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Properties of high-altitude tropical cirrus clouds determined from ACE FTS observations

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[1] During tropical observations made in February and August of 2004, the Atmospheric Chemistry Experiment (ACE) FTIR instrument aboard the Canadian SciSat-1 satellite recorded several extinction spectra in the range of 700–4400 cm^{-1} that exhibit strong signatures of cirrus clouds. We have analyzed these spectra in order to quantify the properties of these high-altitude cirrus clouds. This Letter reports the vertical profiles of the average ice density in the clouds and the particle size distributions. We also discuss the effect of particle shape on the retrieval results.

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1. Introduction

[2] Cirrus clouds, which are composed of small ice particles, cover an average of about 30% of the upper troposphere and their presence leads to a permanent and considerable contribution to radiative forcing [*Baran*, 2004]. The quantitative effect of cirrus clouds on the Earth's radiative budget, however, is uncertain. It has been argued that cirrus clouds contribute to warming or cooling depending on the density, size, and shape of the particles. In addition to the number density, which has a simple direct effect on the propagation of radiation through the clouds, the shape and phase (solid or liquid) of the particles can also strongly affect the transmission of radiation through the atmosphere. The complex spatial density distributions and wide particle size distributions, which range from a few to a few hundred microns, make estimates of the contribution of cirrus clouds to atmospheric radiative transfer highly uncertain.

[3] There is a very large number of publications on a wide variety of cirrus cloud properties. In general, it is agreed that cirrus clouds are formed from non-spherical ice particles and that the shape and size of these crystals depends on the atmospheric conditions such as temperature, pressure and relative humidity that exist during their formation [*Baran*, 2004]. High-level cirrus clouds that form at altitudes above 13 km in the tropics often are composed of small (<15 μm) ice crystals [*Peter et al.*, 2003; *Rinsland et al.*, 1998]. The infrared spectra of such ice particles have broad bands with locations that shift with temperature, particle size and shape [*Clapp et al.*, 1995]. It was shown earlier [*Kahn et al.*, 2002;*Rinsland et al.*, 1998] that these spectral features can be detected in solar occultation observations from space.

[4] Due to their broad bandwidth and high resolution, the spectra recorded by the ACE FTIR instrument [*Bernath*, 2004] contain enough information for retrieval of the phases, densities, and size distributions of cloud and aerosol particles. We have previously reported details of the procedures we use to retrieve this information [*Zasetsky et al.*, 2002; *Zasetsky et al.*, 2004b] and have shown that these methods can provide significant detail on the size distributions for particles that are smaller than about 30 μm, while larger particles are identified only by their contribution to the extinction, since their spectra do not have identifying features that are unique to their sizes. The amount of information that we can retrieve using this method depends to some extent on the signal to noise of the observation and the amount of interference by gas phase components. By exploiting the high resolution of FTIR spectra, however, we can distinguish condensed phase extinctions from pressure broadened and overlapped gas phase absorptions. Typically, this allows us to retrieve the compositions, phases and size distributions for clouds and aerosols in the mid-to-upper troposphere. In the following we report the analysis of 18 tropical cirrus cloud observations by the ACE FTIR instrument in 2004. We present retrievals of the size distributions and vertical density profiles in these high-altitude cirrus clouds.

2. Methodology

[5] The computations use a library of monodisperse reference spectra in a weighted least squares comparison with the observed spectrum. In the present study we use the frequency dependent ice refractive indices of *Clapp et al.* [1995]. A central feature of the present analysis is a weighting procedure that assigns the highest importance to those parts of the spectra that are the least affected by gas phase absorption; an additional weight is also assigned to several microwindows specified by *Rinsland et al.* [1998]. This weighting constrains the solution and makes the retrievals reliable.

[6] It has been reported that high-level tropical cirrus clouds are composed of hexagonal prismatic ice crystals having various aspect ratios [*Pruppacher and Klett*, 1998] so we explored the use of both spheres and hexagonal prisms with various aspect ratios as reference spectra. (The closest agreement was obtained with a ratio of 2.8 and we have used this value in the analysis below.) The Bohren and Huffman procedure [*Bohren and Huffman*, 1983] and Discrete Dipole (DD) method [*Draine and Flatau*, 1994; *Draine and Flatau*, 2003] were used to compute the extinction spectra for spheres and hexagonal prisms, respectively. Taking into consideration the symmetry of

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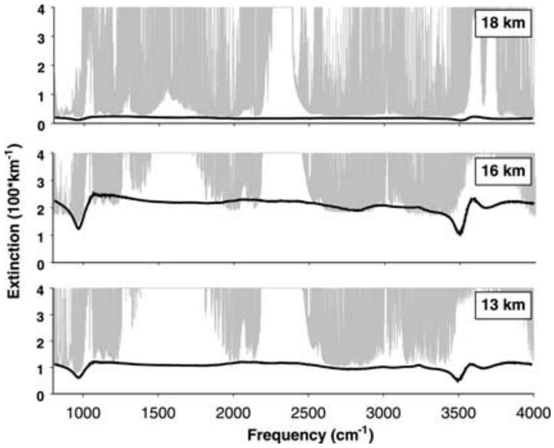

Figure 1. ACE FTS spectra (gray lines) together with Mie fits (black lines).

hexagons, the final extinction intensities for the hexagonal prisms were computed as averages over 60 orientations. The number of point dipoles was chosen to satisfy the criterion $2\pi d|n^*|/\lambda < 1$, in which d is the inter-dipole separation, n^* is the complex index of refraction and λ is the wavelength [*Draine and Flatau*, 2003]. The number of dipoles was approximately 5×10^4 for 12 μm diameter particles.

[7] We compared spectra of spherical particles computed using Mie theory with spectra of hexagonal right prisms having an aspect ratio of 2.8 and equivalent radii (radii of spheres having the same volume) up to 12 μm. Although the spectra differ for the larger particles, the uncertainties in the ACE spectra caused by measurement noise and gas phase interference are greater than the differences in the spectra of the two shapes. Thus hexagonal prisms cannot be distinguished from spheres in remote sensing measurements of this kind. Moreover, many of the shape-related differences in the remote sensing spectra are likely reduced because the particles may be present in a variety of shapes and habits in the long path length of the observation. As a result of this, we could not justify the selection of a particular non-spherical model such as hexagonal prisms for the retrievals. We plan to investigate this aspect in more detail at a later time, both with laboratory spectra and high-resolution FTIR remote sensing measurements, after we have obtained enough of the latter to allow averaging of a large number of observations taken under similar conditions.

[8] The inversion procedure was performed for a number of tangent altitudes and the recorded spectra were interpolated on a 1 km grid. We used an onion-peeling algorithm to improve the estimates of the vertical profiles of the volume density. To eliminate the influence of gas continuum (particularly water continuum) absorptions, which have broad-band features similar to condensed phase extinction, we used the MT_CKD continuum model [*Mlawer et al.*, 2004]. We subtract the modelled continua due to water vapor, nitrogen, oxygen, carbon dioxide, and ozone before implementing our retrieval procedure. The gas phase densities, pressures and temperatures and the tangent heights were taken from the ACE level 2 retrieval data (C. D. Boone et al., Retrievals for the Atmospheric Chemistry Fourier Experiment, submitted to *Applied Optics*, 2005).

3. Results and Discussion

[9] During the periods between 01 and 09 February and 02 and 27 August 2004, when the tangent points of the measurements occurred in the tropics (between 25N and 25S), the ACE FTS recorded several spectra with pronounced ice cloud signatures. The first five of these occurred in February and the next thirteen in August.

[10] An example of ACE spectra containing the features of a cirrus cloud recorded on 2004-02-05 at 3.42S/122.17W is shown in Figure 1. The observed high-resolution spectrum is shown by the faint grey curve. The condensed phase spectrum obtained using our inversion procedure is the heavy black line. In the inversion procedure, we first process the observed spectrum using a wavelet filter and a weight function based on smoothness criteria. Then the retrieval is carried out using a constrained linear least squares procedure involving the comparison of the smoothed, weighted spectrum with a library of 96 monodisperse reference spectra covering the radius range of interest [*Zasetsky et al.*, 2002; *Zasetsky et al.*, 2004a, 2004b]. The condensed phase spectrum shown in Figure 1 is the combination of reference spectra that provides the best fit. Note that this inversion procedure does not involve any modelling of the discrete gas phase spectra.

[11] The extinction feature centered at 800 cm^{-1} is the librational mode; the OH vibrational mode is evident near 3500 cm^{-1}. The shape of these features shows that a significant number of the particles are smaller than 15 μm. The full retrieval procedure confirms this (vide infra).

[12] Temperature retrievals based on the ACE gas phase measurements for these same occultations show that the mean tropopause temperatures for February and August were 192 K and 194 K, respectively. These are consistent with GPS radio occultation measurements made from 2001 to 2003 [*Schmidt et al.*, 2004]. These temperatures were below the freezing point for micron-sized water particles [*Zasetsky et al.*, 2004a] throughout the altitude region from 10 to 17 km. In view of this, we included only ice spectra in our reference database for these retrievals and did not include liquid water spectra. This improved the signal to noise of the retrieval.

[13] Figure 2 shows vertical profiles of the cloud volume densities (lower limits) for the tropical cirrus clouds, retrieved in the manner described below. The grey area gives the background, obtained by averaging 26 ACE FTIR observations in the same geographical region but in the absence of cirrus clouds. The uncertainties become very high for altitudes below 10 km due to strong gas phase interference, thus the retrievals below this altitude are not considered further in this work. This figure demonstrates, however, that cirrus clouds are easily detectable at higher altitudes where the ice volume densities obtained from our procedure are well above background levels.

[14] The maximum altitude in the clouds observed in February is about 16 km, whereas it is 13 km for those observed in August. Since these are occultation measurements, densities are reported as if the clouds are uniformly distributed over the atmospheric path length of the obser-

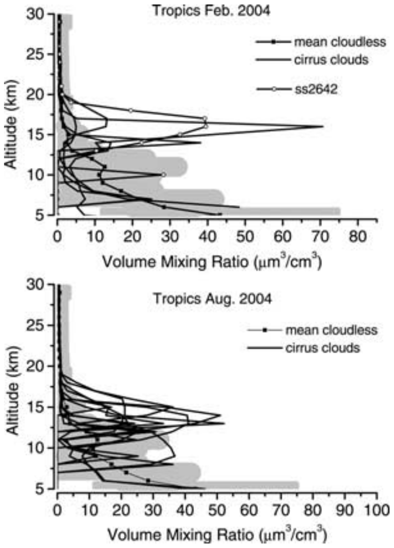

Figure 2. Vertical density profiles of the cirrus clouds together with the mean "cloudless" profile in the tropics observed (top) in February and (bottom) in August.

vation. The reported densities, therefore, should be considered as lower limits to the actual densities in the clouds.

[15] The size distributions in Figure 3 correspond to the cirrus cloud marked by open circles in Figure 2 (top). Each point represents the contribution to the observed spectrum of the reference (monodisperse) spectrum having the indicated radius. The number density is dominated by ice particles smaller than 30 μm. At the lowest altitude, the distribution is broad and monomodal, with a tail that extends up to about 20 μm. At higher altitudes, the distributions become bimodal, with a smaller mode that peaks at about 3 microns and a larger one that appears to peak between 10 and 20 μm. Since this cloud is associated with a tropical convection system, we speculate that the development of this bimodality might be the result of dynamical processes occurring during the rapid upwelling of the humid air. From this single measurement it is not possible to determine whether the smaller mode is the result of additional nucleation during the convective rise or of growth of the larger particles and evaporation of the smaller ones. Similar bimodality has appeared in some of the other observations as well [*Knollenberg et al.*, 1982; *Zhang et al.*, 1999], however, and we are presently undertaking a more thorough investigation of this aspect by means of modelling based on the meteorology.

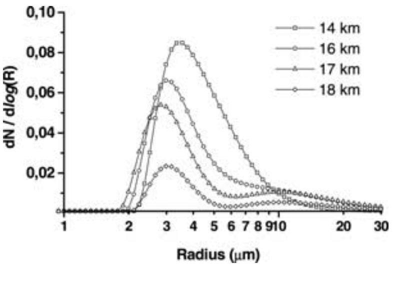

Figure 3. Number distributions as a function of tangent altitude for the ss2642 observation (17N, 162E) made in February 2004.

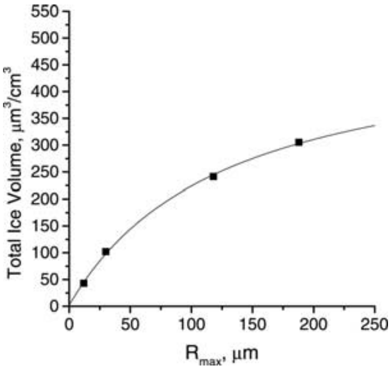

Figure 4. Total volume density at 16 km as a function of maximum particle radius used in the cirrus cloud retrieval for the ss2642 observation (17N, 162E), February 2004.

[16] The number of particles in the larger mode decreases with increasing size, but the observations do not permit us to determine the maximum size directly from the inversion. We can, however, obtain an estimate of the total volume density of the ice particles by finding the dependence of the total retrieved volume on the maximum radius used in the retrieval.

[17] In order to estimate the total volume arising from the particles larger than the limiting size used in our retrieval, we have applied the following procedure. The extinction per unit volume in the simplest case of uniformly distributed spherical particles is

$$C_{ext}/V = A + Sc/r_c \tag{1}$$

where A and Sc are the total absorption and scattering cross sections, and r_c is the cut-off radius (the largest size used in our retrieval procedure).

[18] This relation follows from the fact that absorption is proportional to the total volume $V = 4\pi r^3/3$, while scattering is proportional to the cross-sectional area of the particle. Thus, the total volume as a function of r_c can be written as

$$V = \frac{C_{ext}}{A + Sc/r_c} \tag{2}$$

Having computed the values of the total volume as a function of the cut-off radius r_c for several values, we then fitted these data using expression (2). The best fit (ACE spectrum at 16 km in Figure 1) is shown in Figure 4. The asymptotic limit ($r_c \to \infty$) gives the upper limit of the total volume, which is about 500 μm³/cm³ in the present case. Using this approach, we can obtain the upper limits of the volume density of ice in cirrus clouds. Typical values of the total ice volume density in the high-altitude tropical cirrus clouds observed by the ACE instrument from 01 to 09 February and from 02 to 27 August 2004 are between 10 and 700 μm³/cm³.

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