GEOPHYSICAL RESEARCH LETTERS, VOL. 28, NO. 12, PAGES 2385-2388, JUNE 15, 2001

Stratospheric NO_x enhancements in the southern hemisphere vortex in winter/spring of 2000

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Abstract. POAM III data show unusually large increases in stratospheric NO_2 throughout the late winter and spring at high southern latitudes during 2000. Using HALOE CH_4 data as a tracer of vertical descent, we conclude that excess NO_x was created by particle impacts in the upper atmosphere and descended in the polar vortex during the winter. We speculate that these NO_x enhancements were due to the solar proton event that occurred on 14-15 July 2000, and show that they caused reductions of up to ~45% in middle stratospheric ozone mixing ratios. Comparison of HALOE and POAM data in 2000 to data from 1991-1999 suggests that the 2000 NO_x enhancements were the largest ever documented by satellite in the southern hemisphere middle stratosphere. Also, based on H_2O data, we conclude that NO_x-enriched air observed in the south polar vortex from 1991-1999 originated in the mesosphere, not the thermosphere as is often assumed.

Introduction

It has long been suggested that NO_x ($NO+NO_2$) created in the mesosphere or thermosphere via energetic particle impacts can descend in the polar night to the stratosphere [e.g., *Solomon et al.*, 1982]. In recent years, satellite observations have shown that this is a fairly regular occurrence in the southern hemisphere (SH) polar stratospheric vortex during late winter and spring [*Callis et al.*, 1996; *Rinsland et al.*, 1996; *Randall et al.*, 1998; *Siskind et al.*, 2000 (hereafter referred to as R98 and S00, respectively)]. The Polar Ozone and Aerosol Measurement (POAM) instrument is ideally suited for studying this phenomenon, since it makes measurements of NO_2 in the SH polar vortex throughout the austral winter and spring. In this paper we use POAM and Halogen Occultation Experiment (HALOE) data to show that in 2000, the annual NO_2 enhancement was at its highest level since UARS was launched. This was most likely because of a solar proton event (SPE) that occurred in July and created additional NO_x at or above the stratopause.

POAM and HALOE are both solar occultation instruments, making ~15 measurements around a circle of nearly constant latitude on each day in each hemisphere. HALOE covers latitudes from ~75°S to 75°N in ~40 days, whereas POAM measurements are made from 55-72°N and 63-88°S. In this analysis we use version 6.0 POAM II data [R98], version 3.0 POAM III data [*Lucke et al.*, 1999; *Randall et al.*, 2001], and version 19 HALOE data (http://haloedata.larc.nasa.gov/). POAM SH measurements occur at local sunset (ss) from mid-September through mid-March, and at local sunrise (sr) from mid-March through mid-September. HALOE measurements are made at high southern latitudes in the late winter and early spring at both local sr and ss (each at different times for roughly 10-day periods). However, to avoid complications from the diurnal sequestration of NO_x in the N_2O_5 reservoir, we primarily use data acquired at local ss.

Paper number 2000GL012746.
0094-8276/01/2000GL012746$05.00

Results

Analysis of POAM III data follows the procedures in R98. Figure 1 shows the POAM III daily average NO_2 mixing ratios at 850 K (~28 km) inside the inner edge of the SH vortex (in-V), as defined by *Nash et al.* [1996]. All data with potential sunspot or aerosol artifacts were omitted [*Randall et al.*, 2001]. NO_2 in 2000 reached values two to three times higher than in 1998 or 1999. The POAM III data are also compared to POAM II data in 1994. R98 showed that due to creation of NO_x in the upper atmosphere, and subsequent descent to the stratosphere, in-V NO_2 in 1994 was significantly higher than in 1995-1996. Figure 1 indicates that at its maximum, the NO_2 observed in 2000 exceeds that in 1994 by about 50%.

Figure 2 compares two-week average NO_2 profiles for all in-V points from 1998-2000 and 1994. In all time periods, the NO_2 in 1998 and 1999 is lower than in 2000. By early September, NO_2 above 900 K in 2000 is more than a factor of two higher than in 1998 and 1999. NO_2 enhancements were observed in the middle stratosphere in 1994 earlier than in 2000, so NO_2 in 2000 is generally lower than in 1994 in August and early September. By the end of September, however, NO_2 from ~700-1100 K increased to levels exceeding those in 1994, and remained higher than in 1994 throughout October. Peak NO_2 in 2000 is about 40-50% higher than in 1994.

Clearly, in-V NO_2 was significantly higher at POAM latitudes in 2000 than in any other year during which POAM measurements have been made (late 1993-1996, 1998-2000). Two mechanisms which can result in the larger 2000 NO_2 enhancements are mixing of NO_x-rich air from lower latitudes, and descent of NO_x-rich air from higher altitudes. To separate these two effects, we have examined HALOE measurements of CH_4. In the wintertime polar stratosphere,

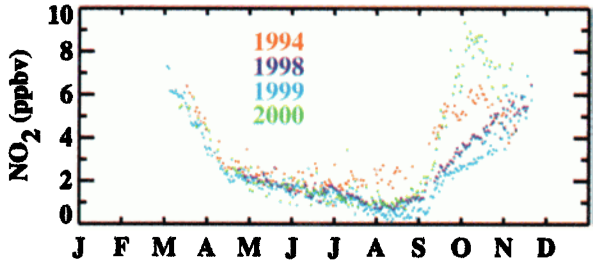

Figure 1. In-V POAM III NO_2 at 850 K, 1998-2000. The POAM III data are compared to POAM II data from 1994, which has been scaled up by a factor of 1.15 to account for the low bias in POAM II NO_2 measurements (relative to HALOE [R98]).

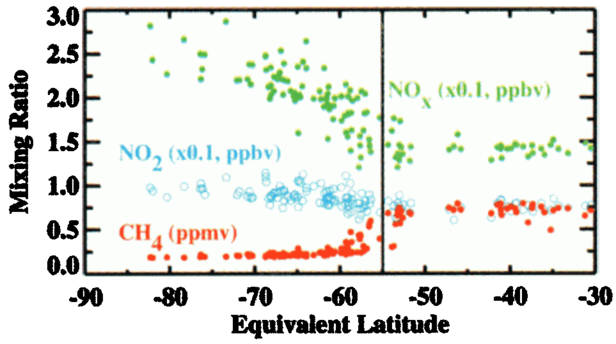

Figure 3. HALOE sunset CH_4, NO_x, and NO_2 at 1000 K from September 4-13, 2000, vs. Equivalent Latitude [Butchart and Remsberg, 1986]. HALOE was measuring at geographic latitudes between 50°S and 62°S at this time. The average inner edge of the Nash vortex is indicated by the vertical line (~55°S).

CH_4 mixing ratios less than ~0.3 ppmv indicate the presence of air from above the stratopause. This characteristic has been used previously to confirm the descent of NO_x-rich air to the stratosphere [e.g., *Callis et al.*, 1996; S00]. We plot HALOE measurements at latitudes from 50-62°S in early September, 2000 in Figure 3. At this time, NO_x measured by HALOE at 1000 K (~33 km) increased dramatically in the vortex, where low values of CH_4 indicate the descent of NO_x-enriched air from the mesosphere; increases in NO_2 are also apparent. The HALOE measurements cannot be directly compared to the POAM data since POAM was at latitudes between 82°S and 87°S at this time, and a significant latitudinal gradient in NO_2 exists because of different solar exposure. Nevertheless, since the POAM equivalent latitudes were between about 60°S and 90°S in early September, we conclude from Figure 3 that the air in which POAM observed the NO_2 enhancements descended inside the vortex from above the stratopause. In the absence of NO_x enhancements due to the impact of energetic particles in the upper atmosphere, NO_x and NO_2 are generally lower in the mesosphere than in the middle and upper stratosphere. That descent of air in the stratospheric polar vortex results in increasing NO_2 implies a particle-induced source.

Interest in transport of enhanced NO_x to the stratosphere lies primarily in the fact that the main loss mechanism of O_3 in the middle stratosphere is the NO_x catalytic cycle. Figure 4 shows that in early to mid September, in-V O_3 at 1000 K

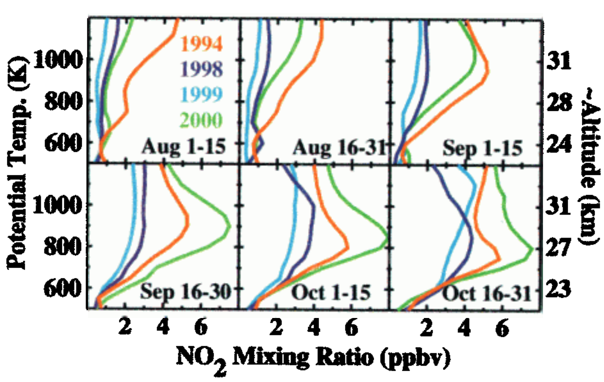

Figure 2. Two-week average in-V NO_2 profiles during 1994 (POAM II, scaled by a factor of 1.15) and 1998-2000 (POAM III).

reached values near 2.2 ppmv in 2000, compared to 4 ppmv in 1999. This suggests that the NO_x enhancements in 2000 caused a reduction of about 40-45% in O_3 mixing ratios. The situation changes at lower altitudes. For instance, O_3 at 700 K (24 km) in early October of 2000 is larger than in 1999 by about 12%, even though NO_2 at this altitude and time is about twice as high in 2000. This may be due to interference with the ClO or BrO ozone loss cycles in 2000 because of the excess NO_2 [e.g., *Jackman et al.*, 2000a], but this hypothesis requires more study.

HALOE data allow late winter/early spring NO_x enhancements to be examined back to 1991. However, interannual comparisons using HALOE data are complicated by a number of issues. First, the HALOE latitude coverage varies from year to year. HALOE ss measurements occurred at high southern latitudes in late October of 1991, and have gradually progressed such that in 2000 they occurred in early September. At the same time, the most poleward latitude sampled progressed from ~80°S in 1991 to ~62°S in 2000. Second, the timing of the production of NO_x in the upper atmosphere by energetic particle impacts, as well as descent rates in the polar region can vary from year to year, changing the altitude at which the NO_x enhancements will appear at any given time during the year. Thus, seasonal, latitudinal and altitude variations in NO_x (e.g., variations in NO_x/NO_y) must be considered. Figure 5a shows profiles of NO_x measured by HALOE at high southern latitudes for the time periods and average latitudes indicated, including only in-V measurements. This figure strongly suggests a larger enhancement in 2000 than in all other years, but because of the complications listed above, it is not definitive. S00 showed that from 1991-1996, the maximum column enhancements in in-V NO_x occurred in 1991. Fortuitously, the POAM measurements in late October of 2000 occurred at approximately the same latitudes as the HALOE measurements made in late October of 1991. The in-V POAM NO_2 concentrations in late October of 2000 are significantly higher than the 1991 HALOE data above 22 km, with peak differences of about 30% at 25 km.

Discussion

The results above show that the NO_2 enhancements observed by POAM in 2000 were caused by the descent of

NO_x-rich air inside the SH vortex, and were larger than in any year since HALOE was launched. There are two possible reasons for this: particle impacts and dynamics. When investigating the interannual variability in NO_x enhancements from 1991-1996, R98 and S00 showed reasonable correlations with the A_p index. The average A_p index in June and July of 2000 was higher than in the corresponding month for any other year since 1992, and the maximum daily A_p index in July of 2000 was higher than in any other July since 1991. Furthermore, *Jackman et al.* [2000b] have shown that the SPE that occurred on July 14-15, 2000 produced more NO_y in the upper atmosphere than any other SPE since 1989. While we cannot separate out magnetospheric (electrons) and solar (protons) sources of NO from the POAM data, these particle results are consistent with the large NO_x enhancements observed in 2000.

Another factor contributing to interannual variability in the observed NO_x enhancements is the amount of mixing which occurs as air descends into the stratosphere. Figure 5b shows the CH_4 profiles corresponding to the NO_x profiles plotted in Figure 5a. Based on the differences in these profiles, the years 2000 and 1992 stand out as anomalous years in terms of dynamical effects above ~900 K (30 km). Even compared to 1999, the year closest to 2000 in terms of measurement parameters, CH_4 above 900 K is relatively low. Mixing with low-latitude air will increase CH_4, so this observation suggests that descent occurred with less mixing in 2000 than in previous years. Plots of HALOE CH_4 vs. equivalent latitude (not shown) confirm that the vortex edge is better defined (less mixing) in 2000 than in 1991-1999. More mixing in 1991-1999 will contribute to the smaller NO_x enhancements observed in these years: lower-latitude, mesospheric air will be NO_x-poor because of the presence of sunlight, and will dilute the NO_x-rich air. S00 showed that smaller NO_x enhancements were observed in the SH vortex in 1992 than were expected based on the A_p index. Figure 5b suggests that this may have been due to more mixing in 1992, consistent with *Kawamoto and Shiotani* [2000].

We now discuss the altitude at which particle impacts first produced the NO_x that descended to the stratosphere in 1991-2000. It has been suggested that both mesospheric and thermospheric processes can affect stratospheric NO_y, but seldom are these effects differentiated [e.g.,*Solomon et al.*, 1982; *Russell et al.*, 1984; *Callis et al.*, 1996]. At issue, in the case of electron impact production, is the energy of the dissociating and ionizing electrons. It is well known that relatively low energy (< 10 keV) electrons associated with the visible optical aurora produce copious amounts of NO above 100 km. Recently *Codrescu et al.* [1997] have shown that medium energy electrons (MEE, 10-100 keV) produce substantial NO in the 65-95 km region (see also *Callis et al.* [1998]). In principle, one could use CH_4 to define the altitude from which the NO_x-rich air descends. But its altitude gradient in the mesosphere is very small, so mixing can obscure any signature due to thermospheric air. On the other hand, H_2O, another tracer, exhibits a sharp gradient above 70 km, so it should be a more sensitive indicator of this initial altitude. On average, high-latitude, late-fall HALOE H_2O maximizes from ~40-60 km near 5-6 ppmv, then rapidly decreases to 2 ppmv or less by 80 km. At altitudes above 80 km, we expect the H_2O to fall to near zero. There is no indication in Figure 5c, which shows HALOE in-V H_2O, that this steep decrease was mirrored in the descending air that reached the stratosphere. Thus, we cannot definitively identify any air parcels which originated in the upper mesosphere (or higher).

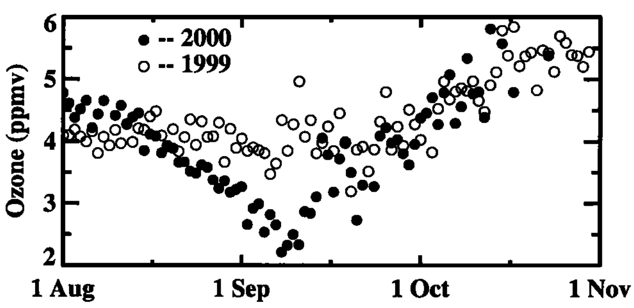

Figure 4. POAM III In-V ozone at 1000 K (31-32 km).

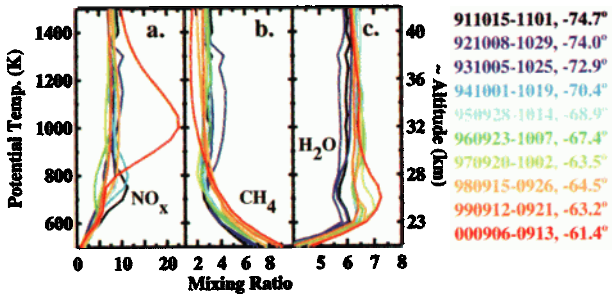

Figure 5. Average in-V HALOE NO_x (ppbv, a), CH_4 x10 (ppmv, b), and H_2O (ppmv, c) profiles for the time periods and latitudes shown.

Assuming that air originating in the lower thermosphere would preserve some signature of low H_2O, these results show that the NO_x-rich air observed in the SH vortex in 1991-2000 originated in the mesosphere, not the thermosphere. This is consistent with descent rate calculations relevant to the polar vortex [*Fischer et al.*, 1993, *Bacmeister et al.*, 1995]. NO_x enhancements of mesospheric origin are expected in 2000, based on the peak energy deposition altitude estimated by *Jackman et al.* [2000b] for the July SPE. In other years when SPEs may not have played a role, our conclusions have direct implications for the initial energy of the particles responsible for the NO_x enhancements [e.g., *Callis*, 1997]. They imply that the initial altitude for the NO production is at the lower end of the possible range defined by observations of the mesosphere and lower thermosphere. This suggests that the MEE population identified by *Codrescu et al.* [1997] is responsible for the stratospheric NO_x enhancements discussed here. Note that even with this conclusion, significant mixing must still have occurred to dilute the low H_2O values present where the NO was first produced. *Summers et al.* [1997a] discuss this possibility.

To summarize, large NO_x enhancements were observed by POAM and HALOE in September through October of 2000 in the SH polar vortex. The excess NO_x caused ozone loss of up to 45% at 1000 K (33 km) in early September. We conclude that the enhancements were due to high NO_x production by particle impacts in the mesosphere, such as the large SPE on 14-15 July, 2000. The NO_x enhancements in the SH polar vortex are the largest observed since the launch of UARS in 1991. It is likely that the NO_x enhancements observed in the 2000 SH polar vortex were the largest since the very large SPE in August of 1972 [*Jackman et al.*, 2000a]. The only other stronger SPE that has occurred since 1972 was in October of 1989 [*Jackman et al.*, 2000b], so it would not have affected the SH vortex. We have used H_2O to investigate the altitude of the source region for SH stratospheric NO_x enhancements observed by satellite. We conclude that in every year since 1991, NO_x enhancements observed in the SH polar vortex resulted from mesospheric NO_x production.

Acknowledgments. We thank C. Jackman, D. Rusch and M. Schoeberl for useful discussions, and A. Kochenash for calculating equivalent latitudes. POAM data are available at the NASA Langley Research Center EOSDIS DAAC. This work was supported by NASA ACMAP grant NAG1-2187 and the NASA data buy program.

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(Received December 7, 2000; revised March 23, 2001; accepted April 4, 2001.)