Properties of Polar Stratospheric Clouds from ACE-FTS Extinction Measurements
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Abstract

We present a method for the retrieval of the particle size distributions and compositions of polar stratospheric clouds (PSCs) from ACE-FTS extinction measurements. We show the results of PSC observation on Feb. 27, 2011 at 80.7°N/77.7°E. The observed PSC consists primarily of ternary particles of radii of 0.05-1.0µm. Larger ternary particles (≥5µm) and ice (≥5µm) also account for a significant volume of the cloud.

Introduction

At sufficiently low temperatures (<195K) in the Arctic and Antarctic stratosphere, the condensation of water vapor and nitric acid (HNO\(_3\)) occurs resulting in the formation of PSCs [9]. At lower stratospheric temperatures, clouds consisting primarily of ice particles may serve as nuclei for the deposition of nitric acid in the form of nitric trihydrate (NAT). PSCs may also well contain droplets of a supercooled ternary solution of H\(_2\)SO\(_4\), HNO\(_3\), and H\(_2\)O. The composition of which has been well documented under stratospheric conditions by Carlaw et al. [3]. Both the bulk and surfaces of PSC aerosols provide sites on which inert chlorine species such as chlorine nitrate (ClONO\(_2\)) and hydrogen chloride (HCl), are converted to active forms. Photolysis of active chlorine produces Cl and ClO radicals promoting the depletion of ozone.

Many satellite based studies on PSCs have been performed such as POAM, SAGE, CLEAS and MIPAS, however only a small fraction of PSCs that form have been observed [7]. In addition to the lack of measurements of PSCs, varying physical and chemical properties from one winter to the next make the modeling a significant challenge. Many measurements have been made to aid the modeling of PSC formation. In this study we aim to observe and determine the composition and phase of PSCs for the 2010-2011 Arctic winter. The 2010-2011 Arctic winter showed unprecedented Arctic ozone loss [5], hence a significant presence of PSCs. Observations during this time period will allow for a number of comparable PSC measurements to be made.

Methods

On the assumption that the extinction contributions of all atmospheric constituents are additive, the total extinction is then given by

\[ \tau_{\text{tot}}(\lambda) = \tau_{\text{ext}}(\lambda) + \int K_{\text{ref}}(\lambda, r) P(r) \, dr \]

Where:

- \( \tau_{\text{ext}}(\lambda) \): gas-phase extinction
- \( K_{\text{ref}}(\lambda, r) \): extinction cross-section of condensed-phases
- \( P(r) \): particle size distribution
- \( r \): radius
- \( n(r) \): complex refractive index

The temperature dependent concentration and the complex refractive indices of STS from Biermann et al [2] are used. The temperature dependent concentration and the complex refractive indices of STS from Biermann et al [2] are used. The extinction cross-sections \( K_{\text{ref}}(\lambda, r) \) are computed by Mie theory for spherical particles for 96 mono-disperse radii between 0.05 and 12µm. Rewriting (1) in matrix form and inverting the least-squares problem:

\[ \chi^2 = \min \left\{ W^{-1} \left( \tau_{\text{ext}} - K \cdot P \right)^2 \right\} \]

Where:

- \( W \): weighting function
- \( \tau_{\text{ext}} \): measured extinction spectrum
- \( K \): reference spectra
- \( P \): particle size distribution

Gas-phase contributions are first reduced by the calculation of a residual spectra. The contributions from the gas-phase components are calculated and removed from the observed spectrum. This calculation also includes the MT_CKD continuum model to eliminate the N\(_2\), O\(_3\), O, and H\(_2\)O continuum. The remaining residual spectrum is that of the condensed-phase as shown in Fig. 2.

Results

The results shown are that of a PSC observation from occultation ss40615 at 20.0km on Feb. 27, 2011, 80.7°N/77.7°E. The retrieved temperature at 20.0km was 190K and the composition of STS was chosen to be 42% HNO\(_3\) and 3% SO\(_2\) at 20.0km.

Figure 2: Observed spectra from ACE-FTS and calculated residual spectra are shown for occultation ss40615 (Feb. 27, 2011, 80.7°N/77.7°E). Spectra at 20.0km have PSC signatures present, while the spectrum of 24.0km is PSC free.

Spectral noise is reduced by the use of a wavelet filter to generate the weight function \( W \). The least-squares problem of Eq. 2 is then solved by a non-negative least-squares fitting procedure to yield \( P \). The particle size distribution \( P \) is subject to a smoothing constraint to ensure \( P \) does not contain any unphysical sharp features.

The superposition of individual chemical components is chosen to form the reference spectra \( K \) for least-squares fit. The composition of the observed PSC is determined by the superposition of reference spectra which minimizes the value of \( \chi^2 \) and provides the best fit. The solution of the least-squares problem \( P \) provides the particle size distribution of each component.

Conclusion

Our retrievals provide a means of determining the composition and particle size distributions of PSCs. In the case shown of occultation ss40615 at 20.0km, the observed PSC was determined to be composed of a significant number of ternary particles of radii of 0.05-1µm. Larger ternary particles (≥5µm) and ice (≥5µm) also account for a significant volume of the cloud.

With the retrievals methods presented, we will continue to process ACE-FTS measurement showing PSC signatures for the 2010-2011 Arctic winter. The significant occurrence of PSCs during this winter will allow for a sufficient number of comparable observations to be made.

References


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Instrument

The Atmospheric Chemistry Experiment (ACE) Fourier Transform Spectrometer (FTS) aboard SCISAT-1 measures extinction through the atmosphere by solar occultation as in Fig. 1. For each occultation, measurements are made at various tangent heights allowing for a vertical resolution of about 1.2km.

The ACE-FTS instrument has a spectral range of 750-4400 cm\(^{-1}\) at a resolution of 0.02 cm\(^{-1}\) allowing for the retrieval of aerosol particle sizes from 0.05 to 12µm.

Figure 1: Schematic diagram of an ACE sunset occultation.

The ACE-FTS instrument also contains a visible/near infrared imager with two filtered channels at 525 and 1000nm with the purpose of monitoring clouds and aerosols.

Temperature profiles are retrieved from the relative intensities of CO\(_2\) lines from ACE-FTS [1].

The combination of the filtered images and temperature profiles allow for the diagnostic observation of PSCs.