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Millimeter wave spectroscopic measurements over the South Pole, 1. A study of stratospheric dynamics using N_2O observations

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Abstract. Millimeter wave measurements of N_2O and O_3 [*Cheng et al.,* 1995], along with several other trace gases, have been made nearly continuously from February 1993 through early January 1994 at the Amundsen-Scott Station, South Pole. In order to separate chemical and dynamical effects, this paper uses the observations of the long-lived tracer N_2O to study stratospheric dynamics. The main emphasis is on the synoptic evolution of the polar vortex over an entire winter period, and quantitative results are given for various times and altitudes. Diabatic descent rates derived for different altitude levels showed the strongest descent in austral fall at high altitudes, agreeing fairly well with model predictions by *Rosenfield et al.* (1994). Subsidence was observed to continue until late October, well after polar sunrise. The breakdown of the vortex occurred first in the upper stratosphere, marked in the intrusion of N_2O rich air at these altitudes, consistent with trajectory calculations. Our calculated descent rates are not consistent with the idea that the polar vortex is a "flowing processor", but instead should be viewed as an isolated system.

Introduction

Nitrous oxide (N_2O), which has only tropospheric sources, currently has a constant mixing ratio of about 315 ppb in the troposphere, where its lifetime is in excess of 100 years. Transported into the stratosphere in the tropics, it is primarily destroyed by photolysis at high altitudes, resulting in a stratospheric lifetime of >1 year below 33 km. The mean distribution of N_2O therefore shows maxima at low altitudes and low latitudes, and decreases poleward and with altitude [*World Meteorological Organisation*, 1986]. Its relatively long lifetime, especially in the polar night, makes it a valuable atmospheric tracer which has been used in a number of studies of atmospheric transport processes.

Ground-based millimeter wave measurements of N_2O at McMurdo Station (78°S) in 1986 [*Parrish et al.*, 1988a] gave the first modern estimates of strong subsidence inside the polar vortex, and have been used to monitor vortex movement in the Arctic [*Emmons et al.*, 1994a]. N_2O has also been measured over Antarctica during the Airborne Antarctic Ozone Campaign (AAOE) [e.g., *Loewenstein et al.*, 1989; *Strahan et al.*, 1989] and used to derive descent rates inside the polar vortex from the downward shift of the N_2O [*Schoeberl et al.*,1992]. However, these data have been limited in space (~72°S and altitudes <20 km) and time.

Recent satellite measurements of N_2O have been carried out by the Improved Stratospheric And Mesospheric Sounder (ISAMS) and the Cryogenic Limb Array Etalon Spectrometer (CLAES) on board the Upper Atmosphere Research Satellite (UARS) launched in September, 1991. The observations from these instruments have been used, for example, to test global circulation modelling [*Randel et al.*, 1994; *Sutton et al.*, 1994], the structure of the vortex edge [*Ruth et al.*, 1994], stratospheric warmings [*Manney et al.*, 1994a], or to seperate chemical and dynamical effects [*Manney et al.*, 1994b]. However, they have been restricted to ≤ 80° south in observing the Antarctic vortex, and to alternate monthly intervals due to the 36-day yaw maneuvers of the satellite. These instruments stopped working in August 1992 (ISAMS) and May 1993 (CLAES).

The ground-based millimeter wave measurements presented in this work fill the gap in monitoring the N_2O profile over almost a full annual cycle at the South Pole, in the heart of the Antarctic vortex. Coverage includes the formation of the polar vortex, winter subsidence, and vortex breakdown. These measurements were also intended to help in the interpretation of our other trace gas measurements (O_3, HNO_3, ClO, and NO_2) which were carried out in the same time period.

Although the end result of significant subsidence within the Antarctic vortex was first observed in the late 1980s [*Parrish et al.*, 1988a; *Loewenstein et al.*, 1989], there is little quantitative information during periods of actual descent. Both high (100 m/d ≡ 1.75 K/d) [*Proffitt et al.*, 1989] or low values (35 m/d ≡ 0.25 K/d) [*Schoeberl et al.*, 1992] have been deduced based on observations near the final stages of vortex evolution in August to September 1987. The altitude range for both values is 16-20 km. These rates are intimately connected to the question of how isolated the air inside the polar vortex is and whether the vortex is a flowing processor or a containment vessel [*Randel*, 1993]. Recent model studies by *Manney et al.* [1994c] and *Rosenfield et al.* [1994] have tried to calculate diabatic descent carefully, but lacked a comparison of their models with quasi-continous observations in the southern vortex. After describing the measurement method and presenting the general features of our

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observations we will focus on a comparison of measured descent rates with the predictions of these two models.

Measurement Technique and Data Retrieval

The pressure broadened spectral line of N_2O at 276.328 GHz has been measured using a cryogenical cooled millimeter wave spectrometer described in detail elsewhere [*Parrish et al.*, 1988b]. Since the earlier measurements [*Parrish et al.*, 1988a], the spectrometer bandwidth has been doubled to 512 channels, while keeping a resolution of 1 MHz per channel, allowing significantly more accurate retrieval of low altitude N_2O. Data were taken approximately every third day starting on February 9, 1993, through to January 6, 1994. The measured single-day N_2O spectra have a typical noise level of 0.01 K compared to a line amplitude of 0.3-0.7 K. (Values are given in degrees Kelvin, using the nearly linear Rayleigh-Jeans approximation to relate emission intensity to an equivalent blackbody temperature at millimeter wavelengths.) The influence of nearby ozone lines in the observed spectra is removed by subtracting a background computed from ozone measurements [*Cheng et al.*, 1995] made within a day or two of a given N_2O observation. Sample spectra are presented in Figure 1 showing a change in line shape during polar winter as subsidence carries N_2O lower and increases pressure broadening of the line.

N_2O mixing ratio profiles were retrieved from the pressure-broadened line shapes using the Chahine-Twomey inversion technique. In the retrieval process combinations of four different starting profiles and two sets of weighting functions, eight permutations in all, were used to minimize any bias induced by these. All starting profiles are set to a nominal 315 ppb in the troposphere (≤8 km), using measurements from the South Pole and taking a yearly increase of 1 ppb [*Rasmussen and Khalil*, 1986]. In Figure 2, representative profiles are shown as averaged values of all eight daily retrievals, and error bars represent the 1 σ variations resulting from these different retrievals. Data are reliable between 15 km, where the lowest altitude weighting function increases to >50 full amplitude, and 45 km where the

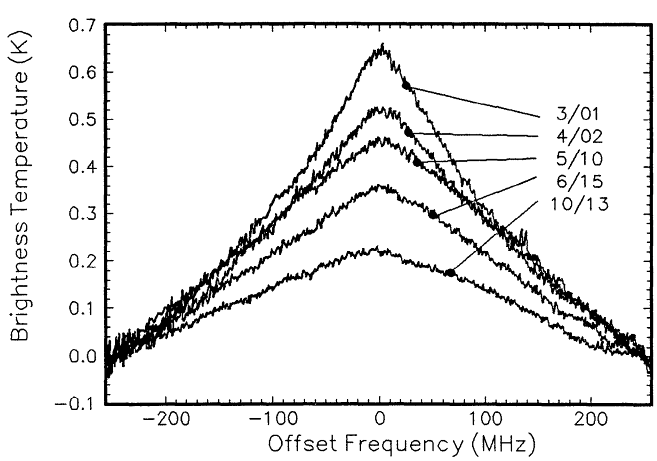

Figure 1. Observed N_2O spectra for different days between March 1, 1993, and October 13, 1993, at the South Pole. Intensity, as equivalent blackbody radiation in Kelvin, is plotted versus relative frequency in MHz. Center frequency is 276,328 MHz, and the resolution of the filter-bank spectrometer is 1 MHz. All spectra have been arbitrarily normalized to zero offset at the edges of the spectral window.

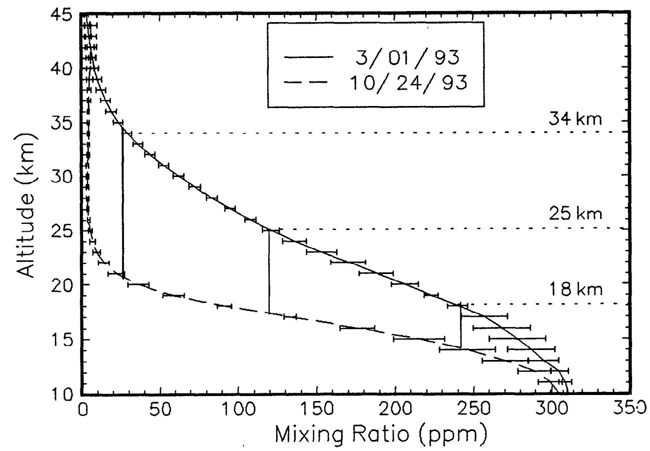

Figure 2. Retrieved N_2O profiles for March 1, 1993, and October 24, 1993, at the South Pole. Error bars indicate the variations caused by different starting profiles and weighting functions.

N_2O mixing ratios begin to assume the same magnitude as their error. A more detailed error analysis [*Emmons et al.*, 1994b] including uncertainties from spectral noise, calibration and limited knowledge of temperature, pressure and pressure broadening coefficient yields an overall uncertainty of < 40 ppb below 20 km, reducing to < 20 ppb for altitudes higher than 25 km.

Observations

The general behaviour of N_2O over the South Pole during the observation period is first displayed via column densities, shown in Figure 3. Because the altitudes below 15 km contribute strongly to the total column density but have a high uncertainty, as mentioned above, the values in Figure 3 were calculated for the column between 15 and 50 km. From Figure 3 it is obvious that the column density decreases rapidly, starting about polar sunset. Because of the typical decrease of N_2O with altitude, this indicates a downward shift of the profile (e.g., subsidence) at this time. Even after polar sunrise, the column amount continues to decrease a little farther, to only 20 % of its March value. The arrival of lower latitude air at the South Pole can be seen in the increase of column densities starting in the end of October (day 300). First the values increase slowly by about a factor of 2 until the beginning of December (December 1 = day 335), when a rapid increase to the prewinter levels occurred.

Changes in the vertical N_2O distribution derived from approximately 100 individual profiles at ~3 day intervals are shown in Figure 4. In order to focus on diabatic events, potential temperature is used as a vertical coordinate. Two major events which will be separately discussed in more detail below are obvious: (1) The downward trend of the isopleths by polar sunset, which occurs in early April at these altitudes, showing the rate of subsidence as a function of altitude inside the polar vortex. Also, around sunset stratospheric temperatures decrease strongly (e.g., see plot by *Cheng et al.*[1995]) and the formation of the polar vortex can be seen in National Meteorological Center (NMC) temperatures potential vorticity maps. (2) The recovery of N_2O at the end of October, starting with a second maximum at high altitudes. Some minor effects like the uplifting of the N_2O isopleths below 600 K in mid-May (day=130) can be observed. This feature can also be seen in temperature profiles.

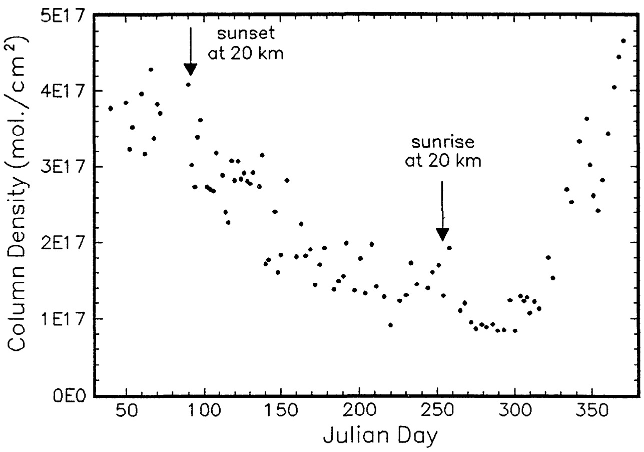

Figure 3. Column density of N_2O between 15 and 50 km altitude during the observation period 1993-1994 over the South Pole.

However, it has to be noted that the errors in retrieved N_2O profiles at altitudes lower than 15 km (between 380 and 440 K depending on the season) are large and interpretation of features below this margin are better avoided.

Subsidence in the Polar Vortex

To illustrate the descent of stratospheric air within the polar vortex, N_2O profiles measured over the South Pole near the beginning of vortex formation (March 1) and the end of polar winter (October 24) are contrasted in Figure 2. At the end of the winter, the N_2O profile has shifted significantly downward, and hardly any N_2O remains above 20 km. It is obvious that mixing ratios representative of the prewinter upper stratosphere (for example, 34 km) can be found more than 10 km lower at the end of the winter, while mixing ratios in the lower stratosphere (for example, 18 km, descending by ~4 km) show a much smaller downward shift. The essential feature is that the downward shift increases with increasing altitude. The larger rates in the upper stratosphere are consistent with data from the Halogen Occultation Experiment (HALOE) [*Russell et al.*, 1993], which show strong descent for the upper stratosphere and mesosphere, and the lower stratospheric rates are consistent with the ones derived from AAOE aircraft data (<20 km) [*Schoeberl et al.*, 1992]. A recent reanalysis of the HALOE data [*Schoeberl et al.*, 1995] using a revised version of the data and more sophisticated analysis methods, gives similiar results to the ones derived from AAOE data. Because of the very low N_2O mixing ratios at altitudes higher than 40 km, however, which are on the order of the retrieval uncertainty, nothing can be derived from our data set about this region or about the link to the mesosphere. A study by *Cheng et al.* [1995] based on our South Pole ozone observations concludes that although ozone appears to furnish a valid measure of vertical transport in the lower stratosphere, it is unreliable above ~40 km, even in the polar night, because of complications arising from zonal transport accompanied by diurnal change as mesospheric air moves poleward and downward into the mid- to upper stratosphere. At 35 km, however, the descent rates derived from N_2O and ozone agree rather well.

To describe the subsidence as a function of time during polar winter, the change in potential temperature for two levels of constant N_2O mixing ratio is shown in Figure 5. Using the same method, *Bauer et al.* [1994] derived diabatic descent rates for the Arctic polar vortex in 1991-1992 of 98 and 114 m/d from the downward shift of the 50 and 100 ppb N_2O levels, respectively. In order to reduce data noise as well as short-term atmospheric variations, the data are fitted with a polynomial of tenth order. Obviously, the curves show a strong downward trend around sunset and flatten out toward the end of winter, but still show some subsidence after polar sunrise, as does the total column in Figure 3. Table 1 gives quantitative values for the calculated geometrical vertical velocities at different altitudes as 2-month mean values. Because of the strong subsidence at high altitudes in austral fall, the mixing ratios in the upper stratosphere decreased to such low values that we cannot extract information at these altitudes for later time periods.

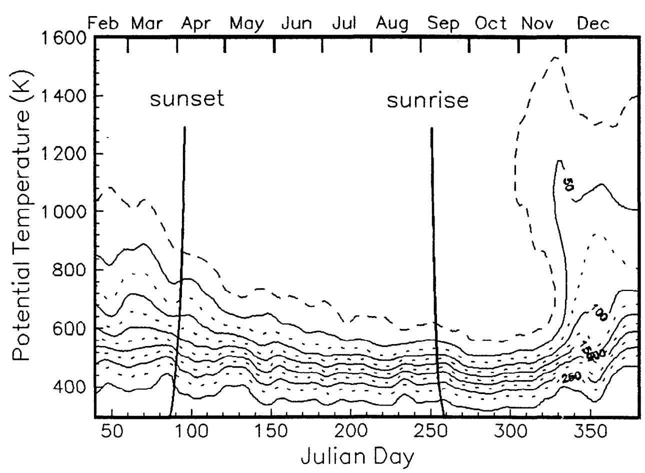

Figure 4. Vertical N_2O distribution using potential temperature as vertical coordinate over the South Pole during the observation period from February 9, 1993 (day=40), to January 6, 1994 (day=371).

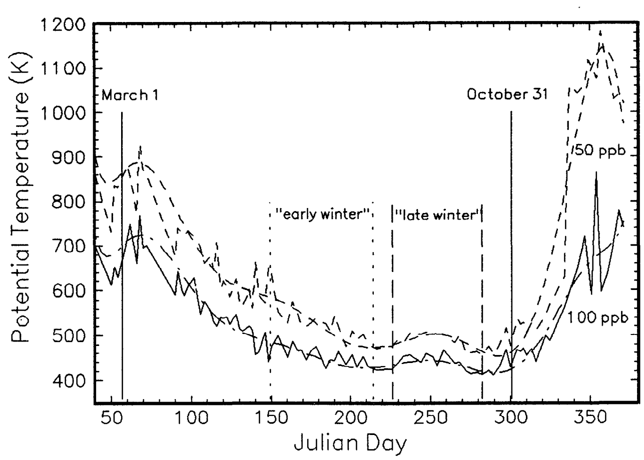

Figure 5. Potential temperature of the 50 and 100 ppb N_2O levels during 1993-1994 over the South Pole. Original data and a tenth order polynomial fit are shown. Times are indicated for the model run by *Rosenfield et al.* [1994] (March 1 through October 31) and by *Manney et al.* [1994] ("early winter": June 1 - August 6; "late winter": August 17 - October 11).

Table 1. Geometrical Vertical Velocities for Different Altitude and Time Periods

	z=20 km	z=25 km	z=30 km	z=35 km
March/April	20	45	75	100
May/June	40	33	45	
July/August	0	25	50	
Sept./October	44	50		

Geometrical vertical velocities are in meters per day.

The Breakdown of the Polar Vortex

At the end of October the spectra start to show a different shape (Figure 6), having a narrow peak at the line center. This immediately indicates an increase in high altitude (low pressure, hence weakly broadened) N_2O emission while the rest of the spectrum is almost unchanged. The wings of the lines do not change until early December when the whole line intensity increases rapidly, which is also reflected in the rapid increase in column density (Figure 3). The profiles retrieved from the spectra shown in Figure 6 are presented in Figure 7. The measurements show an intrusion of N_2O rich air between October 24 and 27 at altitudes of about 35 km, indicating a breakdown of the polar vortex. This was confirmed by several back trajectory calculations showing that an air parcel at 38 km on October 27 over the South Pole would have been at latitudes lower than 45° S only 3 days earlier. In the altitude range between 15 and 25 km, the profile is nearly unchanged, showing a still isolated airmass. Between November 30 and December 3, the final breakdown of the vortex occurs at about 25 km, increasing the N_2O mixing ratio by a factor of ~4. This is again consistent with backward trajectories but cannot be seen so easily from potential vorticity maps. These results show that lower stratospheric air within the vortex stayed isolated until early December, consistent with the model calculations by *Manney et al.* [1994c] for the 1993 winter.

Comparison with Model Results

For comparison with our data the times of different model runs have been marked in Figure 5. At first we focus on the

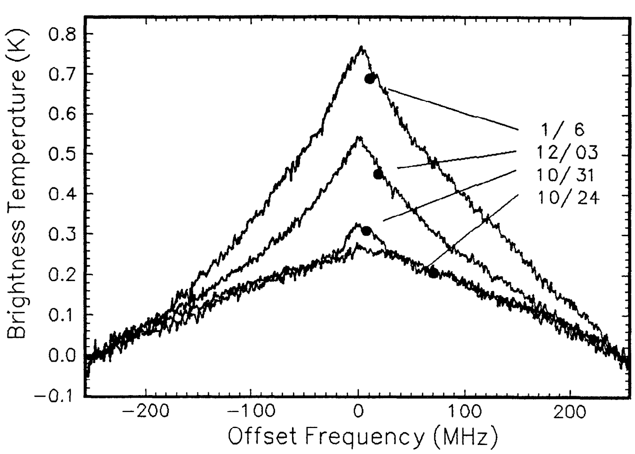

Figure 6. Measured millimeter wave spectra of N_2O for different dates during the breakdown of the polar vortex displayed as in Figure 1.

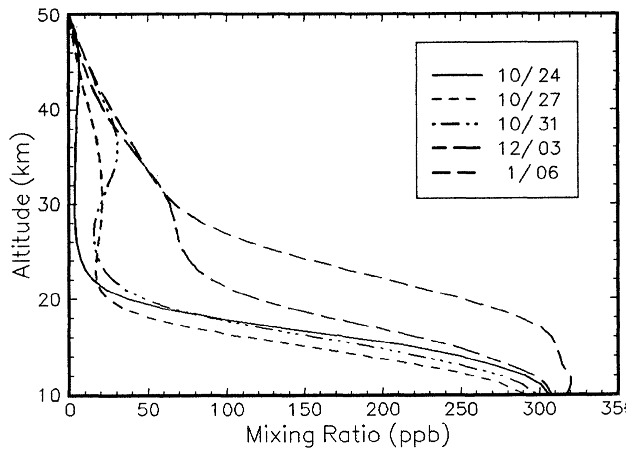

Figure 7. N_2O profiles retrieved from the spectra shown in Figure 6.

model by *Rosenfield et al.* [1994] in which vortex averaged descent rates were calculated using a radiation model together with NMC temperatures for the Antarctic winter 1987 and 1992 over the time period from March 1 to October 31. Because of the small difference in their results for both years which was also seen by *Manney et al.* [1994c] we justify a comparison with our 1993 data. Furthermore, we compare our local measurements in the heart of the polar vortex with a vortex average. In the model, air parcels initialized in March show the same behaviour (their Figure 8) with potential temperature over the polar winter as the N_2O mixing ratio isopleths shown here in Figure 5: both model and data suggest that most of the subsidence (more than 70 %) has already occurred in austral fall by the beginning of June. Encouraged by this qualitative good agreement, we use Figure 2 for a more quantitative comparison. The total descent from March to the end of October given in Table 2 shows excellent agreement, with clearly increasing numbers at higher altitudes from both data sets: air parcels located in the mid-stratosphere (34 km) in March descended more than 10 km during the 8-month period while parcels located in the lower stratosphere (18 km) underwent a much smaller descent of 3-4 km.

In order to look more closely at the relation of diabatic descent versus altitude, we compare our data again with *Rosenfield et al.* [1994]. Figure 8 shows the mean descent rates expressed in degrees per day for different potential temperature levels for both model and experiment over the whole winter from March to October. We have chosen to display results in these units to point on the nearly linear relation between potential temperature and cooling rate. Only the lowest data point of the millimeter wave measurements, at ~ 400 K, departs

Table 2. Descent from March to October Derived From Figure 2 (Geometrical and Diabatic) and Model Results by *Rosenfield et al.* [1994]

Starting Altitude, km	Millimeter Wave		Model
	Geometrical	Diabatic*	
18	4	4	3
25	7	5.5	5-7
34	13	10	10-12

Descent is measured in kilometers.

* corrected for the change in potential temperature with altitude.

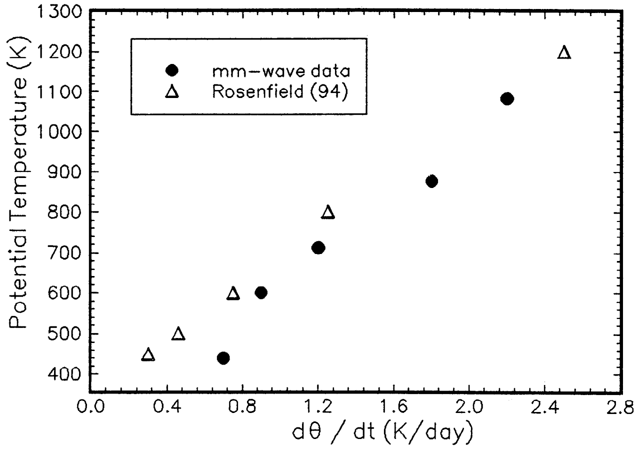

Figure 8. Comparison of mean descent rates over the period March 1 to October 31 derived from millimeter wave measurements over the South Pole 1993 compared with model results by *Rosenfield et al.* [1994] for the southern winter 1992.

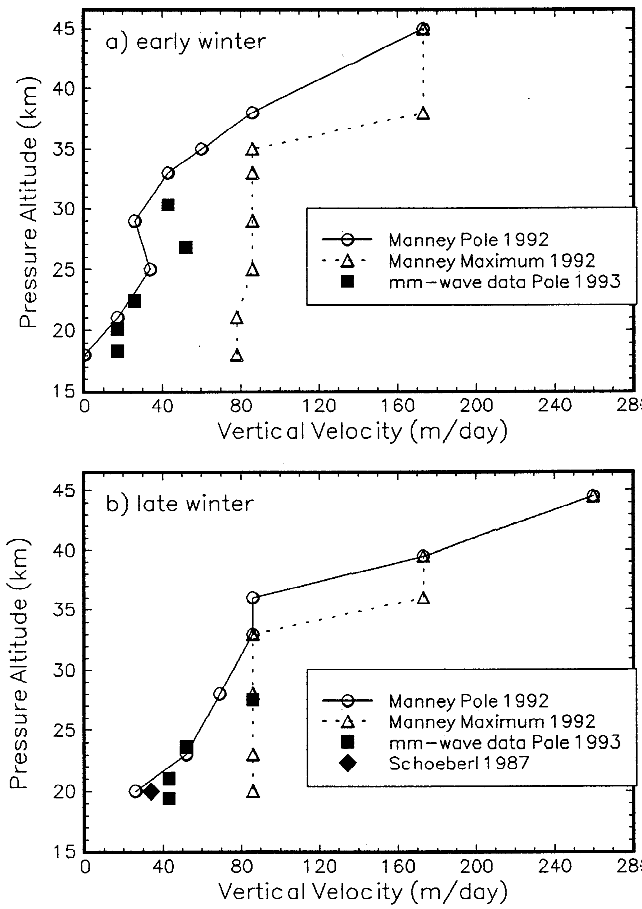

Figure 9. Comparison of measured descent rates over the South Pole 1993 with model results by *Manney et al.* [1994] for a) "early" and b) "late winter" 1992 as defined in Figure 5. The model results are shown for the South Pole and the maximum rate inside the vortex. The data are shown on a log-pressure scale using z=7[km]·ln (1000/p).

a bit off the line, where the error is largely due to the low altitude. Also, at that altitude the vortex can become much weaker and lateral transport may be more effective in altering the relationship prevailing at higher altitudes. From Figure 8, it can also be seen that the average cooling rates required for the vortex to act as a flowing processor, namely 1.7-2.5 K/d [*Tuck*, 1989, *Rosenfield et al.*, 1994] can only be observed in the upper stratosphere and not in the lower stratosphere as *Tuck* [1989] has proposed.

We next consider the model results of *Manney et al.* [1994c] in which trajectories for approximately 40,000 parcels in each run were used to calculate descent rates for "early" and "late winter" in the southern vortex of 1992. As can be seen from Figure 5, the times covered in that study do not include the period of strongest subsidence in the austral fall. From their three-dimensional results they derived the strongest descent rates in the lower stratosphere close to the vortex edge with lower values at the pole. Figure 9 gives mean descent rates for different altitudes from *Manney et al.* during "early" and "late winter" calculated for the pole as well as for the maximum descent inside the vortex, compared with those derived from our South Pole profiles. The model and the data show also a significant increase in descent rate with altitude for both "early" and "late winter" period. Maximum model descent rates were about the same for both periods, while at the pole the model results as well as our data increase slightly from "early" to "late winter". Above 30 km altitude, very little can be concluded from our data during these periods because the N_2O mixing ratio has become too low to allow reliable conclusions to be drawn. Generally, our South Pole data are closer to the values given for the pole by *Manney et al.* than the ones given for the maximum descent rate, and considering the relatively large uncertainty in both data and model, the agreement is very good.

In Figure 9, the descent rate at 20 km is also shown, as derived for the vortex interior by *Schoeberl et al.* [1992] for August to September 1987 from the AAOE flights, which went to about 72° S. This value (0.04 cm/s or 0.25 K/d) is close to our experimental data from the pole and is much lower than the results from *Manney et al.* for maximum descent, which they believe to be close to the vortex edge. It should be mentioned, however, that Figure 9 shows results of three different years, and some of the differences may be attributable to year-to-year variability.

Conclusions

A unique time series of the vertical N_2O distribution measured at about 3-day intervals has been obtained with a millimeter wave spectrometer operated for nearly a year at the South Pole. The descent rates derived from the downward shift of the N_2O profile have shown a strong variation from the polar fall to winter and for different altitudes. Comparing these data over the same time intervals and altitudes with recent model results [*Manney et al.*, 1994c; *Rosenfield et al.*, 1994] gives excellent agreement. The model of *Rosenfield et al.* [1994] shows the strongest vortex averaged subsidence as occurring in the very early state of the vortex in austral fall, as does our data. During March and April we derived a mean descent rate of ~100 m/d at 35 km, which decreases with altitude to ~20 m/d at 20 km. A comparison with the model by *Manney et al.* [1994c] for "early" and "late winter" gives good agreement between the model values calculated for the pole and our data, both showing rates lower than 50 m/d below 25 km.

Regarding the question of whether the vortex could be a flowing processor, our data show mean diabatic descent rates which are much lower than the ones required for the "flowing processor" theory [*Randel*, 1993]. These rates should be about 1.2-1.4 K/d at 16 km for the vortex average, and values this large could only be observed during April when winter subsidence started. As temporal mean values, they could only be observed in the upper stratosphere at altitudes above ~28 km (800 K).

The millimeter wave receiver has recently (February 1995) been set up at the South Pole again and we hope to obtain a similar data set for 1995 in order to get information on the year-to-year variability.

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