is still meaningful to ask the question: Given the fact that the temperature has changed from year to year for the period 1979–1985, what, if any, dynamical effect would this have on Antarctic and global ozone concentrations? This forms the focus of the present investigation.

3. THE ROLE OF EDDIES

Even though the polar vortex over Antarctica is more zonally symmetric than its northern counterpart and the Antarctic lower stratosphere during winter and early spring is closer to radiative equilibrium than probably anywhere else in the Earth's atmosphere, it is still incorrect to assume that eddies are unimportant in forcing the zonal mean circulation there. It is precisely because the Antarctic lower stratosphere is so close to radiative equilibrium that the direction of its weak meridional circulation is sensitive to even weak eddy forcing. Furthermore, the polar vortex becomes very "leaky" (to poleward eddy transports of heat and ozone from lower latitudes) in October.

The net radiative heating rate, $\overline{Q}(\overline{T})$, where an overbar denotes zonal average, is here expanded in a Taylor series about the radiative equilibrium temperature, $T_e(y, z, t)$, and approximated as

$$\overline{Q} \simeq \alpha [T_e - \overline{T}(y, z, t)]$$

where $\alpha = -\partial\overline{Q}/\partial\overline{T} |_{T_e}$ is the coefficient of Newtonian cooling.

Tung et al. [1986] suggested that near the end of the long polar night (marked as $t = 0$ for the present discussion) inside the polar vortex, the temperature of the Antarctic lower stratosphere approaches the radiative equilibrium value, $T_e(y, z, 0)$. As the Sun returns, $T_e(y, z, t)$ increases in time for $t > 0$ due to the absorption of solar radiation by ozone. *Tung et al.* [1986] were led by some preliminary data, which showed that $\overline{T}(y, z, t)$ did not in general increase significantly, to hypothesize that $\overline{T}(y, z, t)$ remains close to $\overline{T}(y, z, 0)$ because it can "adjust only on a longer dynamical time scale, due to 'dynamical inertia'." If and when this situation happens, T_e will exceed \overline{T} sometime in the early Antarctic spring, leading to a net heating, i.e. $\overline{Q} > 0$. This in turn will lead to an upward zonal-mean (residual) velocity $\overline{w} \simeq \overline{Q}/\Gamma$, where Γ is the static stability.

Tung et al. [1986] did not state explicitly the causes of the so-called "dynamical inertia," alluding only to the fact that the temperature evolution is constrained by the prognostic zonal momentum equation.

Irreversible eddy forcing, in the form of an Eliassen-Palm flux divergence [see *Edmon et al.*, 1980], can, in principle, control the evolution of the zonal mean momentum, $\partial\overline{u}/\partial t$, and, through the thermal wind relationship, the evolution of zonal mean temperature, $\partial\overline{T}/\partial t$. This cause of "dynamical inertia" does not appear to have been investigated before in the present context.

This aspect of the problem has been obscured by the fact that even the zonally symmetric circulation has a weak dy-

namical inertia due to the so-called "flywheel effect:" i.e., dynamical inertia in the absence of eddy driving [*R. Snieder and S. B. Fels*, unpublished, as quoted by *Mahlman and Fels*, 1986]. The work of Snieder and Fels showed that the reversed circulation (with $\overline{w} > 0$), which develops because of the "flywheel effect," is too weak (5–10 m/day) and too late (sometimes in October) to play a significant role in the Antarctic ozone hole phenomenon.

Since the Antarctic lower stratosphere during spring is characterized by weak meridional circulations and weak "flywheel effects," the (weak) eddy forcing should not be ignored in our consideration of the zonal-mean meridional circulation and transport.

The temperature evolution, $\partial\overline{T}/\partial t$, can be shown to be governed by the following equation, obtained by combining the zonal-mean momentum, energy, and continuity equations in the quasi-geostrophic limit, with $\nabla \cdot F$ being the eddy Eliassen-Palm flux divergence:

$$\begin{aligned} & \left[\frac{\partial}{\partial y} \frac{\cos^2 \phi}{f^2} \frac{\partial}{\partial y} + \frac{\partial}{\partial z} \frac{1}{\rho_0} \frac{\partial}{\partial z} \frac{\rho_0 H}{R\Gamma} \right] \frac{\partial}{\partial t} \overline{T} \\ & = \frac{\partial}{\partial z} \frac{1}{\rho_0} \frac{\partial}{\partial z} \frac{\rho_0 H}{R\Gamma} \overline{Q} - \frac{\partial^2}{\partial z \partial y} \left(\frac{H}{\rho_0 f R} \nabla \cdot F \right) \end{aligned}$$

where $y \equiv a \sin \phi$; a is the Earth's radius, ϕ is latitude, $z \equiv H \ln(p_{00}/p)$, H is scale height, and $\rho_0(z) \equiv \rho_0(0)e^{-z/H}$. This equation shows that eddy forcing (the $\nabla \cdot F$ term) has a direct influence on the time evolution of the zonal-mean temperature. This is in addition to the effect of the (weak) imbalance between the time dependence of the radiative heating, in $T_e(y, z, t)$, and the time scale of radiative relaxation $\tau_R = \alpha^{-1}$ of temperature \overline{T} , as in $\overline{Q} = (T_e(y, z, t) - \overline{T})/\tau_R$. In the "flywheel effect," only the second mechanism is taken into account. The increase in radiative heating induces a temperature increase with nearly the same time scale (i.e., $\partial\overline{T}/\partial t \simeq \partial\overline{T}_e/\partial t$), so that $\overline{w} \simeq \overline{Q}/\Gamma - \partial\overline{T}/\partial t$ remains close to zero. This constraint is broken in the presence of eddy forcing.

Dunkerton [1988] showed that by imposing a $\nabla \cdot F$ consistent with the observed climatological distribution of Mechoso et al. [1985] in the upper troposphere, an upward mean residual velocity of a few times that due to the "flywheel effect" can be induced that extends into the lower stratosphere. Although there is considerable uncertainty associated with the specification of the eddy Eliassen-Palm flux divergence, a poorly determined quantity from observational data in the southern hemisphere, Dunkerton's work suggests that wave forcing of zonal-mean circulation should not be ignored, even in the Antarctic environment.

4. THE PRESENT APPROACH

In light of the discussion in section 3, the logical quantity that one needs to know in order to characterize the zonally averaged circulation and transport is the eddy Eliassen-Palm flux divergence, in particular, its seasonal evolution and year-to-year change. Knowing this quantity, one can prognostically determine the zonal momentum from the momentum equation, and the zonal-mean temperature \overline{T} from the thermal wind (or gradient wind) relationship. After \overline{T} is determined, a radiative calculation then yields the radiative heating \overline{Q} , which together with the calculated $\partial\overline{T}/\partial t$, determines the zonal-mean residual vertical and horizontal

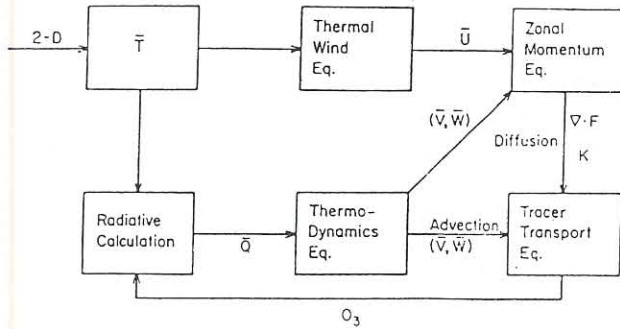


Fig. 4. Schematic algorithm for the coupled two-dimensional model of transport, radiation, and chemistry.

velocities with the aid of the continuity equation. This forms the advective part of the transport for the zonal-mean ozone mass mixing ratio. The diffusive part of the ozone transport is deduced using the eddy Eliassen-Palm flux divergence by considering the horizontal flux of Ertel's potential vorticity. (This procedure was illustrated in Figure 2 of *Tung* [1987]).

Unfortunately, the kind of detailed information on the observed Eliassen-Palm flux divergence is not presently available. We shall consequently adopt an alternative, diagnostic approach, which, although less intuitive, is nevertheless equivalent to the first approach as far as two-dimensional models are concerned. The input to the model is the two-dimensional quantity \bar{T} (and $\partial\bar{T}/\partial t$), from observational data. From this, the radiative heating \bar{Q} is calculated and the Eliassen-Palm flux divergence is deduced from the zonal-mean momentum equation as the amount of eddy forcing needed to maintain the observed (input) zonal-mean temperature departure from radiative equilibrium. The procedure is illustrated in Figure 4.

In Figure 5 we show the magnitude of the eddy Eliassen-Palm flux divergence ($-\nabla \cdot F$) in the Antarctic lower stratosphere, deduced from the monthly mean October National Meteorological Center (NMC) temperatures kindly made available to us by Paul Newman (see also *Newman and Schoeberl*, [1986]), assuming that the radiative property of the atmosphere has not changed (see the discussion in sec-

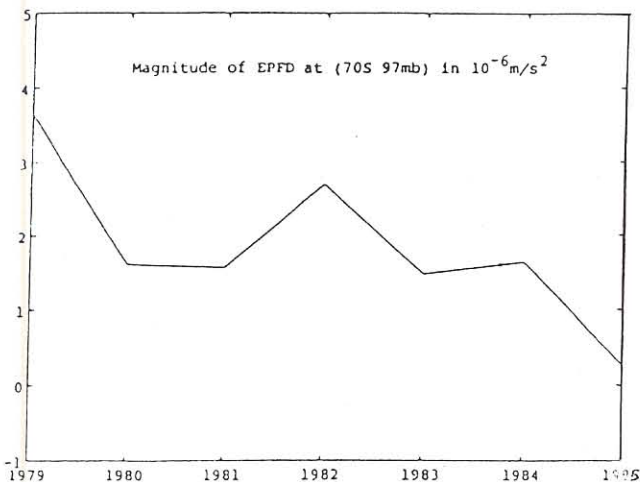


Fig. 5. The negative of the eddy Eliassen-Palm flux divergence at 70°S at 100 mbar deduced from the monthly mean October NMC temperatures for 7 years.

tion 11). It shows an order of magnitude reduction of the eddy forcing from 1979 to 1985. Such a change will undoubtedly have a significant impact on the Antarctic circulation and the ozone transport, as will be shown later. However, it should be noted that most of the reduction for this period occurs only during 2 years, from 1979 to 1980 and from 1984 to 1985. In other words, the large dynamical component of the change in the period 1979–1985 is probably due largely to the fact that eddy activity in 1979 was abnormally active and in 1985 was abnormally quiet. For the intervening years, eddy driving varies by less than a factor of 2 and does not show any systematic year-to-year downward trend. An assessment of the effect of changes in the radiative properties of the atmosphere on our model results will be given in section 11.

5. THE NUMERICAL MODEL

The model used is the so-called “third-generation” zonal-mean model of tracer transport, in which all transport parameters are self-consistently calculated from a prescribed zonal-mean temperature $\bar{T}(y, z, t)$, fitted into Fourier series in space and time from observational data. The formulation of the transport equation in isentropic coordinates is given by *Tung* [1982, 1984] and *Ko et al.* [1985a]. The coupled model dynamics is discussed by *Tung* [1986ab, 1987]. The model has recently been modified in the troposphere with the use of σ -like coordinates to handle the underground isentropes.

The aim of the present exercise is to “hindcast” the ozone change during the period 1979–1985, given the (observed) temperature as an input. The temperature input fixes all the transport parameters in this model. (No tunable transport parameter remains.) By changing the temperature from year to year, as well as from season to season, the transport parameters are changed and ozone is affected as a consequence. We would like to quantify the amount of ozone change corresponding to this temperature change. This we call the dynamical component of ozone change. The technical details of the model are described in the appendix. Here the equations used are listed.

The Zonal-Mean Equations in Isentropic Coordinates

Define a log- θ coordinate z_{\uparrow} as [see *Holton*, 1986]

$$z_{\uparrow} = H\kappa^{-1} \ln\left(\frac{\theta}{\theta_0}\right) \quad (1)$$

where θ_0 is a constant. This coordinate is used for the model stratosphere ($\theta \geq 350$ K). The use of this coordinate in the troposphere leads to the well-known problem of coordinate surfaces intersecting the ground. This problem can be solved by using the method of “underground isentropes” [*Lorenz*, 1955; *Andrews*, 1983], but the remedy is computationally uneconomical because grid points are wasted underground. Our solution is to modify θ_0 to be the (variable) surface mean potential temperature for θ below 350 K. To simplify the discussion in this section, the equations are given in terms of (1) with $\theta_0 = \text{constant}$. (See the appendix for the exact formulae involving the “ σ -like” coordinates used in the numerical model.)

In this coordinate system the column density of ozone will be given by

$$\Omega = \int_0^\infty \sigma \chi dz_\dagger$$

where

$$\sigma \equiv \rho \frac{\partial z}{\partial z_\dagger}$$

is the "density" of air in the z_\dagger -coordinate system, and χ is the mass mixing ratio of ozone. In terms of the concentration of ozone per unit "volume," $f \equiv \sigma \chi$, we have the following equations:

Zonal-Mean Transport

$$\begin{aligned} \frac{\partial \bar{f}}{\partial t} + \frac{\partial}{\partial y} \left(\frac{\bar{V} \bar{f}}{\bar{\sigma}} \right) + \frac{\partial}{\partial z_\dagger} \left(\frac{\bar{W} \bar{f}}{\bar{\sigma}} \right) \\ - \frac{\partial}{\partial y} K_{yy} \cos^2 \phi \frac{\partial \bar{f}}{\partial y} = \bar{S} \end{aligned} \quad (2)$$

where $y = a \sin \phi$, a is the Earth's radius, ϕ is latitude, and $(\bar{V} = \overline{\sigma v} \cos \phi, \bar{W} = \overline{\sigma z_\dagger})$ is the mass circulation. The coefficient K_{yy} is the so-called isentropic mixing coefficient.

The chemical source term, \bar{S} , is approximated by

$$\bar{S}(\bar{f}) \simeq \bar{S}(0) + \frac{\partial \bar{S}(0)}{\partial \bar{f}} \bar{f} + \frac{1}{2} \frac{\partial^2 \bar{S}(0)}{\partial \bar{f}^2} \bar{f}^2 + \dots \quad (3)$$

The quadratic term is small (for standard ozone chemistry) (M. Ko, personal communication, 1987). We have experimented (see the appendix) with several forms of parameterized chemistry schemes and found our result to be insensitive to the form chosen. This is because at high latitudes during winter and spring (the period of interest in the present study) the chemical relaxation time is longer than a year (in terms of homogeneous chemistry) and so chemistry should play a secondary role. The form used here is similar to that of *Ko et al.* [1985b], with a minor modification which limits the chemical relaxation time to 1.5 years, affecting mainly the region near the tropopause and over the poles during the polar night. The original version used by *Ko et al.* [1985b] has no chemical loss in these regions.

Thermodynamics

The thermodynamic equation takes the form of

$$\bar{W} = \bar{\sigma} \bar{Q} / \Gamma \quad (4)$$

where $\Gamma = \kappa \bar{T} / H$, and

$$\bar{Q} = \alpha (T_e - \bar{T}) \quad (5)$$

is the Newtonian form of the net diabatic heating rate. The Newtonian cooling coefficient α is constructed by fitting an analytic function of latitude, height, and season to data of *Kiehl and Solomon* [1986] and *Wehrbein and Leovy* [1982] (see the appendix). The radiative equilibrium temperature T_e is from S. B. Fels and M. D. Schwartzkopf (manuscript in preparation, 1986), as cited by *WMO* [1986], fitted to an annual cycle. No year-to-year variation in T_e and α is incorporated. The effect of the lack of feedback between ozone reduction and radiative heating will be estimated in section 11.

Mass Conservation

$$\frac{\partial}{\partial t} \bar{\sigma} + \frac{\partial}{\partial y} \bar{V} + \frac{\partial}{\partial z_\dagger} \bar{W} = 0 \quad (6)$$

Gradient Wind

$$(2\Omega + \frac{\bar{u}}{a \cos \phi}) \sin \phi \frac{H}{\kappa} \frac{\partial}{\partial z_\dagger} \bar{u} = -\cos \phi \frac{\partial}{\partial y} \bar{T} \quad (7)$$

Zonal Momentum

$$\bar{\sigma} \frac{\partial \bar{L}}{\partial t} + \bar{V} \frac{\partial \bar{L}}{\partial y} + \bar{W} \frac{\partial \bar{L}}{\partial z_\dagger} = \nabla \cdot F \quad (8)$$

where

$$\bar{L} = [\bar{u} + \Omega a \cos \phi] \cos \phi$$

is the absolute angular momentum (divided by a) per unit mass, and $\nabla \cdot F$ is the eddy Eliassen-Palm flux pseudo-divergence, as defined by *Tung* [1986b]. This quantity is equal to the isentropic flux of Ertel's potential vorticity [*Tung*, 1986b], which is treated as another tracer and is thus parameterized as [see *Tung*, 1986b, 1987; *Andrews et al.*, 1987]:

$$\nabla \cdot F = K_{yy} \cos^2 \phi \bar{\sigma}^2 \frac{\partial}{\partial y} \frac{1}{\bar{\sigma}} \frac{\partial \bar{L}}{\partial y} \quad (9)$$

Hydrostatic Ideal Gas

$$\bar{\sigma} = -\frac{1}{g} \frac{\partial}{\partial z_\dagger} \bar{p} \quad (10)$$

with

$$\bar{p} = p_{\infty} \left(\frac{T}{\theta} \right)^{1/\kappa} \cong p_{\infty} e^{-z_\dagger/H} \left(\frac{T_e}{\theta_o} \right)^{1/\kappa} \left(1 + \kappa^{-1} \frac{\bar{T} - T_e}{T_e} \right)$$

The algorithm for solving these coupled equations is as follows. Referring to the flow-chart in Figure 4, we input the zonal-mean temperature, from which \bar{u} is diagnosed using the gradient wind relationship (7), and $\bar{\sigma}$ is calculated using the hydrostatic ideal gas formula (10). The thermodynamics equation (4) yields the vertical mass flow rate \bar{W} , and the mass conservation equation (6) then gives the horizontal mass flow rate \bar{V} . These form the advective transport for the tracer equation (2). The diffusive transport is calculated using the same input \bar{T} via the zonal momentum equation (8), and the parameterization (9), yielding K_{yy} .

6. INPUT DATA

The input data consist of monthly mean zonal-mean NMC temperatures [*Newman and Schoeberl*, 1986] for the southern hemisphere for the months of August, September, October, and November for each of the years in the period 1979–1985. Over the northern hemisphere, and also for the southern hemisphere during nonspring seasons, the data were taken from the climatology of *Geller and Wu* [1987], for the months of January, April, July, and October based on 4 years of National Oceanic and Atmospheric Administration (NOAA)/NMC data for the period from December 1978 to November 1982. No year-to-year changes were incorporated for the northern hemisphere and for nonspring months in the southern hemisphere. The two data sets are extrapo-

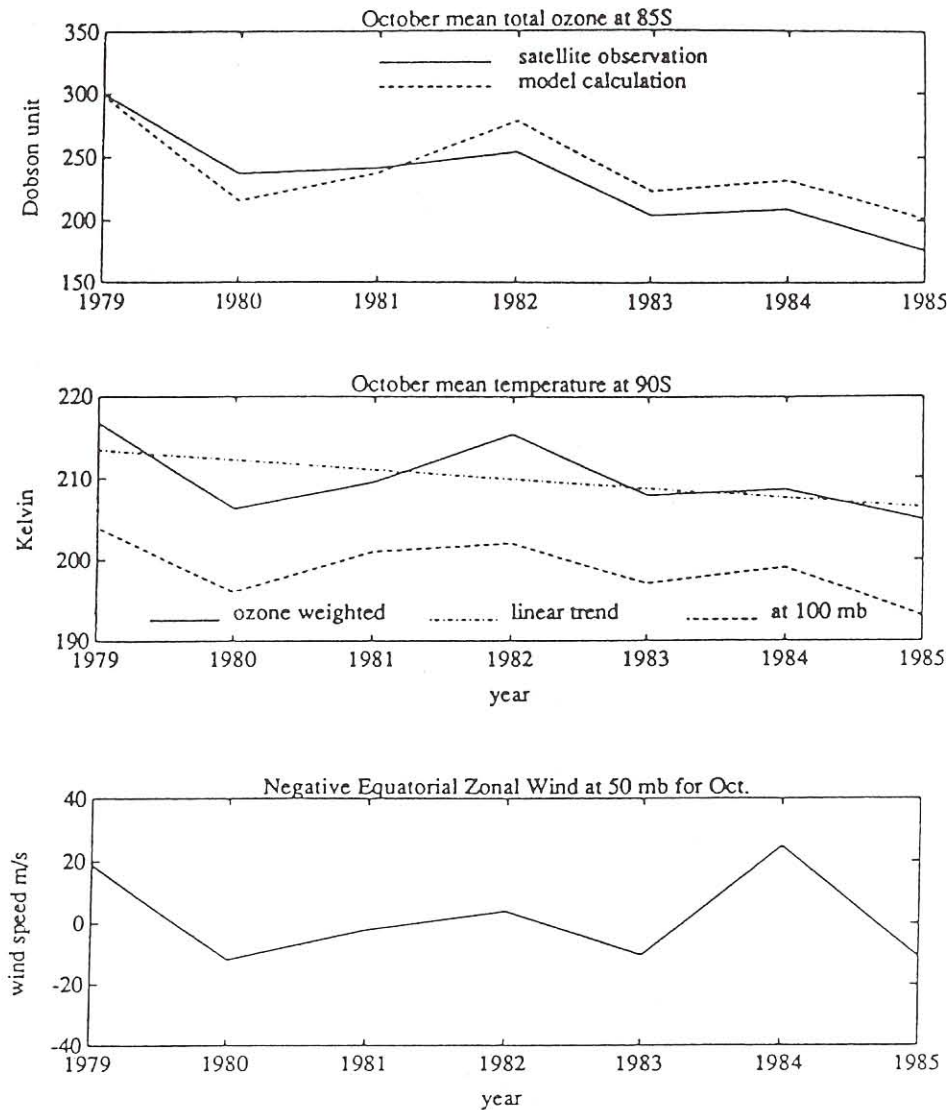


Fig. 6. (Top) The October mean, zonal-mean column ozone at 85°S. Solid curve shows TOMS data (from M. R. Schoeberl, personal communication, 1987); dashed curve indicates present model. (Middle) The October mean, zonal-mean temperature at 90°S: temperature at 100 mbar (dashed curve); temperature field weighted by the observed ozone partial pressure (solid curve); linear trend of the weighted temperature (dash-dot curve). (Bottom) The negative of the equatorial zonal wind at 50 mbar [from Garcia and Solomon, 1987].

lated across the equator and are fitted into a Fourier series in both space and time. (The zonal averages of Newman and Schoeberl were performed on isobaric surfaces, while the model requires zonal averages to be performed on isentropic surfaces. We have ignored such differences for \bar{T} here. The difference should be small for temperature changes with large vertical scales.)

In the calculation for K_{yy} , regions where the zonal-mean gradient of Ertel's potential vorticity changes sign may appear if unprocessed data are used. In the real atmosphere, these would indicate that the zonal flow is unstable to barotropic, baroclinic, or inertial instabilities. Unstable waves presumably will grow at the expense of the zonal-mean shear, which would presumably adjust to a neutral or stable configuration. In our procedure the mean zonal wind is simply smoothed by discarding the highest few Fourier components in \bar{u} , so that no localized unstable region appears in the model.

In the easterly zonal wind region, negative K_{yy} are set to zero with the assumption that isentropic mixing is predominantly caused by stationary planetary waves, which do not propagate into the easterly zonal wind region. The eddy deceleration of the easterly jets are presumably caused by other waves, such as breaking gravity waves [Lindzen, 1981], which mix across isentropes. The effect on column ozone is minimal, as this occurs above 35 km.

7. THE QUASI-BIENNIAL OSCILLATION SIGNAL

The temperature input data for the southern hemisphere appear to have a quasi-biennial signal. Garcia and Solomon [1987] pointed out that both the Antarctic minimum temperature at 100 mbar and the minimum column ozone values were correlated with the phase of the equatorial QBO, as indicated by the sign of the 50-mbar zonal wind over Singapore, for the period examined, 1979–1986. In Figure 6

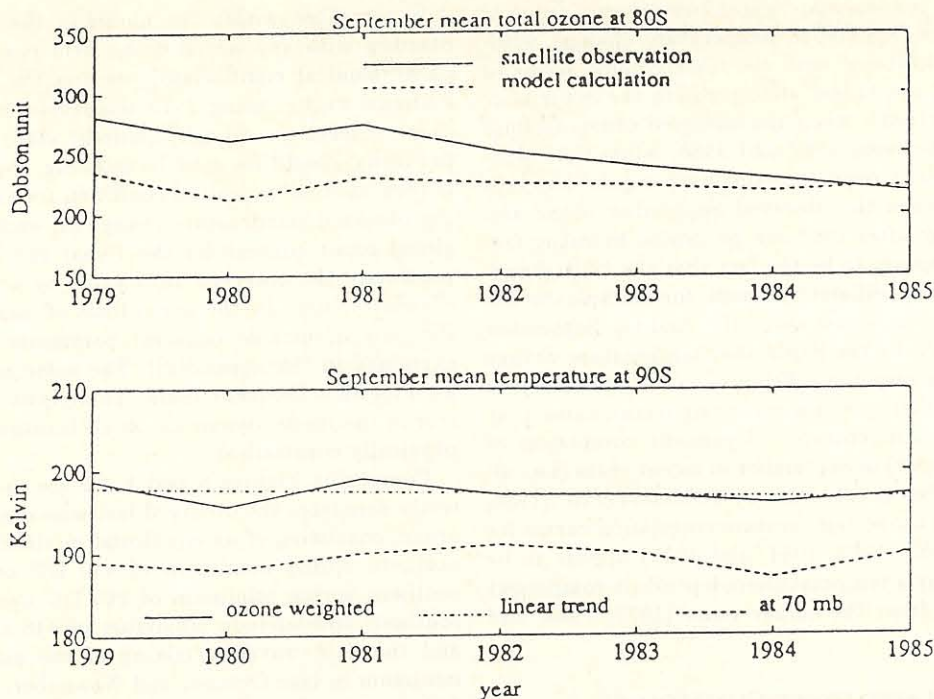


Fig. 7. (Top) The September mean, zonal-mean column ozone at 80°S . Solid curve indicates TOMS data (from M. R. Schoeberl, personal communication, 1987); dashed curve indicates present model. (Bottom) The September mean, zonal-mean temperature at 90°S : temperature at 70mbar (dashed curve); temperature field weighted by the observed ozone partial pressure (solid curve); linear trend of the weighted temperature (dash-dot curve).

we have plotted with the same format the zonal-mean temperature at 100 mbar [from Newman and Schoeberl, 1986] and zonal-mean column ozone at 85°S [from Schoeberl *et al.*, 1986]. The QBO signal in the zonal-mean quantities appears to be even more pronounced than in the minimum quantities.

There is as yet no definitive measure for the phase of the equatorial QBO. The 50-mbar wind direction used by Garcia and Solomon [1987] may be an imperfect measure because during some years the zonal winds in the equatorial lower stratosphere may have different signs at different levels. Dunkerton *et al.* [1988] suggested using the depth of the easterlies as a measure. For the period under consideration, 1979–1985, the two indices appear consistent with each other.

Although it has not been established definitively that the high-latitude quasi-biennial signals in temperature and ozone are caused by the equatorial QBO of the zonal winds, our present work does not depend on there being such a causal relationship. The question we will address is: Given that there is such a year-to-year fluctuation in the observed zonal-mean temperature field, how much of the year-to-year fluctuation in column ozone can be attributed to the dynamical effects of the temperature change?

Tung and Yang [1988] showed analytically that in the absence of chemical changes, one should expect that the column ozone changes be correlated quantitatively with the temperature changes weighted by the partial pressure of ozone. In Figure 6 we have also plotted such a weighted average of the observed temperature field using the August 19, 1986, ozone partial pressure observed by Komhyr *et al.* [1988] as the weighting function. The correlation between the year-to-year variation of this temperature change

and the observed (and model-calculated) ozone change is remarkable. All three curves increase and decrease together and in phase with the equatorial QBO for all the years examined (1979–1985). We will return to a more detailed discussion of Figure 6 in a later section.

There has been some controversy and confusion concerning the temperature trend (see the review by Solomon [1988]). The debate concerns mainly the fact that the decline of 18°K in October between 1979 and 1985 reported by Newman and Schoeberl [1986] appears to be larger than those reported by other authors, which are generally in the 5 to 10°K range. It appears to us that the "discrepancy" is mainly a result of comparing different quantities. The 18°K decline reported by Newman and Schoeberl is not a linear trend but is a simple difference between October 1979 and October 1985. It is furthermore at a higher level, approximately at 10–30 mbar. Other reports usually refer to the 100-mbar level.

At the 100-mbar level, Newman and Schoeberl's data give about 11°K as the simple difference between October 1979 and October 1985 (see Figure 6). The more important quantity, the temperature weighted by the observed ozone partial pressure, has a simple difference for the two Octobers of 12°K (see Figure 6) and a linear trend for the seven Octobers of 7°K , in the range reported by most authors. Since we are more interested in year-to-year fluctuations, the linear trend is not emphasized in the present study.

Figure 6 shows that the October mean column change is correlated with the October mean temperature change. It has often been mentioned that the correlation for September is much less. The September temperature change is smaller than the October change. It is often in phase with the October change (except in 1984 and 1985) but has very little linear trend for the period 1979–1985 (see Figure 7).

The calculated year-to-year September column ozone is correlated with the September temperature change, when the calculation is initiated with the same column ozone in August. However, the model underpredicts the September ozone decline after 1982, when the observed ozone declines by about 30 DU between 1982 and 1985, while the calculated September mean ozone remains unchanged. The lack of correlation between the observed September ozone and temperature change after 1982 can be caused by many factors. One cause appears to be the fact that the "initial condition" for each year is different (caused, for example, by different temperatures in winter and fall). And the September ozone change caused by the September temperature change is too small to overcome the differences in initial conditions for each year. Another, perhaps more important, cause may be that there is a temperature-independent component of change (i.e., chemistry) in September in recent years (i.e., after 1982). It has been pointed out by Schoeberl *et al.* [1986] that October mean ozone-temperature correlation curves for the later years (1982, 1983, 1984, and 1985) appear to be shifted downward by a temperature-independent component (of about 30 DU) from the earlier years (1979, 1980, and 1981).

8. SIMULATED OZONE CLIMATOLOGY

Figure 1 shows the observed climatology of Bowman and Krueger [1985] of the zonal-mean column ozone using satellite data. The climatology is based on the 4-year period from October 1978 to September 1982. A spring ozone minimum over Antarctica (the "ozone hole") is already apparent in these years, although the seasonal minimum reached is not as low as that attained in the recent few years.

In Figure 8 our model climatology is presented. It is based on the 4-year period from January 1979 to December 1982. (We do not have the temperature data for 1978). Since we do not have the ozone initial condition for our model runs,

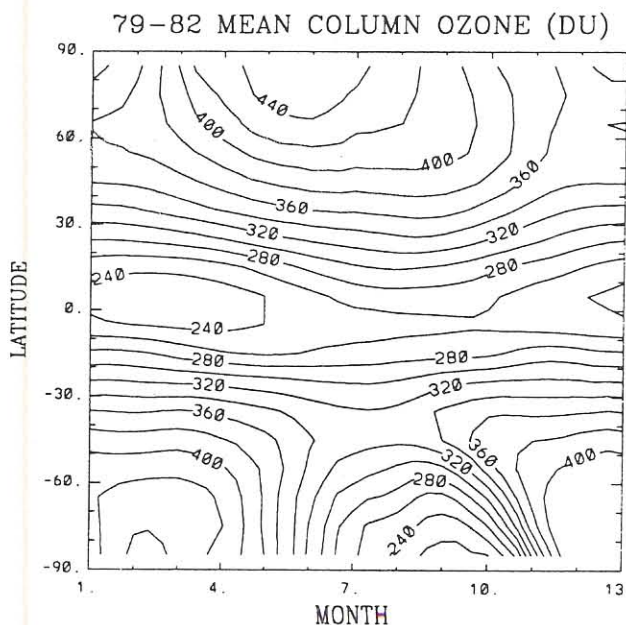


Fig. 8. The model climatology of zonal mean column ozone density, as a function of time and latitude, based on the 4-year average of 1979–1982 in the 7-year time-dependent run.

it is generated within the model in the following manner. Starting with any initial ozone field (we used that due to photochemical equilibrium), we run the model through 4–7 annual cycles, using 1979 temperatures, until the model ozone reaches an annually periodic state (and so "forgets" the initial condition used in spinning up the model). This is then used as our initial condition for the 7-year run with the observed temperature change for each year. The initial global ozone content for the 7-year run is adjusted to approximate the observed 1979 value by adjusting the model chemistry (namely, the upper limit of chemical loss lifetime, the only adjustable constant parameter in the model, as explained in the appendix). The same model chemistry is used for all subsequent years. There is no adjustable parameter in the model dynamics, as all transport parameters are physically constrained.

Comparing Figures 8 and 1, we see that our model correctly simulates the observed latitudinal gradient of column ozone, consisting of an equatorial minimum of 240 DU, the northern spring maximum of 440 DU over the pole, the southern spring minimum of 240 DU over Antarctica, the southern mid-latitude maximum of 340 DU in September, and the final warming/mixing of the maximum with the minimum in late October and November. The main model deficiency involves the duration of the late spring maximum over both poles after the final warming. The model maxima last about 2–3 months too long compared with the observed climatology. This is apparently due to the lack of time resolution of our temperature input data and the smooth extrapolation used during these periods of rapid seasonal change. We plan to remedy this situation in the near future with daily temperature inputs. Since our emphasis is on the early spring in the southern hemisphere, most of our time resolution is placed in this period, and consequently, the seasonal time variation in the early spring appears to be adequately simulated up to and including the early phase of the final warming.

An examination of the model diabatic circulation (see Figure 2) and of the associated ozone transports appears to confirm the validity of the mechanisms discussed in section 2, concerning the causes of the seasonal and latitudinal variations of the observed column ozone over the globe.

In Figure 9 the model-calculated net radiative heating rates for January, July, and April are shown. In Figure 10 the same quantity for the four individual Octobers in the period 1979–1982 are presented. The vertical coordinates have been resorted to an approximate log-pressure coordinate, using the relationship:

$$\theta \approx \bar{T} \left(\frac{p_{00}}{p} \right)^{R/c_p}$$

In Figure 11 the observed (SBUV) ozone mixing ratio in the latitude-height plane for four seasons in 1979 is shown [from McPeters *et al.*, 1984]. Our model-calculated ozone mixing ratio for 1979 is shown in Figure 12. The comparison is reasonably favorable.

9. SIMULATED YEAR-TO-YEAR CHANGES

The observed latitudinal distribution of zonal-mean, October mean column ozone for the 7 years 1979–1985 [from Schoeberl *et al.*, 1986], has been shown in Figure 3; the

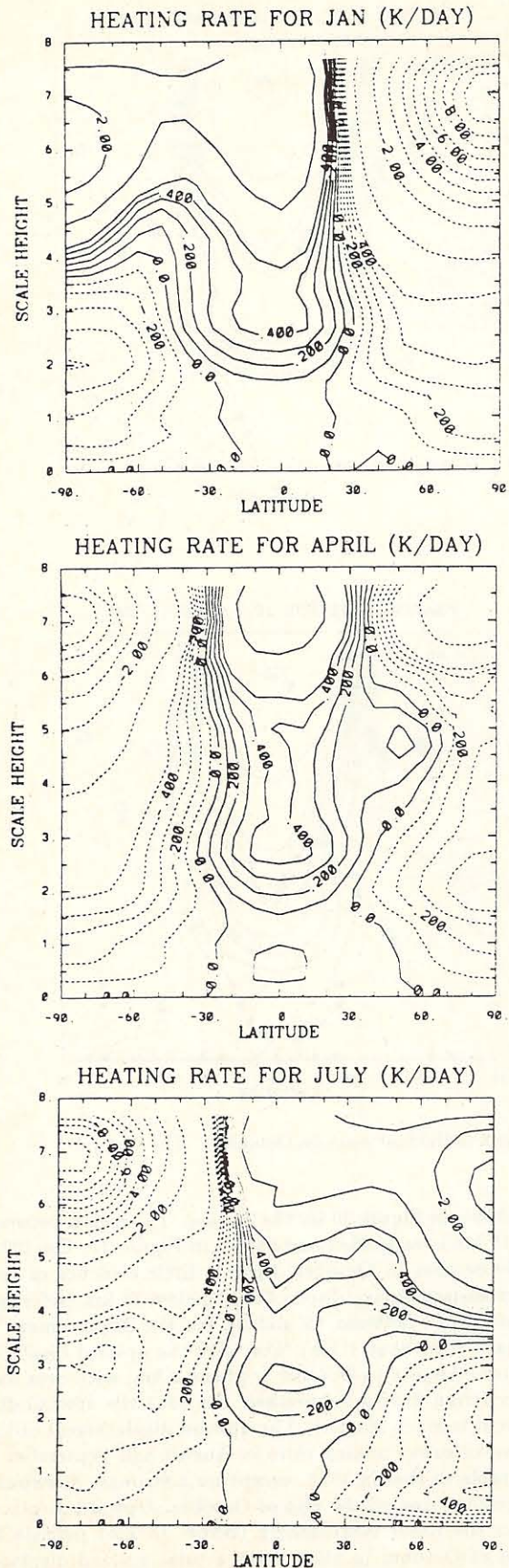


Fig. 9. The model-calculated net radiative heating rates for January, April, and July, based on the 4-year climatological temperature of Geller and Wu [1987].

corresponding quantity simulated by our model is given in Figure 13. In general, the large-scale nature of the ozone decline from 1979 to 1985 is simulated. In particular, the large negative difference between October 1985 and October 1979 over the pole and mid-latitudes, and the small positive difference over the tropics is well simulated. At 85°S the model ozone decreases by about 100 DU, from 300 DU in 1979 to 200 DU in 1985, while the observed zonal-mean ozone dropped by 125 DU, from 300 DU in 1979 to 175 DU in 1985. Over the middle latitudes the model yields declines of about half that over the pole. At 60°S the model ozone declines by 50 DU, from 375 DU in 1979 to 325 DU in 1985. TOMS data showed a decline of 85 DU, from 410 DU in 1979 to 325 DU in 1985. Over the pole the underprediction of the observed decline is caused by the model ozone falling short of the observed low values in 1985 by 25 DU, while over the middle latitudes the underprediction of the observed difference is caused by the weaker simulated latitudinal gradient in 1979. The observed value of 325 DU at 60°S in 1985 is well simulated by the model.

The comparison of model and observed column ozone for individual years (Octobers) has been shown in Figure 6, which displays the good agreement in the quasi-biennial oscillations of the simulated and observed ozone.

The simulation of individual seasonal cycles from August to November for the 7 years is shown in Figure 14 for 85°S and in Figure 15 for 75°S. The way our model is initiated yields no significant differences in August 1 ozone values. Published data on the observed zonal-mean seasonal cycle for the individual years (other than the climatology shown in Figure 1) was given by Schoeberl *et al.* [1986] for 1979–1983. The zonally averaged TOMS data for later years have been provided to us by Mark Schoeberl of NASA. The seasonal behavior for the ozone map minimum was described by Schoeberl [1987].

Compared with the TOMS data, the simulation for 1982 is the worst of all the years examined (see Figure 16). The over-prediction (of about 25 DU) of the October mean polar column ozone for this year, as can be seen in Figure 6, is found to be due to an early warming and mixing (in both the model and the real atmosphere), from which the atmosphere recovered but the model does not, because of a lack of time resolution in the input temperature data. This problem can be corrected for 1982 using 5-day mean temperature data instead of the monthly mean data used here (not shown).

The simulated seasonal behavior of zonal-mean ozone for 1983 is plotted in Figure 17 between the TOMS' data for the minimum over all longitudes and the maximum over all longitudes, from Stolarski *et al.* [1986]. Good agreement is found between the model zonal-mean column ozone and the TOMS zonal mean. The agreement becomes excellent if the simulated ozone is shifted by 5 days to the right.

Figure 17 also serves to illustrate the confusion that can be caused by the (incorrect) comparison of the zonal-mean result from a two-dimensional model to the observed minimum over all longitudes. The observed seasonal decline of the zonal mean from September 1 to the lowest value of slightly less than 200 DU in the middle of October is less severe than that occurring in the minimum regions.

Figure 6 shows that the observed October means of 1984 and 1985 are both lower by about 25 DU than the simulated values. However, the cause for the overprediction is different for the 2 years. An examination of the simulated seasonal

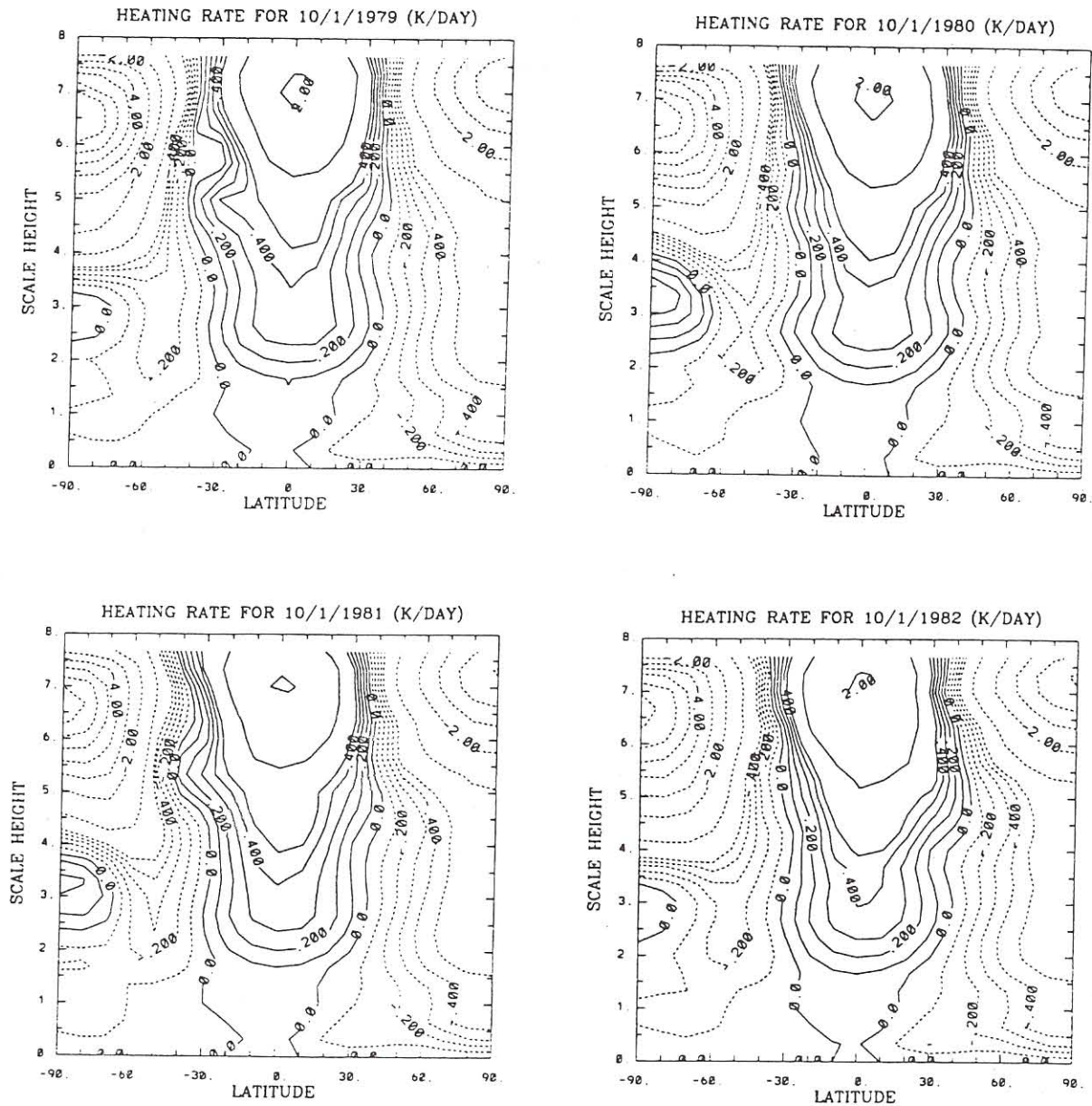


Fig. 10. The model-calculated net radiative heating rates for the 4 individual years on October 1. The vertical coordinate in pressure scale heights is approximate.

behavior for the zonal-mean column ozone shows that for 1984 (Figure 18) the observed minimum value reached in the beginning of October is correctly simulated. However, during the second half of October the model final warming occurs earlier and the mixing is stronger than that observed, leading to an overprediction of the October monthly mean. For 1985 (Figure 19) the timing of the occurrence of the seasonal minimum and the occurrence of the final mixing event are correctly simulated. The overprediction seen in Figure 6 can be attributed to an underprediction of the September decline, suggesting that perhaps the neglect of heterogeneous chemistry might have become increasingly incorrect in September in recent years.

10. VERTICAL DISPLACEMENTS

The seasonal evolution of the zonal-mean model ozone mass mixing ratio at 85°S from August to the end of Octo-

ber is shown in Figure 20 for the 7 years. The region between 8 and 25 km is magnified and shown in Figure 21. For 1979, from September to October, there is little evidence of vertical (cross-isentropic) displacement below 18 km (referring to Figure 21). Between 19 and 22 km the displacement is downward (by about 1 km). For 1980 the upward displacement from August to October is about 2 km, with over half of it occurring during September. For 1981 the upward displacement is much smaller. The upward displacement of the zonal-mean ozone mixing ratio in August and September is comparable to that in 1980, except for a stronger downward displacement toward the end of October. Upward displacement in the lower stratosphere (below 18 km) persists to the end of October in 1983, with a total upward displacement of between 2 and 3 km. For this year the displacement in September for the whole region between 8 to 25 km, while for 1984 the displacement is upward below 18 km and slightly downward above. For 1985 the upward dis-

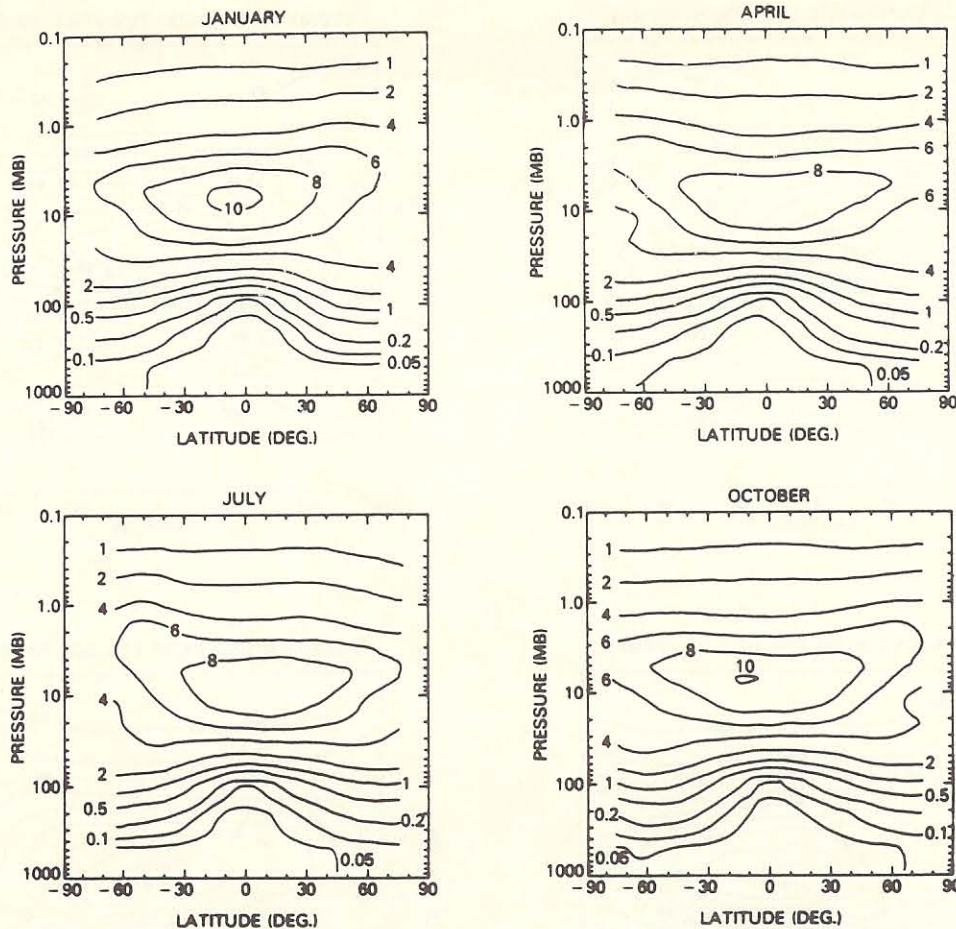


Fig. 11. The observed (SBUV) zonal mean ozone mixing ratio in latitude-height plane for 4 months in 1979 [from *McPeters et al.*, 1984].

placement is about 2–3 km and persists to the beginning of November below 18 km.

In Figure 20 we show the model mass mixing ratio for an expanded altitude region between 0 and 56 km. Above 30 km the vertical displacement is larger than below and has large seasonal and year-to-year fluctuations. (But density is lower in these regions and so the effect on column density is smaller.)

In Figure 22 the zonal-mean vertical velocity (in isentropic coordinates) of the diabatic circulation responsible for the above-mentioned ozone displacement is shown. Figure 22 shows that the vertical velocity in September and October becomes positive in the lower stratosphere (reaching a maximum value of about 30 m/day) for all years except 1979, when it becomes weakly positive only in October. The vertical velocity is strongly downward above the 30-km level (the magnitude of the vertical velocity being about almost 2 orders of magnitude stronger than that in the lower stratosphere).

Figure 22 further shows that prior to September the vertical velocity is downward throughout the model stratosphere and troposphere. This vertical motion is partly responsible for the so-called “preconditioning” of the lower stratosphere, where the mixing ratio profiles of many long-lived species are depressed downward relative to their mid-latitude counterparts. In the northern hemisphere, comparable and even stronger downward velocity also exists in-

side the polar vortex. However, the preconditioning is not as pronounced because of the presence of horizontal mixing from the mid-latitudes. In the southern hemisphere, mixing of mid-latitude air into the polar vortex is not as strong. Consequently, the air found inside the polar vortex in the lower stratosphere in winter and early spring may represent “aged” air from the upper stratosphere inside the polar vortex.

11. DISCUSSIONS

Our understanding of the behavior of ozone in the Antarctic lower stratosphere is admittedly limited. The present study is a preliminary attempt at quantifying the ozone change during the period 1979–1985 by dynamical causes only, without incorporating the additional radiative effects of volcanic aerosols and polar stratospheric clouds and without heterogeneous chemistry.

The dynamical mechanism that leads to the year-to-year decrease in column ozone in the Antarctic region is the cooling that has occurred in the lower stratosphere between 1979 and 1985 (about 12°K). This cooling increases the value of the difference $T_e - \bar{T}$, and favors a net heating

$$\bar{Q} \approx \alpha(T_e - \bar{T}) > 0$$

in spring when T_e increases, which then yields a stronger up-

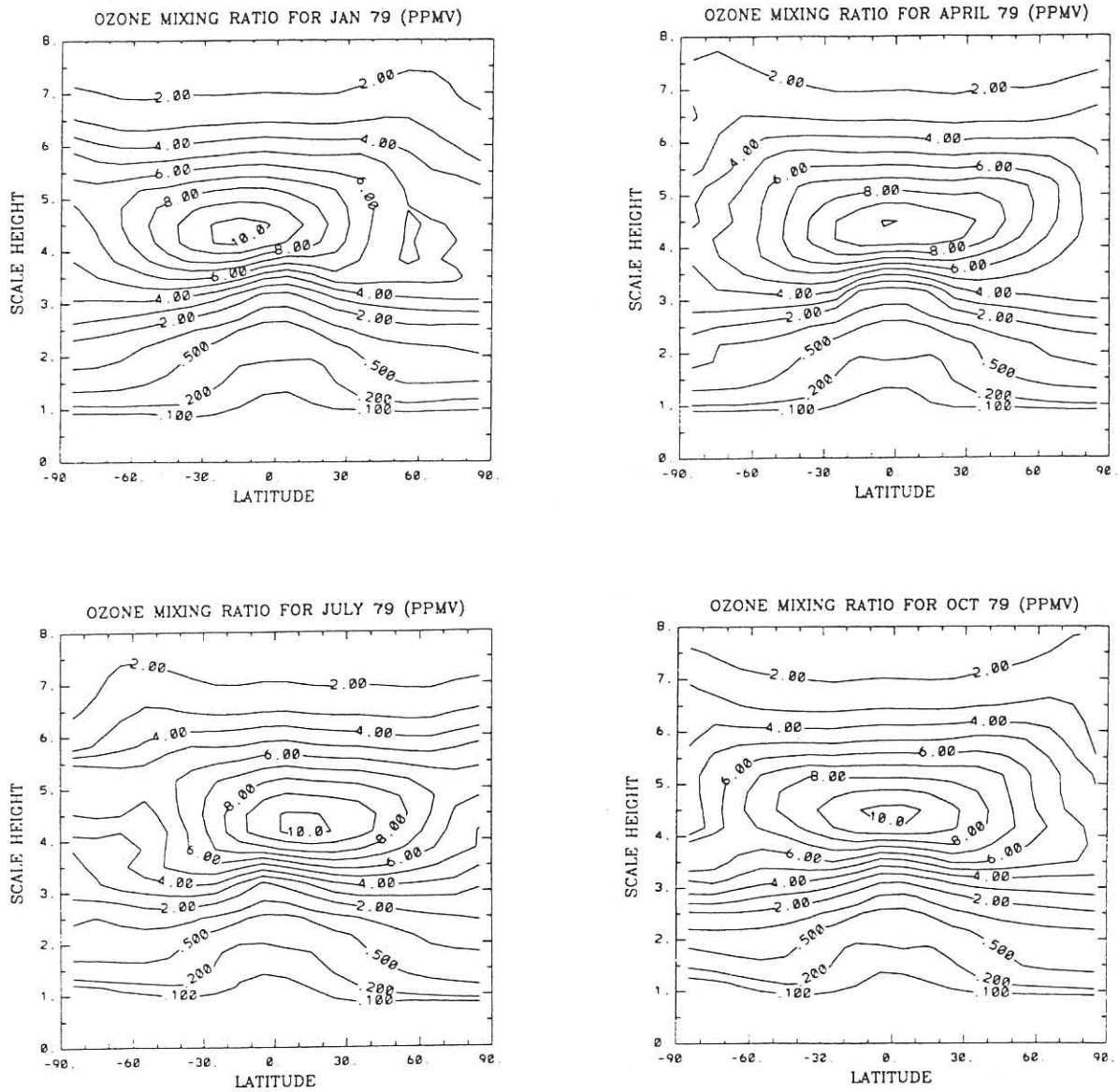


Fig. 12. Model-calculated zonal-mean ozone mixing ratio for 4 months in 1979. The vertical coordinate in pressure scale heights is approximate.

welling. A stronger upwelling in turn produces a reduction in the column ozone amount over the south polar region.

In the mid-latitude region, temperature cooling leads to a weaker downward branch by reducing the difference $\bar{T} - T_e$ and hence reducing the net cooling. In the tropical regions the warming of the lower stratosphere reduces net heating there and hence weakens the equatorial upwelling. The net effect is a weakening of the equator to mid-latitude cell, which thereby reduces the efficiency of transport of ozone from the photochemical production region at the low latitudes into the mid-latitudes, as hypothesized by *Mahlman and Fels* [1986]. The seasonal deepening of the Antarctic minimum cannot, however, be produced by dynamics without the existence of the reverse circulation.

The quantitative conclusions from the present model study should not be stated without a reiteration of the model weaknesses and uncertainties: (1) the crude approximation (Newtonian cooling) to radiative transfer; (2) the simple (Newtonian) chemistry used; (3) the uncertainties in the

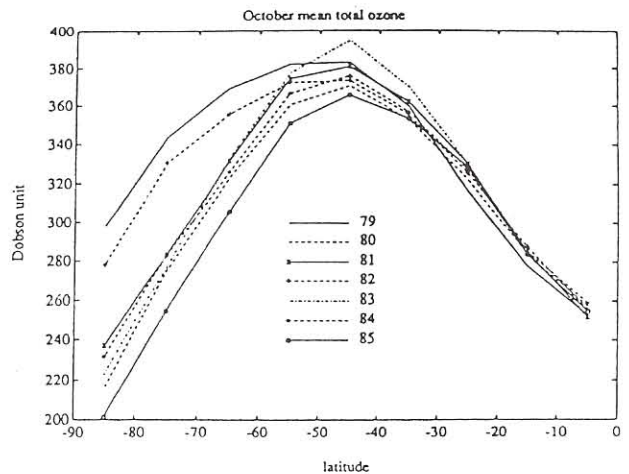


Fig. 13. Model-calculated zonal-mean, October mean ozone column density as a function of latitude for 7 years.

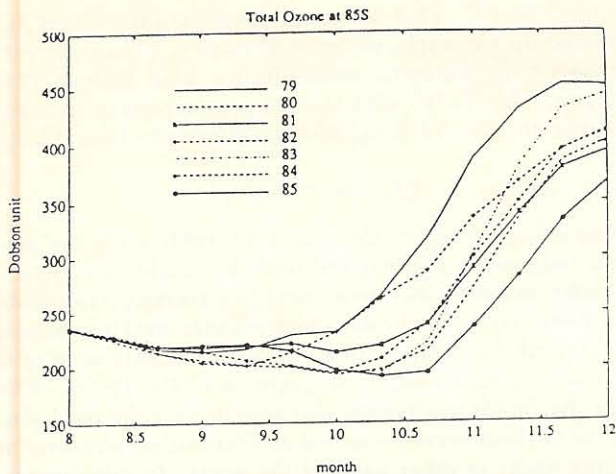


Fig. 14. Model-calculated zonal-mean column ozone from August to November for 7 individual years, at 85°S.

temperature input; and (4) the lack of time resolution in the input data fields.

Full ozone chemistry is essential for perturbation studies. For our present purpose of studying the dynamical changes in the Antarctic region, simple chemistry is used by design. In any case, the relaxation time during polar winter and spring is over 1 year long (with homogeneous ozone chemistry only) and so (homogeneous) chemistry plays a secondary role in our model at high latitudes in the season of interest.

Our input data are monthly mean temperature fields and so events with time scale less than 1 month cannot be resolved. Also, with the use of monthly means as input data on the fifteenth of each month, with the other dates extrapolated by a smooth Fourier series, the timing of events may be off by up to half a month. Another problem associated with the use of monthly mean temperatures fitted into a Fourier series in time is that only the low-frequency portion of temperature variability is treated in this study. Since the actual vertical velocity on isobaric surfaces is the difference between the diabatic heating term and the adiabatic ($\partial\bar{T}/\partial t$) term, a smooth temperature change obtained here will yield a smaller temperature tendency in periods of rapid

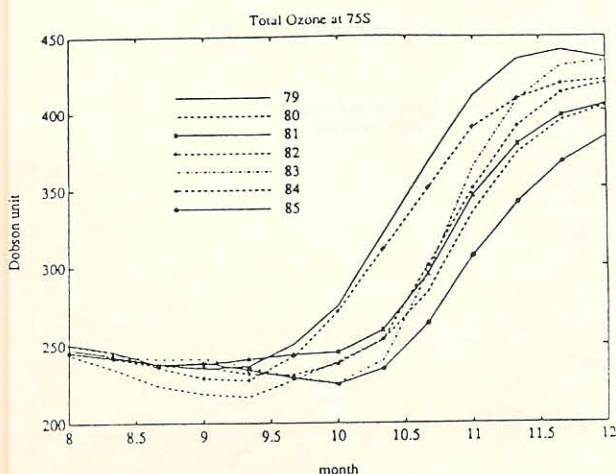


Fig. 15. Same as Figure 14, except at 75°S.

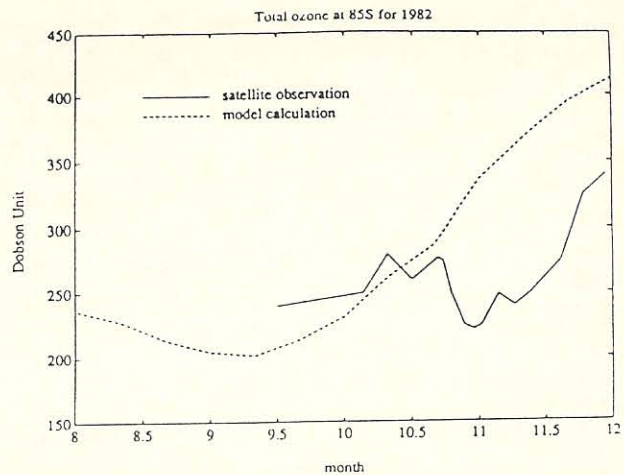


Fig. 16. Comparison of the calculated zonal-mean ozone seasonal cycle (dashed curve) with the observed (solid curve), for 1982.

temperature increases, resulting in a more positive vertical velocity.

The use of Newtonian cooling instead of detailed radiative transfer calculations is potentially the most problematic of all the approximations adopted. In particular, we may have overpredicted the dynamical component of the seasonal decline of Antarctic ozone, because the effect of reduced ozone solar heating due to reduced ozone concentration in recent years has not been incorporated. This problem would be especially severe if the recent ozone decline is caused by chemistry.

We have repeated the calculations with a comprehensive radiative transfer code and less-severely filtered temperature data [Yang, 1988], and the results will be published in a forthcoming paper. Here an approximate assessment of the effect of ozone-temperature feedback is given, based on a recent general circulation model (GCM) experiment by Kiehl *et al.* [1988].

Kiehl *et al.* [1988] performed two GCM experiments with

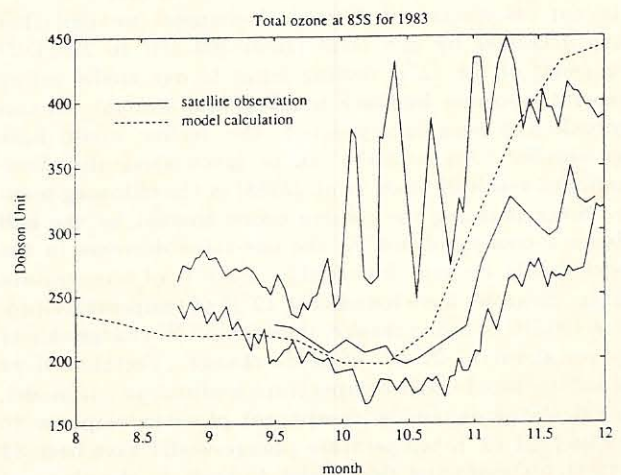


Fig. 17. Comparison of the calculated zonal mean ozone seasonal cycle (dashed curve) with the observed TOMS values (solid curve) for 1983 is shown in the middle two curves. Top curve shows maximum TOMS column ozone for all longitudes; bottom curve shows minimum TOMS column ozone for all longitudes [from Stolarski *et al.*, 1986], for the latitude band 74°–78°S.

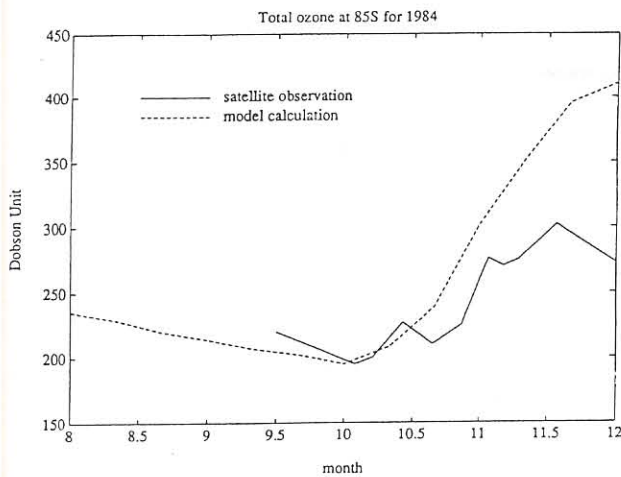


Fig. 18. Same as in Figure 16, except for 1984.

prescribed ozone. One experiment, the "control" run, has no ozone minimum in austral spring. The other, the "ozone hole" run, has a prescribed minimum value in September of 160 DU, about half that in the control run. The depletion is assumed to be caused by changed chemistry. The ozone hole run produces a temperature field which is colder than the control run by (as reported by the authors) 7°K in the lower stratosphere (100- to 20-mbar region). A close examination of the temperature evolution shown in their Figure 4 reveals that this rather large 7°K cooling occurs only in November, later than the period of interest for seasonal ozone decline. The temperature difference in September is negligible. Toward the end of September and in early October, the cooling is about 2°K , increasing to 6°K by the end of October. We estimate the GCM October monthly mean difference, weighted by ozone partial pressure, to be between 3 and 4°K . The observed October mean temperature difference, according to the NMC data of *Newman and Schoeberl* [1986] is 12°K in this region, when weighted by ozone partial pressure (see Figure 6). Therefore at least 8°K of the observed October temperature change cannot be explained by radiative cooling of chemically reduced ozone.

In our calculation of dynamical changes, we find that ozone decreases by one third (from 300 DU to 200 DU) as a result of the 12°K cooling input to our model before temperature-ozone feedback is taken into account. If this feedback had been incorporated, the decline would have been smaller. An estimate can be given using the aforementioned result of *Kiehl et al.* [1988] in the following manner. Since reducing the column ozone amount by one half induces a cooling of $3\text{--}4^{\circ}\text{K}$, the one-third decrease in the model should account for roughly $2\text{--}2.7^{\circ}\text{K}$ of temperature change. Since we have found that 12°K of temperature produces 100 DU of ozone change, then a $2\text{--}2.7^{\circ}\text{K}$ change should produce about 17–22 DU of ozone change. Therefore if we had incorporated ozone-temperature feedback in our model, our calculated dynamical component of ozone response to the observed 12°K temperature change would have been 83 to 78 DU. Comparing this model decrease to the observed ozone decrease from 1979 to 1985, which is 110 DU according to *Schoeberl et al.* [1986], or 125 DU after recent reprocessing (M. R. Schoeberl, personal communication, 1987), we conclude that our model underprediction of the observed October mean, zonal-mean decline from 1979 to 1985 can

be up to 50 DU. This would still leave at least 60% of the observed decline explainable by dynamics. (The underprediction of the Antarctic ozone decline from 1979 to 1985 is found to be 35 DU with the temperature-ozone feedback incorporated into a full radiative transfer code *Yang* [1988].)

12. CONCLUSIONS

Our results suggest that prior to 1984, both the seasonal and year-to-year variability of the zonal-mean Antarctic ozone minimum and the surrounding maximum can probably be accounted for by dynamics without invoking changes in chemical composition (e.g., chlorine content) or special chemistry (e.g., reactions on particles of the PSCs). The same dynamical mechanism also accounts for the good simulation of the observed seasonal and latitudinal structure of column ozone in other parts of the world. In other words, the existence of the Antarctic spring minimum, which was present in climatologies based on data from as early as the 1960s, is probably a natural phenomenon. Our model however appears to underpredict the September ozone decline in recent years, leading to an underprediction of the recent minimum values in the Antarctic ozone hole.

For the period 1979–1985, which we have examined, our model-generated column ozone closely follows the observed quasi-biennial oscillation of temperature, consistent with the observed behavior of column ozone. While the observed difference between October 1985 and October 1979 zonal-mean column ozone densities is about 125 DU, the model yields a difference of 100 DU as the dynamical component of the changes induced by temperature change during August, September, and October only (with no PSC radiative or chemical effects included). The discrepancy appears in the model sometime after 1982 and persists to the last year of the model run (1985) and amounts to about 25 DU of underprediction of the observed zonal-mean decline. We estimate that this discrepancy can be up to 50 DU in 1985, when the effect of ozone-temperature feedback is incorporated.

In an earlier version of this paper, we attributed this discrepancy to the effect of additional radiative heating caused by El Chichón in 1983 and by PSCs for the later years (see also *Mahlman and Fels* [1986]; *Shi et al.* [1986]; *Tung, [1986a]*), assuming that about $0.5^{\circ}\text{K}/\text{day}$ of additional heating can be provided by these aerosols. *Shi et al.* [1986] esti-

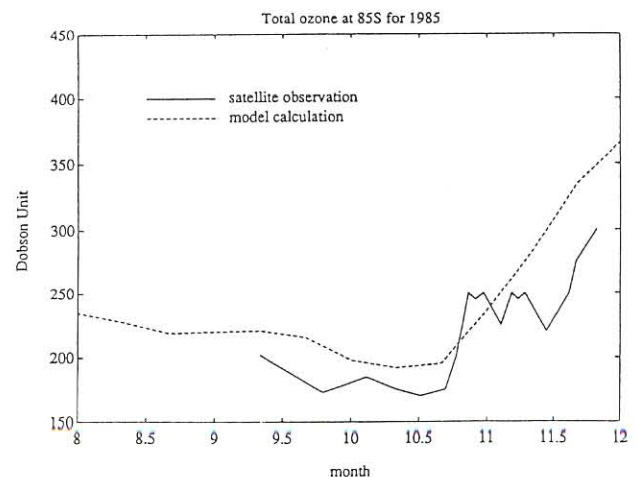


Fig. 19. Same as in Figure 16, except for 1985.

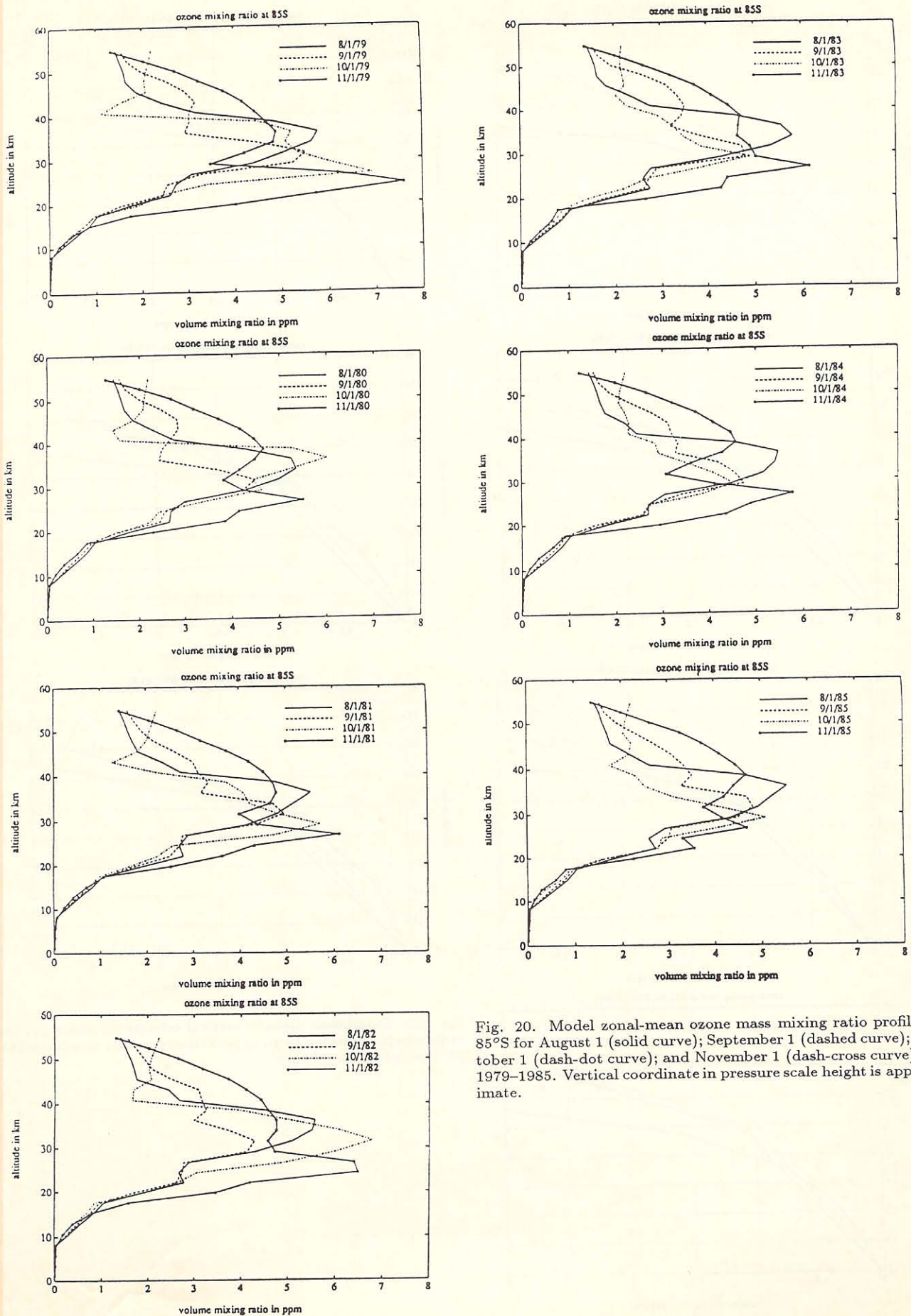


Fig. 20. Model zonal-mean ozone mass mixing ratio profile at 85°S for August 1 (solid curve); September 1 (dashed curve); October 1 (dash-dot curve); and November 1 (dash-cross curve) for 1979-1985. Vertical coordinate in pressure scale height is approximate.

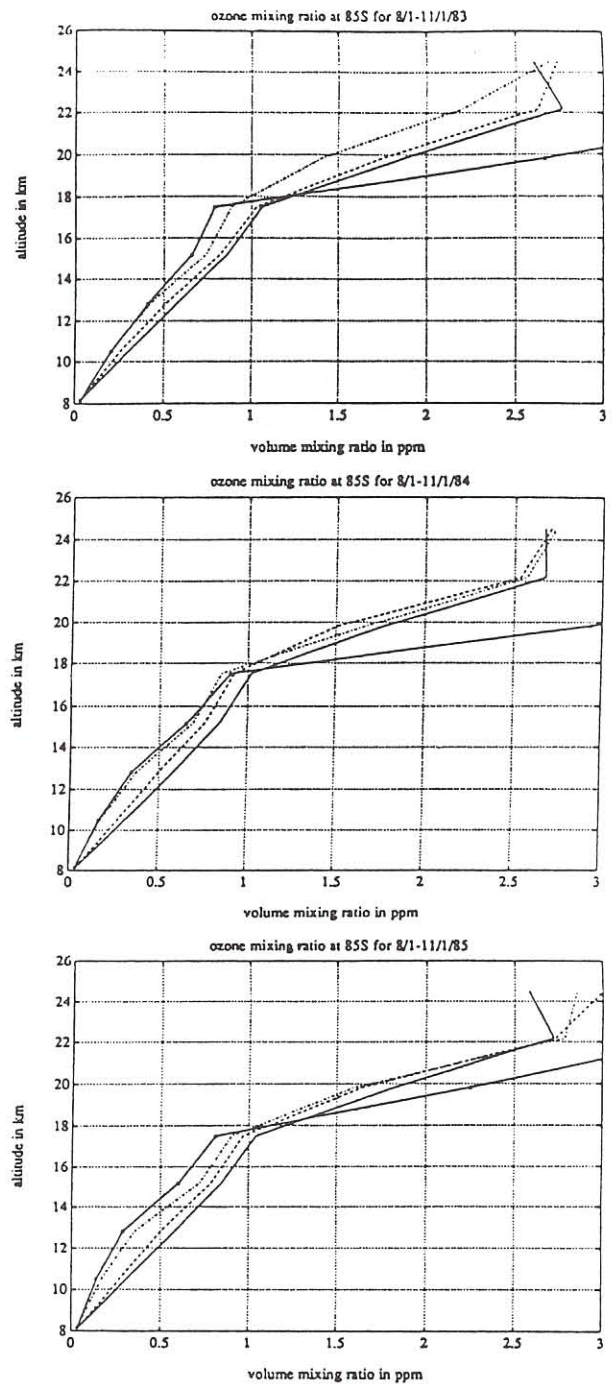
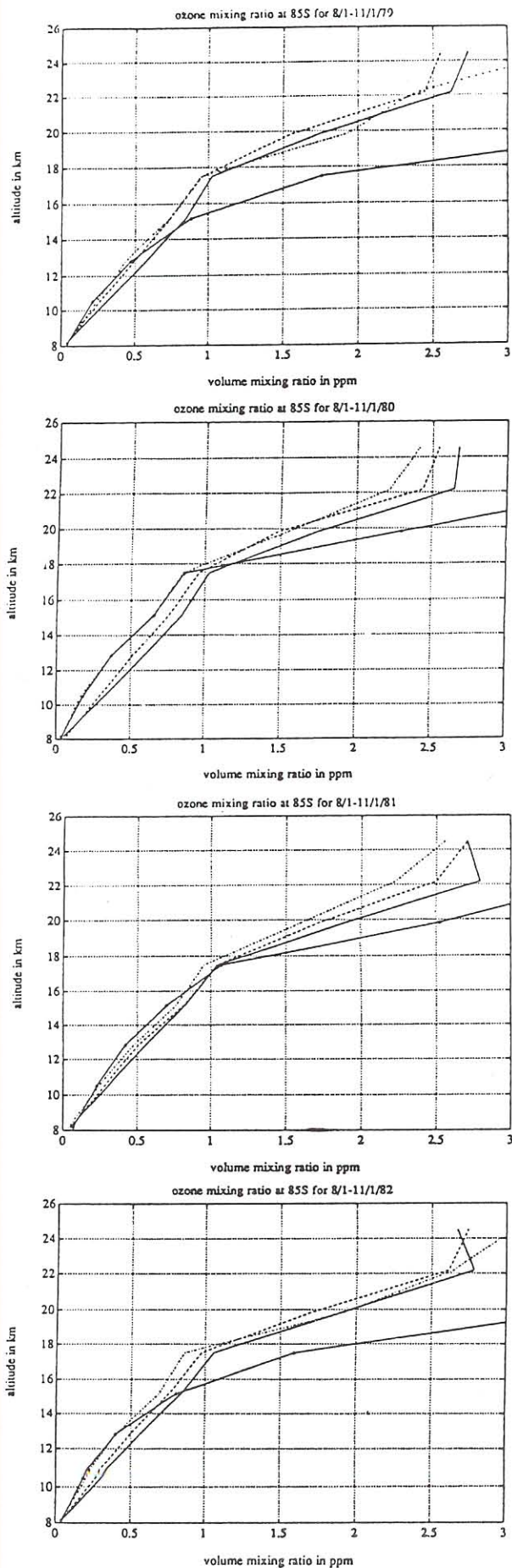


Fig. 22. Zonal-mean diabatic vertical velocity (in isentropic coordinates but resorted to an approximate pressure coordinate) at 90°S as a function of time.

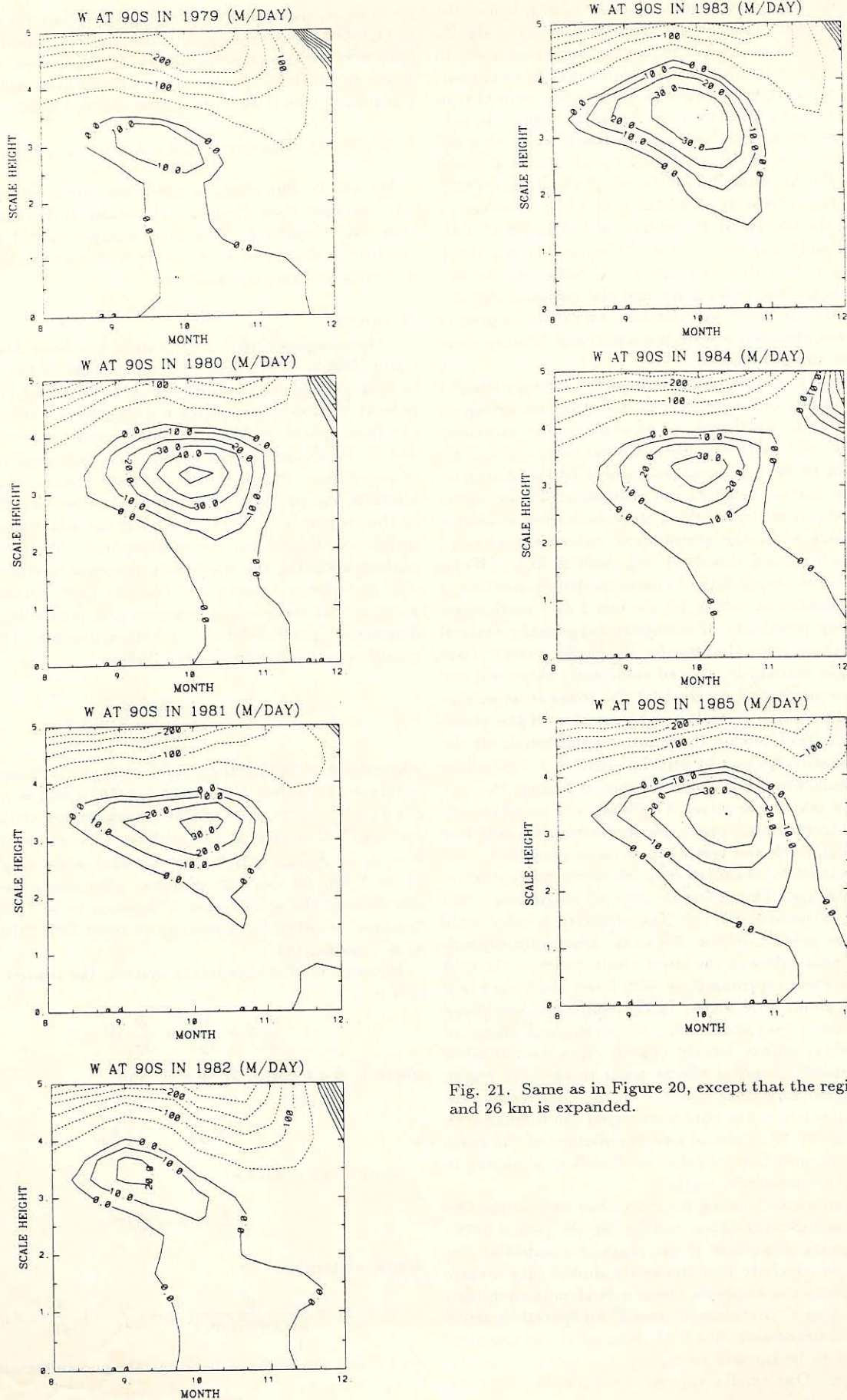


Fig. 21. Same as in Figure 20, except that the region between 8 and 26 km is expanded.

mated that for the Syowa station (69°S) in 1982, before the El Chichón aerosols reached the high latitudes in a significant amount, the aerosol heating in spring can amount to about 0.25 °K/day (subject to uncertainties). Aerosol heating would presumably be larger in 1983, when aerosol concentration reached a peak over the Antarctic region. In subsequent years, background volcanic aerosols have been observed to decay steadily from the 1983 peak, but at a slower rate than in the Arctic region [McCormick and Trepte, 1987]. Because of the increasing abundance of PSCs observed in later years, the combined abundance of volcanic and PSC aerosols probably has not decreased [Iwasaka et al., 1986]. However, since the radiative properties of both volcanic and PSC aerosols are not well known and the radiative calculations are subject to great uncertainties, we cannot at present state with any confidence that the additional heating mentioned above can be realized.

Results from recent Antarctic campaigns strongly suggest an important role of heterogeneous chemistry occurring on surfaces of PSCs. Furthermore, no evidence for upwelling was found in 1986 and 1987 observational campaigns. Theoretical work on the chemical component of the Antarctic ozone decline further suggests the possibility of a switch-on process for chemical depletion of ozone occurring sometime in recent years when the atmospheric chlorine level (e.g., HCl) reaches a certain threshold (e.g., half of NO_x) [Wofsy et al., 1988]. The plausibility of a catastrophic destruction of ozone by chemistry occurring for the last 2 or 3 years raises the interesting possibility of chemistry suppressing vertical motions for these years (but not for the earlier years). That is, if the ozone amount is reduced sufficiently there will not be enough ozone to radiatively heat the lower stratosphere when the Sun returns in spring. This portion of the atmosphere may instead remain in radiative equilibrium. If the vertical transport is close to zero for 1986 and 1987, then in earlier years when the ozone amount is higher, the vertical velocity may be positive. This issue will be addressed further in a forthcoming paper, incorporating full radiative transfer calculations and temperature data after 1985.

The present study examines only the ozone concentration in a zonal average. The minimum over all longitudes is not predicted. As pointed out by Tung [1986a], a large-scale thermal wave present in the Antarctic lower stratosphere can create ozone values in the lower stratosphere in the cold phase of the wave approximately 40% lower than the zonal average and about 40% higher in the warm region. When integrated over a longitude circle, the dynamical effects of the wave perturbations largely cancel. However, if additional irreversible chemical effects occur in the cold region due to preferred formation of PSCs in the cold region, the ozone concentration in the minimum region can become even lower. The 25–50 DU units of underprediction of the zonal average by our model may be due to this effect occurring in the temperature minimum region.

With dynamics accounting for more than 60% of the October mean, zonal-mean ozone decline (for the period 1979–1985), it appears reasonable at this stage of quantitative investigations to conclude that dynamics should play a more important role in the Antarctic ozone hole phenomenon than merely providing a “containment vessel” for special chemical destruction of ozone and that both dynamical and chemical changes have to be invoked to explain the recent Antarctic ozone changes. Our results appear to suggest that dynam-

ics is more important in affecting the year-to-year changes in October monthly means, while the role played by chemistry may have become important in September after 1982. These conclusions are, of course, subject to considerable uncertainties, which we hope can be reduced in future studies.

APPENDIX: MODEL SPECIFICATION AND SENSITIVITY

We use in this study a nonlinear, nongeostrophic, zonally averaged (two-dimensional) model of dynamics, radiation, and chemistry. The three components of the model are briefly described here, along with some details on the numerics and initial conditions.

Dynamics

The nongeostrophic formulation has been described by Tung [1986b, 1987], and the calculation of the isentropic mixing coefficients has been described by Yang [1988]. The present version incorporates a σ -like coordinate system for the troposphere (below the 350°K isentrope), while above 350°K, log- θ coordinates are used, as described in the text of this paper. This hybrid coordinate system is adopted to alleviate the problem of isentropes intersecting the ground in the troposphere, while retaining the advantages of isentropic coordinates for the stratosphere. We have decided against adopting the previously proposed remedy of defining “underground isentropes” [Lorenz, 1955; Andrews, 1983], as we do not want to put precious grid points into the “underground” portions of the numerical domain. The vertical coordinate is defined as [Yang, 1988]

$$z_{\uparrow} = H \frac{\ln(\theta/\theta_o)}{\ln(350^\circ K/\theta_o)} \quad (A1)$$

where for $\theta > 350^\circ K$ (i.e., $z_{\uparrow}/H > 1$), θ_o is chosen to be a constant ($\theta_o = 263^\circ K$) so that $\ln(350^\circ K/\theta_o) = \kappa \equiv R/c_p$. For $\theta < 350^\circ K$, θ_o is a variable corresponding to the zonally averaged value of θ at the surface. Therefore $z_{\uparrow} = 0$ for $\theta = \theta_o = \bar{\theta}_{\text{surface}}$. H is the standard scale height, taken to be 7 km for our calculations. (For the present set of calculations the scale height H is taken to be 7 km and the “surface” is taken for simplicity to be at 1000 mbar, and so $\theta_o = \bar{T}_{1000\text{mbar}}(y)$.)

In terms of this coordinate system, the continuity equation is

$$\frac{\partial}{\partial t} \bar{\sigma} + \frac{\partial}{\partial y} \bar{V} + \frac{\partial}{\partial z_{\uparrow}} \bar{W} = 0 \quad (A2)$$

where $V \equiv \sigma v \cos \phi$, $W \equiv \sigma \dot{z}_{\uparrow}$, and

$$\sigma \equiv \rho \frac{\partial z}{\partial z_{\uparrow}} = -\frac{1}{g} \frac{\partial \bar{p}}{\partial z_{\uparrow}}$$

The energy equation,

$$\frac{d}{dt} \ln \theta = Q/T$$

is now written as

$$\dot{z}_{\uparrow} = \frac{H}{\ln(350^\circ K/\theta_o)} \left[\frac{Q}{T} + \left(\frac{z_{\uparrow}}{H} - 1 \right) \frac{d}{dt} \ln \theta_o \right]$$

where Q is the diabatic heating rate per unit mass divided by c_p . Therefore

$$\bar{W} \simeq \bar{\sigma} \left[\frac{\bar{Q}}{\Gamma} + \min(0, \frac{z_{\uparrow}}{H} - 1) \left(\frac{\bar{Q}}{\Gamma} \right)_{z_{\uparrow}=0} \right] \quad (\text{A3})$$

where

$$\Gamma = \frac{\bar{T}}{H / \ln(350^\circ K / \theta_o)}$$

Note that at the surface, $z_{\uparrow} = 0$, (A3) gives $\bar{W} = 0$.

The meridional momentum equation becomes

$$\begin{aligned} \left(2\Omega + \frac{\bar{u}}{a \cos \phi} \right) \sin \phi \bar{u} = -\frac{1}{a} \frac{\partial}{\partial \phi} \bar{\Phi} \\ + c_p \bar{T} \left(1 - \frac{z_{\uparrow}}{H} \right) \frac{1}{a} \frac{\partial}{\partial \phi} \ln \theta_o \end{aligned} \quad (\text{A4})$$

where the Montgomery streamfunction, $\bar{\Phi}$, is related to the temperature T by

$$\frac{\partial \bar{\Phi}}{\partial z_{\uparrow}} = c_p \frac{\partial}{\partial z_{\uparrow}} T + g \frac{\partial z}{\partial z_{\uparrow}} = \frac{c_p T}{H} \ln \left(\frac{350^\circ K}{\theta_o} \right) \quad (\text{A5})$$

The zonal momentum equation takes the same form as that stated in the text (equations (8) and (9)). The assumption is made there that in the lower stratosphere, large-scale diffusion of Ertel's potential vorticity and conservative tracers is predominantly along isentropic surfaces. An additional assumption is made here that such diffusion takes place along constant z_{\uparrow} -surfaces, which in the troposphere are nearly parallel to the surface contour.

Radiation

We adopt the Newtonian cooling parameterization for the net radiative heating rate Q :

$$\bar{Q} = \alpha (T_e - \bar{T}) \quad (\text{A6})$$

The Newtonian cooling coefficient has the following form:

$$\begin{aligned} \alpha = \{ \exp[-(\frac{z_{\uparrow}}{H} - 7)^2 / 10 - 1.8] - \exp[\frac{3z_{\uparrow}}{H} - 25.9] \\ + 0.002(8 - \frac{z_{\uparrow}}{H}) \} * [1 - \frac{\phi}{270^\circ} \cos(\frac{2\pi t}{360 \text{ days}})] \end{aligned} \quad (\text{A7})$$

The first part is a function of vertical coordinate z_{\uparrow} only and is constructed to fit the result of *Wehrbein and Leovy* [1982]. The second part gives a factor of 2 variation in both latitude and time [*Kiehl and Solomon*, 1986]. \bar{W} is set to 0 at the upper boundary ($z_{\uparrow}/H = 8$) by setting $\alpha = 0$ there. At the lower boundary, which is a coordinate surface $z_{\uparrow} = 0$, $\bar{W} = 0$ by definition.

The analytic formula for the radiative equilibrium temperature in January is, in degrees Kelvin,

$$\begin{aligned} T_e = 200 + 80 \operatorname{sech} \left[0.07 \left(\frac{z}{H} - \frac{48}{7} \right) \right] + 105 \operatorname{sech} \left(0.16 \frac{z}{H} \right) \\ - 20 \operatorname{sech} \left[0.15 \left(\frac{z}{H} - 2 \right) \right] - 10 \operatorname{sech} \left[0.12 \left(\frac{z}{H} - \frac{80}{7} \right) \right] \\ + \sum_{i=1}^7 a_i \operatorname{sech} [b_i (\phi - c_i)] \operatorname{sech} [d_i (\frac{z}{H} - \frac{e_i}{7})] \sin^2(\phi \pi / 180) \end{aligned} \quad (\text{A8})$$

where ϕ is latitude in degrees, $z \equiv H \ln(p_{oo}/p)$ is the log-pressure coordinate, and the other parameters are listed in Table 1. The formula was constructed to fit the result of S. B. Fels and M. D. Schwarzkopf (as cited by *WMO* [1986]), with minor adjustments applied so that the net radiative

TABLE 1: Parameters Used to Fit T_e

a_i	b_i	c_i	d_i	e_i
24	0.03	-90	0.05	48
-30	0.05	20	0.03	60
-120	0.03	90	0.06	48
-30	0.06	20	0.04	40
-35	0.02	-90	0.17	4
-35	0.02	90	0.18	0
-20	0.00	00	0.18	0

heating rate calculated using our Newtonian cooling parameterization and the temperature field of *Geller and Wu* [1987] agrees with that of *Rosenfield et al.* [1987], which is a full radiative transfer calculation using the same temperature data. The radiative equilibrium temperature for July is taken to be the same as T_e for January, except with poles reversed. These fields are then extrapolated to get an annually periodic function, so that the value at other times in the year can be obtained.

The boundary conditions at the poles require $\bar{V} = 0$ there. This then imposes a condition of mass conservation via (A2) as

$$\int_{-a}^a (\bar{W} - \frac{1}{g} \frac{\partial}{\partial t} \bar{p}) dy = 0 \quad (\text{A9})$$

With an imposed, instead of calculated, temperature field, (A9) is not always met. For each level where (A9) is not met, the radiative equilibrium temperature T_e is further adjusted by adding or subtracting a (constant) residue at that level, so that (A9) is strictly enforced.

Chemistry

We have performed several sensitivity experiments with the adopted parameterized chemistry

$$\bar{S} = \bar{\mu}(f_e - \bar{f}) + \text{a small quadratic term} \quad (\text{A10})$$

to test the validity of the statement that the observed seasonal behavior of column ozone is determined predominantly by the seasonal variation of the transports and not by the seasonality of the photochemical source and sinks. We have symmetrized $1/\bar{\mu}$ and f_e to remove any seasonal dependence. The resultant column ozone exhibits the same seasonal behavior as that shown in Figure 8. Removing the small quadratic term in \bar{S} (A10) also introduces very little difference in the calculated column ozone.

As mentioned in the text, we have introduced an upper limit on the chemical relaxation time $1/\bar{\mu}$. Without such a limit, there is no chemical loss of ozone during the polar night and near the tropopause at all latitudes. The calculated ozone content is high globally compared with recent climatology [see *Ko et al.*, 1985b]. By adjusting the upper limit on $1/\bar{\mu}$, the global ozone content can be adjusted to the observed value for any particular year. This single constant adjustment does not affect the latitudinal structure of column ozone. By taking this limit to be longer than one year, the seasonal structure also is not affected. We have taken the value $1/\bar{\mu}_{max} = 18$ months in the present study. The same value is used for all 7 years (i.e., the global ozone content is not adjusted for individual years).

There was concern that although the seasonal behavior of ozone is not affected by a $1/\bar{\mu}_{max}$ that is longer than 1 year, year-to-year behavior may be affected in the 7-year

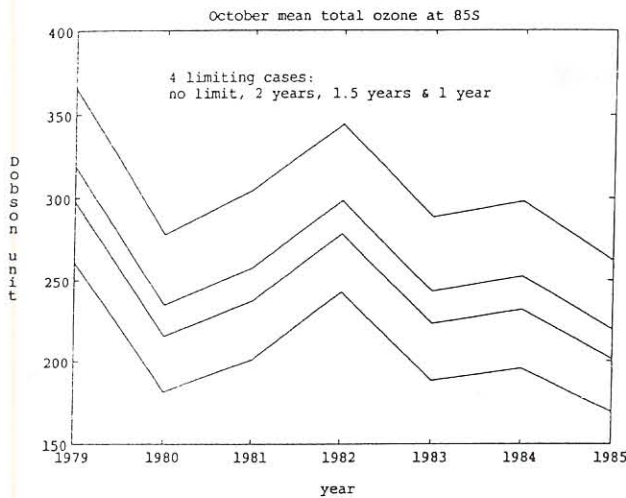


Fig. 23. Model-calculated zonal-mean, October mean column ozone at 85°S for the 7 years. From bottom to top, the maximum chemical loss time is 1 year, 1.5 years, two years, and infinite.

run. Consequently, we have performed several runs with $1/\bar{\mu}_{max} = 1$ year, 1.5 years, 2 years, and ∞ years. The result is plotted in Figure 23. Figure 23 demonstrates that the relative changes between years in all the 7-year runs are unaffected by the value of $1/\bar{\mu}_{max}$. The same 100 DU decline in the October mean column ozone is obtained between 1979 and 1985 even in the $1/\bar{\mu}_{max} = \infty$ case. It is seen that choosing $1/\bar{\mu}_{max}$ has the effect of imposing an "initial condition" on the 1979 ozone content.

Numerics

The numerical domain is eight scale heights, divided into 25 levels, in the vertical. In the horizontal the domain is from 90°S to 90°N with 10° increments in latitude. The time step is 6 hours. The numerical scheme of Smolarkiewicz [1983] is used. The boundary conditions are $\bar{v} = 0$ at $y = -a$, a and $\bar{w} = 0$ at $z_f = 0, 8H$. The concentration of ozone at the lower boundary is fixed at $\bar{f}/\bar{\sigma} = 20$ parts per billion by volume (ppbv) at the lowest grid points to simulate fast surface chemical destruction. Vertical mixing with $K_{zz} = 0.5$ m²/s is added in the model for the lowest 4 grid points to simulate tropospheric mixing processes not included in our parameterization for the stratosphere.

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