Polar processing and development of the 2004 Antarctic ozone hole: First results from MLS on Aura

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[1] The Microwave Limb Sounder (MLS) on Aura is providing an extensive data set on stratospheric winter polar processing, including the first daily global observations of HCl, together with simultaneous measurements of ClO, HNO_3, H_2O, O_3, N_2O, and temperature (among others). We present first results charting the evolution of these quantities during the 2004 Antarctic late winter. MLS observations of chlorine deactivation and ozone loss during this period are shown to be consistent with results from the SLIMCAT chemical transport model. **Citation:** Santee, M. L., G. L. Manney, N. J. Livesey, L. Froidevaux, I. A. MacKenzie, H. C. Pumphrey, W. G. Read, M. J. Schwartz, J. W. Waters, and R. S. Harwood (2005), Polar processing and development of the 2004 Antarctic ozone hole: First results from MLS on Aura, *Geophys. Res. Lett.*, *32*, L12817, doi:10.1029/2005GL022582.

1. Introduction

[2] Aura, NASA's latest Earth Observing System (EOS) satellite [*Schoeberl et al.*, 2004], was launched 15 July 2004. One of its four instruments is the Microwave Limb Sounder (MLS), a greatly enhanced follow-on to the MLS instrument onboard the Upper Atmosphere Research Satellite (UARS) [e.g., *Waters et al.*, 1999]. EOS MLS improvements include better horizontal resolution, better precision and/or vertical resolution, and continuous global coverage (from 82°N to 82°S on every orbit). Perhaps most importantly, EOS MLS makes several measurements not available from UARS MLS, including the first daily global HCl profiles. Other EOS MLS observations relevant to polar process studies include ClO, HNO_3, H_2O, O_3, N_2O (also a new measurement for MLS) and temperature (T). Aura launched in time for MLS to observe the 2004 Antarctic late winter. Here we present first results from this period, with a focus on chlorine deactivation and ozone loss. Because of yaw maneuvers and other data gaps, UARS MLS never captured the complete chlorine deactivation period in the Antarctic. Studies using UARS data were also hampered by the lack of colocated ClO and HCl data, since MLS and the Halogen Occultation Experiment did not sample inside the Antarctic polar vortex simultaneously [*Douglass et al.*, 1995; *Santee et al.*, 1996].

[3] MLS data shown here are preliminary; refinements and extensive validation efforts are underway. Provisional estimated accuracies are about 2 K, 30 ppbv, 2 ppbv, 1 ppmv, 0.2 ppbv, 0.2 ppbv and 0.5 ppmv for T, N_2O, HNO_3, H_2O, HCl, ClO, and O_3, respectively; all have vertical resolution of 3–5 km.

2. The 2004 Antarctic Late Winter

[4] Although lower stratospheric minimum temperatures were below average over most of the 2004 Antarctic winter, they rose and remained near average values after MLS began science operations in mid-August (Figure 1). The lower stratosphere warmed rapidly in September, halting further heterogeneous processing of vortex air by the end of the month.

[5] An overview of the changes over the 2004 Antarctic late winter in key polar processing parameters measured by MLS at 520 K potential temperature (~21 km) is shown in Figure 2. The late-winter evolution of T, HNO_3, HCl, ClO, and O_3 at 550 K (~22 km) is presented in an animation provided as auxiliary material[1]. By 13 August, the first full day that MLS was operated in science mode, confined diabatic descent inside the polar vortex has led to low values of the long-lived tracer N_2O, but high values of species such as HNO_3, H_2O, HCl, and O_3, whose concentrations increase with altitude in the lower stratosphere. In the coldest part of the vortex, however, gas-phase HNO_3 and H_2O have been substantially depleted as polar stratospheric clouds (PSCs) have formed. Heterogeneous chemical reactions on PSC particle surfaces convert chlorine from reservoir species, such as HCl, into active forms, primarily ClO. Virtually all vortex HCl has been depleted, leading to significantly enhanced ClO. However, because measurements on the "day" (ascending) side of the orbit are made in late afternoon (~15:00–18:00 local solar time) at high southern latitudes, MLS is not capturing maximum ClO abundances, which occur around midday. ClO is not enhanced in the vortex core, which is still in polar night. ClO is also not enhanced along the vortex edge, a characteristic pattern at 520 K seen in all southern hemisphere winters observed by UARS MLS [*Santee et al.*, 2003]. Finally, O_3 is significantly depleted in a ring roughly coinciding with the sunlit area of high ClO; this is consistent with studies showing that O_3 loss propagates poleward with the terminator, with little mixing between the vortex edge and core at this time [e.g., *Lee et al.*, 2000].

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[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2005GL022582.

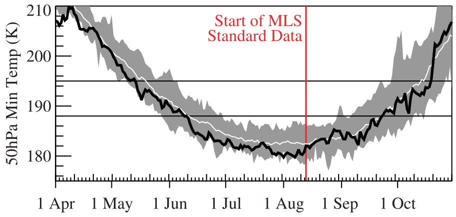

Figure 1. 50 hPa minimum temperatures poleward of 40°S from the National Centers for Environmental Prediction/Climate Prediction Center analyses during the 2004 Antarctic winter (black line). Grey shading shows the range over 1979–2003, with the white line the average. Horizontal lines denote approximate thresholds for polar stratospheric clouds.

[6] The region cold enough for PSCs has shrunk substantially by 6 September, and HNO_3 and H_2O have partially recovered. Some PSCs may have evaporated and returned HNO_3 and H_2O to the gas phase; continuing descent also contributes to the observed increases. HCl remains very low throughout the vortex, and ClO has increased in the vortex core with exposure to daylight. Ozone depletion has progressed, since mixing ratios have decreased despite replenishment by descent. By 12 days later, on 18 September, chlorine partitioning has changed considerably: HCl has started to recover over most of the vortex, and ClO is much less enhanced.

[7] The decrease in N_2O along the vortex edge by 1 October is attributable to continuing descent in this region [e.g., *Manney et al.*, 1994]. Even though temperatures have risen above PSC thresholds, HNO_3 remains depleted (and does not recover substantially as long as the lower stratospheric vortex stays intact), implying the occurrence of denitrification. No signature of widespread dehydration is seen at this level. Similar differences in the springtime distributions of HNO_3 and H_2O have been observed previously [e.g., *Manney et al.*, 1999]. Moreover, measurements from Polar Ozone and Aerosol Measurement III also indicate atypically large stratospheric H_2O abundances during this winter (G. Nedoluha, personal communication, 2004). Lastly, chlorine has been almost completely deactivated, with only a hint of enhanced ClO, and large HCl mixing ratios throughout the vortex.

[8] The vertical extent of polar processing is shown in Figure 3. On 6 September, during the time of peak chlorine activation, almost no HCl remains inside the vortex below ~600 K. Very good correspondence is seen in the vertical extent of depleted HCl and enhanced ClO. Depletion in gas-phase HNO_3 (not shown), indicating PSC activity, extends over a similar vertical range. That ClO enhancement more completely fills the vortex at lower altitudes is consistent with behavior seen in UARS MLS ClO data [*Santee et al.*, 2003]. The dip in O_3 contours across the vortex edge in the lower stratosphere arises in part because in the southern hemisphere descent is strongest in this region [e.g., *Manney et al.*, 1994, 1999]. By 1 October HCl has recovered, nearly

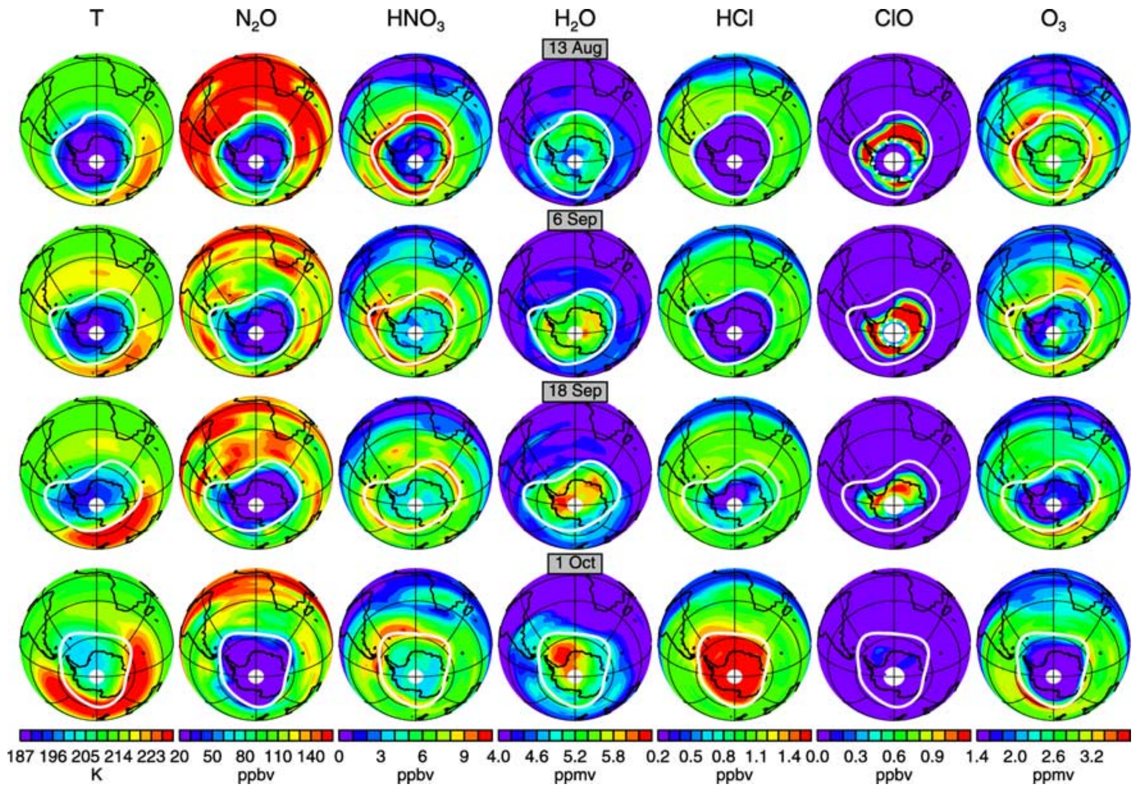

Figure 2. Maps of MLS T, N_2O, HNO_3, H_2O, HCl, ClO, and O_3 for selected days during the 2004 Antarctic late winter, interpolated to 520 K using Global Modeling and Assimilation Office Goddard Earth Observing System (GEOS-4) temperatures. Solid white lines show the 0.5 × 10^{-4} K m^2 kg^{-1} s^{-1} GEOS-4 potential vorticity (PV) contour, approximating the polar vortex boundary. Only data from the "day" (ascending) side of the orbit are shown for ClO; the dashed white circle on ClO maps demarks the edge of daylight.

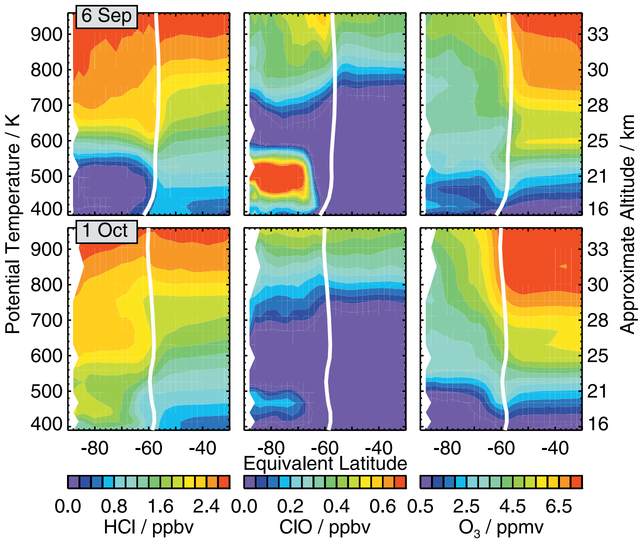

Figure 3. Equivalent latitude (EqL, the latitude enclosing the same area between it and the pole as a given PV contour)/potential temperature cross sections of MLS HCl, ClO, and O_3. Only ascending data are used for ClO. A contour of scaled PV [e.g., *Manney et al.*, 1994] outlines the vortex edge (white line).

all ClO has disappeared above 500 K (although weak enhancement persists at lower altitudes), and the ozone hole has developed.

[9] Figure 4 shows changes in MLS HCl, ClO, and O_3 at 520 K over the late-winter period in two equivalent latitude (EqL) bands representing the vortex interior and edge regions. In the vortex interior, ClO continues to increase until early September, reflecting the change in solar zenith angle. In mid-September ClO declines and HCl increases steeply. Rapid ozone loss begins in late August and levels off in early October. In contrast, near the vortex edge ClO is relatively constant in mid-August, decreasing thereafter. HCl recovery starts earlier and is more gradual than at higher EqLs. The timing of the recovery, with almost all active chlorine converted to HCl by mid-October, agrees well with that seen previously in ground-based infrared column measurements [*Liu et al.*, 1992; *Kreher et al.*, 1996]. Ozone loss, which most likely started in this region well before mid-August [e.g., *Lee et al.*, 2000], appears to be less severe than that in the vortex interior, although it is also being compensated to a greater extent here by stronger descent [e.g., *Manney et al.*, 1994, 1995].

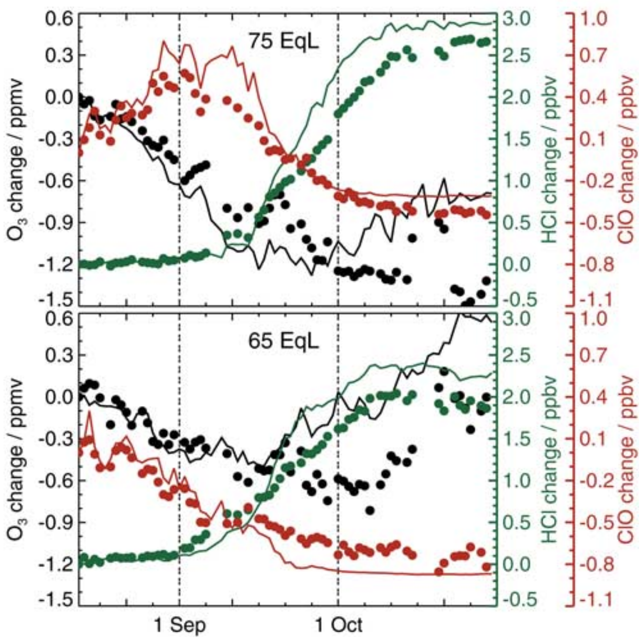

Figure 4. Changes in MLS HCl, ClO, and O_3 at 520 K over the late-winter period (filled circles). Daily averages were calculated in 5°-wide EqL bands, and values for the initial day were then subtracted. Day-night differences are shown for ClO. Symbols mark days for which retrievals are available; MLS made measurements on most days in this period, but not all have been run through the data processing system. Also shown (solid lines) are results from the SLIMCAT model, sampled at the MLS measurement locations/times.

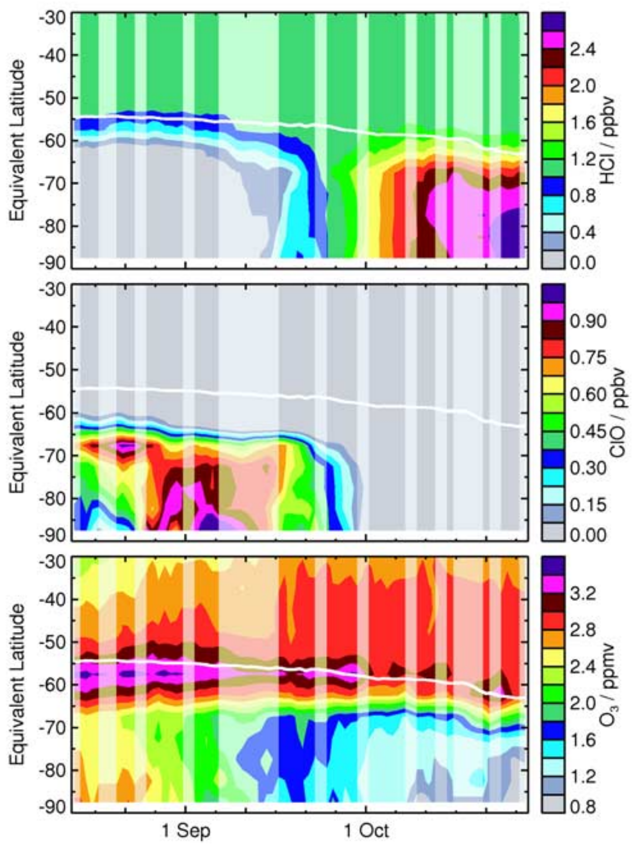

Figure 5. Time series of MLS HCl, ClO, and O_3 versus EqL at 520 K. Only ascending data are used for ClO. To fill in data gaps, Kalman smoothing has been applied to the daily averages; paler colors denote regions where the estimated precision of the interpolated values is poor [see, e.g., *Santee et al.*, 2003]. A PV contour (white line) demarks the vortex edge.

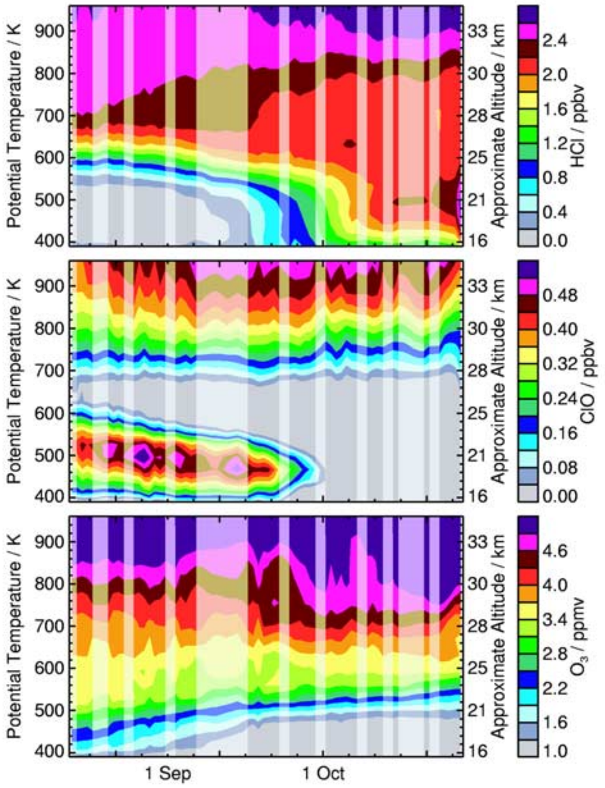

Figure 6. Time series of vortex-averaged (calculated within the 1.4×10^{-4} s^{-1} contour of scaled PV) MLS HCl, ClO, and O_3 versus potential temperature. Only ascending data are used for ClO.

[10] Figure 4 also shows results from the SLIMCAT chemical transport model [*Chipperfield*, 1999] forced by meteorological fields from the U.K. Met Office. The model used a spectral resolution of T42, with 50 isentropic levels from near the tropopause to ~60 km. For each MLS measurement an equivalent sample, interpolated to the same location, was taken from the model at the nearest available time (within 15 min). Overall agreement between measured and modeled values is very good, especially for the first half of the period. Although variations in MLS ClO are generally reproduced well, the model attains larger peak abundances in the vortex interior; model HCl at both EqLs also recovers to larger values in the spring than those measured by MLS. These discrepancies, to be investigated in future studies, may arise from uncertainties in MLS or meteorological data and/or deficiencies in model chemistry. Measured and modeled O_3 decreases match quite well until late September; the divergence thereafter is likely related to strong sensitivity to dynamical effects in spring, when vertical transport slows or reverses and the vortex weakens.

[11] A broader view is given in Figure 5, which shows that the spatial and temporal extent of chlorine activation are consistent in the ClO and HCl fields. Inside the vortex, O_3 decreases steadily. That O_3 continues to drop at the highest EqLs even after chlorine deactivation may indicate the onset of diabatic ascent in the vortex core [e.g., *Manney et al.*, 1994]. MLS H_2O and HNO_3 data (not shown) are also consistent with significant ascent in the vortex core by mid-October.

[12] Figure 6 shows that the vertical extent of ClO enhancement is consistent with that of HCl depletion, as is its decay over time. Figure 6 also shows the late-winter downward progression of the ClO profile peak, seen previously in UARS MLS [*Santee et al.*, 2003] and ground-based [*de Zafra et al.*, 1995; *Solomon et al.*, 2002] measurements. O_3 loss is clearly visible up to ~580 K but likely extends to higher altitudes; studies have indicated that chemical destruction takes place up to ~650 K but is at least partially compensated by the effects of descent above 520 K [e.g., *Manney et al.*, 1995]. SLIMCAT results (not shown) exhibit qualitatively very similar behavior.

[13] MLS on Aura is providing a much more complete picture of polar processing than was previously available from any single set of simultaneous measurements. MLS observations of chlorine deactivation and ozone loss have been shown to be consistent with results from the SLIMCAT model during the 2004 Antarctic late winter.

[14] **Acknowledgments.** Thanks to Y. Jiang and R. P. Thurstans for their help in producing the MLS animation, S. A. Sena for making Figure 1, and M. P. Chipperfield for developing and sharing the SLIMCAT model. Work at the Jet Propulsion Laboratory, California Institute of Technology, was done under contract with the National Aeronautics and Space Administration.

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