

MICROWAVE LIMBSOUNDER HNO₃ OBSERVATIONS: IMPLICATIONS FOR POLAR STRATOSPHERIC CLOUD COMPOSITIONS

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The catalytic destruction of O₃ in the polar lower stratosphere is initiated through heterogeneous chemical reactions on the surfaces of polar stratospheric cloud (PSC) particles. Accurate theoretical predictions of O₃ loss require knowledge of the phase and composition of these PSC particles. The canonical model of Type I PSCs [1] assumes, following thermodynamic stability arguments [2], that they are composed of solid nitric acid trihydrate (NAT) particles. Recently, however, several observational, laboratory, and modeling studies [3] have suggested a liquid HNO₃/H₂O (ternary) solution as a more likely state for the Type I PSC particles.

To gain insight into the issue of PSC composition, we compare model results and measurements of gas-phase HNO₃ made by the Microwave Limb Sounder (MLS) aboard the Upper Atmosphere Research Satellite (UARS) during the southern hemisphere winters of 1992 and 1994. During the MLS south-looking period in late May/early June 1994, temperatures were below the threshold for the onset of NAT PSCs (~195 K) for two full weeks before MLS observed any reduction in gas-phase HNO₃ at 465 K (on 7 June, as temperatures approached the ice frost point). Loss of gas-phase HNO₃ continued throughout the MLS south-looking period; by the beginning of July there was essentially no gas-phase HNO₃ remaining within the region of lowest temperatures. A time series of MLS HNO₃ mixing ratios averaged over the region of low temperatures on the 465 K surface is compared to predicted behavior assuming various models of PSC formation. Specifically, we use NMC temperatures, an assumed water vapor mixing ratio of 4.5 ppbv and the formulae of Hanson and Mauersberger [2] and Worsnop *et al.* [4] to calculate the saturation vapor pressures of HNO₃ over NAT and NAD (nitric acid dihydrate), respectively. Both sets of calculations predict low HNO₃ vapor concentrations that do not correspond well with the MLS HNO₃ observations. We also use an aerosol physical chemistry model (APCM) [5] for the ternary system, assuming 10 ppbv of HNO₃ and background sulfate aerosol conditions. In this case, excellent agreement is found between the MLS measurements of gas-phase HNO₃ and the HNO₃ partitioning predicted by the APCM assuming a ternary solution composition for the PSCs.

These results can be contrasted to MLS HNO₃ observations from 1992 (although the observing period is not exactly the same, data exist for 2 June in 1992 and 1994). In both years the HNO₃ values on 2 June are high in the portion of the vortex where temperatures are not low, due to descent of HNO₃-rich air from above. However, in 1992 there is a deficit in gas-phase HNO₃ coincident with the region where temperatures are below 195 K, conforming to the expected pattern for NAT PSC formation, whereas in 1994 HNO₃ values are high throughout the vortex, even inside the region of low temperatures. Trajectory calculations reveal a more rapid cooling rate for the parcels within the 195 K temperature contour in 1992 than in 1994 during the 20 days prior to 2 June. We are investigating the possible correlation between temperature history and PSC composition as inferred from MLS measurements of gas-phase HNO₃. We are also examining the differences between the 1992 and 1994 southern vortex MLS observations in relation to the changing sulfate aerosol loading as the stratosphere recovers from the Mt. Pinatubo eruption.

[1] e.g., Turco *et al.*, GRL 94, 16,493 (1989).

[2] Hanson and Mauersberger, GRL 15, 855 (1988).

[3] e.g., Zhang *et al.*, J. Phys. Chem. 97, 8541 (1993); Carslaw *et al.*, GRL 21, 2479 (1994); Drdla *et al.*, GRL 21, 2475 (1994); Tabazadeh *et al.*, GRL 21, 1619 (1994); Stefanutti *et al.*, GRL 22, 2377 (1995); Toon and Tolbert, Nature 375, 218 (1995).

[4] Worsnop *et al.*, Science 259, 71 (1993).

[5] Tabazadeh *et al.*, JGR 99, 12,897 (1994).