

## Change in ozone trends at southern high latitudes

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[1] Long-term ozone variations at 60–70°S in spring are investigated using ground-based and satellite measurements. Strong positive correlation is shown between year-to-year variations of ozone and temperature in the Antarctic collar region in Septembers and Octobers. Based on this relationship, the effect of year-to-year variations in vortex dynamics has been filtered out. This process results in an ozone time series that shows increasing springtime ozone losses over the Antarctic until the mid-1990s. Since approximately 1997 the ozone losses have leveled off. The analysis confirms that this change is consistent across all instruments and is statistically significant at the 95% confidence level. This analysis quantifies the beginning of the recovery of the ozone hole, which is expected from the leveling off of stratospheric halogen loading due to the ban on CFCs and other halocarbons initiated by the Montreal Protocol. **Citation:** Yang, E.-S., D. M. Cunnold, M. J. Newchurch, and R. J. Salawitch (2005), Change in ozone trends at southern high latitudes, *Geophys. Res. Lett.*, 32, L12812, doi:10.1029/2004GL022296.

### 1. Introduction

[2] The Antarctic ozone hole phenomenon has been an unabated international concern ever since it was first recognized from the ground-based measurements at Halley Bay [Farman *et al.*, 1985]. Based on intensive research over the past two decades, the details on the Antarctic ozone hole are well established [e.g., *World Meteorological Organization (WMO)*, 2003, and references therein], Bodeker *et al.* [2002] and Newman *et al.* [2004] have shown that the size of the Antarctic ozone hole, with total column ozone less than 220 DU, steadily increased in the 1980's and that the growth slowed down in the 1990's. These changes have been related to long-term changes in halogens in the stratosphere based on Effective Equivalent Stratospheric Chlorine (EESC) [e.g., Newman *et al.*, 2004, Figure 2]. Coupled chemistry-climate simulations suggest that the minimum ozone column may either have already occurred or should occur within the next decade [WMO, 2003, chap. 3].

[3] The ozone hole size is particularly sensitive to temperature and chemistry at the inside edge of the vortex in September when temperatures rise above 195 K [Newman *et al.*, 2004; Lee *et al.*, 2001]. Year to year temperature

variations, produced by dynamical forcing, result in 10–20% changes in ozone hole size [Newman *et al.*, 2004]. Here we examine the consistency of ozone changes between 60° and 70°S measured by a number of instruments. We filter out effects of year-to-year dynamical variability using the correlation between temperature and ozone in the collar region [e.g., Newman *et al.*, 2004]. Long-term changes in filtered ozone are then related to changes in EESC.

### 2. Data

[4] Our analysis uses data from four different sources. The first consists of measurements of total ozone column from the Dobson spectrophotometers at Vernadsky (65.2°S) and Syowa (69.0°S), (<http://antarctica.ac.uk/met/jds/ozone> and <http://www.msc-smc.ec.gc.ca/woudc>). These data cover almost every day in September and October starting from about 1957. The second data source is ozonesonde measurements at Syowa, available almost once per week. Ozone columns between 12–25 km are selected because most ozone depletion occurs in this altitude range; this is slightly different from the 12–20 km range selected by WMO [2003, chap. 3]. The third data source consists of profiles obtained by the Stratospheric Aerosol and Gas Experiment (SAGE) II and Halogen Occultation Experiment (HALOE), used to obtain the stratospheric ozone columns starting from 2 km above the tropopause (<http://www-sage2.larc.nasa.gov> and <http://haloedata.larc.nasa.gov>). To ensure that all the time series cover the 1979 to 2004 period, these data have been supplemented by an approximation to stratospheric columns derived from Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) measurements from 1979–1984 (before the SAGE II measurements). For this purpose TOMS Level 2 data with reflectivity less than 15% (clear-sky conditions) were adjusted by subtracting the climatological differences between the SAGE II stratospheric column ozone and the TOMS total column ozone for Septembers and Octobers in 1984–1993. The fourth data source is the total ozone column time series for 1979–2003 based on merged TOMS/SBUV version 8 data ([http://code916.gsfc.nasa.gov/Data\\_services/merged](http://code916.gsfc.nasa.gov/Data_services/merged)). NCEP temperature data at 100 hPa (<http://www.cdc.noaa.gov/cdc/reanalysis>) have also been used.

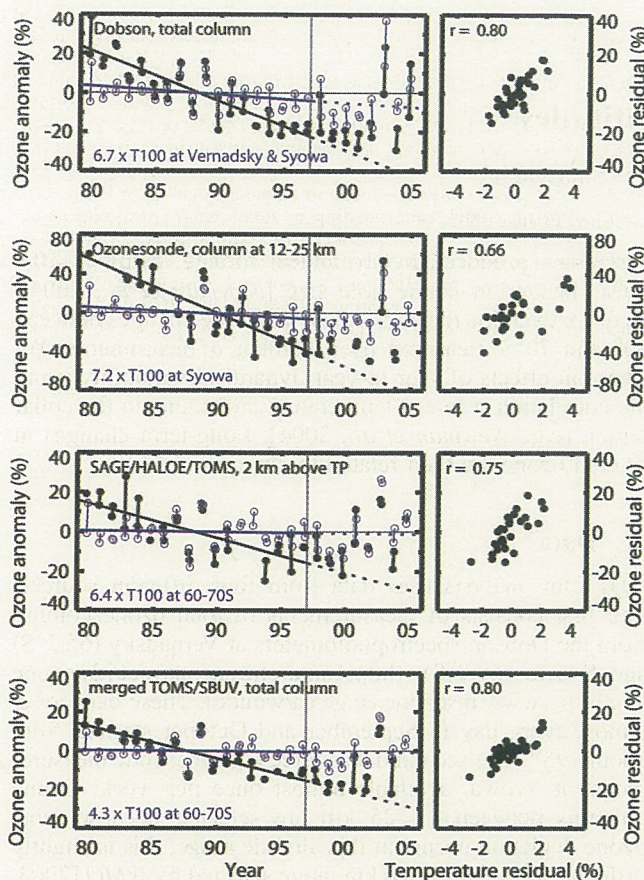
### 3. Temperature and Ozone Relations

[5] Ozone and temperature anomalies have been calculated in % departures from their climatological monthly means. The ozone (black filled dots) anomalies in Septembers and Octobers are shown in the left panels of Figure 1 (therefore, two data points each year for each data set). The level of ozone anomalies in Figure 1 seems to have been flat since the mid-1990's but with large year-to-year variance. To precisely define long-term ozone variations due to the change in halogen loading, it is important to remove the

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**Figure 1.** Monthly mean ozone (black filled circles) and ozone equivalent temperature (blue open circles) anomalies for Septembers and Octobers (left) and scatterplots of ozone residuals against temperature residuals at 100 hPa for 1979–1996 (right) for the Vernadsky (65.2°S) and Syowa (69.0°S) Dobson total ozone columns (averaged, first panels), ozonesonde ozone columns from 12–25 km for Syowa (second panels), SAGE/HALOE/(TOMS) stratospheric ozone columns at 60–70°S (third panels), and merged TOMS/SBUV total ozone columns at 60–70°S (fourth panels). The ozone and temperature residuals plotted for 1979–1996 result from subtracting the 1979–1996 trend line from their anomalies. The ozone equivalent temperature anomalies are the temperature anomalies after multiplication by 6.7, 7.2, 6.4, and 4.3 respectively for each data source. They represent the ozone variations attributable to the temperature perturbations.

variations due to dynamics. The right panels of Figure 1 show the ozone residuals versus the temperature residuals for 1979–1996, where residuals are defined herein as detrended anomalies. Tight correlation is found between ozone and temperature residuals at 100 hPa with correlation coefficients of 0.80, 0.66, 0.75, and 0.80 for the Dobson total, ozonesonde, SAGE/HALOE/(TOMS), and merged TOMS/SBUV total ozone columns, respectively. A similar correlation coefficient ( $-0.79$ ) between 50-hPa temperatures at 55–75°S and hole size has been reported by Newman *et al.* [2004].

[6] Here we further describe the temperature anomalies. Figure 1 shows time series of the temperature anomalies,

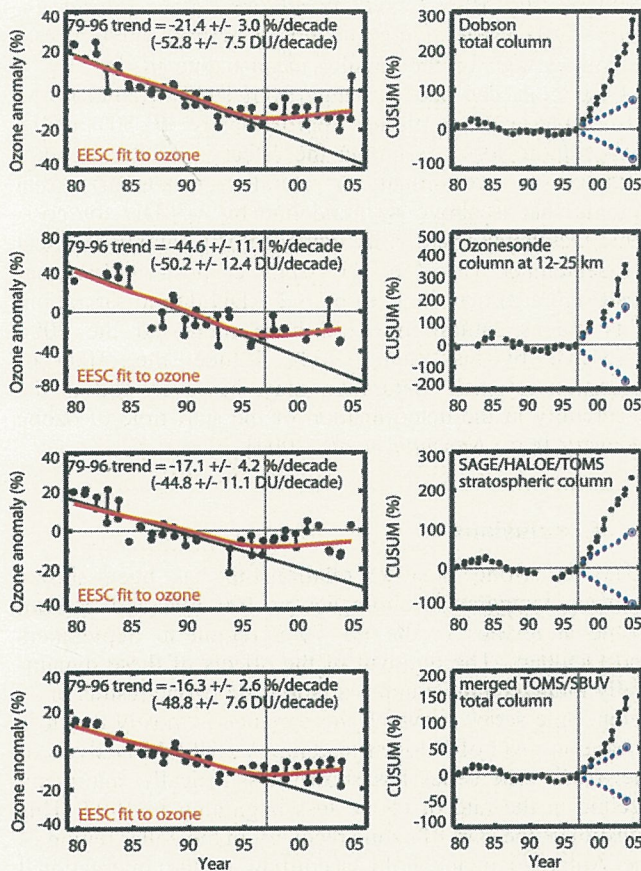
converted to “ozone equivalent anomalies” (blue open circles), that are found by multiplying the temperature anomalies by 6.7, 7.2, 6.4, and 4.3. These numerical values are the slopes in the scatter plots of the ozone versus temperature residuals, for the four respective data sets (as indicated). The smaller slope (4.3) found for the merged TOMS/SBUV data results from the uniform coverage of the 60–70°S region, compared to the other data sets that have coverage weighted towards higher latitudes within this 10° band. Indeed, the sensitivity of ozone response to temperature is found to increase to 7.4 at 70–80°S for the merged TOMS/SBUV data. These ozone equivalent anomalies represent the variations in ozone attributable to temperature perturbations. Most importantly, these time series exhibit no significant trends over the time periods of interest.

[7] The excellent correlation between ozone and temperature at 60–70°S in September and October is explained by heterogeneous chemistry occurring inside the polar vortex, which is associated with enhanced PSC formation under colder conditions [e.g., Bodeker *et al.*, 2002; Newman *et al.*, 2004]. This positive temperature-ozone relation has been found, based on our trajectory calculations, to be increased by transport and mixing of mid-latitude and polar air masses at the polar vortex edge. The contribution from two air masses with different origins is consistent with a temperature-ozone relation with no time lag in ozone response, because any memory would have been lost when the air masses mixed together. As a result, both reasons for temperature-ozone correlations are consistent with a direct (almost zero lagged) relationship between the September (October) temperatures and the September (October) ozone.

#### 4. Temperature Adjusted Ozone

[8] The left panels of Figure 2 show time series of ozone columns after removal of the ozone equivalent anomalies (blue open circles in the left panels of Figure 1). The variances of the ozone time series in Figure 2 are now significantly less than those in Figure 1. The black lines in Figure 2 show the linear ozone trend for 1979–1996. The selection of the turnaround point (end of 1996, or 1997.0) is somewhat flexible, within approximately  $\pm 1$  year, because ozone trends seem to change slowly around the turnaround point. The trend estimates and their two standard errors are indicated both in %/decade and DU/decade. The trend estimates for 1979–1996 are  $-52.8 \pm 7.5$ ,  $-50.2 \pm 12.4$ ,  $-44.8 \pm 11.1$ , and  $-48.8 \pm 7.6$  DU/decade for the Dobson total columns, ozonesonde ozone columns from 12–25 km, SAGE/HALOE/(TOMS) stratospheric ozone columns, and merged TOMS/SBUV total columns, respectively. These estimates are consistent with the  $-59 \pm 12$  DU/decade trend in the Antarctic ozone minima from 1979–2000, reported by WMO [2003, chap. 3].

[9] The ozone columns since 1997 in the left panels of Figure 2 are nearly flat and show systematically positive values over the 1979–1996 trend lines, which can be explained by a decline in stratospheric halogen loading (e.g., EESC) [Montzka *et al.*, 1999]. The red lines in the left panels of Figure 2 show fits to ozone of an EESC time series (from WMO [2003, Figure 1–23]). By definition, EESC is lagged by three years relative to the tropospheric measurements of total chlorine [WMO, 2003,



**Figure 2.** Temperature-adjusted ozone anomalies (left) and cumulative sum (CUSUM) of ozone residuals (right) in % for Dobson total ozone columns (first panels), ozone-sonde ozone columns at 12–25 km (second panels), SAGE/HALOE/TOMS stratospheric ozone columns at 60–70°S (third panels), and merged TOMS/SBUV total ozone columns at 60–70°S (fourth panels). The temperature-adjusted ozone anomalies are obtained by subtracting the ozone equivalent temperatures (blue open circles in the left panels of Figure 1) from the ozone anomalies (black filled dots in the left panels of Figure 1). The black line indicates the ozone trend calculated from observations for 1979–1996 and forecasted linearly afterwards. Linear trends and 95% confidence intervals for 1979–1996 are listed in %/decade and DU/decade. The red line shows the EESC fits to the temperature-adjusted ozone. The blue lines indicate the 95% confidence envelopes of departure from natural variability and trend model uncertainty.

section 1.2.5]. The EESC fits evidently explain most of the long-term changes in the temperature-adjusted ozone. Therefore, the flat ozone residuals since 1997 are a response to the leveling off of the stratospheric halogen loading since 1997.

[10] A cumulative (or consecutive) sum (CUSUM) of ozone residuals is a measure to detect systematic departures of ozone from the trend line [Reinsel, 2002; Newchurch *et al.*, 2003]. The ozone residuals after 1997, following subtraction of the projected 1979–1996 trend line, show systematically positive values. The 95% confidence limits for unbiased random walks about the projected trend line

appear as the blue traces in the right panels of Figure 2, where the limits also include uncertainty in trend model estimates. All CUSUM values are beyond the 95% uncertainty envelopes, indicating that the ozone trend after 1997 is different from the 1979–1996 trend at the 95% confidence level. Based on the definition of recovery put forth by the International Ozone Commission, “a statistically significant slowing of the downward trend” <http://ioc.atmos.uiuc.edu/documents/Statement-QOS2004.pdf>, the data shown in Figure 2 constitute the first stage of recovery for Antarctic ozone.

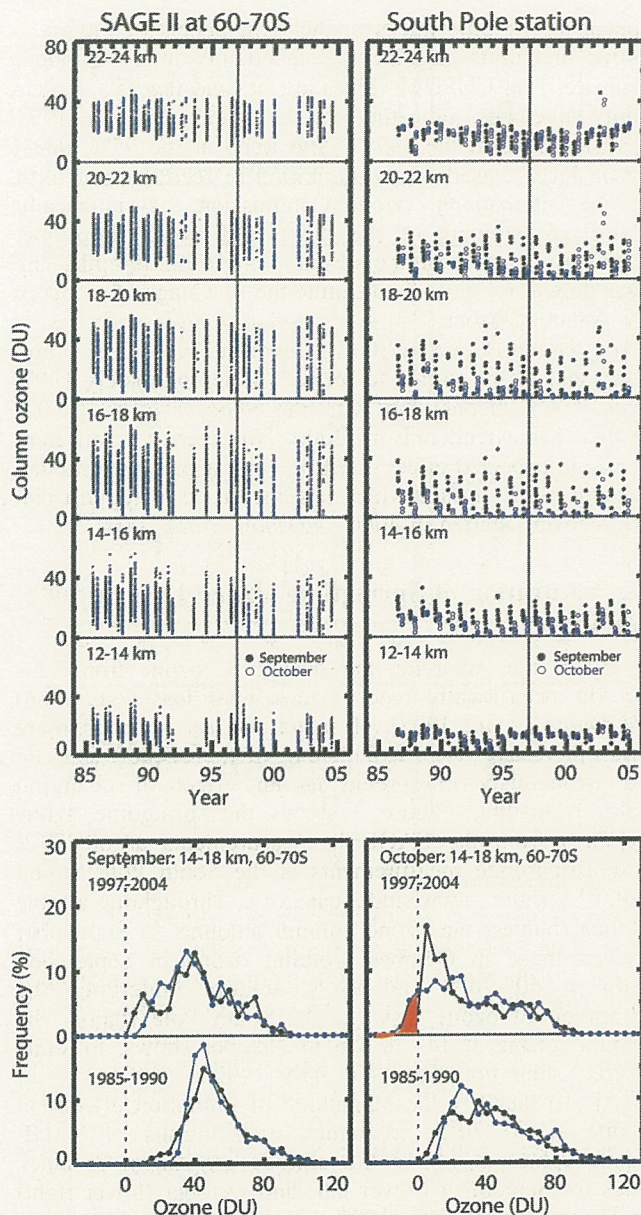
[11] Based on the EESC fit shown in Figure 2, it would seem that ozone started to recover in approximately 1997. This is not inconsistent with the Bodeker *et al.* [2002] results that extend only to 2000. However, it is different from the expected effect of EESC with a six year time lag relative to the surface which has been used to suggest that the recovery started in 2000 [Newman *et al.*, 2004].

## 5. Saturation of Springtime Ozone Loss in the Antarctic

[12] In the Antarctic, the springtime ozone from 12–20 km occasionally reaches near total loss (saturation) [Hofmann *et al.*, 1997]. If saturation has occurred more often in recent years, a flattening of the ozone trend can be erroneously interpreted as an effect of changing halogen loading. Figure 3 shows the springtime ozone columns from the SAGE II measurements at 60–70°S and ozone-sonde measurements at the South Pole station (90.0°S) (<http://www.cmdl.noaa.gov>). Throughout all the vertical ranges, the ozone column amounts in September exceed those in October. Column ozone in September, both at 60–70°S and 90°S, seldom undergoes the saturation of ozone loss. At the South Pole station, the column ozone at 14–20 km in October shows, to some degree, saturation since the early 1990’s.

[13] To quantify the magnitude of saturation effects on ozone at 60–70°S, frequency distributions of SAGE ozone values at 14–18 km altitude are shown as black lines for September (lower left) and October (lower right) in Figure 3, for 1985–1990 and 1997–2004. The lower tail of each frequency distribution shows a cutoff above zero except for the October ozone in 1997–2004. The enhanced lower-tail seen in the October ozone for 1997–2004 is probably due to the saturation effects. The effects are estimated from the differences between the lower tails of the ozone and the temperature based frequency distributions which are obtained by adding ozone equivalent temperature residuals to the mean ozone. The latter are obtained from scatter plots, similar to those in Figure 1 (right) for SAGE/HALOE, but for the 14–18 km altitude range. The slopes are approximately 2 DU/K. The frequency distributions for the adjusted temperatures (now in DU) are shown by the blue lines in the lower panels of Figure 3. The shaded area in red in October for 1997–2004 then represents the data points in which the ozone column at 14–18 km should have been less than zero based on the temperatures. These points instead show up as part of the broad peak in ozone around 10 DU.

[14] Now we estimate the effect of saturation on the observed ozone trends. The impact on the mean ozone of



**Figure 3.** Vertical ozone columns from 12 to 24 km with a thickness of 2 km for SAGE II at 60–70°S (upper left) and ozonesonde measurements at the South Pole station (upper right). The black (blue) dots represent ozone columns in September (October). The frequency distribution for the SAGE ozone values at 60–70°S is also shown as black lines for September (lower left) and October (lower right) at 14–18 km in two periods, 1985–1990 and 1997–2004. The frequency distributions for temperature have been converted to ozone units (see text) and are shown in blue in the lower panels. The shaded area in red in October for 1997–2004 represents the data points in which the ozone columns at 14–18 km should have been less than zero based on the relationship with temperature.

using the negative values in place of an equal number of 10 DU values is approximately 5% (1.5 DU). A slightly smaller estimate of the saturation effect is obtained by overlaying the ozone frequency distribution for 1985–

1990 on the 1997–2004 distribution after shifting the 1985–1990 distribution to the left by approximately 15 DU to compensate for the smaller mean ozone in the 1997–2004 period. The ozone change from 1987 to 2000 in the 14–18 km layer contributes approximately 30–50% of the total column loss. Assuming the effect is similar at other altitudes (an overestimation) saturation results in a total column that is above its trend line by  $\sim 5$  DU for post-1997 Octobers. There is no evidence of a saturation effect for September. Therefore, the Sept./Oct. post-1997 anomalies (Figure 2) may be high by  $\sim 2.5$  DU due to saturation. This means saturation could have increased the 2004 CUSUMs by less than  $\sim 13\%$ . Hence, the effect of saturation is small. Saturation may, however, add to the uncertainty in the determination of the start time of ozone recovery [e.g., Newman *et al.*, 2004].

## 6. Conclusions

[15] A strong positive relationship has been shown between temperature anomalies at 100 hPa and column ozone anomalies in the 60–70°S region in Septembers and Octobers. The removal of the effects of these dynamically induced temperature variations on ozone results in an ozone time series in which the variance is mostly due to a change in level of halogen loading. A CUSUM analysis of the ozone time series has shown a statistically significant decline in the rate of ozone loss beginning in 1997. This constitutes the start of ozone recovery in the collar region of the Antarctic ozone hole, according to the International Ozone Commission definition. The SAGE data show that the saturation of ozone loss is not the cause of the change in ozone loss rate. Both ground-based and satellite measurements consistently confirm the flattening of ozone loss rates in the Antarctic collar region. Dynamical variability can still lead to large ozone holes in individual years and long-term temperature decreases predicted by some climate models might offset, to some degree, the future effects of decreasing halogens [WMO, 2003].

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