Arctic PSCs observed with ILAS during the winter of 1996/1997: Analysis of temperature history and inference of the chemical composition of particles

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Abstract. The Improved Limb Atmospheric Spectrometer (ILAS) captured many Polar Stratospheric Cloud (PSC) events during the winter and early spring of 1997 in the Northern Hemisphere [Hayashida et al., 2000]. To infer the chemical composition of the observed PSC particles, the ILAS aerosol extinction coefficient data and nitric acid data were compared with the predicted values assuming the formation of super-cooled ternary solution (STS), nitric acid dihydrate (NAD) or nitric acid trihydrate (NAT) particles. The intensive analysis suggested the formation of type Ia and Ib PSC particles during the operation of the ILAS. The temperature histories of all of the observed PSC particles were examined, and we found some events whose histories were theoretically required for the formation of liquid or solid particles.

Introduction

It is necessary to know the phase and chemical composition of PSC particles in order to estimate the amount of ozone loss exactly. Ravishankara and Hanson [1996] and Borrmann et al. [1997] suggest that liquid particles may convert inactive chlorine to an active form more efficiently than frozen particles. As well as playing a role in heterogeneous reactions, solid particles also play an important role in the process of redistribution of NOy and water vapor following the sedimentation of the particles.

The ILAS on board the Advanced Earth Observing Satellite (ADEOS) successfully observed the profiles of stratospheric ozone and related species, such as aerosol extinction, nitric acid and water vapor, in both polar regions from November 1996 through June 1997 [Sasano et al., 1999]. The ILAS captured more than 60 PSC profiles in the Northern Hemisphere during the winter and early spring of 1997, and their temporal and special distributions have been reported [Hayashida et al., 2000]. Our recent analysis revealed that the ILAS also observed many PSC events in the Southern Hemisphere during the early winter of 1997.

This study shows the possibility of inferring the phase and chemical composition of the observed PSC particles from the ILAS data and the trajectory analysis. Using the simultaneously obtained nitric acid and water vapor data as well as the extinction data will give us detailed information on the chemical composition of the PSC particles.

PSC identification

Our approach to identifying PSCs is similar to that of Poole and Pitts [1994] and Fromm et al. [1997]. All the extinction data inside the polar vortex were averaged for each altitude interval and each 10-day period when the collocated temperature was above 200 K. A '5-sigma selected PSC' was an event with an extinction exceeding the threshold value defined as the average plus five standard deviations.

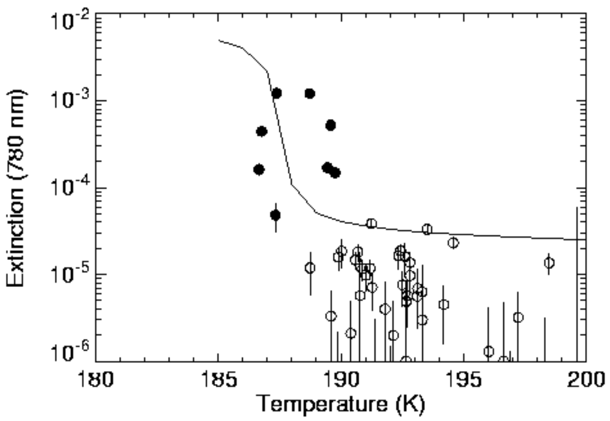
Comparison of the ILAS data with theoretical prediction

Comparison of the ILAS data and the values predicted from theoretical considerations of PSC composition enabled us to infer the chemical composition of the observed particles. The particle volumes were predicted theoretically, utilizing the Carslaw's analytic expression assuming the formation of super-cooled ternary solution (STS) particles [Carslaw et al., 1995]. The volumes were converted into the 780-nm extinction applying Mie theory and the results were compared with the ILAS extinction data. The calculated fraction of nitric acid remaining in the gas phase after uptake into STS particles was compared with the ILAS nitric acid data. The total amounts of nitric acid, water vapor and sulfate required for the calculation with the Carslaw's analytic expression were estimated from the ILAS data. The total amounts of nitric acid and water vapor were determined from the background average observed with the ILAS, while the total amount of sulfate was that which fit the background aerosol profiles.

Furthermore, the ILAS nitric acid data were also compared with the calculated vapor pressures of nitric acid in equilibrium over nitric acid dihydrate (NAD) and nitric acid trihydrate (NAT) based on the work of Hanson and Mauersberger [1998] and Worsnop et al. [1993]. The ILAS water vapor data were used to calculate the vapor pressures of nitric acid over NAT and NAD.

Results and Discussion

Figure 1 shows scatter-plots of the extinction coefficient vs. temperature (a) and the nitric acid vs. temperature (b). The theoretically predicted curves for STS (solid line), NAT (dotted line) and NAD (dots and dashes) are also indicated. Figure 1 illustrates that both the extinction data and the nitric acid data show good correspondence to the theoretically predicted values for STS at 20-25km in mid-January, suggesting that some STS particles were observed with the ILAS during this period. The temperature histories for all of these events were also closely examined using trajectory analysis. Before observation, they experienced a monotonic decreasing temperature to several degrees below T_{NAT}, as required for the formation of liquid particles [Tabazadeh et al., 1995; Larsen et al., 1997]. On the other hand, some data correspond to the theoretically predicted values for NAT rather than STS in other periods. We also found some temperature histories suggesting the formation of the solid particles [Tabazadeh et al., 1995; Larsen et al., 1997]. The data suggesting the existence of type Ia PSCs are still being analyzed.



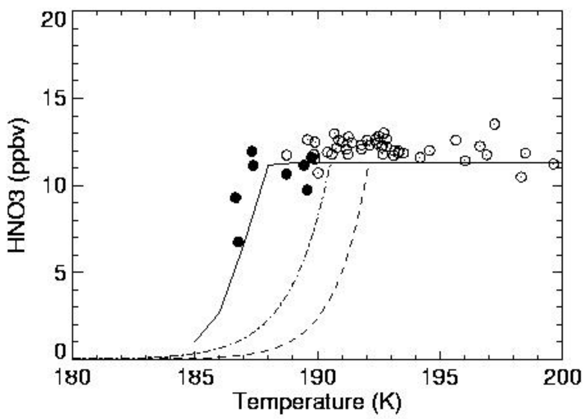


Figure 1

Scatter-plots of the ILAS extinction coefficient versus the collocated temperature (a) and the ILAS nitric acid versus the collocated temperature (b) for mid-January 1997 at 24 km. The theoretically predicted curves for STS (solid line), NAT (dotted line) and NAD (dots and dashes) are also indicated in the figure. Black points represent ‘5 sigma-selected’ events. Error bars are also shown with the ILAS data.

This study showed that combined analysis of trajectory and the observed ILAS data when compared with the theoretically predicted NAD/NAT/STS formation will allow us to reveal the chemical composition of PSCs. Furthermore, the trajectory analysis may give us information about the formation and breakup of PSC. Preliminary results for the Southern Hemisphere will also be shown.

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