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Decadal evolution of the Antarctic ozone hole

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Abstract. Ozone column amounts obtained by the total ozone mapping spectrometer (TOMS) in the southern polar region are analyzed during late austral winter and spring (days 240-300) for 1980-1991 using area-mapping techniques and area-weighted vortex averages. The vortex here is defined using the -50 PVU (1 PVU = 1.0 x 10^{-6} K kg^{-1} m^2 s^{-1}) contour on the 500 K isentropic surface. The principal results are: (1) there is a distinct change after 1985 in the vortex-averaged column ozone depletion rate during September and October, the period of maximum ozone loss, and (2) the vortex-averaged column ozone in late August (day 240) has dropped by 70 Dobson units (DU) in a decade due to the loss in the dark and the dilution effect. The mean ozone depletion rate in the vortex between day 240 and the day of minimum vortex-averaged ozone is about 1 DU d^{-1} at the beginning of the decade, increasing to about 1.8 DU d^{-1} by 1985, and then apparently saturating thereafter. The vortex-average column ozone during September and October has declined at the rate of 11.3 DU yr^{-1} (3.8%) from 1980 to 1987 (90 DU over 8 years) and at a smaller rate of 2 DU yr^{-1} (0.9%) from 1987 to 1991 (10 DU over 5 years, excluding the anomalous year 1988). We interpret the year-to-year trend in the ozone depletion rate during the earlier part of the decade as due to the rise of anthropogenic chlorine in the atmosphere. The slower trend at the end of the decade indicates saturation of ozone depletion in the vortex interior, in that chlorine amounts in the mid-1980s were already sufficiently high to deplete most of the ozone in air within the isolated regions of the lower-stratospheric polar vortex. In subsequent years, increases in stratospheric chlorine may have enhanced wintertime chemical loss of ozone in the south polar vortex even before major losses during the Antarctic spring.

I. Introduction

The dramatic decrease of the total column of ozone over Antarctica in southern late winter and spring (August through November), a phenomenon known as the Antarctic ozone hole, was first described using observations made with Dobson spectrophotometer on the ground [*Farman et al.*, 1985] and later by satellite observations [*Stolarski et al.*, 1986]. It is now understood that the Antarctic ozone depletion is caused by the catalytic chlorine chemistry in the lower stratosphere, where low temperatures prevail due to the special geography and meteorology of the Antarctic regions (e.g., Plate 1). This cold air in the lower stratosphere at high southern latitudes permits the formation and persistence of polar stratospheric clouds (PSCs). Heterogeneous chemistry [*Solomon et al.*, 1986] on these ice clouds, followed by the action of sunlight, converts reservoir species of chlorine (HCl and $ClONO_2$) into very reactive radicals such as Cl and ClO [*McElroy et al.*, 1986a, b; *Molina et al.*, 1987b; *Toon et al.*, 1986; *Tolbert et al.*, 1987]. The presence of these chlorine radicals [*de Zafra et al.*, 1987], particularly of ClO and its anticorrelation with O_3, has been confirmed by in situ measurements from aircraft [*Anderson et al.*, 1989a, b, 1991] and by remote sensing from the Upper Atmosphere Research Satellite (UARS) [*Manney et al.*, 1993b; *Waters et al.*, 1993a, b]. While there is little doubt of the key role played by heterogeneous chemical reactions involving stratospheric chlorine, the quantitative aspects of the mechanisms involved in producing the Antarctic ozone hole are still the subjects of intense study and observation. This is due principally to the effects of dynamics on the temperature (and thus the ice cloud microphysics) and on the distributions of chemical trace gases and their exposure to sunlight [*Solomon et al.*, 1986; *Bojkov*, 1986; *Mahlman* and *Fels*, 1986; *Tung et al.*, 1986; *Garcia* and *Solomon*, 1987; *McIntyre*, 1989; *Solomon*, 1990; *Schoeberl et al.*, 1991, 1992; *Tung* and *Yang*, 1994a, b; *Yang* and *Tung*, 1994]. These effects vary from year to year and thus complicate attempts to interpret the observed 30% to 60% decline in total column ozone amounts during the last decade. A thorough discussion of the Antarctic ozone hole was given by *Solomon* [1988, 1990] and *World Meteorological Organization* (*WMO*) [1988, 1989, 1991].

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In this paper we examine the record of total column ozone variations observed by the total ozone mapping spectrometer (TOMS) over the period 1980 - 1991. Past analyses of the Antarctic ozone depletion have tended to use the ozone minimum values or zonal averages when examining decadal trends [*Bowman*, 1985, 1988, 1990; *Herman et al.*, 1991, 1993; *Krueger et al.*, 1992; *Stolarski et al.*, 1991, 1992]. Other analyses have focused on interannual variations in the ozone minimum values and have found potential links to phenomena associated with the Quasi-Biennial Oscillation (QBO) [e.g., *Garcia* and *Solomon*, 1987; *Lait et al.*, 1989]. More recently, *Randel* and *Wu* [1995] examined trends in ozone by first averaging TOMS data along potential vorticity (PV) contours in order to account for displacement and longitudinal asymmetries of the polar vortex. Another natural coordinate for these purposes is ozone itself. Here we will use the ozone area-mapping technique first applied to Antarctic ozone by *Yung et al.* [1990] to reexamine the decline rates of Antarctic springtime column ozone from 1980 to 1991, emphasizing both general and year-to-year changes over that period. A key modification of the previous application of this technique is to constrain the ozone area mapping to the south polar vortex using values of PV derived from National Meteorological Center (NMC) data for the same period.

In section 2, we first briefly describe the data we used. The area-mapping method is described in section 3. Then, in section 4, we present our main results obtained from the TOMS ozone data using the area-mapping method in combination with the use of PV and of area-mapped NMC temperatures in the polar vortex. Two important features emerging from the analysis are a distinct change in the decline rate of vortex-averaged column ozone after 1985 and the anomaly represented by the 1988 Antarctic spring.

2. TOMS Ozone Column Abundance, PV and Temperature

The Nimbus 7 spacecraft was launched into a local-noon, Sun-synchronous, near-polar orbit on October 24, 1978. Its TOMS instrument has measured the spatial distribution of total ozone from 1978 until May 6, 1993, by scanning across the track of the satellite to obtain data between successive satellite orbital tracks. Total column ozone is retrieved from the measured differential absorption of backscattered ultraviolet irradiance using observations taken with solar zenith angles up to 88°. No measurements are available in regions of polar night. In our analysis we used the daily TOMS gridded ozone data of version 6.0 [*Herman et al.*, 1991] from NASA (on a 1° x 1.25° grid in latitude and longitude) from 1980 to 1991. The 1979 data were not used in our analysis mainly due to frequently missing data (e.g. days 277, 278, 279) in this year. The more recent version 7 with data coverage till 1994 has not been officially released yet.

The other two data sets used are NMC temperatures and Rossby-Ertel PV [*Manney* and *Zurek*, 1993a] derived from NMC analyses of temperature and geopotential heights [e.g., *Gelman et al.*, 1986]. The PV and temperature fields are gridded at 2.5° x 5.0° in latitude and longitude. In our study we will use temperature and PV fields on the 500 K isentropic surface to help interpret variations in the total column ozone. This level is in the lower stratosphere near the ozone number density peak in early August, prior to the formation in recent years of the ozone hole during the austral spring.

3. Area-Mapping Method

The ozone area-mapping method used here was described by *Yung et al.* [1990] and is similar to that used in area mapping of Ertel's potential vorticity on an isentropic surface [*Butchart* and *Remsberg*, 1986; *Baldwin* and *Holten*, 1988]. Let $A(t, \Omega^*)$ be the area enclosed by any contour value $\Omega^*(\vec{r}\ t)$ of ozone column abundance, as a function of time (t) and position ($\vec{r}$) on the surface of the Earth. A convenient unit for the area bound by an ozone isopleth is the area of the southern hemisphere, $2\pi R^2$ (R is the radius of Earth). For comparison, the areas out to the Antarctic circle and 60°S latitude circle are 0.079 and 0.13, respectively, while the area of the Antarctica continent, including the ice shelf, is 5.5% of the area of the southern hemisphere.

Since total column ozone generally changes monotonically outward from a minimum in the interior to the edge of the polar vortex, an area increase (decrease) of a total column ozone isopleth with time implies that the ozone hole (i.e., the region of lowest column ozone) is expanding (shrinking). This result holds no matter where the center of ozone hole is. Thus, to first order, this method filters out planetary-scale (i.e., low longitudinal wavenumber) perturbations of the Antarctic polar vortex such as distortions of the vortex or displacements away from the pole. We assume that the ozone column abundance in the south polar night region is smaller than that in the daylight region and only include daytime data in the area averages, adjusting the area weighting to include just the sunlit portions of the south polar regions. This has little effect on our analyses as we focus on the late winter and spring period when the area with no TOMS measurements is very small (e.g., Plate 1).

The assumption of monotonic variation breaks down beyond the edge of the polar vortex, in the region of the total column ozone maxima in high midlatitudes sometimes called the polar "collar" region. For some contours near the vortex boundary, area-mapping would mix data points from outside of the vortex with that inside the vortex. Vortex-averaged quantities have been shown to be useful in examining the relationships among, for instance, ozone, temperature and ClO at various heights in the lower stratosphere [e.g., *Manney et al.*, 1994b]. *Randel* and *Wu* [1995] recently mapped column ozone by averaging the TOMS data around PV contours at 520 K. The issue is how to apply a vortex constraint given that a definition of the polar vortex in terms of PV contours will vary with height and from year to year.

Because the total column ozone value is heavily weighted to the lower stratosphere [e.g., *Manney et al.*, 1994a; *Froidevaux et al.*, 1994], we have chosen to define the vortex using PV values there. By inspection we have chosen a single value, namely, -50 PVU (1 PVU=1.0 x 10^{-6} K kg^{-1} m^2 s^{-1}). In later years (1992 and 1993, not considered here), *Chen* [1994] found that this value, which was toward the poleward edge of the region of large PV gradients, defined an isolated inner vortex at 500 K. For the years and season considered here the -50 PVU contour is typically in the middle of the region of strong PV gradients on the 500 K isentropic surface and is outside the region of large ozone loss (Plate 1), which makes it suitable for limiting the ozone area-mapping. Interannual variations of the area within this PV contour are representative of changes in vortex size and reflect at least indirectly variations in planetary wave activity, including minor warmings; these can affect both ozone transport and heterogeneous chemistry. To assess these effects more quantitatively requires consideration of vertical as well as horizontal variations in both ozone and PV and is beyond the scope of this paper.

4. Area Mapping of Ozone, PV, and Temperature: Results and Discussion

4.1. Average Column Ozone: The Ozone Hole Period

We will first investigate variations of area-averaged ozone column abundance inside the polar vortex. Figure 1 shows the temporal variations of the area-weighted average of ozone column abundance inside the polar vortex (solid line), as constrained by the location of the -50 PVU contour at 500 K. The time interval chosen for each year is from day 240 (August 28 for nonleap year) to day 300, corresponding to the period of maximum ozone depletion in each year. These vortex-constrained area averages of column ozone are significantly larger than the minimum ozone column abundance [e.g., *Lait et al.*, 1989, Figure 2a], as they include large regions of the vortex close to the boundary, where ozone values are higher (Plate 1).

The dotted lines in Figure 1 indicates the area-weighted average of ozone column abundance in the region between 60°S and 90°S. Differences between this zonal and the vortex-limited average reflect the longitudinal asymmetry of the polar

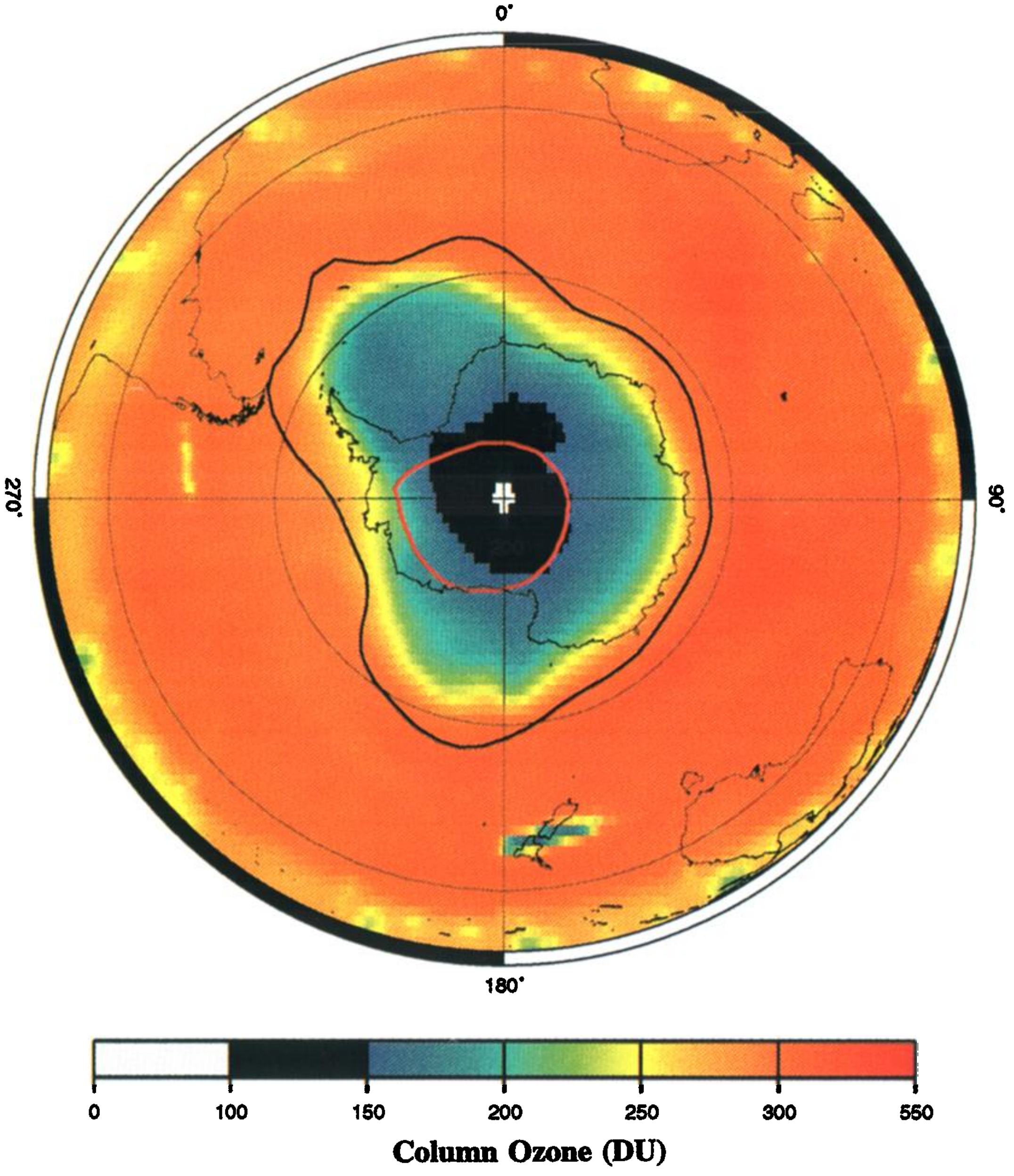

Plate 1. Southern polar orthographic projection of ozone column abundance (DU) on day 275 (October 2) 1987. There are no TOMS data in the white area. The area bound by the 300-DU contour is 13% of the area of the southern hemisphere. The smaller and larger circles are located at 60°S and 30°S latitudes, respectively. The thick red line indicates regions where the temperature is below 195 K on the 500 K isentropic surface and polar stratospheric clouds (PSCs) may form. The -50 PVU contour of potential vorticity (PV) (thick dark line) at 500 K, used to define the polar vortex, is also shown.

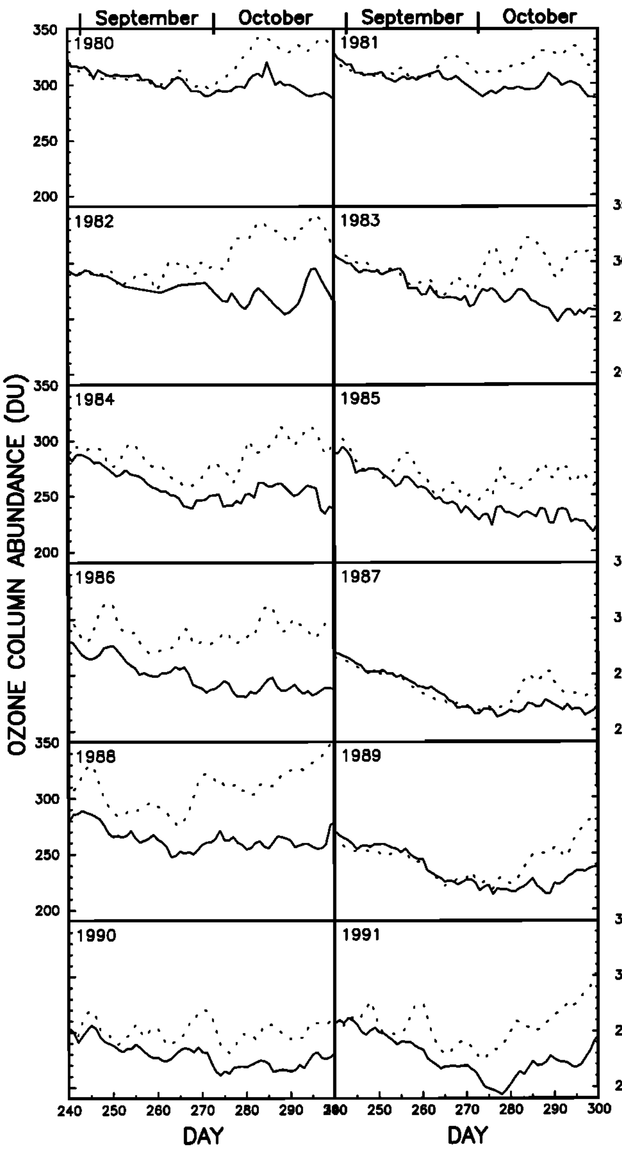

Figure 1. Area-weighted average ozone column abundance (DU) inside the polar vortex bound by PV contour of -50 PVU from days 240 to 300 for 1980 - 1991. Dotted lines denote area-weighted average ozone column abundance (DU) between 60°S and 90°S.

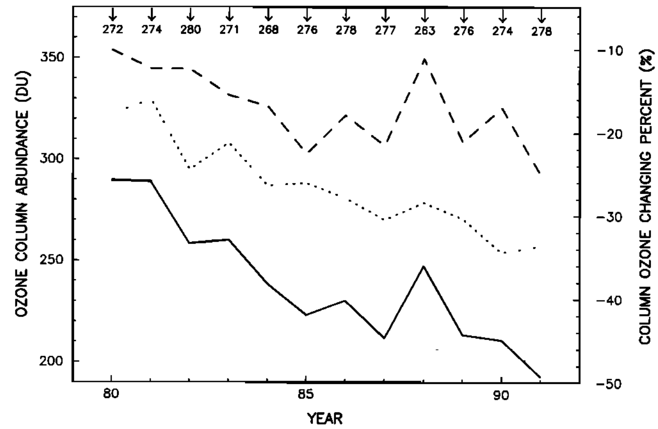

Figure 2a. The area-weighted average ozone column abundance (DU) on day 240 of each year (dotted curve); same for the minimum value attained in each year between days 240 and 290 (solid curve). Dashed line gives the percent loss of ozone (relative to day 240). The days on which the minimum value for the area-weighted average ozone column abundance occurred are indicated.

vortex and its seasonal decrease in area during southern spring. The latter is evident as a tendency for the zonal and vortex delimited averages to separate after September. As the vortex shrinks seasonally, the zonal average includes more of the higher ozone values outside the vortex. The short-period fluctuations (i.e., of the order of a few to several days) are more pronounced in the high-latitude zonal average. Similar but smaller amplitude fluctuations appear in the vortex-limited area-weighted column ozone. These fluctuations, often correlated with those of the zonal average, suggest that synoptic-scale disturbances of the vortex, of ozone in the vortex, or of both are not filtered out by averaging within the vortex.

Both averages show the decline of column ozone in September - October over the last decade. The years 1986 and 1988 are notable in that the 60°S latitude and vortex-limited averages differ even in early September. These years were dynamically active [*Krueger et al.*, 1989; *Schoeberl et al.*, 1989; *Kanzawa* and *Kawaguchi*, 1990]: the vortex and ozone hole were, by southern hemisphere standards, often greatly distorted and displaced away from the geographic south pole. By contrast, the polar zonal and vortex-averaged column ozone averages are remarkably similar in 1987 and 1989, indicating that in those years the vortex was nearly zonally symmetric and extended at least to 60°S.

Similar differences extend to other years as well, with the polar zonal and vortex area-weighted averages of column ozone agreeing more closely in September during odd than during even years. Furthermore, as seen in Plate 2, the vortex-limited average of column ozone tends to decrease significantly from 1981 to 1982 and 1983 to 1984, but with the column ozone averages relatively unchanged from 1980 to 1981, 1982 to 1983, and until October 1984 to 1985. A different pattern follows in 1986, with ozone average values similar to those in 1985. These are followed by a significant decrease in September - October 1987 but substantially increased values in 1988, when vortex-limited column ozone values are as high or higher than those for the same months in 1984. After the anomaly of 1988 the vortex-limited ozone average again declines, with the lowest averaged values occurring in October 1991. In September the years 1990 and 1991 are reminiscent of the pattern in vortex-limited averaged column ozone before 1986. The lowest vortex-averaged ozone column abundance occurs in 1991. This result is consistent with that based on an analysis of the ozone minimum value in October [*Krueger et al.*, 1992]. *Hofmann et al.* [1992] argued that the penetration of volcanic aerosols from Pinatubo and Hudson into the polar vortex might have enhanced the depletion of ozone. However, *Schoeberl et al.* [1993] showed that the polar vortex and the midlatitude regions were nearly isolated from each other during the late August period. The issue of what caused the unusually low ozone in 1991 remains unresolved.

Figure 2a shows the vortex-averaged value of column ozone on day 240, the minimum value before day 290 (mid-October), and the day on which the minimum occurred for each year. As seen here and in Table 1, the minimum vortex-averaged

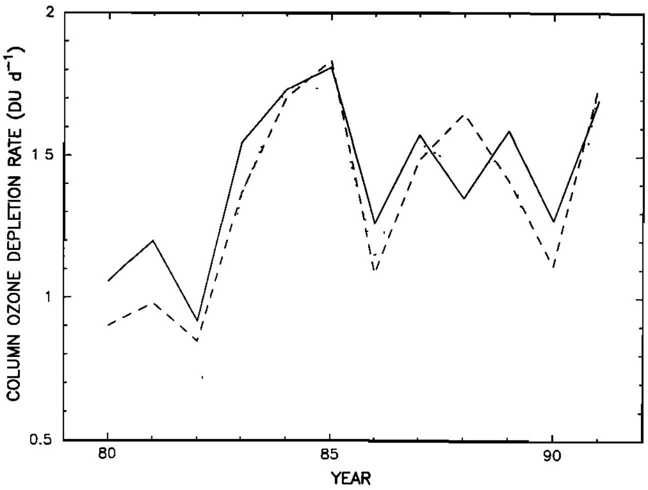

Figure 2b. Mean ozone column depletion rate (DU d^{-1}) in the polar vortex. The solid line denotes the rate computed using ozone values on day 240 and the day of minimum ozone. The dashed line is based on a 5-day average of the initial value of ozone from days 240 to 244. The dotted line is based on a linear least squares fit to the average ozone values between day 240 and the day of minimum ozone. Details are summarized in Table 1.

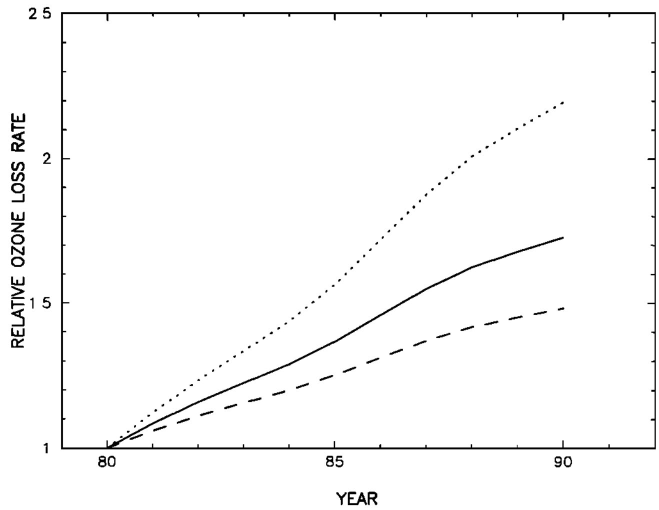

Figure 2c. The relative rate of ozone loss in October (solid line) as estimated using the model of *Sander et al.* [1989]. The total chlorine (Cl_y) in the stratosphere from 1980 to 1990 is taken from the model of *Weisenstein et al.* [1992]. The dashed and dotted lines give the rate of ozone loss if it were proportional to Cl_y and Cl_y^2, respectively. All values are normalized to that of 1980, the year with Cl_y = 1.44 ppbv.

column amount occurred in all years during the last week of September and the first week of October, except for 1988 when it occurred nearly a week earlier in mid-September.

The divergence of the day 240 and the minimum value curves indicates that the average loss of ozone during this season early in the decade is about half that at the end of the decade, except for the anomalous year 1988. The "initial value" of vortex-average column ozone on day 240 has declined by 20% over the decade. Recent observations by the Microwave Limb Sounder (MLS) onboard UARS of higher ClO and lower ozone as early as mid-July [*Waters et al.*, 1993a, b; *Santee et al.*, 1995] indicate that some chlorine activation and photochemical ozone loss occur in southern midwinter. The depletion of ozone at this early time (while the geographic south pole is still in the dark) can occur because the south polar vortex is sufficiently large that its outer regions are in sunlight even during the period of polar night. Air there may be exposed to polar stratospheric clouds in the same regions or can move into the sunlit regions following exposure to PSCs in areas of polar night. The ensuing chemical loss of ozone in the sunlit portions of the vortex due to chlorine photochemistry could account, then, for the downward trend of vortex-average column ozone at the end of August.

Figure 2b shows the decadal variation in the mean ozone depletion rate from day 240 to the day when the minimum area-averaged column ozone is found in the polar vortex (solid line). In the first three years the ozone depletion rate during this September-October period is only about 1 DU d^{-1}. After 1982 the ozone depletion rate increases to a value of 1.8 DU d^{-1} in 1985. Since then, the ozone depletion rate has fluctuated around 1.5 DU d^{-1} and shows a possible QBO signature. The dashed line in Figure 2b is obtained in the same way as for the solid line, except that the mean of 5 days (days 240 - 244) is used for the initial value. The dotted line is obtained by using a linear least squares fit to the data. Detailed information related to the computation of various rates is summarized in Table 1. The depletion rates are particularly difficult to estimate in 1988 and 1990, because the variation during the season is more nonlinear in those years (Plate 2).

The likely explanation for the rapid increase in the ozone depletion rate is the rapid rise of total chlorine (Cl_y) in the lower stratosphere in the early part of the decade. The dominant effect of chlorine chemistry on ozone loss is quadratic in ClO [*Molina* and *Molina*, 1987a; *Sander et al.*, 1989; *Anderson et al.*, 1991], with smaller contributions from coupling to bromine and HO_x chemistry. The latter effects are linear in ClO. Figure 2c (solid line) shows the rate of ozone loss (relative to 1980) as estimated using the model of *Sander et al.* [1989]. The values of Cl_y in the stratosphere are taken from a model [*Weisenstein et al.*, 1992], which is based on the release of chlorofluorocarbons, atmospheric observations, and modeling. This shows that the chemical forcing on the ozone hole has increased from 1980 to 1985 by about 40%, which is about 60% of the increase in ozone depletion rate in the same period (see Figure 2b). For comparison, we also compute the chemical forcing if it were linear in Cl_y (dashed line) or quadratic in Cl_y (dotted line). The BrO mixing ratio was set equal to 5 (pptv) in all these calculations [*Sander et al.*, 1989]. We should point out that despite the dominance of the Molina-Molina catalytic cycle that is quadratic in ClO, the chemical forcing is not quadratic in Cl_y. The reason is that as Cl_y increases, part of the reactive chlorine is sequestered in the Cl_2O_2 dimer, thus lowering its efficiency for the destruction of ozone.

After 1985 the depletion of ozone apparently reached saturation, in that the ozone depletion rates did not increase with increasing chlorine and, in fact, were probably moderated in later years by dynamical processes. These could horizontally modulate the region of photochemical loss within the vortex (i.e., the chemically perturbed region) or the area of

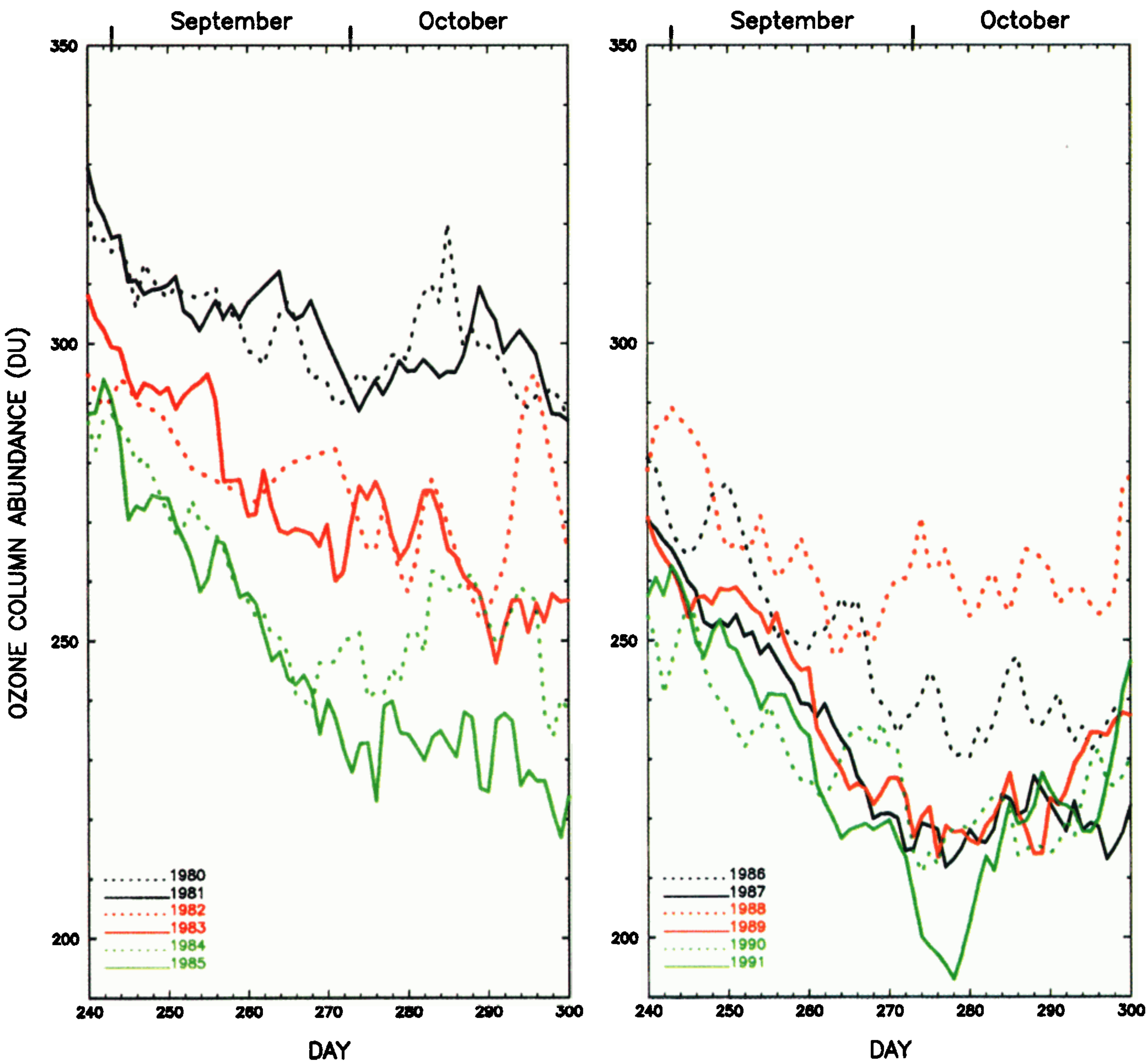

Plate 2. Alternative display of the solid lines from Figure 1. The solid lines are for the odd years; the dotted lines are for the even years.

the vortex itself; they could also affect the rate of vertical motion and so the seasonal downwelling of air with higher ozone mixing ratios from the middle stratosphere in the polar vortex [*Manney et al.*, 1995].

The moderated depletion rate after 1985 for column ozone may also be due to the fact that the loss rate depends on ozone itself, and ozone amounts during this later period are already very low during this season. This effect can be illustrated by dividing for each year the difference between the day of minimum vortex-averaged column ozone and day 240 by the vortex-averaged amount on Day 240. As seen in Figure 2a (dashed line), the 1985 loss rate is sustained in later years, with 1988 again being a most exceptional year.

We can compare the loss rates of the vortex-average area-weighted column ozone of Figure 2b to those computed by *Lait et al.* [1989]. They used the minimum TOMS column ozone value found poleward of 30°S on any given day to estimate linear decline rates D for the late August to September period. They then estimated the change over the period 1979 - 1987 by least squares fitting of the September variations and obtained:

$$D\ [\mathrm{DU\ d^{-1}}] = -0.19(\mathrm{year} - 1980) - 0.65$$

This can be compared to comparable linear least squares fitting of the values shown in Figure 2b for 1980 - 1987:

$$D\ [\mathrm{DU\ d^{-1}}] = -0.14(\mathrm{year} - 1980) - 0.71$$

As noted above, the distinct change in decline rates for the vortex-averaged column ozone occurs after 1985. A linear least squares fitting for the September decline rates for 1980 - 1985 yields

$$D\ [\mathrm{DU\ d^{-1}}] = -0.25(\mathrm{year} - 1980) - 0.51$$

Table 1. Column Ozone Depletion Rate (DU d^{-1}) As Computed by Three Methods

	Method 1[a]				Method 2[b]		Method 3[c]
Year	Ω (DU) (240)	Ω (DU) (Minimum)	Day of Minimum	Depletion Rate	Ω (DU) (240)	Depletion Rate	Depletion Rate
1980	324	293	272	1.06	320	0.90	0.76
1981	332	291	274	1.20	323	0.98	0.64
1982	297	261	280	0.92	293	0.85	0.63
1983	309	262	271	1.55	303	1.37	1.36
1984	288	241	268	1.73	288	1.70	1.74
1985	289	225	276	1.81	292	1.83	1.73
1986	286	233	278	1.26	277	1.09	1.14
1987	271	214	277	1.57	269	1.49	1.55
1988	283	251	263	1.35	289	1.65	1.45
1989	271	217	276	1.59	266	1.42	1.50
1990	256	213	274	1.27	251	1.11	0.84
1991	259	198	278	1.70	262	1.75	1.76

Mean column ozone depletion rate (DU d^{-1}) in the polar vortex.

[a] Method 1 uses ozone values on day 240 and the day of minimum ozone.

[b] Method 2 is based on a 5-day average of the initial value of ozone from days 240 to 244.

[c] Method 3 is based on a linear least squares fit to the ozone values between day 240 and the day of minimum ozone.

which translates into an increase over those years by a factor of 3 or more. This would reduce to a factor of 2 using the decline rates computed in Figure 2b as simple differences. Both of these factors are larger than that suggested by the growth in stratospheric chlorine (Figure 2c).

Lait et al. [1989] found that the residuals of the September decline rates of minimum column ozone values from the interannual linear fit showed a distinct QBO signature. This is consistent with the September decline rates of the vortex-averaged column ozone shown in Figure 2b for years before 1986, except for 1984. Oscillations in September decline rates after 1985 have the character and magnitude of those during September in 1980 - 1982. However, the distinctive QBO-like pattern shown in Plate 2 for vortex-averaged column ozone in 1980 - 1985 is not apparent in later years.

Figure 3 shows the vortex-average area-weighted column ozone for the period when the ozone hole is deepest from 1980 to 1991. The change in trend from year to year for the years before and after 1986 is clearly seen, as is the anomaly of the 1988 column ozone variation. The area-averaged ozone column abundance decreases with a slope of 3.8% (11.3 DU) per year before 1987 and 0.9% (2.0 DU) per year after that (the year 1988 is anomalous and is not included in the trend analysis). The average rate of decline is 25% per decade from 1980 to 1991. These rates of decline of the vortex-averaged column ozone during one month (mid-September to mid-October) of lowest ozone may be compared to the value 30% per decade obtained on the basis of zonal averages in the southern polar regions from 1979 to 1990 [*Herman et al.*, 1993]; similar rates of decline were also obtained when column ozone was mapped as a function of PV within the Antarctic polar vortex for September over the same period [*Randel* and *Wu*, 1995]. However, the distinct break in trend after 1985 has not been previously noted.

4.2. Growth of the Ozone Hole

The results presented above utilized area-weighted averages of column ozone over the entire polar vortex. Here the area-mapping technique is used to examine the evolution of the ozone hole in more detail. Plate 3 shows the area enclosed by contours of ozone column abundance, again plotted versus enclosed area in units of $2\pi R^2$. As before, the period covered is from days 240 to 300 for 1980 - 1991, and the domain of integration is confined inside the polar vortex by PV contour of -50 PVU at 500 K. In these plots we have used 5-day smoothing to remove high-frequency variations. Since there are no ozone data for the part of the vortex in the dark, the bottom left parts of these figures are blank. Note that the areas bounded by column ozone contours less than 250 DU increase

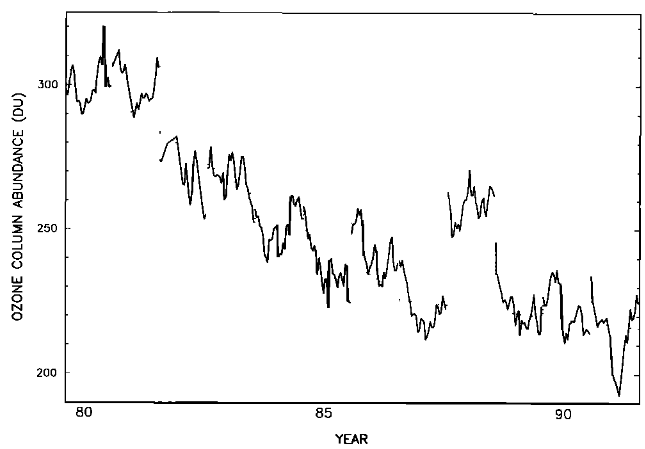

Figure 3. The vortex-averaged area-weighted column ozone abundance (DU) in the vortex from days 260 to 290 (mid-September to mid-October), the season of minimum column ozone each year. The steep dotted line is the linear least squares fit to the data during this month from 1980 and 1987. The slope is 3.8% (11.3 DU) per year with the averaged ozone value 300 DU during this time in 1980 as reference. The less steep dotted line is a similar fit to the data from 1987 to 1991 but excluding 1988. The slope is 0.9% (2.0 DU) per year by using the averaged ozone value 222 DU during this time in 1987.

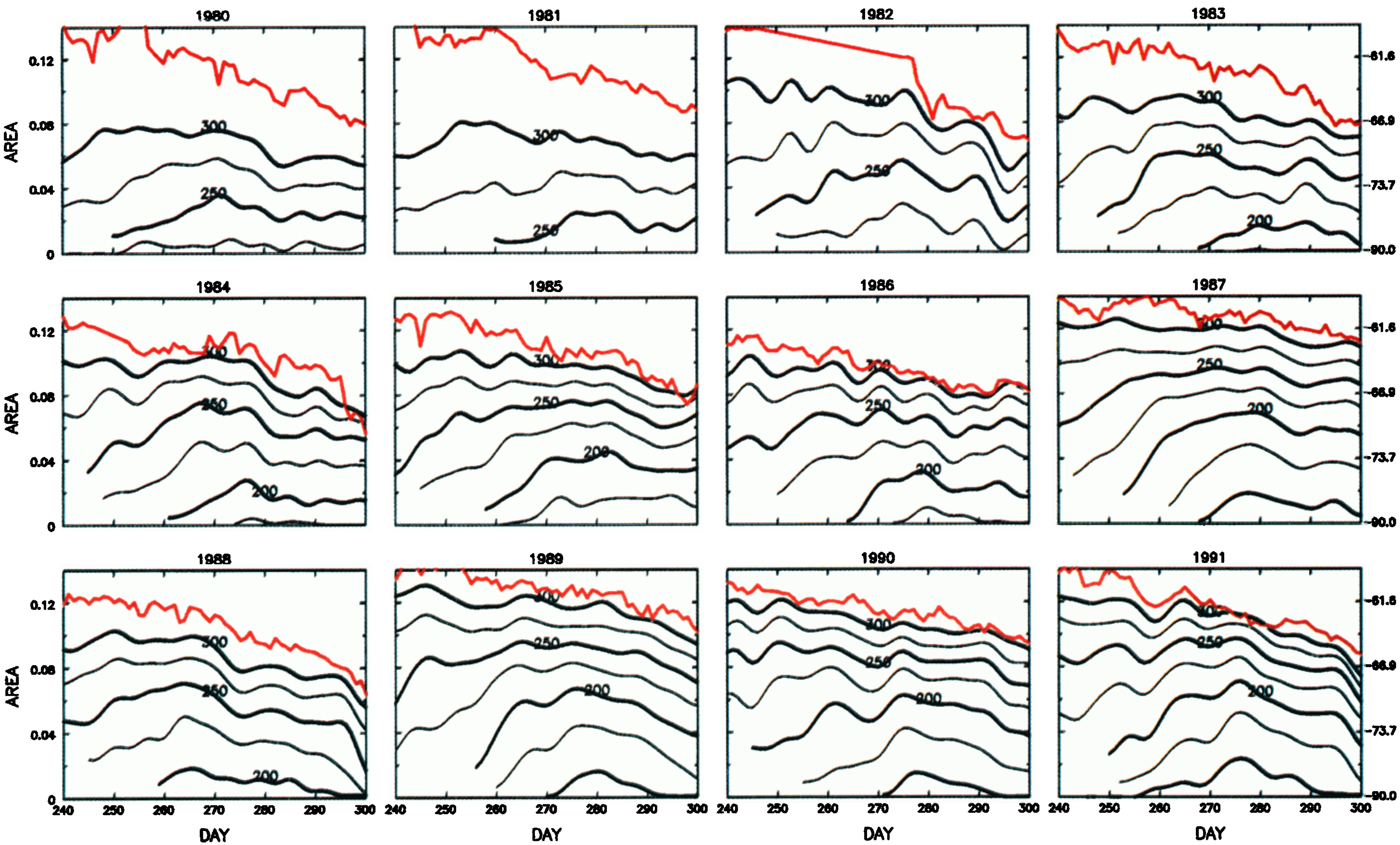

Plate 3. Contours of ozone column abundance (DU) plotted versus enclosed area in units of $2\pi R^2$ (R is the radius of Earth) from day 240 to day 300 for 1980 - 1991. The area mapped is confined within the vortex as defined by the -50 PVU contour (red line). Areas greater than the vortex area reflect computational uncertainty due to using different grids for PV and O_3 and to different gradients in these fields at the vortex edge. The blank space in the bottom left corner is caused by the lack of data. Positive slopes indicate ozone decrease; negative slopes indicate ozone increase. Contour increment is 25 DU. The equivalent latitude is indicated in the right-hand axis.

rapidly around day 260 (mid-September), implying that the ozone hole is expanding and the column ozone is decreasing inside the vortex. By day 280 (early October) the ozone hole has reached its greatest extent and depth. Typically, the area remains constant then for a few days before it begins to shrink. This reflects first the warming of the vortex air and eventually the seasonal weakening of the vortex itself, which has generally broken up by November.

As can be seen from Plate 3, the ozone column abundance contour of 200 DU does not appear until 1983 but occupies a maximum area of 7% of the southern hemisphere in 1991. The 150-DU contour appears in 1987. These regions are well inside the polar vortex. On the vortex boundary, where the ozone column abundance is usually greater than 300 DU, the seasonal change of area reflects the area of the vortex. In this region (e.g., the 300-DU contour) there is a little increase during the seasonal formation of the ozone hole, but mostly the area is constant until the vortex begins to decrease in area. As seen in Plate 3, the ozone hole develops during late August to mid-September and is largely independent of day-to-day variations near the vortex boundary. These different trends and the general steepening of the gradients at the vortex boundary indicate that the air within the vortex is isolated in the lower stratosphere from air at similar levels outside the vortex. The steepening of ozone gradients at the vortex boundary results from the spatial expansion as well as the deepening of the ozone hole over that period.

The anomalous behavior of 1988 is clearly seen by comparing the areas bound by the 200-DU contour in the successive years 1987, 1988, and 1989. The maximum areas in 1987 and 1989 exceed 6% of the southern hemisphere, whereas in 1988 the maximum area is only about 2% of the southern hemisphere. Similar area mapping results were obtained by *Stolarski et al.* [1990, Figure 3] for these years.

In Plate 3 the increasing area in successive years of the ozone contours inside the vortex on day 240 indicates that the ozone column abundance must have already begun to decrease earlier in the season. The area mapping (not shown here) of column ozone from days 220 to 320 (early August to mid-November) shows ozone depletion even before day 220. This is consistent with the trends in the vortex-averaged column ozone (Plate 2, Figure 2a) and, as discussed earlier, with recent observations by UARS.

Area mapping provides a convenient measure of the magnitude of maximum ozone depletion each year. Consider the area bounded by a low-ozone contour, such as the 250-DU contour (Plate 3). Figure 4 shows the maximum area enclosed by this contour between days 260 and 300 (mid-September to late October) and the days on which the maximum area is attained in each year. Except in 1981 and 1986 the increase of the maximum area before 1987 is roughly linear. The linear least squares fit shows a slope of 8.36 x 10^{-3} per year from 1980 to 1987. The same fitting procedure shows no trend for 1987 - 1991 (with 1988 excluded); the average over the whole period is 5.49 x 10^{-3} per year from 1980 to 1991. From the definition of equivalent latitude θ_e,

$$\Delta A(t, \Omega^*) = \sin(90° + \theta_e)\, \Delta\theta_e,$$

we can express the change in the maximum area of the 250 DU contour as an equatorward expansion at a rate of approximately 1.35° per year before 1987 and at a rate of 0.84° per year for the full 1980 - 1991 period (see Table 2). The

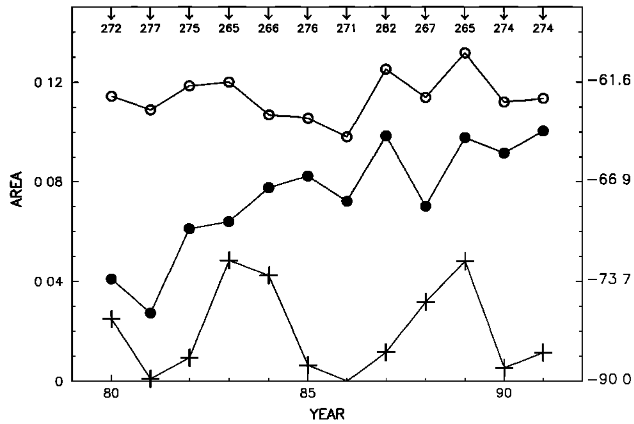

Figure 4. The maximum area (dots) enclosed by the 250 DU contour in each year between days 260 and 300. The days on which the maximum area is attained are indicated. Also shown for the same days are the areas of the vortex, as indicated by the -52 PVU contour (open circle) on the 500 K isentropic surface and of the 195 K isotherm (plus), also at 500 K. The equivalent latitude is indicated in the right-hand axis.

maximum area of the ozone hole usually occurs in late September or early October, consistent with the vortex-average trends discussed earlier, and also near or before the time when the minimum ozone column amounts (not shown) are found.

One may ask whether this rapid growth in the chemical ozone hole is accompanied (or partly driven) by climatological changes in the stratosphere [e.g., *Mahlman et al.*, 1986, 1994; *Angell*, 1993]. To address this question qualitatively, we compute the areas of the polar vortex using the -52 PVU contour and the 195 K isotherm, both on the 500 K isentropic surface. Figure 4 shows these areas on the same days of maximum area of the 250-DU column ozone contour. The vortex (PV) area has little trend over the decade (only 0.05° yr^{-1}; see Table 2), although it fluctuates interannually about a more or less constant value. In 1980 the vortex area at 500 K is nearly twice that of the 250 DU contour, but the latter increases until it fills more than 70% of the entire vortex by 1985. After 1985 the fluctuations in the area of the 250-DU contour reflect those of the vortex area itself, which limits the size of the 250-DU area and thus of the ozone hole.

The lack of correlation in Figure 4 between the areas enclosed by the 195 K temperature, an indicator of the horizontal extent of PSCs, and the areas enclosed the 250-DU column ozone contour, on the days when the latter is a maximum, is not surprising given that much of the processing of polar air would have occurred earlier. Furthermore, stratospheric winds can expose most of the vortex interior air to PSCs, even if the latter occupies only part of the vortex.

Table 2. Decadal Growth Rates (degree yr^{-1}) of Areas Bound by Isopleths of O_3 (250 DU), PV (-52 PVU), and T (195 K)

	1980-1987	1987-1991	1980-1991	Reference
O_3	1.35±0.27	0.00±0.28[a]	0.84±0.16	Figure 4
PV	-	-	0.05±0.10	Figure 4
T	-	-	0.05±0.19	Figure 4

[a]The year 1988 is not included in the calculation.

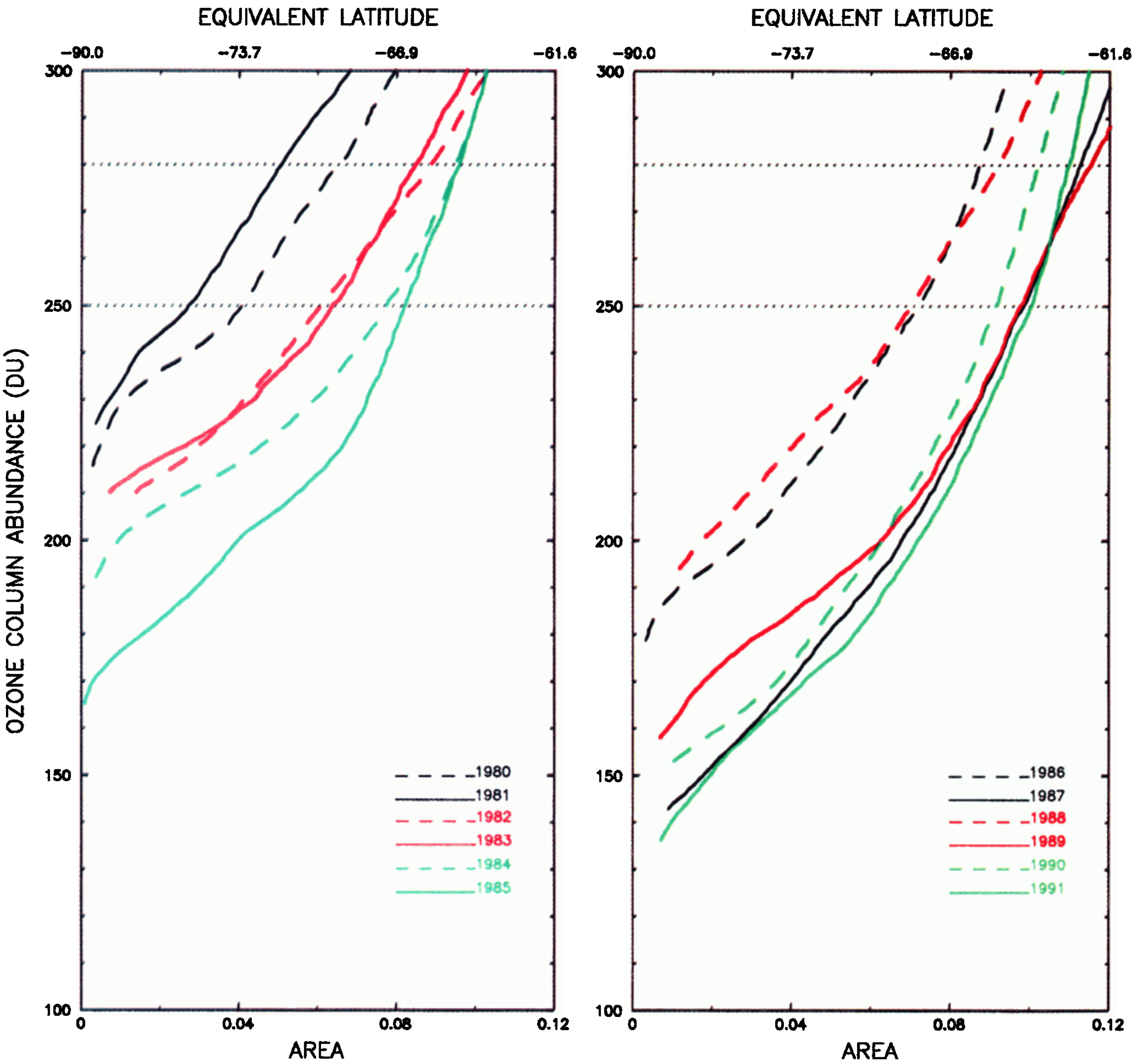

Plate 4. Ozone column abundance versus area on the days shown in Figure 4 for each year for 1980 - 1991. The equivalent latitude is indicated in the top axis.

This has been shown most dramatically for the north [*Waters et al.*, 1993a], where PSCs are not so extensive. The more critical factor may be exposure to sunlight. For short time periods this exposure depends critically on details of the air motion within the vortex, but for periods of a few weeks or more this factor depends more crucially on the vortex extent and location and may be reflected in the vortex area shown in Figure 4. At least to first order, most of the growth of the ozone hole over the last decade appears driven by chemistry alone, due to increasing stratospheric chlorine. This increase has slowed down, not because the stratospheric chlorine loading or activation has stopped increasing but simply because the region of ozone-depleted air now fills (horizontally) much of the polar vortex, whose own fluctuations in area reflect interannual variability in dynamical activity.

This limiting of the ozone loss region to the vortex is reflected in the fact that after 1983, high contour values of column ozone (larger than 300 DU) are near the vortex edge as defined here on the 500 K isentropic surface (Plate 3), except in 1988. In that year the chemically perturbed region was distinctly within the vortex (Plate 3, Figure 4), as described by *Schoeberl et al.* [1989] in their analysis of the 1987 Antarctic campaign (AAOE) data. Plate 3 and Figure 4 show that 1988 is not representative in this regard of all other years following 1985. Thus further chemical ozone loss during this season now depends upon factors that would expand the south polar vortex region horizontally or which would extend the zone of chemical depletion vertically, as happened in 1993 due to heterogeneous chemical effects associated with the entrainment of Mount Pinatubo aerosols into the south polar vortex [*Hofmann et al.*, 1994].

Plate 4 shows the variations in ozone column abundance versus area on the days of maximum area in the 250-DU contour (Figure 4) each year from 1980 to 1991. The greater extent of ozone-poor air in the years after 1985 is again apparent, as is the anomaly of ozone values in 1988. As more ozone gets depleted, the radial gradient of ozone in the vortex gets steeper, as can be seen from Figure 5, which shows the average gradient of the transition region between 250 DU and 280 DU, defined as [280-250(DU)]/[θ_e(280DU)- θ_e(250DU)]. Years since 1984 are typically characterized by larger gradients as a result of greater ozone depletion, as discussed earlier, with notable exceptions in 1988 and 1989. As we have remarked repeatedly, 1988 is anomalous in many respects. The relatively weak column ozone gradient in 1989 is partly due to the fact that the vortex area in 1989 was the largest in the record (Figure 4). Although Plate 4 and Figure 5 show results for a single day, the area-contours of Plate 3 show that the results are representative of the ozone hole period in those years. Finally, as pointed out by previous studies [*McIntyre*, 1989; *Hartmann et al.*, 1989], the steep gradients of PV and ozone indicate relative isolation of the chemically perturbed region from air outside the vortex.

Plate 5 shows the time rate of change of ozone column abundance over a period from days 240 to 300 (late August to late October) for each year from 1980 to 1991. The results for 1987 have been shown previously by *Yung et al.* [1990, Figure 4]; this figure extends that result to 12 years. The average rate of ozone depletion in the vortex is 1 to 2 DU d^{-1}, consistent with that shown in Figure 2b.

Larger depletion rates (5 DU d^{-1}) are prevalent near the center of the vortex (Plate 5). In some of the recent years, such as the dynamically quiescent years 1987 and 1989, large ozone depletion rates can persist for long periods of time. In most years the development of the ozone hole is periodically interrupted by increases of ozone with a period about 10 - 20 days, with the changes being more prominent away from the vortex center. These increases may be due to the action of synoptic-scale systems which are known to produce minor warmings in the midstratosphere, as these systems have a characteristic timescale of 10 - 20 days [e.g., *Fishbein, et al.*, 1993; *Manney, et al.*, 1993a]. These dynamical events affect ozone in the lower stratosphere within the vortex by moving ozone into the vortex in midstratosphere and then down to lower levels [e.g., *Leovy et al.*, 1985; *Manney, et al.*, 1994b].

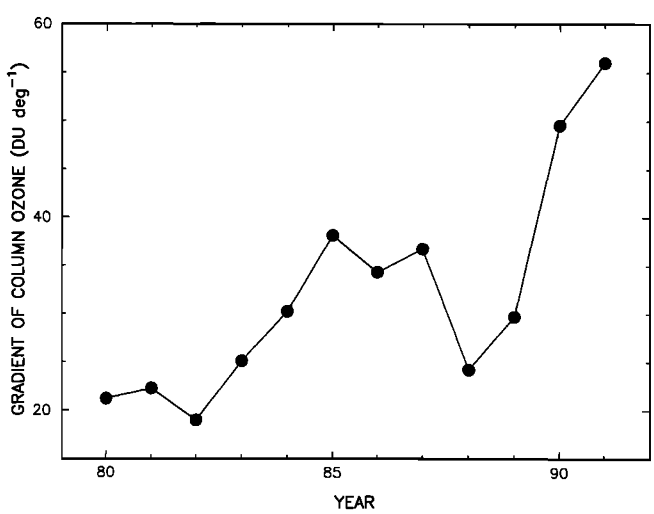

Figure 5. Average ozone column gradient with respect to equivalent latitude, computed as described in the text, for the same days as in Plate 4.

Some care needs to be taken when interpreting the area-mapped fields shown in Plates 3 and 5, as the area mapping tends to filter out the planetary-scale features. However, after the ozone hole reaches its maximum depth around day 280 in early spring, the polar vortex starts to shrink and ozone column abundance starts to recover. Significant warming events and erosion of the vortex are marked in Plate 5 by ozone increases extending nearly to the vortex center. Because of the anomalous circulation and extremely cold temperature of 1987 [*Randel*, 1988] the final warming and the breakup of the ozone hole was delayed 20 days (as compared with climatology). Plate 5 shows that the ozone depletion rate was never so persistent and as extensive as in 1987. This decrease was interrupted by a minor warming around day 280 and then terminated by the final warming after day 300.

In 1988 on the other hand, stratospheric temperatures had warmed significantly by late September, terminating most decreases in column ozone. The maximum total ozone decline is only 15% during September 1988 compared to nearly 50% during September 1987 [*Schoeberl et al.*, 1989; *Krueger et al.* 1989]. This appears to be the direct result of the brevity of the decline through September. Wave activity at the time was anomalously strong [*Manney et al.*, 1991] and the stratospheric sudden warming in late winter was reminiscent of major winter warmings in the northern hemisphere [*Kanzawa* and *Kawaguchi*, 1990]. The 1988 ozone hole began filling on October 19, nearly six weeks earlier than in 1987, and drifted out of the polar regions in mid-November for the earliest breakup since 1979.

5. Conclusions

The area-weighted ozone column abundance averaged over the polar vortex in late August (day 240), often viewed as the beginning of the ozone hole season, exhibits a steady decline from about 320 DU in 1980 to about 250 DU at the end of the decade. This amounts to a total drop of 70 DU in a decade (Table 1) and accounts for a large part of the overall decrease of more than 90 DU in vortex-averaged October ozone column abundance that has occurred over the same period. Most of that drop occurred before 1987, as the decline since then has been much smaller (Figure 3).

Part of this decadal decline may be due to the recurrent presence of the ozone hole every year. Because of the long lifetime of ozone in the lower stratosphere, there is persistence of depleted ozone from one year into the next. The effect, known as the dilution effect, has been modeled by *Sze et al.* [1989], *Prather et al.* [WMO, 1989, Chap. 1] and *Mahlman et al.* [1994]. These models predict a decline of ozone of 5 - 7% at polar latitudes in June due to the presence of an ozone hole, or about 15 - 20 DU. That still leaves 50 - 55 DU in the decadal decline unexplained. It is more likely that the result is due to increasing stratospheric chlorine producing increasing chemical loss of ozone in the south polar vortex even during

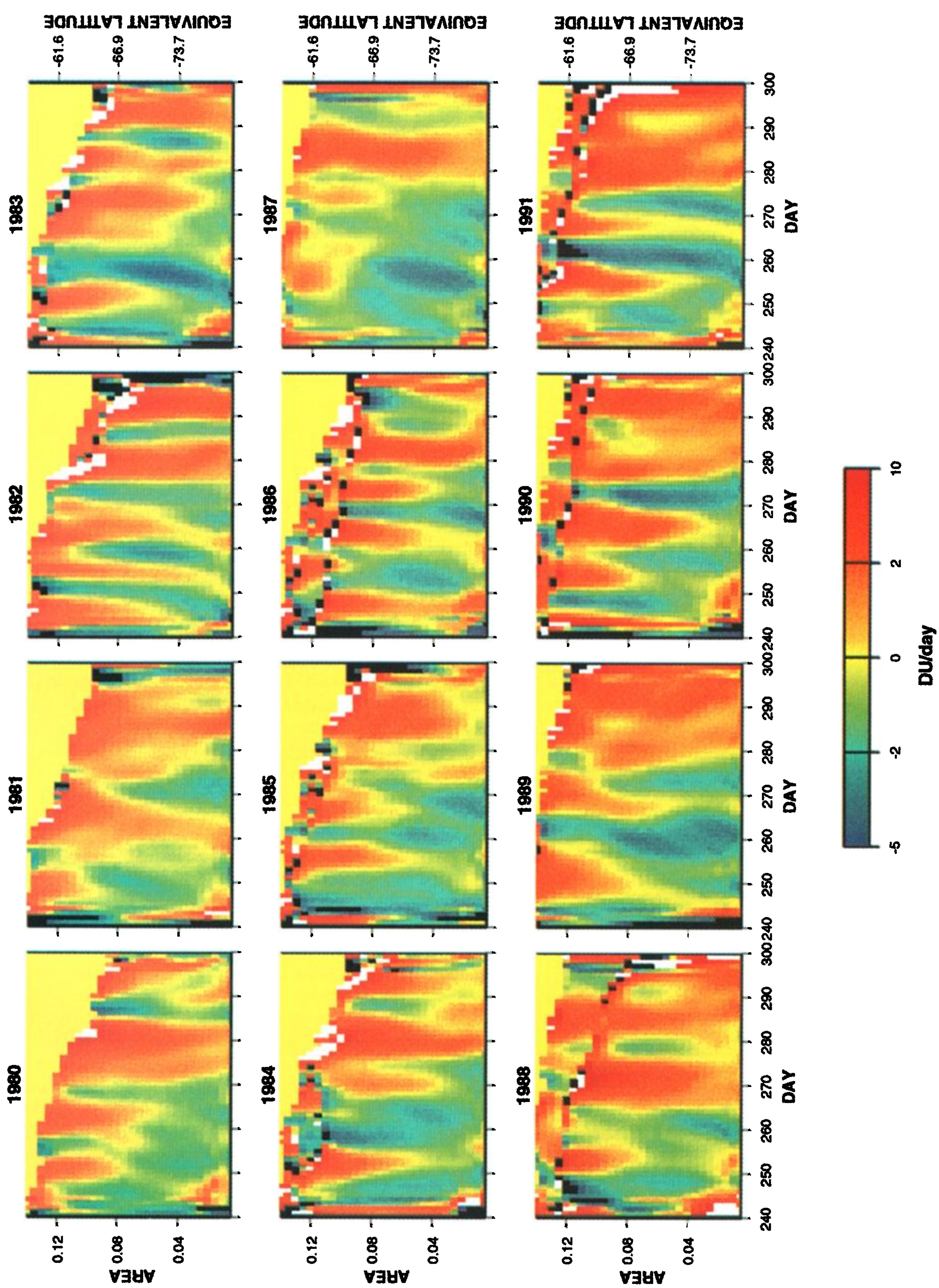

Plate 5. Contour plot of the time rate of change of ozone column abundance ($\partial\Omega/\partial t$), in DU d^{-1}, plotted versus enclosed area from day 240 to day 300 for 1980 - 1991. The horizontal axis is day number and the vertical axis is area in units of $2\pi R^2$ (R is the radius of Earth).

southern winter, as indicated by the recent UARS observations. Photochemical loss can occur because the southern vortex is sufficiently large that it has regions in sunlight even during this period of polar night (such loss could be more heavily weighted in our method because of the area-weighting of our vortex averages).

The mean ozone depletion rate in the vortex between late August (day 240) and the day of minimum vortex-averaged ozone (typically in late September or early October) has changed from about 1 DU d^{-1} at the beginning of the decade, rapidly increasing to about 1.8 DU d^{-1} by 1985, and apparently saturating thereafter (Figure 2b). This change in ozone depletion rates is qualitatively but perhaps not quantitatively consistent with chemical forcing of the ozone hole solely by chlorine (Figure 2c). The character and magnitude of fluctuations in the September depletion rates in 1980 - 1982 are comparable to those after 1985, and it is possible that these reflect the extratropical QBO in ozone. A QBO signature in the depletion rates derived here is perhaps not so strong as obtained by *Lait et al.* [1989] who analyzed minimum column ozone abundance and not vortex averages. The QBO signature is evident in the vortex ozone averages themselves for the years 1980 - 1985 but is not readily apparent in later years. That interannual variations can affect ozone abundance during this season is clearly indicated by the anomalously high column ozone values during September 1988, a very active year dynamically.

Area mapping of various column ozone contours provided some insight into the detailed morphology of the ozone depletion rate in the south polar vortex. The time rate of change of column ozone appears to be greater deeper in the vortex (Plate 5). Whether this reflects the exposure of air parcels to sunlight or some other factor was not investigated here. Many of the regions with the highest rates of change in ozone are cold enough that water-ice PSCs may form; if so, greater dehydration and denitrification of the vortex air may enhance the ozone loss there. Regions near the vortex edge may have greater downwelling of ozone-rich air during this season. The 10 to 20-day modulation of the ozone change rates within the vortex suggests that synoptic activity is affecting the rate of resupply, probably by descent of ozone from midstratospheric levels.

Our results suggest that further increases in stratospheric chlorine after 1985 did not significantly increase the horizontal extent of ozone loss, as the area of loss now filled most of the inner (i.e., isolated) region of the polar vortex in the lower stratosphere and was now limited by the area of the vortex. Thus depletion rates of the vortex-average ozone did not increase further and in fact were not so high as in 1985 until 1991, when volcanic aerosols may have extended the vertical extent of ozone chemical loss.

In summary, our results suggest that chemical loss due to stratospheric chlorine was significant even early in the 1980 - 1991 period studied here, as judged from a vortex average. Increases in ozone depletion rates since 1982 are consistent with increasing chlorine effects approaching saturation in 1985 as the region of ozone loss filled the isolated inner regions of the polar vortex during September. In more recent years the main effect of increased chlorine may have been to increase the winter time chemical loss of ozone, thereby lowering the threshold on which the ozone hole develops in August and September. This early loss would occur despite the polar darkness, due mainly to the great size of the southern winter vortex. The dilution effect discussed above may also play a role. This view is necessarily speculative until more quantitative dynamic-chemical simulations of chlorine activation and ozone loss are done.

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