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## Interhemispheric Differences in Polar Stratospheric $\text{HNO}_3$ , $\text{H}_2\text{O}$ , $\text{ClO}$ , and $\text{O}_3$

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# Interhemispheric Differences in Polar Stratospheric $\text{HNO}_3$ , $\text{H}_2\text{O}$ , $\text{ClO}$ , and $\text{O}_3$

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Simultaneous global measurements of nitric acid ( $\text{HNO}_3$ ), water ( $\text{H}_2\text{O}$ ), chlorine monoxide ( $\text{ClO}$ ), and ozone ( $\text{O}_3$ ) in the stratosphere have been obtained over complete annual cycles in both hemispheres by the Microwave Limb Sounder on the Upper Atmosphere Research Satellite. A sizeable decrease in gas-phase  $\text{HNO}_3$  was evident in the lower stratospheric vortex over Antarctica by early June 1992, followed by a significant reduction in gas-phase  $\text{H}_2\text{O}$  after mid-July. By mid-August, near the time of peak  $\text{ClO}$ , abundances of gas-phase  $\text{HNO}_3$  and  $\text{H}_2\text{O}$  were extremely low. The concentrations of  $\text{HNO}_3$  and  $\text{H}_2\text{O}$  over Antarctica remained depressed into November, well after temperatures in the lower stratosphere had risen above the evaporation threshold for polar stratospheric clouds, implying that denitrification and dehydration had occurred. No large decreases in either gas-phase  $\text{HNO}_3$  or  $\text{H}_2\text{O}$  were observed in the 1992–1993 Arctic winter vortex. Although  $\text{ClO}$  was enhanced over the Arctic as it was over the Antarctic, Arctic  $\text{O}_3$  depletion was substantially smaller than that over Antarctica. A major factor currently limiting the formation of an Arctic ozone “hole” is the lack of denitrification in the northern polar vortex, but future cooling of the lower stratosphere could lead to more intense denitrification and consequently larger losses of Arctic ozone.

The severe depletion of stratospheric ozone over Antarctica (1) in late winter and early spring (the so-called Antarctic ozone “hole”) is now known (2) to be caused by chlorine chemistry. Recently, there has been heightened concern about the possibility of a similar feature appearing in the Northern Hemisphere as the abundance of chlorine in the stratosphere continues to increase. Abundances of reactive chlorine, in the form of  $\text{ClO}$ , comparable to those over Antarctica (3) have been observed throughout the Arctic vortex (4), but the accompanying loss of  $\text{O}_3$  over the Arctic has been much less severe (5) than that over Antarctica. The presence of  $\text{HNO}_3$  affects the cumulative amount of chlorine-catalyzed  $\text{O}_3$  destruction in two major ways. First, in the low temperatures of polar winter,  $\text{HNO}_3$  condenses to form type I nitric acid trihydrate (NAT) polar stratospheric clouds (PSCs), which provide surfaces for the heterogeneous activation of chlorine. Second, photolysis of  $\text{HNO}_3$  vapor releases nitrogen dioxide ( $\text{NO}_2$ ), leading to chlorine deactivation through the formation of the reservoir species chlorine nitrate ( $\text{ClONO}_2$ ). Type II water ice PSCs, which form if temperatures drop below the frost point, can also incorporate  $\text{HNO}_3$  vapor releases nitrogen dioxide ( $\text{NO}_2$ ), leading to chlorine deactivation through the formation of the reservoir species chlorine nitrate ( $\text{ClONO}_2$ ). Type II water ice PSCs, which form if temperatures drop below the frost point, can also incorporate  $\text{HNO}_3$  vapor releases nitrogen dioxide ( $\text{NO}_2$ ), leading to chlorine deactivation through the formation of the reservoir species chlorine nitrate ( $\text{ClONO}_2$ ).

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ments of  $\text{HNO}_3$  have an estimated precision and absolute accuracy of  $\sim 2$  parts per billion by volume (ppbv) and  $\sim 15\%$ , respectively, at 46 hPa, and of  $\sim 2$  ppbv and  $\sim 50\%$ , respectively, at 100 hPa, and a vertical resolution of  $\sim 5$  km. The Cryogenic Limb Array Etalon Spectrometer (CLAES) on UARS also measures  $\text{HNO}_3$  (16). Initial comparisons of MLS and CLAES lower stratospheric  $\text{HNO}_3$  show good agreement, generally to better than 30% at 46 hPa (15).

The evolution of MLS  $\text{ClO}$ ,  $\text{O}_3$ ,  $\text{HNO}_3$ , and  $\text{H}_2\text{O}$  in the Southern Hemisphere in 1992 is illustrated in Fig. 1. These particular days were selected to represent conditions before, during, and after the period in which stratospheric temperatures were low enough for PSCs. The MLS data are displayed on an isentropic [constant potential temperature (17)] surface to remove the effects of adiabatic vertical motions. We show the 465 K surface ( $\sim 19$  km) near the level of maximum  $\text{O}_3$  depletion (12); surfaces at 520 K ( $\sim 21$  km) and 585 K ( $\sim 24$  km) have also been examined and exhibit similar behavior. Steep gradients in potential vorticity (PV) (18) on an isentropic surface demarcate the boundary of the polar vortex, so the proximity of the two PV contours in the maps indicates vortex strength. The southern polar vortex is already well established by 28 April. Relatively large concentrations of  $\text{HNO}_3$ ,  $\text{O}_3$ , and  $\text{H}_2\text{O}$  are evident in the vortex interior at this time and can be attributed to the diabatic downward transport of air with higher mixing ratios of these species (19). Temperatures are still above the PSC formation threshold, and chlorine has not yet been activated.

Chlorine activation has begun by the time MLS looks south again (20) on 2 June, as evidenced by the large  $\text{ClO}$  values in the sunlit portion of the vortex. Abundances of  $\text{O}_3$  and  $\text{H}_2\text{O}$  inside the vortex have increased, consistent with continuing diabatic descent. Altitude changes in CLAES  $\text{N}_2\text{O}$  (21) mixing ratio contours between late April and mid-June confirm that descent is ongoing [ $\text{N}_2\text{O}$  is a dynamical tracer that develops horizontal gradients on isentropic surfaces in the presence of diabatic processes (22)]. Although diabatic descent is also expected to cause an increase in  $\text{HNO}_3$  abundances, a deficit in gas-phase  $\text{HNO}_3$  has developed, coincident with the region in which temperatures have fallen below the formation threshold (6) ( $\sim 195$  K) of NAT PSCs. Whether this deficit is indicative of the permanent removal of  $\text{HNO}_3$  from the lower stratosphere (denitrification) or just the temporary sequestration of gas-phase  $\text{HNO}_3$  in NAT PSCs cannot be ascertained from the MLS observations alone.

por (6, 7). Removal of gas-phase  $\text{HNO}_3$  from the lower stratosphere, either temporarily through condensation or permanently through the sedimentation of type I or type II PSC particles (denitrification), reduces the availability of  $\text{NO}_2$  and allows chlorine to remain activated. Measurements of gas-phase  $\text{HNO}_3$  have recently been obtained from the Microwave Limb Sounder (MLS) on the Upper Atmosphere Research Satellite (UARS) (8). Because MLS also provides data on  $\text{ClO}$ ,  $\text{O}_3$ , and  $\text{H}_2\text{O}$ , we now have a global set of simultaneous, commonly calibrated measurements directly addressing the correlations between denitrification, dehydration, chlorine activation and deactivation, and ozone destruction.

The MLS instrument (9) acquires stratospheric measurements that are not degraded by PSCs or aerosols (10); MLS observations of  $\text{ClO}$  (4, 11),  $\text{O}_3$  (4, 11–13), and  $\text{H}_2\text{O}$  (14) have been reported previously. Improved accuracy and an extended vertical range for the  $\text{H}_2\text{O}$  data reported here were achieved with a nonlinear, iterative retrieval scheme, yielding an estimated precision and absolute accuracy of  $\sim 0.2$  parts per million by volume (ppmv) and  $\sim 15\%$ , respectively, at 46 hPa (1 hPa = 100 Pa = 1 mbar), and of  $\sim 1.3$  ppmv and  $\sim 100\%$ , respectively, at 100 hPa. The measurement of  $\text{HNO}_3$ , a minor contributor in the 205-GHz spectral bands used to measure  $\text{O}_3$  and  $\text{ClO}$ , was a secondary MLS objective (10). Vertical profiles of gas-phase  $\text{HNO}_3$  are now being retrieved for the lower stratosphere and have improved the fits to the radiometric data (15). The MLS measure-

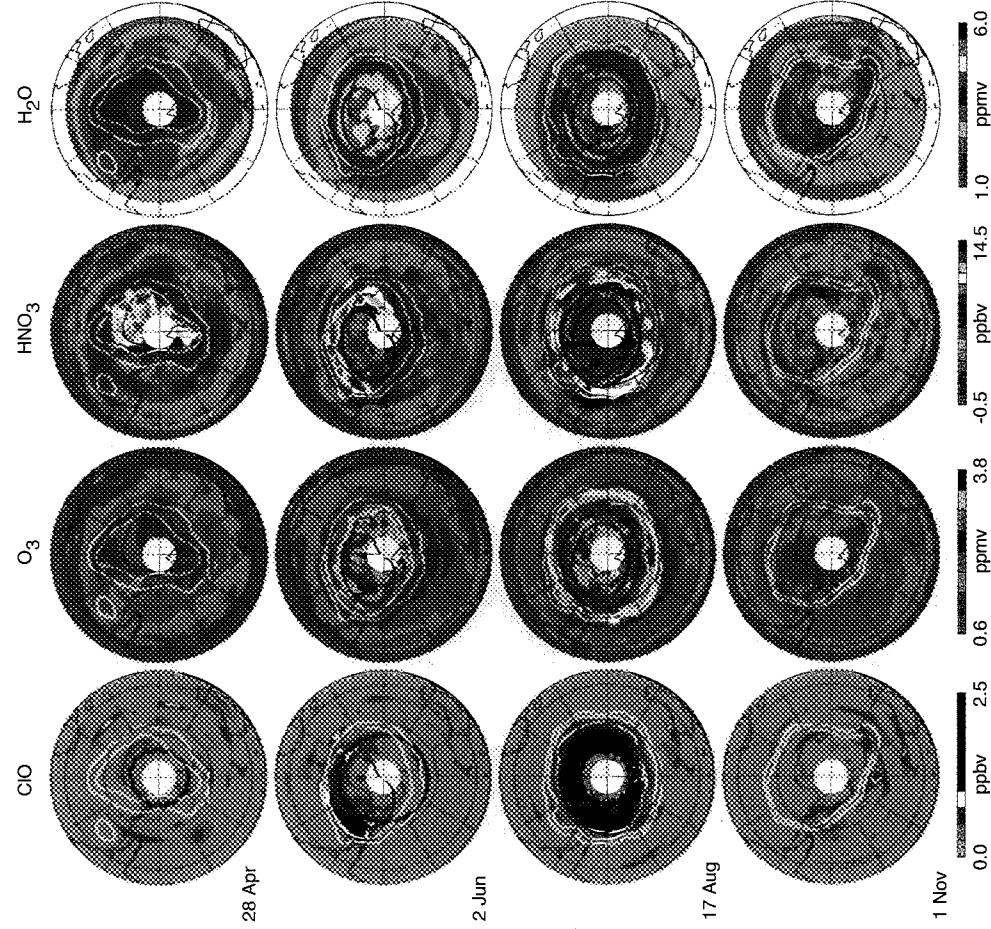
This is the earliest that  $\text{HNO}_3$  condensation has been observed in southern winter. Because temperatures remain above the frost point at this time, a comparable decrease in  $\text{H}_2\text{O}$  vapor is not expected and indeed is not present in the MLS data. Although in situ observations of correlated low concentrations of water vapor and total reactive nitrogen over Antarctica (23) have been taken to imply that the processes of denitrification and dehydration are coupled, other observations (24) have been interpreted to suggest that denitrification can occur without significant dehydration. Theoretical studies (7, 25) have found that the sedimentation rate of  $\text{HNO}_3$  particles can be sufficiently rapid—given low cooling rates or cooling to just below the NAT condensation point—for substantial  $\text{HNO}_3$  loss to take place over short time scales, independent of the formation of water ice clouds. On the basis of temperature data collected during the 1987 southern winter, Salawitch *et al.* (25) calculate that extensive denitrification could have occurred by the first or second week in June, with significant dehydration occurring later. The MLS instrument does not detect  $\text{HNO}_3$  or  $\text{H}_2\text{O}$  in condensed phase, but the significant decrease observed in gas-phase  $\text{HNO}_3$  without a corresponding decrease observed in gas-phase  $\text{H}_2\text{O}$  is a necessary (albeit not sufficient) condition for the decoupling of these processes.

The MLS observations on 17 August (11) show  $\text{ClO}$  values of  $\sim 2$  ppbv in most of the sunlit area poleward of  $60^\circ\text{S}$  and a ring of depleted  $\text{O}_3$  coincident with the largest  $\text{ClO}$  abundances. Temperatures are below the formation threshold (6) of type II PSCs over most of the Antarctic continent, and gas-phase  $\text{HNO}_3$  values are extremely low ( $\leq 2$  ppbv) throughout this region. A “collar” of high  $\text{HNO}_3$  ( $\geq 10$  ppbv) is seen just outside the low-temperature region, consistent with previous aircraft observations (23, 26) and colocated CLAES measurements (16). The outer boundary of enhanced  $\text{ClO}$  roughly corresponds to the inner edge of the  $\text{HNO}_3$  collar region, as expected from conversion of  $\text{ClO}$  into  $\text{ClONO}_2$  through reaction with  $\text{NO}_2$  supplied by  $\text{HNO}_3$  photolysis. Depletion of  $\text{H}_2\text{O}$  vapor has been in progress since at least shortly before the end of the previous MLS south-looking period in mid-July (27); by mid-August, there is extensive loss of  $\text{H}_2\text{O}$  vapor within the low-temperature zone, where values range from about 1.5 to 3.5 ppmv, in good agreement with earlier aircraft observations inside the vortex in August and September (28). Again, the MLS data do not distinguish between the temporary formation of ice clouds and the permanent removal of  $\text{HNO}_3$  or  $\text{H}_2\text{O}$ . However, NAT PSC particles can grow

large enough (7, 25) to experience rapid fallout, and the larger ice crystals of type II PSCs [which can also incorporate  $\text{HNO}_3$  (6, 7)] sediment more readily. It is therefore likely that the Antarctic lower stratosphere has undergone both denitrification and dehydration by this time.

There are no MLS measurements over Antarctica from mid-September through the end of October (20). By the time southwesterly resumes on 1 November, chlorine over Antarctica has been largely deactivated. However, the  $\text{O}_3$  deficit that developed in September (4, 11, 12) persists. The deficits in gas-phase  $\text{HNO}_3$  and  $\text{H}_2\text{O}$  also per-

sist, with mixing ratio values less than 6 ppbv and less than 3 ppmv, respectively, throughout most of the vortex. Similar  $\text{H}_2\text{O}$  values were measured by the UARS Halogen Occultation Experiment (HALOE) in mid-October 1992 (29). The strong PV gradient indicates that the vortex is still intact, inhibiting mixing between polar and mid-latitude air. Lower stratospheric temperatures rose above the NAT PSC formation threshold the last week in September (30). The fact that gas-phase  $\text{HNO}_3$  and  $\text{H}_2\text{O}$  values remain depressed long after the last PSCs would have been expected to evaporate strongly implies that irreversible re-



**Fig. 1.** Maps of MLS  $\text{HNO}_3$ ,  $\text{ClO}$ ,  $\text{O}_3$ , and  $\text{H}_2\text{O}$  for selected days during the 1992 southern winter, interpolated onto the 465 K potential temperature (17) surface with U.S. National Meteorological Center (NMC) temperatures. The maps are polar orthographic projections extending to the equator, with the Greenwich meridian at the top and black circles at  $30^\circ$  and  $60^\circ\text{S}$ . They were produced by linear interpolation of measurements taken over a 24-hour period. The nonlinear  $\text{H}_2\text{O}$  retrieval was performed poleward of  $30^\circ\text{S}$ , and no measurements were obtained in the white area poleward of  $80^\circ\text{S}$ . The thick black contours on the  $\text{ClO}$  maps identify the edge of daylight for the measurements; only data from the “day” side of the orbit are shown for  $\text{ClO}$  (34). Thin black lines show NMC temperature contours of 195 K (outer contour) and 188 K (inner contour), the approximate threshold temperatures (6) for type I and type II PSCs, respectively. Also superimposed (in white) on each of the maps are two contours of potential vorticity (PV) (18), calculated from NMC temperature and geopotential height fields. The outer contour ( $\sim 2.5 \times 10^{-5} \text{ K m}^2 \text{ kg}^{-1} \text{ s}^{-1}$ ) represents the approximate edge of the polar vortex during winter when it is well established (12); inclusion of the inner contour ( $\sim 3.0 \times 10^{-5} \text{ K m}^2 \text{ kg}^{-1} \text{ s}^{-1}$ ) provides information on the steepness of the PV gradient, indicating the strength of the vortex.

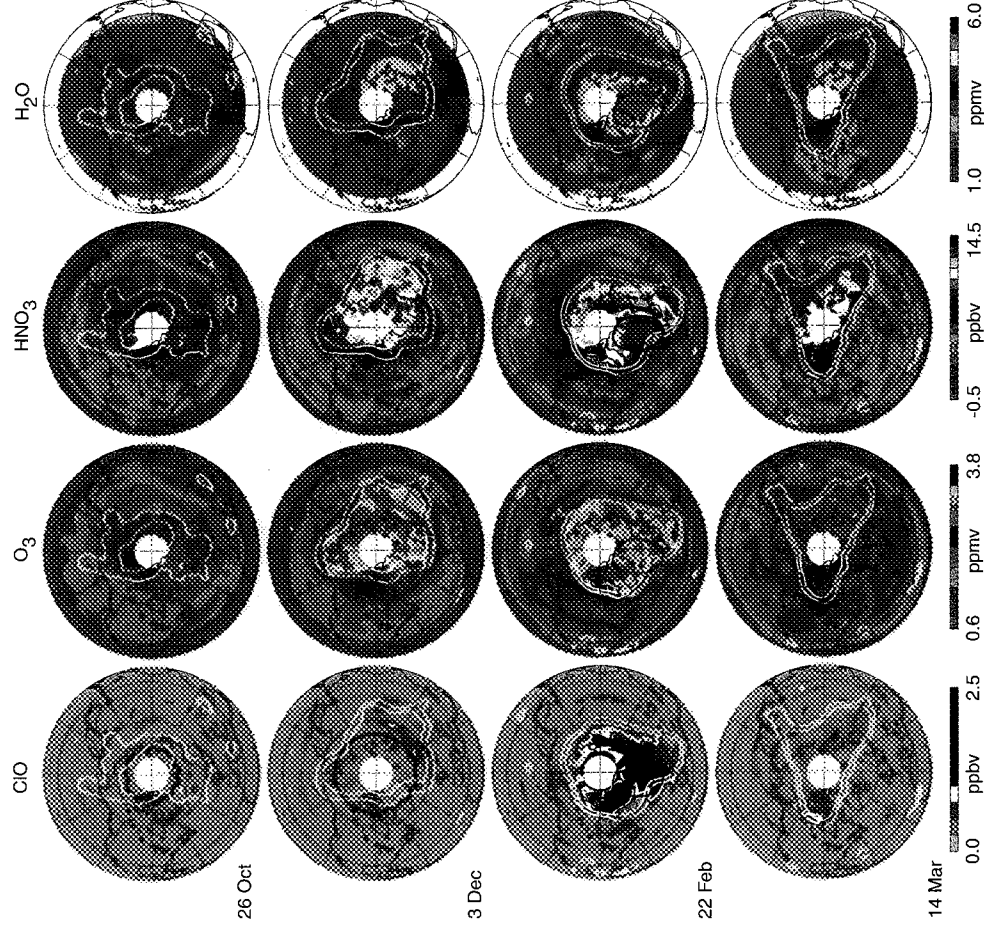
moval (denitrification and dehydration) occurred at this level.

This signature of denitrification and dehydration is not apparent in the 1992–1993 northern polar vortex (Fig. 2). In contrast to the Southern Hemisphere at the comparable season, on 26 October the northern vortex is not well established at 465 K. The PV gradient indicates that the vortex has increased substantially in size and strength by 3 December. Except for a small area over southern Finland, temperatures remain too high for PSCs, and ClO is not enhanced. Mixing ratios of  $O_3$ ,  $HNO_3$ , and  $H_2O$  are relatively large inside the vortex at 465 K, again because of diabatic descent, and there is no evidence that condensation of  $HNO_3$  has begun.

In late winter, there are striking differences between the hemispheres. Whereas the southern vortex in August is nearly centered over the pole, the northern vortex in February is distorted, with the southernmost portion covering most of Europe on 22 February. The ClO abundances are generally enhanced ( $>1$  ppbv) throughout the vortex, but  $O_3$  concentrations remain high,

suggesting that the replenishment by diabatic descent partly masks the destruction by chlorine chemistry (5). Between Greenland and Scandinavia, there is an area with temperatures below the NAT PSC threshold in which gas-phase  $HNO_3$  abundances are low (about 5 to 9 ppbv) relative to the remainder of the vortex (about 10 to 14 ppbv). This low-temperature, low- $HNO_3$  region persists for only a few days. There is no corresponding perturbation in  $O_3$  or ClO, implying that the sequestration of gas-phase  $HNO_3$  in NAT PSCs, rather than intrusion of lower latitude air or uplift of the isentropes, causes the observed  $HNO_3$  decrease. Compared to that for the Antarctic (Fig. 1), the Arctic reduction in gas-phase  $HNO_3$  is less intense, more localized, and more transient, indicating that the Arctic vortex has not undergone significant denitrification. In addition, no significant decrease is observed in gas-phase  $H_2O$ .

Northern Hemisphere winters exhibit a high degree of interannual variability, and the 1992–1993 lower stratospheric vortex is characterized by below-average temperatures and anomalously strong PV gradients



**Fig. 2.** As in Fig. 1, but for selected days during the 1992–1993 northern winter, with the Greenwich meridian at the bottom and positive PV contour values.

(31). Therefore, the decrease in gas-phase  $HNO_3$  in Fig. 2 is probably greater than that in most Arctic winters. By mid-March, after two strong stratospheric warnings (32), the high values of ClO have diminished. Ozone abundances are lower than in February, but the depletion is much less severe than that in the southern vortex between August and September (5). The continued presence of  $HNO_3$  throughout the Arctic winter moderates the destruction of  $O_3$  by providing a source of  $NO_2$  to quench ClO.

Under present climate conditions, Arctic winter temperatures remain below the type I PSC threshold sufficiently long for lower stratospheric chlorine to be substantially converted to reactive form. However, they do not fall low enough or stay low long enough for sedimentation by either type I or type II PSC particles to cause extensive denitrification. Therefore, although ClO is enhanced over the Arctic as it is over Antarctica, the formation of an ozone “hole” over the Arctic has so far been limited by the lack of permanent removal of  $HNO_3$ . Future cooling of the lower stratosphere (for example, caused by increases in greenhouse gases) could intensify the loss of  $HNO_3$  within the vortex and, while the chlorine loading remains high, lead to greater depletion of Arctic  $O_3$  (33).

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17. Potential temperature ( $\theta$ ), defined as the temperature an air parcel would have if it were expanded or compressed adiabatically to a pressure of 1000 hPa, is related to pressure  $p$  (in hPa) by  $\theta = T(1000/p)$ .

- $p)^{0.265}$ , where  $T$  is temperature in kelvin. Potential temperature is conserved under adiabatic conditions. For the low temperatures characteristic of the winter polar vortices,  $\Theta = 465$  K corresponds to about  $p = 50$  hPa and  $\Theta = 585$  K corresponds to about  $p = 22$  hPa.
18. Potential vorticity (PV) is defined as  $PV = -g(f + \zeta)/d\theta/dp$ , where  $g$  is the acceleration due to gravity,  $f$  is the Coriolis parameter,  $p$  is pressure,  $\theta$  is potential temperature ( $f\tau$ ), and  $\zeta$  is the component of relative vorticity orthogonal to the  $\theta$  surface. For adiabatic, frictionless flow, PV is conserved, and contours of PV on potential temperature surfaces comprise the same air parcels.
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35. We thank M. R. Schoeberl for encouraging us to pursue the MLS  $HNO_3$  measurement; A. E. Roche and the CLAES team for  $N_2O$  data and for discussions and comparisons of  $HNO_3$  data; National Meteorological Center (NMC) Climate Analysis Center personnel, especially M. E. Gelman and A. J. Miller, for making NMC data available to the UARS project; two anonymous referees for helpful comments; and our MLS colleagues, especially T. Lungu, R. P. Thurstans, and E. F. Fishbein. M.L.S. gratefully acknowledges a Resident Research Associateship from the National Research Council. The work at the Jet Propulsion Laboratory, California Institute of Technology, was sponsored by the National Aeronautics and Space Administration, and the work at Edinburgh University and Heriot-Watt University was sponsored by the Science and Engineering Research Council.

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