# Trends evolution of ozone between 1980 and 2000 at midlatitudes over the Southern Hemisphere: Decadal differences in trends

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[1] The variability of atmospheric midlatitudinal ozone between 1980 and 2000 over the Southern Hemisphere is discussed. The distribution of ozone and ozone change during the seasonal cycle is discussed using Total Ozone Mapping Spectrometer Nimbus and Earth Probe data binned at 72 (30° longitude by 5° latitude) bins, between 60° and 30°S. Rather than using a standard trend approach, the annual mean time series for each bin were fitted with a cubic polynomial. The results show that in the zonal mean sense there is a sizable, latitude-dependent slowdown of the ozone loss from the early 1990s onward, but when individual bins are considered, significant longitudinal patterns of ozone change appear, with both positive (enhancement) and negative (depletion) changes in total ozone. Thus regional evolution remains important as an indicator both of chemical depletion evolution and the relation with climate. Such longitudinal behavior is limited in the subtropics and grows toward the subpolar edge of the sampled region. For example, a large decrease was observed over southern South America in the 1980s, but during the 1990s there was only a limited change. The analysis for January, June, and October over the 20-year period shows changes in the evolution along the year, both in time and space. Furthermore, such seasonally dependent changes reach a peak in October, as would be expected. The October pattern of interannual variability could be linked to Southern Annular Mode, though there probably are some other processes driving it.

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## 1. Introduction

[2] The global stratospheric ozone decay, between 1980 and the present, is well established. Several authors (*Gleason et al.* [1993], *Herman and Larko* [1994], *Callis et al.* [1997], *Atkinson* [1997], and *Steinbrecht et al.* [2003], among numerous others) have studied the global total ozone changes. *World Meteorological Organization* (*WMO*) [1995, 1999, 2003] provides an excellent overview. An extensive review on the issue can be found as well by *Staehelin et al.* [2001].

[3] Several studies [*Bojkov et al.*, 1990; *Callis et al.*, 1991; *Stolarski et al.*, 1991, 1992; *Niu et al.*, 1992; *Herman and Larko*, 1994] have shown the large midlatitude decay in the total ozone column and in the lower stratospheric ozone between 1979 and 1980 and the mid-1980s. *Fioletov et al.* [2002], who carried out a comparison of six data sets and extended the analysis, when possible, back to 1964, and


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*Harris et al.* [1997] revised total ozone column seasonal trends at midlatitudes using zonal averages. *Harris et al.* [2001] have further refined the approach used to determine ozone variability and change. Furthermore, *Fioletov and Shepherd* [2003] demonstrated the remnant effect of the Antarctic ozone hole signature in zonal mean total ozone levels over the Southern Hemisphere through the austral summer.

[4] In most studies cited above, it had been customary to use zonal mean values, ignoring all longitudinal effects. Another option has been the use of equivalent potential vorticity (PV) latitude. It is argued that possible longitudinal changes are thought to be mainly related to dynamic processes, while the zonal mean analysis, in real or equivalent PV latitude, can, in principle, better capture the impacts of modified stratospheric chemistry. However, certain features resulting from changes in tropopause height, tropospheric processes, or coupled chemistry-dynamics may not necessarily be distinctly captured in equivalent PV latitude. The changes in response to dynamic variability could occur in response to a number of combined processes, a combination that may not necessarily be linear, could include feedbacks with chemical processes, and could be seasonally dependent as well. Such changes may not be zero when averaged over longitude in either geographic or PV coordinates.

[5] A further option has been to work straightforwardly with the usual geographic coordinates, while trying to determine features which could be linked to dynamic processes. *Bojkov et al.* [1990], *Niu et al.* [1992], *Hood and Zaff* [1995], *Hood et al.* [1997], *WMO* [1999], and *Peters and Entzian* [1999] have discussed the longitudinal dependencies of ozone change. Indeed, *Niu et al.* [1992] estimated the seasonal trends in column $O_3$ as a function of latitude and longitude using Total Ozone Mapping Spectrometer (TOMS) data in latitudes between 70°S and 70°N, for the period 1978–1990 with a 10° latitude by 10° longitude grid. They determined that ozone trends are highly seasonal and dependent on geographic location.

[6] The research referenced above, except *Niu et al.* [1992], *Fioletov et al.* [2002], and *Fioletov and Shepherd* [2003], has primarily focused upon northern midlatitude longitudinal variability and the mechanisms that drive such changes. The coupling between dynamic variability and change over the Northern Hemisphere has appeared in all cases as a significant cause of longitudinal ozone change, e.g., *Peters and Entzian* [1999].

[7] It has thus become clear that ozone change is not limited to chemical processes but is also dependent on dynamical changes [*WMO*, 1999, 2003, and references therein] which can even modify mean latitudinal distribution [*European Commission*, 2003]. Current evidence suggests that dynamically induced changes may be as significant as chemically induced ones. *Appenzeller et al.* [2000] have shown that the local null trend observed at the Rekjavik (Iceland) Dobson station was due to the behavior of the North Atlantic Oscillation during recent decades. Once its effect was taken into account in the trend calculation, the resulting trend estimate agreed well with the expected chemically induced depletion.

[8] It is necessary to describe, understand, and begin to attribute the spatial and temporal variability, particularly over the Southern Hemisphere, where no such studies have been attempted. In the Southern Hemisphere midlatitudes, because of the dominant ocean expanses, there are few ground stations located at the same latitude to perform a longitudinal analysis and even fewer have long enough data records. Therefore TOMS data become crucial to study the geographical, seasonal, and latitudinal dependence of the total ozone column. The 10- and 20-year TOMS linear trends reported by *Malanca et al.* [2003] together with total ozone column (TOC) evolution over different regions of the Southern Hemisphere point to a possible change in the TOC trends near the end of the 1980s and the beginning of the 1990s, with an important longitudinal and latitudinal dependence. The regions most affected by the decrease in total ozone appear to be southern South America and the South Atlantic, in agreement with *Niu et al.* [1992]. Nevertheless, certain significant changes are observed in the longer 20-year time series. Both *Malanca et al.* [2003] and *Fioletov et al.* [2002] show that the most negative linear trends at middle and high southern latitudes have shifted eastward from southern South America into the South Atlantic and the western Indian Ocean. *Huth and Canziani* [2003], using Principal Component Analysis, have shown the eastward shift in position of the Antarctic polar vortex over the years for the period 1979–1997.

[9] As *Niu et al.* [1992] have done, we have investigated the latitudinal and longitudinal variation of the total ozone column concentrating on southern midlatitudes between 30° and 60°S. We have extended by 10 years the study performed by Niu (i.e., 1980 to 2000 year), for this region, and have completed the preliminary analysis presented by *Malanca et al.* [2003]. While *Fioletov et al.* [2002] primarily consider seasonal averages and significant latitudinal averaging, the approach in this paper is to use a finer time and space sampling. Our analysis includes a latitudinal and longitudinal study of the annual and monthly variations of the total ozone fields during the last 2 decades at midlatitudes. Section 2 includes a brief description of our data analysis approach. Section 3 presents the results of the analysis of the total ozone latitude-longitude distribution and its evolution, while section 4 discusses the results. Future work will include an analysis of possible relationships of decadal ozone change with physical variables in the atmosphere, e.g., temperature and PV, among others.

## 2. Data Sources and Analysis Method

[10] The TOMS instruments on Nimbus 7, METEOR, and Earth Probe satellites have successively provided a daily record containing high-resolution gridded total ozone column (1° latitude by 1.25° longitude) for most of the period between 1978 and the present day. The uncertainty associated with each daily grid value is not more than ±5 Dobson units (DU) [*Weinberg et al.*, 1996]. Version 7 [*McPeters et al.*, 1996] data were used for the present analysis. Due to the failure of the Nimbus 7 TOMS in 1993, followed in quick succession by the failure of the METEOR TOMS in 1994, there is a gap in the time series until the new operation of Earth Probe TOMS in 1996. Note that the METEOR TOMS data set is not included in this analysis due to the difficulties in using it as a result of the variable local time sampling.

[11] We sorted out the data between 60° and 30°S and 180°W and 180°E in monthly or annual "bins" of daily

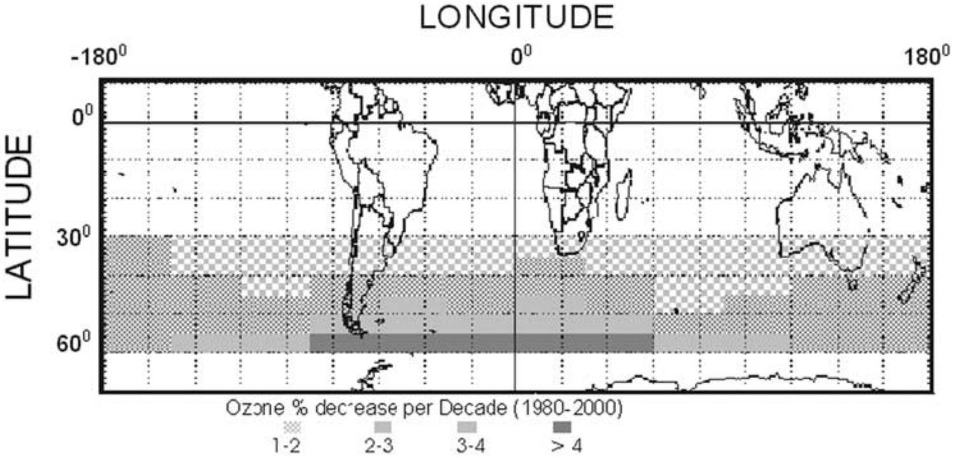


**Figure 1.** TOC 20-year linear trend evolution between 30° and 60°S. A distinct dependence on both longitude and latitude can be observed for the trends.

high resolution gridded TOMS total ozone, with final latitudinal and longitudinal sizes of 5° and 30°, respectively. We imposed the mean value to the center point of each bin (e.g., the data contained between 60° and 55°S and 180° and 150°W are averaged and represented by the center point 57.5°S, 165°W). We thus obtained 72 bins, and the variation of the ozone column was investigated in each one. Monthly mean values were calculated for each bin. An analysis of the 72 bins was carried out in order to establish the latitudinal and longitudinal dependences of the total ozone column between 1980 and 2000.

[12] The determination of the 20-year evolution in the annual mean TOC was carried out for each bin using a cubic polynomial fit rather than a simple linear fit, as applied by *Malanca et al.* [2003]. Such trend analysis procedure is suggested, for example, by *Essenwanger* [1976]. The zonal mean quasi-biennial oscillation (QBO) and solar cycle signatures were filtered out first, by estimating their contribution to the time series using a least squares adjustment of the QBO and sunspot number indexes, respectively. The cubic fit was chosen because in all cases the calculated chi-square coefficient $\chi^2$ yielded its lowest value for the fit. Occasionally, the second-order polynomial fit was almost as good. The $\chi^2$ coefficient increased rapidly for higher-order polynomials. In all cases the values for the $\chi^2$ coefficient showed that the fit was better than the 95% confidence level. It should be noted that the $\chi^2$ coefficients calculated for the cubic polynomial fit were overall better than for a linear fit or even an exponential one. This approach agrees with *Harris et al.* [2001] in their study of selected Dobson station data.

[13] In the case of the monthly mean samples chosen, specifically January, June, and October, for further study of seasonal behavior, no fit was made due to the fact that for certain longitudes and certain seasons, considerable interannual variability was observed, even after the filtering of the QBO and the solar cycle signature. Such features did not warrant the use of a simple linear fit. In many cases the use of a polynomial fit resulted in a reasonable fit quality for higher-order polynomials only, particularly in June, which had the most prominent interannual variability. Furthermore, the order of the polynomial fit could be longitude-dependent as well, further complicating matters. Since no pattern was detected in the occurrence of the order of these higher-order polynomials, their use was not considered at this stage, and no fits are thus presented here for these. Furthermore, from a methodological point of view, given the relative short length of the samples, it is not appropriate to consider higher-order polynomials. The interannual variability was obtained using the differencing of the mean values of the same months in successive years, which act as a high pass filter. Note that the methodology philosophy is to keep the methods used as simple as possible to pick up the robust signals of ozone change and variability so as to avoid, at this stage, difficulties in the interpretation of the results which can happen when more complex statistical approaches are used.

## 3. Results

[14] Figure 1 shows the percent decrease for each bin over the full 20-year period considering a standard linear approach. The analysis reveals the strongest decay in TOC between 60°–55°S and 120°W–90°E. These values show that the longitudinal dependence of the annual mean linear trend is larger at higher latitudes and smoothes out at lower latitudes. In the higher latitudes the largest trends are observed in the longitude band corresponding to South America and the South Atlantic and are most probably due to the ozone hole displacements over the region. Note that trend values within ±2% are nonsignificant. Compared to *Niu et al.* [1992] 1979–1990 trends, present results suggest a weaker overall trend over 1980–2000. *Fioletov et al.* [2002], globally, *Malanca et al.* [2003], for southern midlatitudes, and *Staehelin et al.* [2001], for northern midlatitudes, observed a slowdown in the ozone decrease, when the TOC linear trends in the 1980s and 1990s are compared. Thus the weaker trend of the 20-year sample, shown in Figure 1, when compared with the results by *Niu et al.* [1992], could result from changes in the decadal evolution. Such a possible change, and its decadal evolution, needs to be considered in further detail. Due to the possible remnant effects of the winter and spring ozone depletions during the summer months, which have been

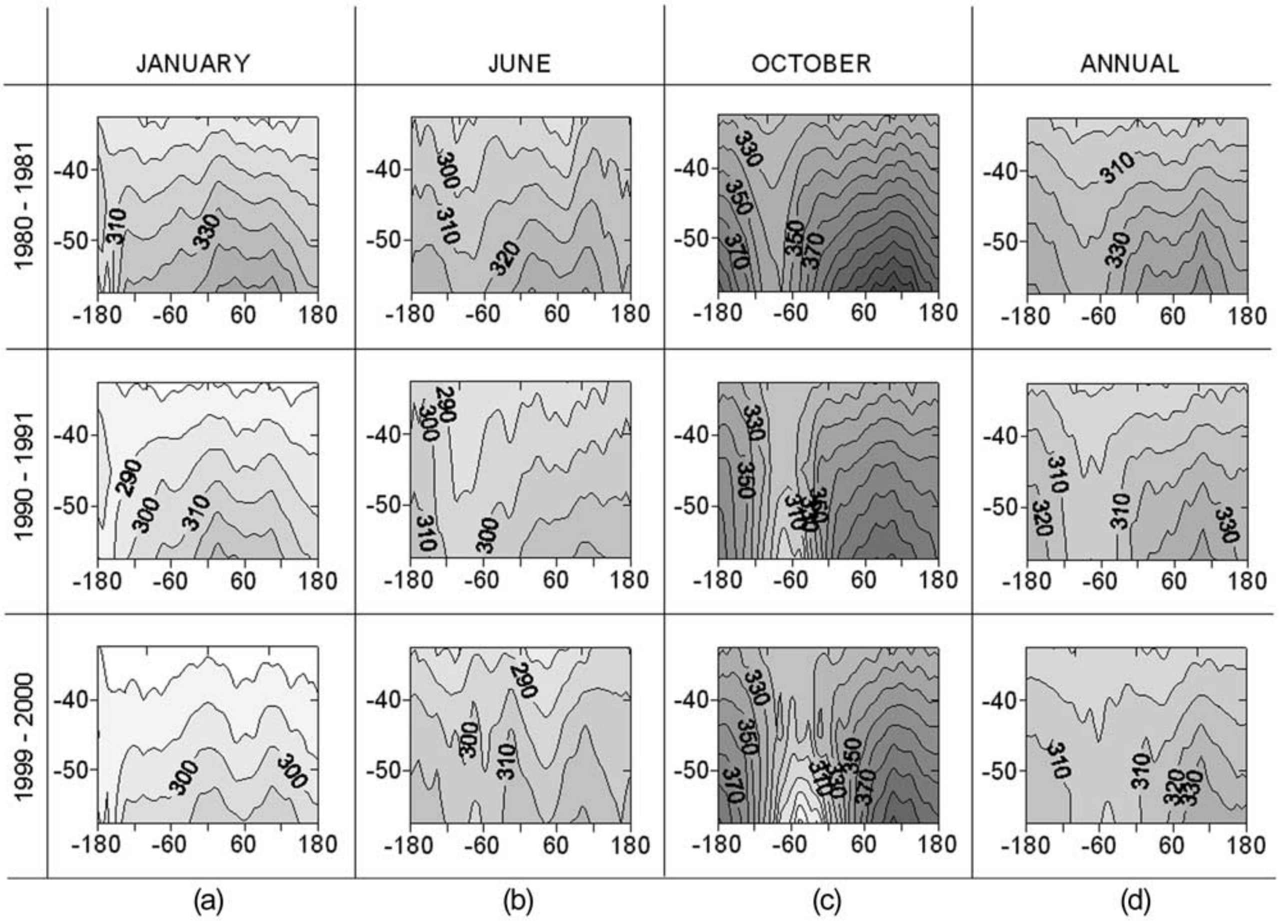


**Figure 2.** Two-year average TOC distribution over Southern Hemisphere midlatitudes, for the months of (a) January (summer), (b) June (winter), and (c) October (spring), and for the (d) annual mean, in Dobson units. The 2-year averages correspond to 1980–1981 (pre-ozone hole period), 1990–1991, and 1999–2000.

observed in the zonal mean sense, this study considers the annual cycle evolution from June to January, rather than the more common chronological approach.

### 3.1. Preliminary Decadal Change Analysis

[15] Before proceeding further with the trend analysis, so as to have a first insight into the spatial and temporal evolution of TOC over the Southern Hemisphere (SH), it is convenient to observe a sequence of maps of TOC distribution between 1980 and 2000. We follow the approach used by *Hood et al.* [1999] for the Northern Hemisphere. Such maps can point to where the significant changes occurred and how the midterm/decadal variability may have evolved. Figure 2 shows the June (winter), October (spring), and January (summer) together with the annual mean midlatitude TOC fields, averaged for three different 2-year periods, i.e., 1980–1981, 1990–1991, and 1999–2000. Such an averaging procedure follows the scheme suggested by *Hood et al.* [1999] to eliminate/minimize possible effects of the QBO in the analysis of long-term variability. Furthermore, the 10-year sampling time step also significantly minimizes solar cycle variability in TOC. Indeed, the first two periods correspond to solar cycle maxima, while the last one is sampled just before the corresponding maximum.

[16] The June plot for 1980–1981 (Figure 2b) shows a large ozone trough, deeper along the western edge of the Andes mountain range, and a weaker one near 60°E suggesting a combined wave 1 and 2 ozone distribution over the region. During June 1990–1991, where a general decline in column ozone can be observed, the eastern hemisphere trough disappeared, and the one over the eastern Pacific became stronger. This change suggests a possible relative enhancement of a wave 1 structure and a weakening of a wave 2 component. Ten years later the eastern trough appears again, significantly stronger and somewhat displaced to the west, while the one to the west of the Andes appears to have broadened and weakened at the same time. Such a behavior suggests the return of a significant wave 2 contribution to the ozone field during the early winter period.

[17] The results for October (Figure 2c) show a dominance of a wave 1 structure throughout the period under study, as would be expected from the Southern Hemisphere stratosphere climatology, and a fairly sustained decrease, especially at high latitudes. Compared with June, there now is a significant trough over the Andes and on their eastern side. This trough becomes deeper, particularly in the higher latitudes, and broader, with an eastward displacement. Originally centered near 75°W through the 1980s, it appears

located, at the end of the 1990s, around 50°–45°W. This agrees with the results in the PV analysis by *Huth and Canziani* [2003].

[18] During January (Figure 2a) the ozone has a trough due east of the dateline, while the larger TOC values are located east of the Greenwich meridian and peak over the southern Indian Ocean. The ozone distribution patterns do not show significant changes in longitude over time, but overall, there is a decrease in ozone levels, suggesting possible ozone depletion and/or mixing of ozone poor air from the polar vortex breakdown. In other words the strong wave 1 pattern remains almost stationary over the 20-year period considered. Note that there appears to be a growing ozone trough around 60°E and a weaker decrease near 60°W. Such a behavior suggests that at least in summer there has not been a major change in the circulation that could have influenced TOC distribution over the hemisphere, except for the apparent growth of higher-order wave structures, most probably dominated by the wave 2 pattern.

[19] The evolution of the October TOC trough is reflected in the annual mean through the poleward widening of the trough's eastern limit. Indeed the TOC values for spring and early summer are the largest, and hence their spatial distribution contributes significantly to the annual mean. The contribution of the long-term varying wave 2 appears to be sufficiently significant to appear, too, even if somewhat weaker in the annual mean.

[20] Figure 3 shows the decadal differences for each of the sequences in Figure 2, giving a coarse estimate of change in TOC, and where the main changes are found over the hemisphere. In general, the largest differences in mean values can be observed between 1980–1981 and 1990–1991, where considerable decreases (~20 DU) can be seen almost at all latitudes and longitudes. Between 1990–1991 and 1999–2000, while there are still decreases, these are not as large (5–10 DU). There are even regions that have not experienced significant changes, and some even present limited enhancement in TOC.

[21] During the month of June (Figure 3b) the regions that have experienced the largest changes during the first decade are southern South America and Australia and adjacent oceans. However, during the next decade both regions seem to have experienced little change or even an enhancement, for example, just off the western edge of the Andes along Chile and Argentina, while most of the decrease is now located over South Africa and the oceans due south. Hence the regions with weak depletion in early winter in the 1980s became the regions with the largest changes in the 1990s and vice versa.

[22] During October (Figure 3c) most of the TOC decrease was located during the 1980s along the South American axis, extending well into the subtropics. This could be due to circulation and maybe some impacts of the polar vortex/ozone hole system, which tends to favor off-pole displacements toward Argentina, Chile, and the South Atlantic [e.g., *Huth and Canziani*, 2003] pushed by the dominant standing wave 1 seasonal feature. During the next decade the rate of change has continued unabated in the vicinity of the polar vortex, with an enhancement in the TOC levels over the higher latitudes of the South Pacific. The latter may be due to enhanced meridional transport and/or a strengthening and shift of the wave 1 feature as a result of interannual variability. Note that the latitudinal extent of the region with decreasing TOC appears somewhat reduced and the region of major change has moved eastward, well into the South Atlantic, in agreement with the PV evolution [*Huth and Canziani*, 2003]. Nevertheless, there remains a sizable depletion extending along the TOC trough and spilling over into the western Indian Ocean, which extends well into the lower latitudes of the sample. It may be argued that some of the large changes observed during the 1990s could be attributed to the exceptional 2000 ozone hole event. Whether this is the case or not will be discussed in section 3.2.

[23] Even if the overall features of TOC distribution did not change significantly in January, TOC shows significant changes (Figure 3a) during the 1980s, throughout the hemisphere, with particularly large changes along the coast of Argentina, the central South Pacific, and the southern Indian Ocean. During the 1990s the major changes were smaller, i.e., less than half the previous decade, but still significant, between 5 and 2%. Fairly sizable decreases still occur east of the Greenwich meridian, south of 40°S and over Patagonia in Argentina and Chile. Note that the decadal changes also show the possible growth of the wave 2 feature, particularly during the second decade.

[24] Finally, the mean annual change (Figure 3d) suggests that during the first decade the most significant changes occurred at high latitudes over the tip of South America and the South Atlantic. Considering the inhabited continental regions, the most affected during this period are the Chilean and Argentine territories, which had a somewhat reduced TOC to start with. During the next decade the situation appears to stabilize over the hemisphere's inhabited regions, and the largest changes are observed over the southern oceans, in particular, the South Atlantic. The spring depletion has, as mentioned above, a significant impact on the mean annual decadal changes.

[25] It is interesting to note that there were some broad similarities between the ozone change geographical distribution in October and January, over the second decade, suggesting an apparent remnant impact of the spring ozone depletion in the region. As previously mentioned, *Fioletov and Shepherd* [2003] discussed this for the zonal mean, and this spatial pattern would imply a similar effect in space as well for TOC changes. Furthermore, these similarities are coincident with the larger and longer- lasting ozone holes that took place during the 1990s. The spatial structure in the differences appears better defined in October than in January, and would, as a first guess, be attributed to dilution after the ozone hole breaks up in late spring. It should be noted that the region of largest change in January, as the main feature of this apparent remanence, is found to the east of the October one, during the second decade, and could be attributed to the continued eastward drift of the dynamic features as they finally weaken and disappear in the austral summer. Yet it is troubling that during the first decade, when large overall ozone deletion took place, such a relationship is not clear. During and after the polar vortex breakup the ozone-poor air rapidly mixes with midlatitude air and, on the one hand, the identity of the vortex air is rapidly lost and, furthermore, there is no evidence in the distribution of ozone or PV in the early

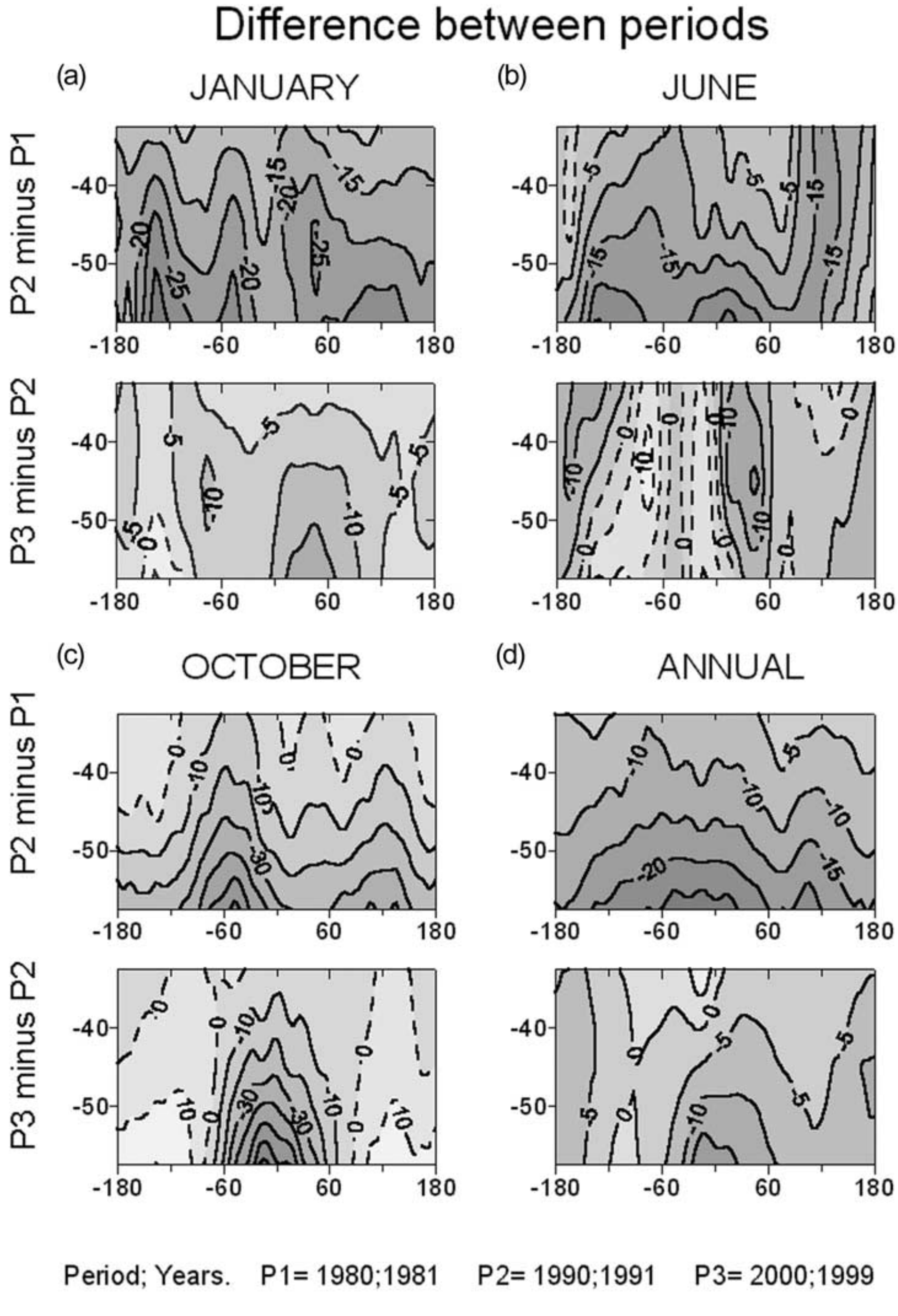


**Figure 3.** Decadal differences of the 2-year average TOC distribution over Southern Hemisphere midlatitudes, for the months of (a) January (summer), (b) June (winter), and (c) October (spring), and for the (d) annual mean, in Dobson units.

summer of polar vortex characteristics [e.g., *Huth and Canziani*, 2003]. In consequence, this feature is not really a spatial summer remnant of the spring depletion. Its origin can be found in the persistence of a low-ozone region in the daily TOC fields at the end of December 1998 and the first half of January 1999. This feature, which has become apparent in the late 1990s, is normally located over or in the vicinity of the South Pole. Due to the presence of a very strong horseshoe ozone structure, that results most probably from the combination of slowly rotating waves 1, 2, and 3, this low-ozone region is displaced off center into the eastern hemisphere and is picked up in the averaging process.

### 3.2. Polynomial Fit of Annual Evolution and Evolving Trends During June, October, and January

[26] Figure 4 shows the evolution of the annual mean TOC after the QBO and zonal mean solar cycle effects were subtracted, over selected bins at 57.5° and 32.5°S, respectively. These bins show the three typical kinds of evolution

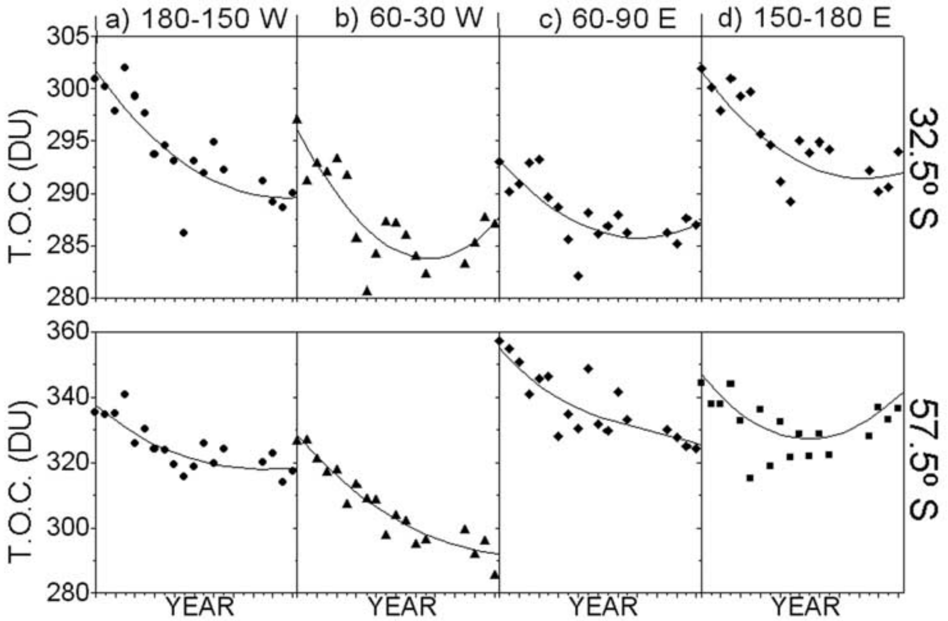


**Figure 4.** Evolution of annual TOC 1980–2000 for a selection of "bins" at (top) 32.5°S and (bottom) 57.5°S at (a) 180°–150°W, (b) 60°–30°W, (c) 60°–90°E, and (d) 150°–180°E. Symbols (circles, triangles, diamonds, and squares) show the mean of the observations for each bin, and the curves show the result of the cubic polynomial fit of those points.

detected in the region. These are located for both latitudes at 180°–150°W (Figure 4a), 60°–30°W (Figure 4b), 60°–90°E (Figure 4c), and 150°–180°E (Figure 4d). The plots also show the cubic function fit (curves). Three main types of evolution can be detected, but in all cases the years around 1990 seem to be turning points in the evolution of annual TOC. The kinds of trend detected are (1) a quasi-stationary state after ~1990, (2) a regional TOC increase around 1990 or in subsequent years, and (3) a weaker but sustained declining trend after 1990. The hemispheric potential vorticity Principal Component Analysis carried out by *Huth and Canziani* [2003] and the microwave sounding unit (MSU) lower stratospheric temperature study by *Compagnucci et al.* [2001] and *Salles et al.* [2001] have also detected a change in the behavior of those variables around the same period. It should be noted also that over some bins this latter kind of evolution suggests a possible enhancement of the TOC decrease in more recent years at high latitudes. As mentioned above, this change could in part be due to shift in position of the polar vortex/ozone hole system.

[27] It is interesting to note that for a given longitude band, different evolutions can be detected at different latitudes. Thus for pairs of plots, e.g., Figure 4b or 4c, opposite temporal behaviors can be observed at different latitudes but similar longitudes. These differences between latitude regions point to the complexity in assessing the regional evolutions of the ozone layer.

[28] Figure 5 shows the longitude-time variations for the smoothed annual trend estimate, with the derivative of the cubic polynomial fit, at 32.5°S (Figure 5a) and 57.5°S (Figure 5b). The annual trend, given as a percentage of the previous year's TOC, was calculated by simple differentiation of the cubic fit formula. At 32.5°S the trends show little longitudinal variability, except over the central Pacific, where peak values are observed, i.e., greater than −0.5% per year or 5% per decade. The trends quickly diminish and become nonsignificant between 1988 and 1990, at all longitudes. During recent years an incipient TOC enhancement can be observed over the western Pacific and South America with trends slightly above 0.2% per year (2% per decade). On the other hand, at higher latitudes (Figure 5b) a far more prominent longitudinal dependence is evident. Again the larger trends are observed in the vicinity of southern South America and the South Atlantic. In the earlier years of the sample, over Patagonia and adjacent oceans, trend values >1.4% per year (14% per decade), can be observed. These decrease around 1990 to ~0.4% per year. After 1990 a slow increase in the negative trend can be observed, again reaching values near 1.2% per year (12% per decade), with a shift in largest trends practically over the Greenwich meridian. At other longitudes, in particular, east of Greenwich, the trends are much weaker, near −0.4/−0.6% per year (−4/−6% per decade) during the 1980s, and there appear to be regional enhancements during the last few years, with a positive trend near 0.4% per year (4% per decade). The largest change in trend was detected near the dateline, over the central South Pacific, with a change in trend from approx. −1.2% per year (−12% per decade) to 0.4% per year (4% per decade) over the 20 years of the sample.

### 3.3. A More Detailed Look at the TOC Evolution During June, October, and January

[29] For reasons explained in section 2, the polynomial fits for the specific months were not calculated. Nevertheless, inspection of the evolution of TOC, during June, October, and January, at selected latitudes, reveals that the main features observed in the preliminary decadal analysis are good representations of the TOC evolution during the

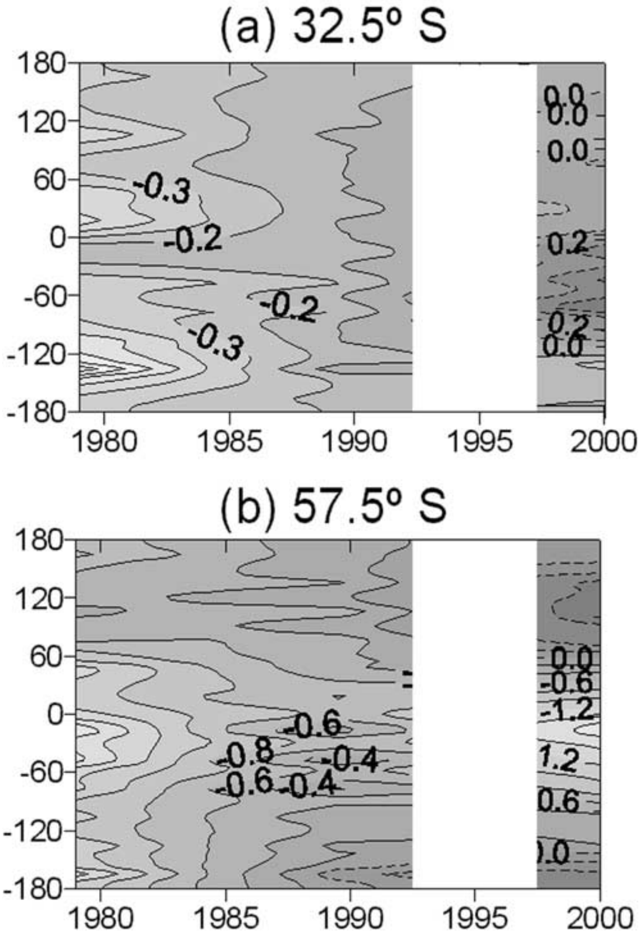


**Figure 5.** Longitude-time variations for the smoothed annual trend estimate, derived from the cubic polynomial fit, at (a) 32.5°S and (b) 57.5°S. Annual trend is given as a percentage change from the previous year's TOC.

1980s and 1990s, i.e., that the preliminary analysis is consistent.

[30] During October and January, despite occasionally large interannual variability, the main structures evolve smoothly over time, as can be seen, for example, in Figure 6a. On the other hand, it is possible to observe in June (Figure 6b) the variability over the Pacific Ocean, which can help explain the appearance of a trough in the region at the beginning and the end of the sample but not in the June 1990–1991 average (Figure 2). Except in June, where the sizable variability in particular renders trend estimates complex, the inference that there was a trend change around 1990 is verified by the full sample TOC evolution, in agreement with the annual mean trend evolution shown in Figure 5.

[31] The longitudinal dependence of ozone change can be observed as well in the relative annual percentage changes between successive months of June, October, and January. These interannual percentage changes were calculated after filtering of the solar cycle effect and the QBO/interannual variability. Such a calculation, by comparing the evolution from one year to the next, acts as a high pass filter. It reveals primarily the structures of interannual evolution rather than the longer-term trends and can help understand why the cubic polynomial approach is no longer a useful approach using month mean samples.

[32] Figure 7 shows the TOC percent change from one year to the next for the months of June, October, and January at 37.5°, 47.5°, and 57.5°S. Note that individual bins may occasionally have somewhat larger extreme values than those shown in the figures, as a result of the inherent smoothing of the plotting procedures. Nevertheless, the primary aim of the figures is to show the spatial structure of TOC midterm variability and its evolution during the different seasons.

[33] Inspection of the first column in Figure 7 shows two different latitude regimes for ozone variability in June. At 37.5°S the June interannual changes evolve in time between −2.5 and 2.5%/yr, and a quasi-zonal behavior dominates the variability. Peaks in variability appear over the Andes and the eastern South Pacific in the 1980s and early 1990s, but toward the end of the sample the pattern loses its quasi-zonal feature. At 47.5° and 57.5°S, there appears to be a sizable midterm variability with a range between 2.5 to −3%/yr and 2.5 to −4%/yr, respectively. The ozone change evolution clearly shows the apparently quasi-decadal nature of the variability, with maximum positive trend maxima

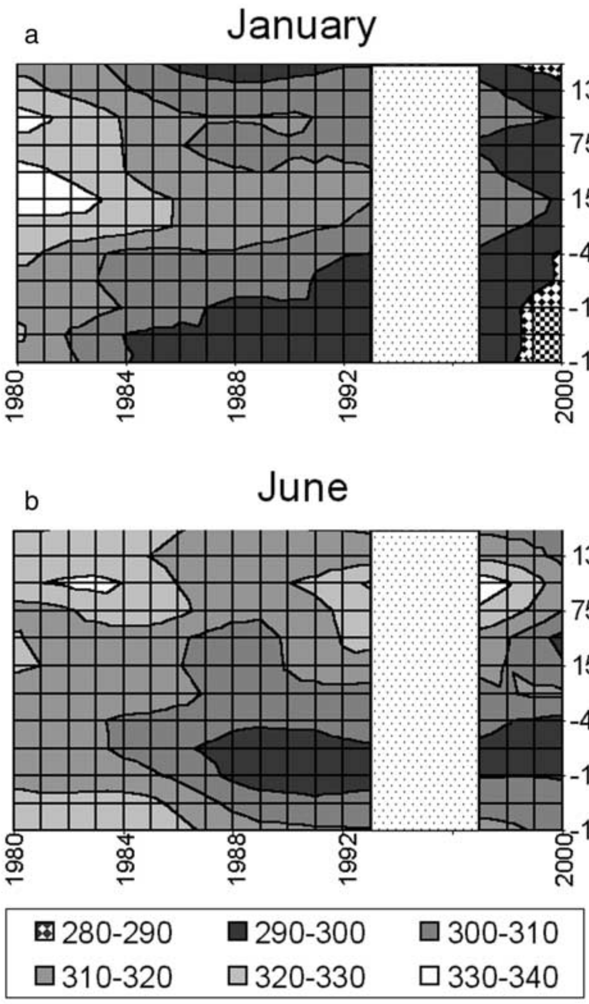


**Figure 6.** TOC evolution between 1980 and 2000 during (a) January and (b) June at 47.5°S.

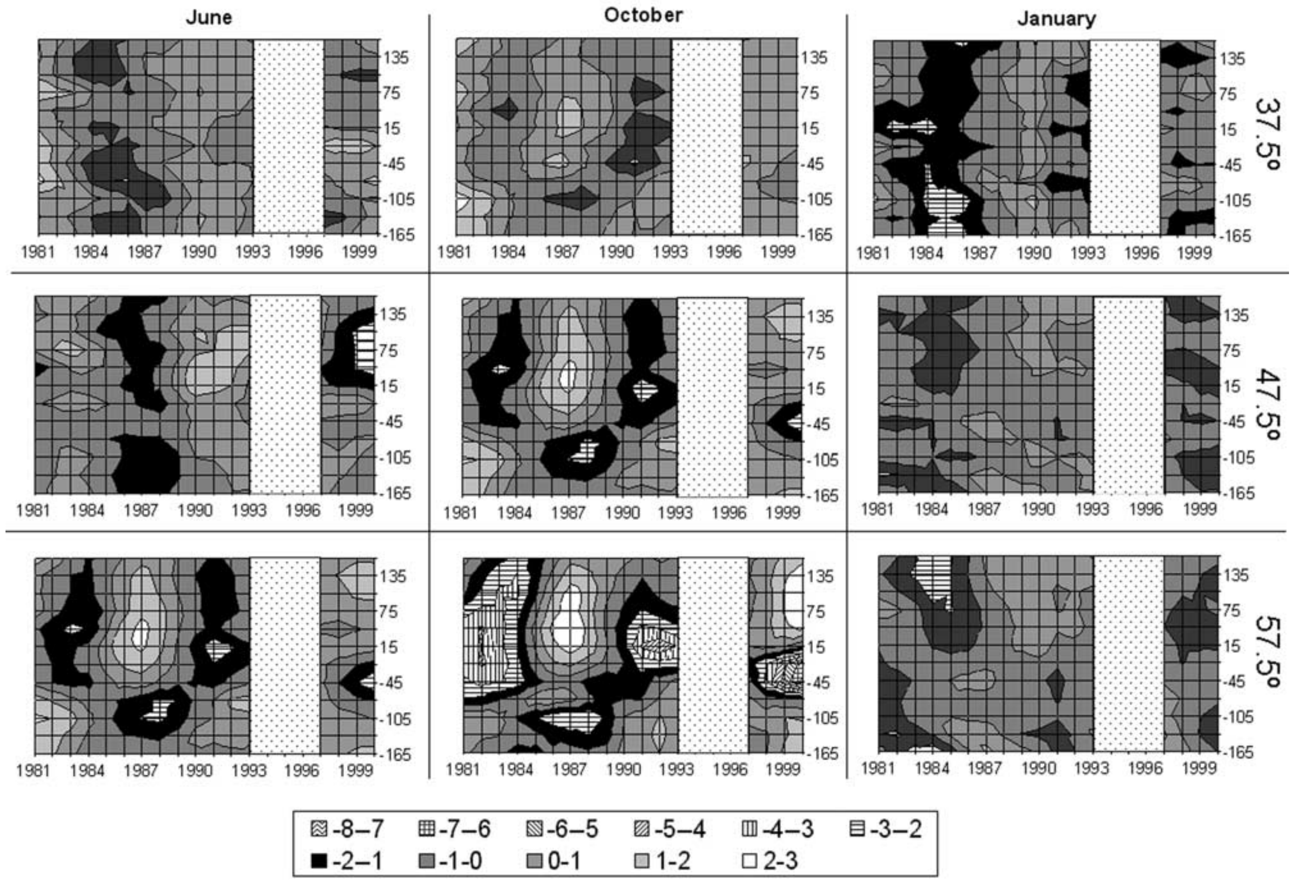


**Figure 7.** Interannual TOC change during June, October, and January at (top) 37.5°S, (middle) 47.5°S, and (bottom) 57.5°S, given as the percentage change from one year to the next, for each month.

around 1982–1983 and 1989–1990 in this region. Note that the term apparent quasi-decadal is used, since, due to the shortness of the sample, it is not possible to determine the repeatability of the phenomenon or exactly define the period of the apparent oscillation. At the higher latitudes, there appears to be a moderate westward phase progression in the ozone change. At both higher latitudes the largest negative trend occurs to the west of the dateline at the end of the samples.

[34] The TOC main trough region over the Atlantic and South America (compare to Figure 2) shows much weaker oscillations, particularly at 57.5°S. Such difference in variability can result in an interannual enhancement or weakening of the longitudinal differences between the TOC ridges and the troughs described above. Thus the evolution of the TOC interannual change in ozone, shown in Figure 7, also distinctly picks up these different regimes between South America/western South Atlantic and the rest of the hemisphere. Note that the quasi-zonal mean changes at 37.5°S during the 1980s and the early 1990s are not quite in phase with ozone change over the South Pacific and Indian Ocean at the more southern latitudes. During the late 1990s, despite the more complex structure, particularly at lower latitudes, these oscillations appear to strongly evolve in phase.

[35] During October (Figure 7, second column), ozone change variability at 37.5°S is fairly zonal to begin with. During middle and late 1980s this quasi-zonal feature is lost due to an antiphase behavior evolution over the eastern South Pacific, to be recovered, though not completely, in the late 1990s. The structure of the variability suggests an eastward phase migration. The variability suggests a periodicity of 6–7 years over the Indian Ocean. At 47.5°S the variability is almost in phase with the evolution at 37.5°S. The periodicity during the 1980s and early 1990s is similar. Defining the axis or node of the longitudinal pattern as the region with weakest perturbations closest to the Greenwich meridian, it appears to drift weakly toward the east over the period of the sample, beginning around 45°W in the early 1980s, and ending near 15°E in 2000. The range of the oscillations is ±2.5%/yr, approximately. At 57.5°S the range of the oscillations grows to +3 to −4%/yr, though extreme values can be as large as −6–7%/yr. The longitudinal pattern is somewhat shifted to the west to begin with since the axis is near 75°–70°W and the axis or node of the longitudinal structure migrates east over time, ending near 15°E, as at 47.5°S. The ozone change anomalies are much broader and deeper over the eastern hemisphere. At the end of the samples the most negative changes are observed over the central South Atlantic.

[36] During January (Figure 7, third column) at 37.5°S the range of interannual perturbations is limited, of the order of ±1–1.5%/yr, and there are no distinct oscillatory features. During the first 10 years, ozone changes are essentially zonal with a slight enhancement around 1990, with weak positive trends (approx. 0.5%/yr). The largest negative

changes occur in the mid-1980s. After that, there is a weak negative trend (close to −0.5%/yr on average) without any spatial pattern. The same applies at 47.5°S, though positive trends are somewhat more frequent at the end of the 1980s and early 1990s, with a quasi-zonal structure. The end of the sample yields negative trends during the last few years. Nevertheless, at 57.5°S the longitudinal varying structure observed during October prevails during the 1980s and early 1990s. There appears to be a western phase migration of the anomalies this time, more prominent to the east of the dateline and during the 1980s. The periodicity of positive and negative trends over the eastern Pacific appears to be of the order of 8 years. Over southern South America and the adjacent Atlantic Ocean, ozone change remains mostly weakly negative over the period. In the last years of the sample, ozone change is dominated by negative change at all longitudes. These results are consistent with the decadal sampling of the preliminary analysis, which yielded limited TOC structural change during the period sampled.

## 4. Discussion

[37] The above results fully confirm the strong longitudinal dependence in the trends and interannual variability of the monthly and annual mean TOC values at mid-southern latitudes, after the filtering of the solar cycle and possible QBO contributions. In agreement with *Niu et al.* [1992] we observe significant seasonal changes in ozone trends as a function of both longitude and latitude. While the change observed over the 1980s is consistent with *Niu et al.* [1992], the annual ozone change rates and the interannual variability are shown to slowly vary over the 20-year sample.

[38] The evolution and changes in decadal trend, reported by *Malanca et al.* [2003], i.e., a reduction in the rate of linear ozone decrease during the 1990s as compared with the 1980s, can be visualized both in Figure 3 and in the data points of Figure 4. The inherent smoothing of the cubic fit, shown in Figures 4 and 5, nevertheless confirms the important change in evolution between the 1980s and the 1990s in the annual mean TOC, even despite the gap due to the lack of TOMS sensors in the mid-1990s. The largest differences in decadal change are observed in June and January, while, despite the variations in the amount of change and even a partial enhancement in some longitude bands at higher latitudes, the October mean TOC overall shows a more limited evolution in the pattern of regional changes.

[39] The annual TOC evolution in the lower latitudes of the sample is fairly homogenous at all longitudes, except at the beginning and the end of the sample. Such a behavior could in part be due to end effects of the sample, since no such large longitudinal differences in ozone change are observed for the first decade of the preliminary analysis. Nevertheless the regions with positive and negative ozone change rates, in the vicinity of southern South America, at the end of the sample appear to be fairly consistent with the decadal changes shown in Figure 3d. At higher latitudes, because the magnitude of the changes, both positive and negative, is much larger, the derived trend is more consistent with the decadal change estimates shown in Figure 3d.

[40] It must be noted that these observed changes are broadly consistent with changes in the behavior of MSU channel 4 temperature retrievals and PV dynamic climatologies over the region [*Compagnucci et al.*, 2001; *Salles et al.*, 2001; *Huth and Canziani*, 2003]. The fact that the longitudinal ozone structure is evolving in ways similar to the changes in temperature and potential vorticity, despite the chemical depletion processes, shows the importance of climate dynamics in determining the full state of the ozone layer at any given period. As pointed out in *WMO* [2003], the magnitude of this contribution needs to be assessed on regional and hemispheric-scale processes, as well as the mechanisms that drive it.

[41] The negative trend enhancement in the later years of the sample is coincident with the longer-lasting larger polar vortex/ozone hole events in the late 1990s and 2000, which appears to affect the central South Atlantic. This feature is observed both in the decadal sampling and in the interannual variability analysis. At the higher latitudes of the sample it can be argued that the fairly frequent passage of the polar vortex/ozone hole system over a preferential longitude region can influence the mean TOC values in the region. Nevertheless, this particular feature extends into the lower midlatitudes during October, despite the well-established highly reduced permeability of the polar vortex during the austral spring. Thus the larger trend observed in the Atlantic trough region cannot be solely attributed to the effects of the ozone hole. Furthermore, this trough appears to act as a node in the longitudinal structure of the interannual ozone change variability. Inspection of Figures 2 and 3 shows that the largest changes in June and January can occur both in the vicinity of or over the TOC trough features as well as in other regions of the sample, but the decadal ozone change features with largest longitudinal structures tend to occur in these steeper gradient regions. In the case of June the disappearance and later reappearance of the secondary trough over the eastern hemisphere is intimately linked to the longitudinal structure of the interannual variability, with quasi-decadal periodicity. Observed ozone change in June (Figure 3b) also suggests that the long-term trend in winter is comparatively weak and that any trend estimate during this time of the year will be strongly dependent on the beginning and end of the sample chosen for the analysis, due to the influence of such interannual variability. The June evolution agrees with the *Fioletov and Shepherd* [2003] conclusion that the Southern Hemisphere winter "erases" the memory of the previous ozone hole season, showing here as well the spatial nature of this atmospheric "memory loss."

[42] The apparently quasi-decadal oscillations observed in June, October, and January, albeit at higher latitudes only, are an interesting feature. On the one hand, they cannot be due to the solar cycle effects, since they are shown to be geographically and seasonally dependent: The features are enhanced at the higher latitudes, with strong longitudinal signatures. Such behavior precludes possible artifacts resulting from satellite orbital effects. It should be noted that similar quasi-decadal features were also observed in the evolution of reanalysis PV fields, carried out using a different statistical approach with an independent data set [*Huth and Canziani*, 2003]. Their

PV anomaly field analysis yields spatial features with some similar characteristics, the apparent periodicities for the most frequent anomaly modes ranging between 7 and 12 years, as is the case with the TOC interannual change. Such a relationship points to the dynamic nature of the interannual ozone change.

[43] It is interesting to note that in October, the western South Pacific, Indian, and Atlantic Ocean regions appear to have more prominent oscillations, and to the west of South America such features tend to be either weak or nonexistent at lower latitudes. At higher latitudes the longitudinal structure develops into a reduced negative (positive) change region and a large positive (negative) region extending at least from the eastern South Atlantic into the western South Pacific. Such a feature is broadly consistent, spatially, and in a preliminary analysis, with the Antarctic or Southern Annular Mode (SAM) [*Thompson and Wallace*, 2000]. This would seem reasonable since ozone change at these scales appears to be related primarily to transport and changes in the transport processes. The weak longitudinal structure in January, only present at higher latitudes, also would agree with the SAM concept. A comparison of the SAM index with the evolution of ozone change suggests that over the 1980s and early 1990s the ozone change is negative during peak positive index values and positive near weakly negative or weakly positive index values. A closer inspection of the present results with the SAM structure and the SAM index points to some differences, however. The longitudinal structures of ozone change during October and to a lesser extent in June show a bodily eastward migration, something not contemplated in the SAM definition. A detailed analysis is not straightforward and is beyond the scope of the present study.

[44] It should be noted, on the other hand, that the shape and location of the ozone change suggests a significant contribution from the standing wave 1 stratospheric feature and maybe wave 2 as well. It is well established that the midlatitude ozone distribution in winter and spring over the Southern Hemisphere is linked to the stationary planetary wave structure. Interannual variability of these waves and the differing changes in amplitude of waves 1 and 2 could help explain such a behavior. An analysis of monthly mean stationary and quasi-stationary waves in the stratosphere and their long-term evolution is necessary to reach a more conclusive answer at this stage.

[45] Finally, note that the axis or node of the interannual variability in June and even more so in October follows closely the displacement of the TOC trough region. Since the eastward evolution has been observed in PV this is clearly a dynamical issue, connecting ozone, PV, and temperature variability. Nevertheless, at this stage it is not possible to infer the mechanisms that are driving such large-scale circulation changes.

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