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UARS Microwave Limb Sounder Observations of Denitrification and Ozone Loss in the 2000 Arctic Late Winter

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Abstract. The UARS Microwave Limb Sounder obtained measurements of ClO, HNO_3, and O_3 inside the Arctic lower stratospheric vortex during two intervals in February and March 2000. The data show evidence of significant chemical processing in February, consistent with the exceptionally cold conditions that prevailed earlier in the winter. Ozone at 465 K decreased by 0.04 ± 0.01 ppmv/day in early February, implying chemical loss rates greater than those observed during the 1995–1996 winter, which was also unusually cold in the lower stratosphere. A persistent depression in the HNO_3 abundances in late March, well after polar stratospheric cloud activity had ceased, suggests a moderate degree ($\sim$20%) of denitrification around the 465 K level at 70°N and higher equivalent latitudes. This is the strongest evidence yet seen for the occurrence of denitrification in the Arctic over spatial scales large enough to be detected in satellite measurements.

1. Introduction

After a series of northern winters during the mid-1990s characterized by below-average stratospheric temperatures [*Pawson and Naujokat*, 1999], the 1997–1998 and 1998–1999 winters were mild, with comparatively little ozone loss [*Braathen et al.*, 2000]. During the 1999–2000 winter, the Arctic once again experienced exceptionally cold conditions, with temperatures below 195 K, the approximate threshold for polar stratospheric cloud (PSC) formation, persisting over a larger area in the lower stratosphere for a longer period of time than during any of the previous 20 years [*Manney and Sabutis*, 2000; *Braathen et al.*, 2000]. The Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS), which had not been operated in more than 6 months, was turned on to measure ClO, HNO_3, and O_3 inside the Arctic vortex during two late-winter periods: 2–13 February and 27–29 March 2000. These MLS data, although spatially and temporally more limited than those from previous Arctic winters, cover a substantial portion of the Arctic vortex (both horizontally and vertically) and show significant chemical processing and denitrification consistent with the meteorological conditions. The MLS observations supplement data from aircraft, balloon and ground-based campaigns conducted this winter.

2. Measurement Description

MLS began measurements of the stratosphere in late September 1991. After several years in orbit the antenna scan mechanism started to exhibit signs of wear; performance degraded over time and the instrument was finally placed in "standby" mode in July 1999. MLS was not powered up again until February 2000, at which time it was operated in a mode such that limb scans were attempted only poleward of 50°N on the "day" side of the orbit, to maximize the probability of obtaining useful measurements in the sunlit polar vortex. Because power from the UARS spacecraft was limited, only the 205-GHz radiometer, providing the primary measurements of ClO, HNO_3, and O_3, was turned on. No MLS antenna scan slips occurred during either observing period. However, a large fraction of the good limb scans were lost during the February interval because problems with the UARS tape recorder forced reliance on real-time data collection, which favors certain geographical regions while excluding others. Data coverage was significantly improved during the March interval by increasing utilization of the onboard tape recorder, scheduling longer communications with the data relay satellites, and performing limb scans on both sides of the orbit.

Results shown here are from the recently-released version 5 MLS retrieval algorithms. Estimated vertical resolutions and single-profile precisions at the levels in the lower stratosphere of interest in this study are about 6 km, 4 km, and 4 km and 1.0 ppbv, 0.3 ppbv, and 0.3 ppmv for HNO_3, ClO, and O_3, respectively. For the averages shown here, uncertainties are typically lower by a factor of 5–10. Further details of the quality of the MLS version 5 data are available from the MLS web site (http://mls.jpl.nasa.gov).

3. Observations and Discussion

Because the data dropouts did not occur in the same locations every day, coverage of the vortex can be extended by averaging several days of data. Fig. 1 shows MLS maps of HNO_3, ClO, and O_3 interpolated to the 465 K potential temperature surface using United Kingdom Meteorological Office (UKMO) temperatures [*Swinbank and O'Neill*, 1994] and averaged over 3-day intervals at the beginning (2–4 February) and near the end (9–11 February) of the first observing period and during the second (27–29 March) ob-

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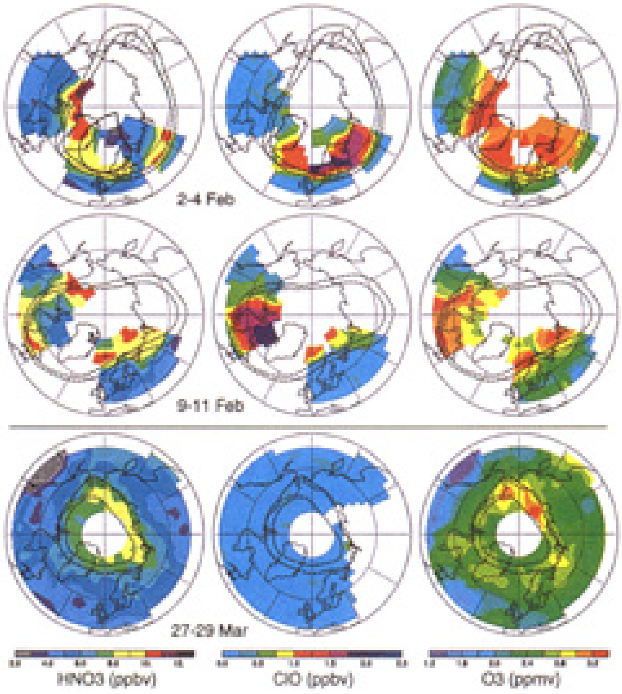

Figure 1. Maps of MLS HNO_3, ClO, and O_3 at 465 K averaged over the periods (top) 2–4 February, (middle) 9–11 February, and (bottom) 27–29 March 2000. The orthographic projections start at 40°N, with a thin dashed black circle at 60°N. Blank spaces represent data gaps. Two contours of UKMO PV (thick dashed black lines) indicate the size and strength of the polar vortex. The solid black line on the HNO_3 maps is the 195 K UKMO temperature contour.

serving period. Contours of UKMO temperature and potential vorticity (PV) are also shown.

By the time MLS began measurements in February, 465-K minimum temperatures had been continuously below 195 K for nearly two months [*Manney and Sabutis*, 2000]. In the absence of chemical processing, unmixed diabatic descent leads to HNO_3 and O_3 mixing ratios that are high throughout the vortex and have strong gradients across its boundary in the lower stratosphere. The early February map shows evidence of gas-phase HNO_3 depletion inside of and just downstream from the low-temperature region, indicating the presence of PSCs. Consistent with activation on PSCs, enhanced ClO filled most of the sunlit portion of the vortex where data coverage extended. Temperatures increased slightly (<5 K) and the low HNO_3 and high ClO abundances shifted position as the vortex sloshed around between the first and second halves of the observing period. Back trajectory calculations (not shown) indicate that the air parcels in the vicinity of the highest ClO over northeastern Canada on 9 February originated from the area of ClO enhancement over northern Russia about a week earlier. Comparison of the maps suggests a reduction in 465-K O_3 mixing ratios inside the vortex by the end of the February observing period.

In mid-February temperatures again dropped well below PSC existence thresholds and remained low until the final warming began in early March [*Manney and Sabutis*, 2000]. By the time of the MLS observing period in late March, although a fairly sizeable vortex remnant still persisted in the lower stratosphere, the barrier to mixing was much weaker (as evidenced by the PV gradient in the maps) and intrusion of extravortex air likely occurred. The decrease in HNO_3 from the previous month probably resulted primarily from greater photolysis as sunlight levels increased together with enhanced mixing of vortex and lower-latitude air [*Santee et al.*, 1999]. However, as discussed below, denitrification may also have contributed to maintaining low HNO_3 concentrations after PSC formation ceased. Although the significant cold spell after the first MLS observing period probably promoted additional PSC processing and O_3 depletion, by late March chlorine had been completely deactivated. Since O_3 mixing ratios were, on average, lower outside the vortex than inside, even in late March, the mixing-in of extravortex air may also have reduced vortex O_3 abundances; thus the observed O_3 decrease between February and March may not have resulted solely from chemical destruction.

Averages of MLS ClO in equivalent latitude/potential temperature (EqL/θ) space over the 3-day intervals in February 2000 (Fig. 2) indicate a smaller vertical range for significant ClO enhancement than was observed in February of either 1996 or 1997 [*Santee et al.*, 1997], when enhanced ClO values extended up to ~650 K. Although earlier in the 1999–2000 winter the area of low temperatures had been much more extensive throughout the lower stratosphere than in previous years, by the time of the MLS observations in February the cold region had all but disappeared above 585 K [*Manney and Sabutis*, 2000], and any ClO that had been present at these levels had apparently already been deactivated. The ClO gradients across the vortex edge appear much weaker in the early February interval than in the later interval or in previous years; however, sensitivity tests on the 1996 data indicate that the weaker gradients in February 2000 may be a consequence of the sparser data coverage.

EqL/θ representations of MLS HNO_3 are shown in Fig. 3. One caveat in interpreting these plots is that the gridding procedure smears out short-lived, highly localized phenomena such as PSC events, with more smoothing occurring as more points are averaged. As with ClO, some of the features seen (or not) in the HNO_3 plots may be artifacts of the data sampling. The regions of low HNO_3 at high EqL in early February coincide with regions of low temperature (not shown) and are therefore likely associated with HNO_3 sequestration in PSCs. Temperatures were higher during the

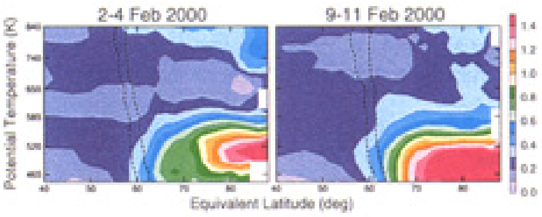

Figure 2. Equivalent latitude/potential temperature (EqL/θ) space representation of MLS ClO (in ppbv) averaged over the 3-day intervals in February shown in Fig. 1. (EqL is the latitude enclosing the same area as a given PV contour.) The dashed lines are contours of UKMO scaled PV.

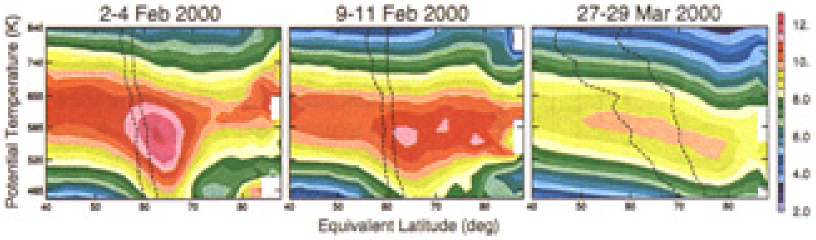

Figure 3. As in Fig. 2, for MLS HNO_3 (in ppbv) averaged over the 3-day intervals shown in Fig. 1.

mid-February interval, and some recovery in HNO_3 is evident; that HNO_3 values remained depressed over the region from 70–90° EqL below 520 K indicates either that some lingering PSCs had not yet fully evaporated or that some HNO_3 was permanently removed. A signature of HNO_3 depletion is still apparent around 465 K at the highest EqLs in late March. Not only were stratospheric minimum temperatures unusually low during the 1999–2000 winter, but the area of temperatures below 195 K was unusually large and enduring [*Manney and Sabutis*, 2000]. Thus air masses were exposed to PSC conditions for longer periods, increasing the potential for denitrification [*Tabazadeh et al.*, 2000]. Similar springtime persistence of low HNO_3 abundances is seen in the 1997 MLS data (not shown). Evidence for denitrification in 1997 was found in air masses sampled in February [*Kondo et al.*, 2000] and April [*Rex et al.*, 1999]. We conclude that the MLS HNO_3 data from the 2000 late-winter period suggest moderate denitrification over a fairly large area (of course local denitrification on spatial scales below those distinguishable by MLS may have been much larger).

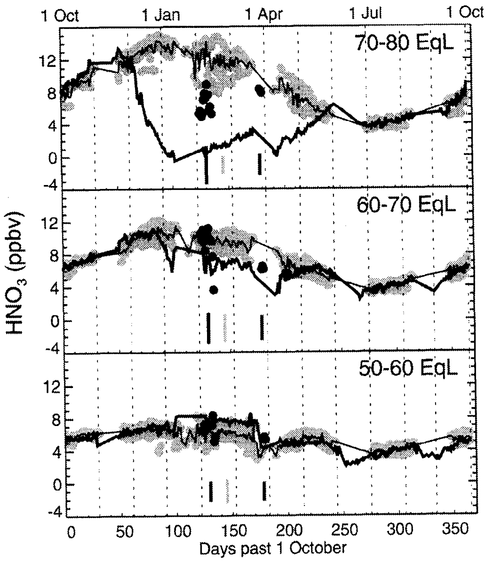

Figure 4. Time series of MLS HNO_3 at 465 K binned into 10° EqL bands and averaged. Black circles represent daily averages for the 2000 observing periods; grey circles represent values for the previous Arctic winters observed by MLS (1991–92 to 1998–99, zonal means for the first 6 years are depicted in separate colors in *Santee et al.* [1999]). Thick vertical lines at the bottom of each panel denote typical error bars for the February and March 2000 data and for the February data from previous years. Overall averages calculated from the daily values in the previous 8 years are indicated by the thin black line. For comparison, overall daily averages calculated from the southern hemisphere data points in each year are overlaid as a thick black line (shifted by 6 months so that comparable seasons are aligned). Dashed vertical lines demark calendar months.

This view is supported by examining daily HNO_3 averages at 465 K computed in several EqL regions (Fig. 4). We focus on the 465-K level since it is in the center of the persistent depression in high-latitude HNO_3; note that averages at different times represent different air masses because of the effects of diabatic descent. The vertical resolution of the MLS data is not sufficient to pinpoint the altitude of maximum denitrification. Although HNO_3 abundances at northern high latitudes exhibit a large degree of interannual variability, on most of the MLS measurement days in February 2000 the HNO_3 mixing ratios in the 70–80° EqL region were considerably lower than those observed in any of the previous 8 years. Since MLS never before observed northern high latitudes in late March, it is impossible to know with certainty whether the 2000 values are exceptional; nevertheless, that the March 2000 points fall below the line connecting the averages from the adjacent periods of coverage (even when the error bars are taken into account) is suggestive of a modest amount ($\sim$20%) of permanent removal at 70–80° EqL. Whereas at 465 K the 60–70° EqL region was inside the vortex or near its edge in February, it was largely outside the vortex in late March (compare the PV contours overlaid in Fig. 3). The March 2000 points also appear to deviate from the average behavior in this EqL range, possibly indicating that denitrification occurred here as well. Alternatively, the below-average values in this region may have arisen through mixing as the vortex decayed with parcels that experienced denitrification at higher EqLs to such a degree that their HNO_3 concentrations were reduced below typical extravor-

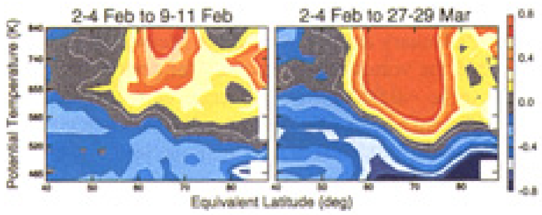

Figure 5. Change in MLS O_3 (in ppmv) between (left) the first and second intervals in February and (right) the first interval in February and the March interval, in EqL/θ space.

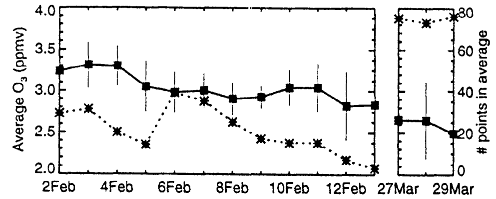

Figure 6. Time series of 465-K daily averages (solid squares) of MLS O_3 measurements obtained inside the vortex. The error bars represent the standard deviations in the averages. Also shown (asterisks) are the number of individual measurements contributing to the average each day.

tex values. In contrast, the 2000 data in the 50–60° EqL range, representing air that was consistently outside the vortex throughout the winter, follow the patterns observed in previous years. Taken together, Figs. 3 and 4 provide the strongest evidence yet seen for the occurrence of Arctic denitrification over spatial scales large enough to be detected in MLS measurements, although the HNO_3 removal is not nearly so severe or widespread as in the Antarctic.

The changes in MLS O_3 between the beginning and end of the February observing period and between the beginning of February and the end of March are compared in Fig. 5. Significant O_3 depletion occurred over the brief interval in February throughout the vortex below ~520 K, with the peak in the O_3 loss at high EqLs at ~465 K. By late March the O_3 values had decreased substantially below 520 K. The pattern of net O_3 decreases at the lowest levels in the stratosphere and net increases above (as the balance shifts between chemical destruction and replenishment via diabatic descent) is consistent with those seen in the MLS data in previous years [*Manney et al.*, 1997]. Although conditions were conducive to chemical destruction of O_3 until mid-March [*Manney and Sabutis*, 2000], after that time mixing processes as the vortex eroded may have contributed to the observed O_3 decrease at 60–70° EqL. Fig. 5 indicates that extravortex O_3 also declined throughout most of the lower stratosphere. Similar behavior was observed in 1997 (but not in previous winters) [*Manney et al.*, 1997].

Daily averages of the O_3 measurements obtained inside the vortex at 465 K are shown in Fig. 6. These are not true vortex averages because of the limited data coverage. A line fit through the O_3 daily averages from February reveals a decreasing trend of 0.04 ± 0.01 ppmv/day. (Because mixing with extravortex air may have significantly affected the March O_3 abundances, they were not included in the fit.) In February transport processes increase O_3 at 465 K and thus mask chemical removal. For the (longer) period between 29 January and 3 March 1996, *Manney et al.* [1996] estimated the vortex-averaged chemical destruction rate at 465 K to be ~0.022 ppmv/day, based on an estimated O_3 replenishment rate of ~0.005 ppmv/day. Assuming that diabatic descent was of similar magnitude in the two years, the 2000 MLS data imply O_3 loss rates greater than those observed during the 1995–1996 winter.

The fact that MLS detected for the first time moderate denitrification over a sizeable fraction of the Arctic vortex, accompanied by large O_3 losses, during a year in which stratospheric temperatures remained lower over a larger area than previously observed, has important implications. Although the exact mechanisms have not been identified, *Tabazadeh et al.* [2000] have shown that exposing air masses to temperatures below PSC formation thresholds for longer periods of time promotes denitrification. In a future possibly colder and more humid lower stratosphere, widespread severe denitrification could significantly enhance Arctic O_3 loss [*Tabazadeh et al.*, 2000]. Thus the results of this study underscore the importance of continued stratospheric monitoring through the EOS Aura (formerly known as CHEM, scheduled to begin operations in 2003) era, when stratospheric chlorine levels will still be high and the frequency and duration of PSC formation may be increasing.

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