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Millimeter wave spectroscopic measurements over the South Pole
4. O_3 and N_2O during 1995 and their correlations for two quasi-annual cycles

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Abstract. In two separate papers we have previously reported observations of stratospheric O_3 and N_2O over the South Pole during the 1993 annual cycle. Here we present (1) new O_3 and N_2O observations at the South Pole in 1995 and (2) correlations between O_3 and N_2O for two 11-month observations during February 1993 to January 1994 and January-December 1995. Strong similarities exist between the two quasi-annual cycles for both O_3 and N_2O. A double-peaked profile again dominates O_3 vertical distribution in 1995 as in 1993. Features such as a pronounced summer-fall decline in mid-stratospheric O_3 followed by an early winter increase, a downward trend in the O_3 contour pattern associated with vertical transport, a transient enhancement of middle to upper stratospheric O_3 just before local sunrise, the timing of the ozone hole onset, and a dramatic increase of stratospheric O_3 during and following vortex breakup all show good consistency between the two annual cycles. N_2O observations show a good agreement between the two 11-month cycles in atmospheric descent rate during fall and winter, and in the timing of N_2O recovery from diminished values during spring. We use O_3-N_2O correlations to further investigate the double-peaked vertical distribution of O_3. During springtime warmings the O_3/N_2O ratio shows a tight coupling between O_3 and N_2O around 20 km, as transport creates the low-altitude O_3 peak. A rapid and systematic decrease of the O_3/N_2O ratio during summer in the 25 to 30 km region (while N_2O is essentially stable) supports the increasingly dominant role of photochemistry in producing the vertical profile for O_3 above ~25 km while leaving a transport-produced layer with a relatively large mixing ratio below ~25 km. The resulting double-peaked O_3 distribution, which persists for many months, can alter the normally negative correlations between O_3 and N_2O in the lower and middle stratosphere, although in measurements of the N_2O/O_3 ratios for polar air these perturbations have often been taken to be a hallmark of catalytic ozone depletion by chlorine. The present analysis should help to clarify the influence of the relatively unique O_3 vertical distribution of polar ozone when interpreting O_3-N_2O correlations.

1. Introduction

The polar stratosphere has been a major subject of atmospheric research since the discovery of the Antarctic springtime ozone hole in the mid-1980s. During the period between February 1993 and January 1994 we initiated a program of ground-based millimeter wave measurements of several stratospheric trace gases at the South Pole, designed to investigate both polar stratospheric dynamics and chemistry of several species over all or most of an annual cycle in this remote region. In collaboration with the Antarctic Submillimeter Telescope and Remote Observatory (AST/RO) program, a continuation of these measurements at the South Pole was carried out during January-December 1995, using the same instrument. The resulting data from the two 11-month series constitute a unique set of simultaneous measurements of profiles of stratospheric N_2O and O_3 deep within the polar vortex above aircraft and/or

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balloon altitude limits. (The extensive data sets from the Upper Atmosphere Research Satellite (UARS) go at most to 80°S latitude and have alternating ~36-day gaps in high-latitude coverage.) Results of the first campaign (1993-1994) include the use of quasi-continuous observations of N_2O to establish the duration and vertical velocity gradient of diabatic descent for stratospheric air within the vortex core during polar fall and winter, as well as the timing of N_2O transport back into the polar region when the vortex breaks down. We have shown that recent results from radiative transfer models are in good agreement with our observational data [*Crewell et al.*, 1995]. Annual evolution of South Pole ozone throughout the stratosphere was presented and analyzed by *Cheng et al.* [1996]. A persistent double-peaked structure in Antarctic stratospheric O_3 and the early seasonal onset of the ozone hole were noted in the 1993 data and interpreted. The seasonal climatology of HNO_3 and its removal from the lower stratosphere by its incorporation into polar stratospheric cloud particles (PSCs) have been detailed by *de Zafra et al.* [1996]. We have also verified the production of new gas-phase HNO_3 in the polar mid-stratosphere during the winter night.

In the present paper we first present analyses of O_3 and N_2O observations from the 1995 field work, followed by a study of correlations between O_3 and N_2O for the two quasi-annual records. Comparisons between the two field experiments highlight several interesting features and provide further support to our earlier findings. Since our ground-based measurement covers an altitude range well above that of aircraft and was located in the core of the southern vortex, the present correlation study complements and extends several earlier studies and furnishes a test for our current understanding of O_3-N_2O behavior in this extreme region.

During portions of the annual polar cycle not affected by chemical ozone depletion, O_3 as well as N_2O may serve as a useful trace species. The O_3-N_2O correlation can be a useful indicator in evaluating roles of various dynamical and chemical processes. In the lower and middle stratosphere, especially in the polar regions, N_2O can be treated as a conserved quantity, and associations between N_2O and O_3 yield information about perturbations and rate of change in O_3 that go beyond information available from observations of O_3 alone. For instance, N_2O has been used as a reference to estimate O_3 chemical loss and as a tracer to determine air origin [*Schoeberl et al.*, 1990, 1992; *Proffitt et al.*, 1990; *Collins et al.*, 1993; *Bregman et al.*, 1995]. The O_3-N_2O relationship in the polar winter lower stratosphere was investigated in several studies with aircraft data, and a negative correlation was established. *Strahan et al.* [1989] have asserted that this negative correlation is a signature of the chemically unperturbed lower to middle stratosphere due to the spatial gradients of O_3 and N_2O. To evaluate using a linear reference relationship between O_3 and N_2O to estimate polar winter O_3 loss from aircraft data, *Proffitt et al.* [1992] have compared two-dimensional model simulations of O_3 and N_2O with northern high-latitude measurements (winter aircraft data and limited data from the NIMBUS 7 satellite). Recently, *Hall and Prather* [1995] have used a three-dimensional photochemical and transport model to interpret observed O_3-N_2O correlations in a theoretical framework throughout the global stratosphere. The model successfully simulated the normal anticorrelation between O_3 and N_2O in the lower stratosphere, in agreement with observations below 20 km (the height limit of the data used for that study). Despite the speculation that negative correlations extend up to 35 km [*Strahan et al.*, 1989] the O_3-N_2O relation has remained untested above 20 km over polar regions. We will show in the present paper that this assumption is frequently invalid.

2. Techniques

Millimeter wave pressure-broadened emission spectra of stratospheric O_3 and N_2O (at 276.923 and 276.328 GHz, respectively) were typically collected at 3- to 5-day intervals in 1993 but on a reduced (once per week or less) basis in 1995. Vertical profiles of mixing ratios have been retrieved through deconvolution techniques [*Twomey et al.*, 1977]. Reliable altitude coverage extends from ~15 to 55 km for O_3 (with profiles for $\leq$ 15 km supplied from balloon ozonesondes) and from ~15 to 45 km for N_2O in summer, although subsidence of N_2O during winter significantly reduces the upper limit of reliable altitude coverage in the latter case. The vertical resolution of the ground-based observing geometry, defined as the ability to resolve two separate emitting layers, is limited to about 1 atmospheric scale height, i.e., ~7 km in the polar stratosphere. Tests show, however, that uncertainty in determining altitude of the peak mixing ratio for a single well-defined layer is $\leq$2 km. Overall accuracy for the recovered mixing ratios in the retrieval-sensitive region (~16-45 km) is estimated to be typically ±15%. Detailed descriptions of the data collection and retrieval are found in earlier publications [*Crewell et al.*, 1995; *Cheng et al.*, 1996]. Meteorological data used in the present study were obtained from the National Meteorological Center (NMC) on a day-by-day basis for the immediate South Pole region.

3. O_3 Vertical Profiles

An example of the double-peaked structure in stratospheric O_3 over the South Pole in 1993 is illustrated in Figure 1. We have interpreted the lower peak at ~20 km [*Cheng et al.* 1996] as being due to transport from ozone-rich regions near the edge of the Antarctic, while the profile from ~25 km upward, composing the "trough" region and the upper peak, is shown to be a result of photochemistry during continuous summer solar exposure. In 1995 the double-peaked O_3 structure again dominates the vertical distribution, as seen

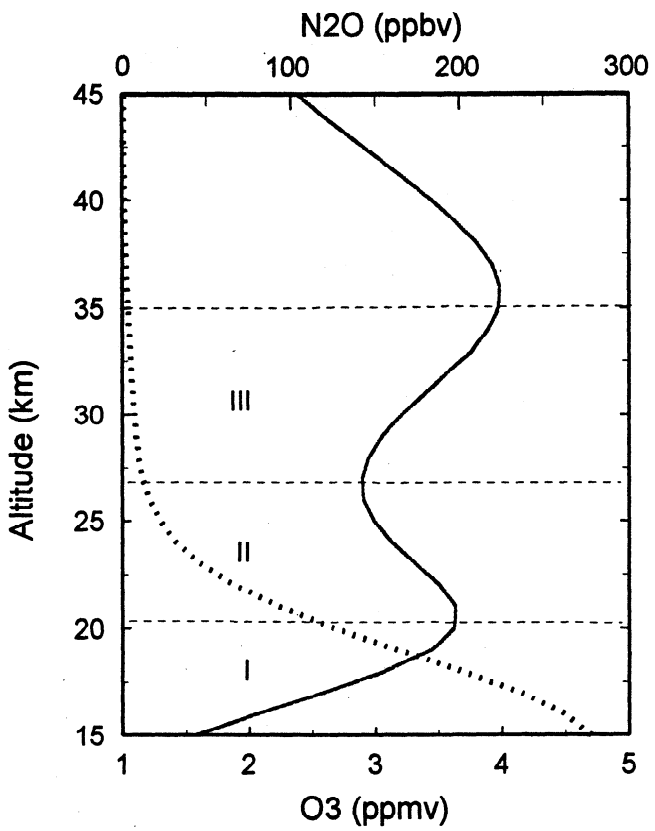

Figure 1. Vertical profiles of millimeter wave O_3 (solid line) and N_2O (dotted line) over the South Pole on May 10, 1993. The negative vertical gradients of both species in the 20-27 km region (Region II) lead to a positive correlation between O_3 and N_2O in this region.

from retrieved vertical profiles. We do not specifically illustrate this feature with the 1995 data here, since the morphology of this double-peaked O_3 distribution closely duplicates what was observed in 1993, as detailed by *Cheng et al.* [1996]. We include Figure 1 here as an aid in clarifying our later discussions of O_3-N_2O correlations as a function of altitude and time.

4. O_3 Contours

Figure 2 presents O_3 contours observed during February 1993 to January 1994 (reprinted from *Cheng et al.* [1996]) compared with new data obtained during January-December 1995. The double-peaked vertical structure is not explicitly seen in these contour maps, because the limited vertical resolution of our instrument cannot always provide the contrast between the peaks and the trough strongly enough to be picked up by the relatively coarse grid contour mapping used here.

A rather sharp March-April mid-stratospheric O_3 decrease is seen to have occurred in both years. This high-latitude summer-fall O_3 decline has been investigated with both observational data and theoretical modeling predictions by *Perliski et al.* [1989]. In that study the model O_3 mixing ratios at 10 mbar and 75° latitudes (the highest latitude at which observations are compared with models for both hemispheres) exhibit an approximately linear O_3 decline caused by the uninterrupted destruction of ozone by NO_x starting in early summer and lasting through fall, in good agreement with observations (satellite measurements and rocket and balloonsonde data). Our data clearly show that this general behavior extends to the South Pole with detailed temporal evolution and full altitude range, although the drop in O_3 appears to occur more abruptly than it does in the model simulations.

The rapid late April-May increase of mid stratospheric O_3 is also evident in 1995. We have interpreted this as being due to the inner vortex mixing process flattening out the summer negatively poleward O_3 gradient [*Cheng et al.*, 1996]. The clear downward trends in

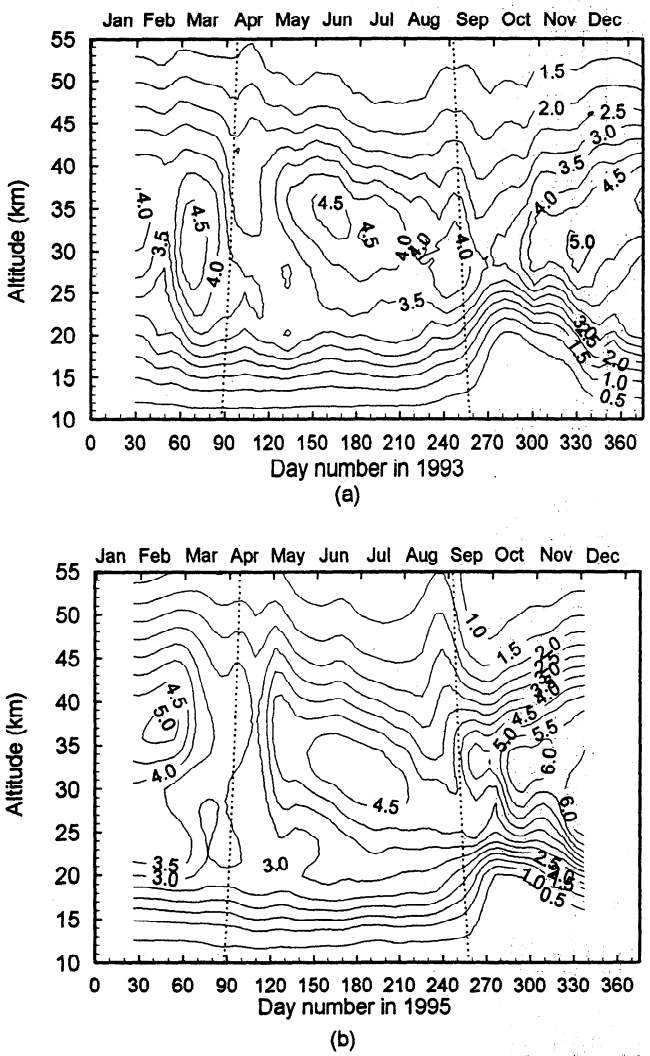

Figure 2. Mixing ratio contours derived from retrieved O_3 vertical profiles at the South Pole. (a) 1993 data. A Gaussian smoothing function with a 5-day half width has been applied to data taken approximately every third day. (b) 1995 data. A Gaussian smoothing function with a 7-day half width has been applied to data taken approximately every week. The dotted lines mark polar sunset and sunrise as a function of altitude.

the O_3 contour pattern in the middle and upper stratosphere associated with diabatic subsidence of air during fall to early spring, and the dramatic increase of lower to mid-stratospheric O_3 during and following vortex breakup (starting in mid-October), all show excellent consistency between the two annual cycles.

In both 1993 and 1995, O_3 in the vicinity of 20 km is seen to begin a decline in mixing ratio in mid-August, well before local sunrise. We believe this is due to poleward transport of O_3-depleted air from outer, sunlit regions of the vortex where O_3 is destroyed by catalytic chlorine species, as discussed by *Cheng et al.* [1996].

A transient increase in middle to upper stratospheric O_3 just before local sunrise in August is clearly seen in each year. We do not see any corresponding disturbance in potential vorticity or temperature from NMC data strong enough to explain a direct dynamical cause. This interesting phenomenon needs further investigation.

Differences between the 1993 and 1995 mid-stratospheric O_3 patterns in late summer (March) suggest that this region exhibits significant interannual variability caused by an interplay of chemistry and dynamics. During late spring, (November to early December) stronger mid-stratospheric transport in 1995 brought in O_3 as high as 6 parts per million by volume (ppmv), as against 5 ppmv in 1993.

5. N_2O Contours

Results from observations of N_2O in both 1993 and 1995 are displayed as contours in Figure 3. The two records show an excellent agreement in atmospheric descent rate (represented by the slope of the contours) during fall and winter. The inferred descent rate as a function of time and altitude from our 1993 N_2O measurements has been given by *Crewell et al.*, [1995] and is in good agreement with results from radiative transfer models of *Rosenfield et al.* [1994] and *Manney et al.* [1994].

The timings of polar middle to upper stratospheric N_2O recovery from the diminished winter values in both springs also agree well with each other. This recovery results from the breakup of the polar vortex. Unfortunately, some detail has been lost in 1995 because of a paucity of measurements in comparison with 1993.

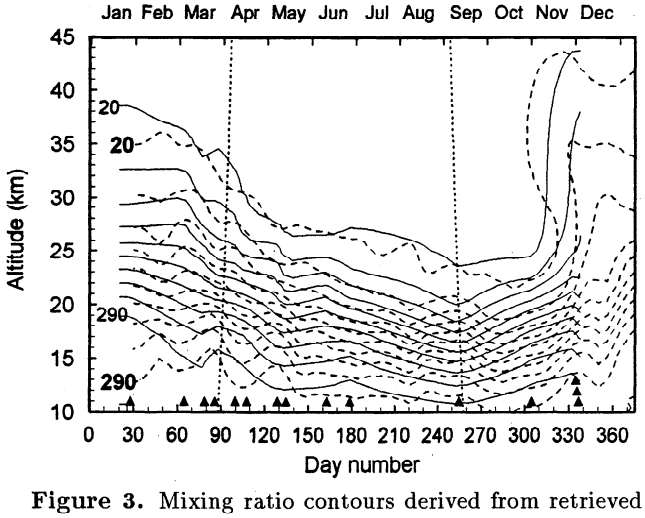

Figure 3. Mixing ratio contours derived from retrieved N_2O profiles at the South Pole. Dashed lines; 1993 data. A Gaussian smoothing function with a 5-day half width has been applied to data taken approximately every third day. Solid lines; 1995 data. Data were taken for the days indicated by solid triangles at the bottom of the plot. No smoothing has been applied to these data. Both data sets use the same contour values from 20 to 290 ppbv at intervals of 30 ppbv. The 1993 data are labeled in bold numbers. The dotted lines mark polar sunset and sunrise as a function of altitude.

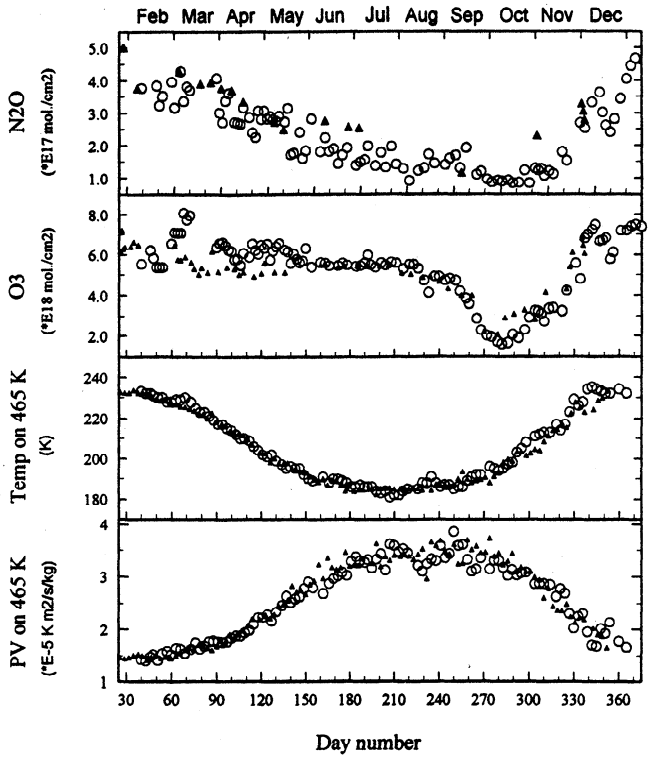

Figure 4. (Panels 1 and 2) Column densities of N_2O and O_3, derived from integrating millimeter wave retrieved profiles from 15 to 50 km. (Panel 3) NMC temperature at 465 K potential temperature surface. (Panel 4) absolute Ertel's potential vorticity (PV) at the 465 K surface over the South Pole. Circles indicate data for 1993, and solid triangles indicate data for 1995.

6. Column Densities

Figure 4 shows the annual evolution of column densities of N_2O and O_3 for the two annual cycles, calculated by integration of vertical profiles from 15 to 50 km. Also included are Ertel's potential vorticity (PV) and NMC temperatures, both at the 465 K potential temperature surface (~19-20 km in winter) over the Pole, obtained from the NASA Goddard Space Flight Center (GSFC) "AutoMailer" meteorological data analysis system. Our observations have a lower limit of ~16 km. We arbi-

trarily choose the 465 K surface here, for comparisons with other data, although other nearby choices would give similar correlations. *PV* is prevailingly negative in the southern hemisphere. For simplicity, absolute *PV* values are used in Figure 4 and the correlation studies discussed below, so that "larger" *PV* means greater in magnitude.

The general behavior in the columns from the two records resemble each other: The stratospheric N_2O column diminishes during winter and early spring; the early onset of the ozone hole is seen during middle to late August; late spring recovery from the ozone hole is seen for O_3, as is recovery of N_2O from extremely low winter values. Consistent with the contour maps shown in Figure 2, significant variability is also evident in O_3 columns during February to April and again beginning in October, the periods during which dynamics tends to dominate O_3 distribution.

Figure 4 shows that changes in column N_2O closely follow the rate of change in temperature and $|PV|$ into August. The onset of the N_2O column recovery in mid-October appears to be significantly delayed with respect to the onset of warming and *PV* decline in the lower stratosphere, however. This delay is a result of the continued subsidence within the still-intact lower vortex during spring, so that the N_2O column density (dominated by altitudes below 20 km) is slower to reflect vortex breakdown than *PV* or temperature at ~20 km. After mid-October, all quantities in Figure 4 resume their close coupling. During this period the weaker vortex presents diminishing barrier effects to mixing across the vortex boundary. O_3 and N_2O are transported into the polar region from lower latitudes, accounting for the rapid increase in column densities, although chemical processes can make small modifications to the column density above ~25 km.

7. O_3/N_2O Ratios and the Double-Peaked O_3 Distribution

Since the N_2O mixing ratio is essentially a conservative quantity, the O_3/N_2O ratio also yields information about transport and chemistry. Figure 5 depicts O_3/N_2O ratio contours as a function of altitude and time, contrasted directly with several N_2O mixing ratio contours that reveal atmospheric subsidence from late fall to early spring and transport during the vortex breakup. The slopes of O_3/N_2O ratio contours closely follow the slope of the N_2O mixing ratio itself from April to August, suggesting that O_3 is also transport-controlled during the period. The breakdown of this O_3-N_2O coupling in September and October is consistent with ozone hole chemistry. It is interesting to note that in the 30-km region the O_3/N_2O ratio drops rapidly from ~190 to ~60 within 1 month around late November after both species initially increase dramatically in the region in early to mid-November, according to Figure 2 (O_3 increase) and Figure 3 (N_2O increase).

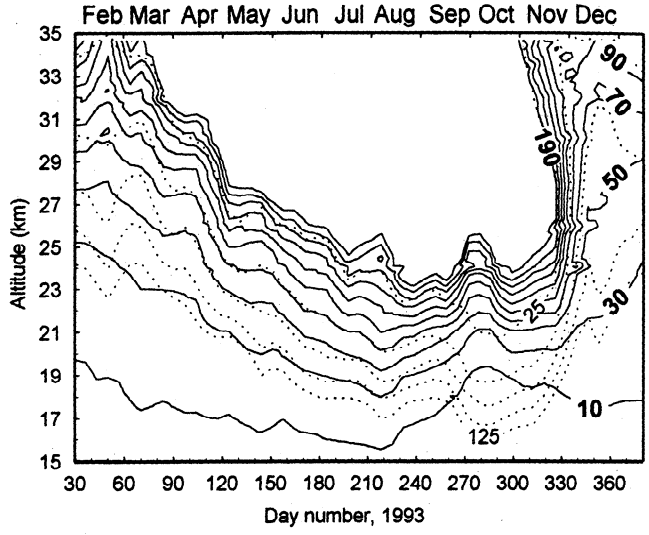

Figure 5. Solid lines; 1993 O_3/N_2O ratio contours as a function of altitude and time, labeled with bold numbers at intervals of 20. Dotted lines; several 1993 N_2O contours at intervals of 25 ppbv.

This early summer decline in the O_3/N_2O ratio implies that O_3 decreases rapidly in relation to N_2O. Further, this decoupling of O_3/N_2O indicates that initial large O_3 mixing ratios created by poleward transport undergo a reduction toward photochemical equilibrium in and above the 30-km region during continuous summer daylight, as illustrated by the strongly polar concentric symmetry of UARS microwave limb sounder (MLS) O_3 data [*Cheng et al.*, 1996] and predicted by summer polar photochemistry [e.g., *Perliski et al.*, 1989]. In the 20-km region, however, the O_3/N_2O ratio stays between 10 and 30 during and after vortex breakup, showing that initial transport dominates, and subsequent photochemistry does not much alter the O_3 mixing ratio in the lower summer stratosphere, where transport tends to dominate over chemistry. In summary, the O_3/N_2O behavior therefore constitutes further evidence for transport as the mechanism that produces the lower O_3 peak at ~20 km, while the entire profile above ~30 km, including the "trough", is a consequence of summer photochemical erosion on transport-enhanced O_3 [*Cheng et al.*, 1996].

8. O_3-N_2O Correlation Plots: Ozone Loss and Transport

In order to emphasize different features of O_3 and N_2O mutual variations, in this section we use correlation plots of O_3 versus N_2O to exhibit seasonal trends. Under typical conditions in the lower and middle stratosphere, N_2O decreases with altitude while O_3 increases, so the correlation between O_3 and N_2O with respect to altitude is negative, as detailed in several previous studies, such as those of *Proffitt et al.* [1990, 1992]. Our South Pole millimeter wave data extend to greater alti-

tudes than those available from aircraft however, and cover seasonal behavior not previously observed. In Figure 6, O_3 versus N_2O correlation plots for different months illustrate seasonal variations in their relationship up to the reliable N_2O retrieval height limit for each year, along with a least squares fit to northern hemispheric extravortex air by *Proffitt et al.* [1990] for a comparison. In deriving each curve, 2-week mean mixing ratios were used whenever more than one measurement was available within the 2-week period. Negative correlations with respect to altitude are seen in Figure 6 for the lower portion of the stratosphere (the section rightward along each curve from the diamonds represents altitudes below 20 km). For altitudes above 20 km in most of the pre-ozone-hole plots (the April plot in 1993, and the January to May plots in 1995, (Figure 6a)), however, the correlation is gradually reversed, becoming positive (both species decrease with increasing altitude). These observed positive correlations are due to the existence of the O_3 trough mentioned in section 3.

As downward transport occurs during austral fall and winter, both low O_3 (from the trough) and low N_2O (due to the negative vertical gradient) between the trough (~30 km) and the lower peak (~20 km) descend to yield a positive correlation (region II of Figure 1). Positive correlations may also occur in this altitude range when wave motions fold vertical distributions of O_3 and N_2O. The strength and duration of the O_3 trough suggest that this positive correlation can be observed both vertically and horizontally on various timescales during polar fall-winter. This phenomenon creates a positive O_3-N_2O correlation in the lower to middle stratosphere having no relationship with O_3 de-

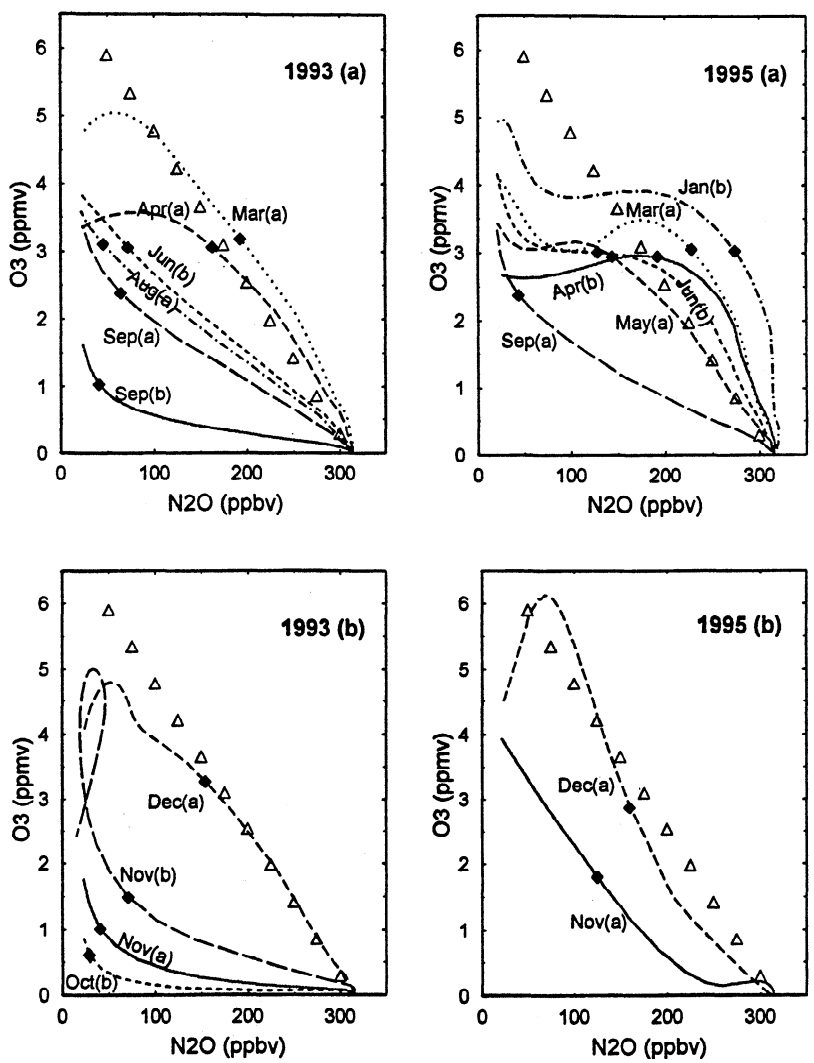

Figure 6. Correlation plot of O_3 versus N_2O for (left) 1993 and (right) 1995 data up to the reliable N_2O height limit, along with a least squares fit (triangles) to northern hemispheric extravortex air by *Proffitt et al.* [1990] shown as a reference curve. Two-week mean mixing ratios are used when more than one measurement is available. An a or b in parentheses following the abbreviated month labels indicates the earlier or later part of that month. Diamonds represent the mixing ratios at 20-km altitude, and the portion of each curve to the right represents lower altitudes for both variables.

pletion from chlorine chemistry. This contrasts with the conclusion derived primarily from aircraft measurements in the late winter vortex by *Strahan et al.* [1989], namely, that positive O_3-N_2O correlations can not be explained by transport (below 35 km) and appear to be a sensitive indicator of ozone loss through chlorine chemistry.

Figure 6a demonstrates that the slope in the O_3 versus N_2O plot is slowly reduced during fall and winter seasons. With the aid of the 20-km altitude symbols and the slopes of the plots relative to the reference (the fit for extravortex air), one sees that variation of the O_3-N_2O correlation behavior in the lower stratosphere is initially dominated by N_2O decrease due to subsidence before August (dynamic cause) but later by rapid O_3 loss during the September-October ozone hole season (chemical cause).

During the vortex breakup, O_3-N_2O correlations can be retrieved up to 45 km, because of stronger N_2O signals associated with the return of N_2O in the middle to upper stratosphere. The O_3-N_2O relation at this time is characterized by a "loop" as shown in the 2-week mean November curve in Figure 6, 1993 (b). This is a dominant feature during November and is caused by high-altitude intrusion of N_2O-rich air from lower latitudes as the vortex weakens from the top downward, leading to an early N_2O peak forming at ~35 km, above the upper O_3 peak that has subsided to ~30 km (see Figures 1a and 2). This interplay of peaks of two gases as extraavortex air returns over the Pole at various altitudes leads to multivalued mixing ratios of one species against a single mixing ratio of the other. A transition from the November behavior toward the extravortex reference correlation occurs in Decembers in Figure 6b, shortly after vortex breakup, indicating arrival of extravortex air. Interannual variability in the correlation behavior is significant. Enhanced late summer lower stratospheric O_3 and N_2O values in 1995 are responsible for the departure of the January and March correlation plots from the reference relationship in Figure 6a, leading to a more prominent double-peaked O_3 structure and positive correlations in the plots.

9. Summary and Conclusions

Millimeter wave spectroscopic measurements over the South Pole exhibit striking similarities between the two quasi-annual cycles for both O_3 and N_2O. The double-peaked structure again dominates O_3 vertical distribution in 1995 as in 1993. Features such as a pronounced summer-fall decline in mid-stratospheric O_3 followed by an early winter increase, a downward trend in the O_3 contour pattern associated with winter air subsidence, a transient increase of middle to upper stratospheric O_3 just before sunrise, the timing of the ozone hole onset, and the dramatic increase of O_3 during and following vortex breakup all show good consistency between the two years. N_2O observations also show a good agreement between the two annual cycles in atmospheric descent rate during fall and winter and the timing of N_2O recovery in spring.

The N_2O column density is highly correlated with lower stratospheric PV and temperature. The observed column densities of O_3 and N_2O and meteorological parameters all show strong correlations with one another in springtime, largely due to dynamical effects. With rapid warmings after mid-October, transport from lower latitudes reestablishes the strong coupling between O_3 and N_2O in the 20-km region, constituting further evidence that springtime transport produces the long-lasting low-altitude O_3 peak typically seen over the south polar region [*Cheng et al.*, 1996]. The decoupling of the initial strong O_3-N_2O correlations during summer in the 30-km region indicates the dominant role of photochemistry in the production of the O_3 profile above 30 km, including the "trough" region. This trough causes positive O_3-N_2O correlations during the fall-winter period prior to chemical depletion of O_3, as demonstrated by the correlation plots. This conclusion contrasts with the one reached by *Strahan et al.* [1989] from analysis of limited aircraft data, namely, that a positive correlation between O_3 and N_2O is a safe indicator of chlorine-initiated O_3 depletion at any altitude up to 35 km. Before August the slope in O_3 versus N_2O plots slowly decreased because of stratospheric subsidence. During the ozone hole season the slope in O_3 versus N_2O plots is greatly reduced because of O_3 chemical loss. The O_3-N_2O relation in the middle and upper stratosphere during the vortex breakup is characterized by a "loop" in the O_3 versus N_2O plot for the November data, caused by a relatively early intrusion of extra-vortex air in the middle stratosphere. Significant interannual variability in the correlation plots is seen by a comparison of the 1993 and 1995 data.

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