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Temperature of mesospheric ice retrieved from the O-H stretch band

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[1] For the first time, the temperature of mesospheric ice particles is retrieved directly from ice infrared extinction spectra measured in the solar occultation regime. The position of ice O-H stretch band peak varies from $3230 cm^{-1}$ at T = 120K to $3246 cm^{-1}$ at T = 155K, which enables the retrieval of ice temperature by fitting a model spectrum to a measured one. The retrieved temperature is independent of cloud vertical and horizontal patchiness and has a random uncertainty of <12K. The retrieval is sensitive to chosen particle shape: cubes, hexagons and spheroids of certain forms give same result. Spheres and rectangular prisms (aspect ratio $\geq$2) change temperature by 3–4K. For nearly 400 ice spectra analyzed the retrieved temperature for cubes ranges from 120K to 150K with the distribution maximum centered at $\sim$135K. The standard temperature for same ice spectra retrieved from the gas phase differs considerably and sometimes reaches values of 200K and above. **Citation:** Petelina, S. V., and A. Y. Zasetsky (2009), Temperature of mesospheric ice retrieved from the O-H stretch band, *Geophys. Res. Lett.*, *36*, L15804, doi:10.1029/2009GL038488.

1. Introduction

[2] Layers of ice particles, the biggest of which form optically detectable Polar Mesospheric Clouds (PMCs), exist near the polar summer mesopause where the temperature is low enough for ice nucleation to occur [e.g., *Thomas*, 1991; *Lübken*, 1999]. For simplicity, we will refer to all mesospheric ice particles as PMCs. It is known that PMCs are composed of crystalline water ice [*Eremenko et al.*, 2005] and their properties are governed mainly by temperature and water vapor pressure and, to a lesser extent, by other atmospheric parameters, such as vertical winds, meteoric influx, and eddy diffusivity [e.g., *Lübken et al.*, 2007].

[3] The importance of temperature (T) for the process of PMC formation follows from the fact that the equilibrium water vapor pressure over ice, P, is an extremely strong function of T. Indeed, as P can be roughly described as: $P = \{(T - T_0)/B\}^{12}$ [*Lide*, 1999], where T_0 is 126 K and B represents all temperature-independent parameters, a small change in temperature would result in a huge shift in water pressure and thus in the rate of ice particle formation.

[4] Studying the formation and variability of PMCs with respect to the properties of surrounding environment, particularly temperature, is important for improving our understanding of physics in the upper mesosphere and also for the analysis of both short and long-term changes in the atmosphere [e.g., *Thomas*, 2003]. This, however, is not trivial due to the remote location of this region. Ground-based and rocket-borne observations are scarce and restricted to specific geographic locations. Satellites can sample the polar region daily, but various instrument limitations often result in noticeable errors in the retrieved temperature and other mesospheric parameters, especially in the presence of PMCs [e.g., *Hervig and Siskind*, 2006].

[5] One of the modern satellite instruments capable of simultaneous measurements of ice volume in PMCs, as well as temperature and water vapor concentration in the upper mesosphere, is the Fourier-Transform Spectrometer on the Atmospheric Chemistry Experiment satellite (ACE-FTS). However, its wide, about 4 km, vertical field-of view (FOV) limits the potential use of these data in studies where the accuracy of temperature in PMCs is essential. In the present work, the intrinsic temperature in PMCs, which is independent of the FOV or cloud horizontal and vertical patchiness, is determined by using the ACE-FTS O-H stretch band infrared spectra between 3200 and 3300 cm^{-1}. As will be shown in the following, these new temperature values seem more reliable for the analysis of mesospheric ice than those provided in the satellite operational dataset.

2. Instrument and Data Description

2.1. ACE-FTS

[6] ACE is a solar occultation satellite launched in 2003 in a circular orbit at 650 km altitude with an inclination of 74° [*Bernath et al.*, 2005]. ACE-FTS has a spectral coverage from 750 to 4400 cm^{-1} and a spectral resolution of 0.02 cm^{-1}. It measures the atmospheric transmittance during sunrise and sunset and produces vertical profiles of temperature, pressure, and volume mixing ratios of more than 30 gaseous species [*Boone et al.*, 2005]. The instrument FOV is 1.25 mrad, which corresponds to about 4 km at the tangent point, and its vertical resolution is 3–4 km. This results in a vertical smoothing of all measured atmospheric parameters and may cause ambiguities in the interpretation of data in the upper mesosphere at PMC altitudes. As the laps rate for temperature at 80–88 km is about 5–7K per kilometer [*Lübken*, 1999], a 4 km vertical smoothing may result in averaging over a temperature range of nearly 30K. Since PMCs respond to much smaller variations in temperature [e.g., *von Savigny et al.*, 2007], the averaging over 30K may cause significant uncertainties in studying the relation between PMC properties and temperature.

2.2. PMC Measurements by ACE-FTS

[7] The presence of water ice particles in the mesosphere results in the absorption and scattering of incident solar radiation, which is detected by ACE-FTS in three spectral

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regions: around 800 cm^{-1} (librational mode), 1600 cm^{-1} (bending mode), and between 3000 and 3500 cm^{-1} (O-H stretching mode). The latter band is more sensitive to the presence of ice particles. A detailed description of ACE-FTS spectra in the O-H stretch band in the presence and absence of PMCs can be found in work by *Eremenko et al.* [2005]. In the present work, we use nearly 400 PMC spectra recorded during the northern hemisphere summer of 2005. We note that the highest latitudes for ACE-FTS measurements during the PMC season do not extend poleward of 70°.

[8] To ensure high quality of results, the data have been selected such that the maximum of absorption to baseline noise ratio in the range of 3200–3300 cm^{-1} is $\geq$5. This implies that weak clouds and/or very small particles that do not contribute much to the observed signal may not be represented in this analysis. The ACE-FTS infrared spectra are sensitive to the volume of ice (or ice mass density) in the mesosphere, but not to the variations in the size of ice particles within the 10–100 nm range. If we assume that the volume of ice particles with radius <10 nm is very small, then it is very likely that such particles will not affect the results of this study.

2.3. Standard Temperature From ACE-FTS

[9] A standard algorithm for the retrieval of vertical profiles of all atmospheric parameters from ACE-FTS is described by *Boone et al.* [2005]. In that approach, temperature and pressure profiles are determined using a non-linear least squares global fit. For temperature retrievals, CO_2 spectral features are fitted using a total of 106 narrow spectral intervals, typically 0.3–0.5 cm^{-1} wide, in the wavenumber ranges 930–940 cm^{-1}, 1890–2450 cm^{-1}, and 3300–3400 cm^{-1}. The HITRAN-2004 spectroscopic database [*Rothman et al.*, 2005] is used in the forward model calculations. The validation results for these temperature retrievals are given by *Sica et al.* [2008] and suggest a systematic high bias of 3–6K at 50–70 km and a possible systematic low bias of about 2K near 23 km. There was no evidence of any systematic bias at PMC altitudes that, according to limited number of comparisons, has a random error of up to 10K. In the following we will refer to these standard temperature values as retrieved from gas phase.

3. Temperature Retrieval From the O-H Stretch Band

[10] It has been shown that the position of the O-H stretch band, caused by the presence of ice particles, varies as the temperature in ice particles changes [*Clapp et al.*, 1995]: it shifts from 3230 cm^{-1} at T = 120K to 3246 cm^{-1} at T = 155K. This is because vibrational modes are typically temperature dependent (although not as strong as rotational modes) due to the changes in the shape of potential energy surface that, in turn, result from changes in the density of ice. This effect offers a more accurate, “target”, approach to retrieve the temperature in PMCs from ACE-FTS spectra. By fitting the observed spectra with those calculated using the T-dependent refractive indices from *Clapp et al.* [1995], we can pinpoint the temperature in PMC particles more accurately in comparison to that retrieved from gas phase. Below we will refer to the temperature derived from the ice O-H stretch band spectra as the solid phase temperature.

[11] In order to model PMC extinction spectra in the O-H stretch band, the shape of mesospheric ice particles is required. Although spheroids as a model for PMC particles have been adopted in some recent studies [e.g., *Hervig et al.*, 2009 and references therein], representing crystalline ice by spheroids may not be the best choice from the thermodynamics and kinetics point of view. Our latest results on the mesospheric ice nucleation process suggest that cubes and/or hexagons are most physically appropriate forms for representing the shape of crystalline ice particles in the mesosphere [*Zasetsky et al.*, 2009]. As there is no reliable method to distinguish cubic ice from hexagonal ice from ACE-FTS spectra, we use both shapes to retrieve the temperature in mesospheric ice particles. For hexagons, the length-to-side ratio (L/S) is taken to be 1.1, similar to that used by *Eremenko et al.* [2005].

[12] The extinction calculations for mesospheric ice (forward model) are carried out with the discrete-dipole approximation method first proposed by *Purcell and Pennypacker* [1973] and further developed by B. T. Draine and P. J. Flatau (User guide for the discrete dipole approximation code DSCAT.6.0, 2003, available at http://arxiv.org/abs/astro-ph/0309069). Using the above method, we obtain scattering and absorption intensities for particles with arbitrary shapes. Here, 2000 point dipoles are used to approximate cubes and hexagons, with the resulting spectra computed by averaging over 216 orientations.

[13] The overall uncertainty in the retrieved solid phase temperature consists of the fitting error, the error in optical constants, and the uncertainty associated with the choice of particle shape. The statistical (fitting) error depends strongly on the intensity of the O-H stretch mode and is less than 4% for clouds discussed in this work. The uncertainty associated with the errors in the chosen optical constants is difficult to estimate; they are reported to be lower than 5% in the O-H stretch peak region [*Clapp et al.*, 1995]. Both particles shapes used here, hexagonal (L/S = 1.1) and rectangular (aspect ratio AR = 1 that is a cube) prisms, give the same result. Based on the recent studies discussed earlier, we believe that these shapes serve as a reasonable approximation for the true shapes of ice particles in the mesosphere, and estimate the associated uncertainty in the retrieved temperature as $\sim$1K. Thus, for PMCs used in this study the highest random uncertainty in the retrieved solid phase temperature for an individual observation is estimated as <12K.

[14] We also examined the effect of other possible shapes on the retrieved PMC temperature. The use of spheroids with AR = 2 [e.g., *Hervig et al.*, 2009] gives same temperature values as those for hexagons and cubes. The use of rectangular prisms with AR $\geq$ 2 shifts the band peak position toward higher wavenumbers (due to worse accuracy of the fit). The assumption of a spherical shape, which also has been widely used in PMC studies [e.g., *Robert et al.*, 2009, and references therein], has the opposite effect – it shifts the position of the band peak toward lower wavenumbers. In both these cases, the difference in the retrieved temperature, as compared to cubes, compact hexagons and spheroids, is about 3–4K.

[15] We note that it would be advantageous to carry out our analyses with the 800 cm^{-1} band, which is rotational in nature and thus more sensitive to the changers in temper-

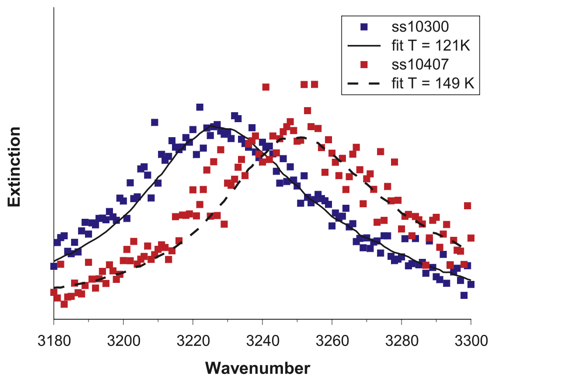

Figure 1. Normalized extinction from PMC ice particles against wavenumber measured by ACE-FTS with the spectral resolution of 0.02 cm^{-1}. The PMC temperature, determined by fitting a calculated spectrum for cubic ice to the observed spectrum, is 121 K for occultation ss10300 (solid blue squares) and 149K for occultation ss10407 (solid red squares).

ature and much less dependent on the particle shape. However, in the case of ACE-FTS spectra, the signal-to-noise ratio in this spectral region is too low, and an additional study is required in order to determine the prospective of combining two bands for the temperature retrievals.

4. Results and Discussion

[16] Figure 1 illustrates two PMC extinction spectra (normalized) where actual ACE-FTS measurements are shown as separate points. For these selected spectra, the solid phase T, which is 121K for occultation ss10300 and 149K for occultation ss10407, is retrieved by fitting a calculated spectrum for cubic ice to a measured spectrum. For comparison, standard (gas phase) temperatures for these two occultations are 156K and 169K respectively.

[17] For PMC observations in Figure 1, the solid phase temperature is lower than the gas phase temperature by 35K for ss10300 and by 20K for ss10407. For the second occultation, the retrieved gas phase temperature in PMCs, 169K, is significantly larger than the ice frost point temperature of ~150K [e.g., *Lübken*, 1999]. This implies that in the case of ACE-FTS observations, the solid phase temperature seems more reliable for PMC studies than the gas phase temperature retrieved from CO_2 rotational bands.

[18] The total volume of ice along ACE-FTS line of sight as a function of temperature for nearly 400 PMCs is shown in Figure 2. The results for both, the solid phase and the gas phase, retrievals are shown. Apparently, the gas phase temperature exhibits a much wider scatter compared to the solid phase temperature, and even values as high as 200K and above can be seen. As noted earlier, a wide FOV of ACE-FTS, about 4 km, makes it very difficult to retrieve the temperature in mesospheric ice layers accurately, especially since the temperature laps rate in this region is large, about 5–7K. High variability of thermodynamic conditions in this region, as well as PMC patchiness in horizontal and vertical directions, also contributes to this effect. The above matters may explain why the gas phase temperature in PMCs is much higher than the frost point temperature of ~150K for many data points in Figure 2. An interesting observation is that the lowest value for gas phase temperature, about 106K, is not seen for the solid phase temperature. Cases like this can be attributed to instances where PMCs are seen in the lower part of a 4 km FOV while its upper part observes a PMC-free region with lower temperatures. There is also a cut-off of solid phase temperatures at about 150 K, which is a climatological frost point temperature in the summer upper mesosphere [*Lübken*, 1999], that seem to be natural and result from the underlying physics.

[19] The temperatures retrieved from solid phase (black squares in Figure 2) have a narrow distribution centered at about 135K with minimum and maximum near 120K and 150K respectively. To our knowledge, the most accurate data on temperatures in the upper mesosphere are provided by either in-situ measurements (rockets and falling spheres), or by ground-based lidars and radars. Although such data are still scarce, available results on temperatures in PMCs located equatorward of 70°N yield values similar to those reported here. Ground-based lidar and radar observations at 54°N revealed temperatures less than 150K in PMCs and 154–162K right below PMCs [*Gerding et al.*, 2007]. *Lübken et al.* [2002] reported that the temperatures in mesospheric ice layers, between 82 and 90 km, measured by falling spheres at latitudes 68° and 69°N typically range from 120 to 150K. According to *Rapp et al.* [2002], the temperatures in PMCs measured by sounding rockets at ~69°N range from 120 to 150K.

[20] Results reported here also generally agree with those obtained from a statistical comparison of PMC brightness and closely coincident upper mesospheric temperatures observed independently by two satellite instruments [*Petelina et al.*, 2005]. In the above work, temperatures measured by the Sounding of the Atmosphere using Broadband Emission Radiometry (SABER) instrument on the Thermosphere Ionosphere Mesosphere Energetics and Dynamics (TIMED) satellite were compared to PMCs measured by the Optical Spectrograph and Infrared Imager System (OSIRIS) on Odin. As a result, temperatures in PMCs were between 112 and 142K. However, as noted by *Petelina et al.* [2005] and thoroughly studied by *Kutepov et al.* [2006], the SABER temperature retrievals used, version

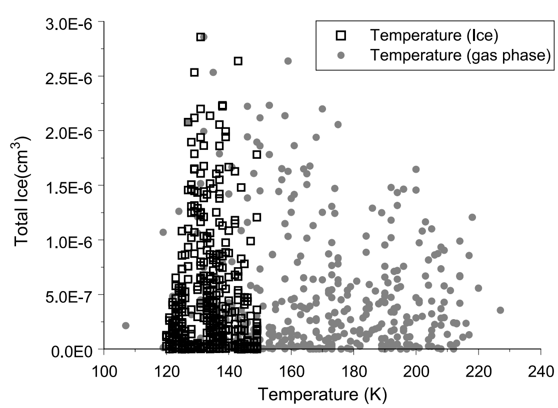

Figure 2. Total volume of ice (cm^3) for nearly 400 PMCs measured by ACE-FTS in July 2005 as a function of temperature retrieved from solid phase (black squares) and from gas phase (grey circles).

1.03, were systematically lower by ~10K compared to other data. If a crude correction of +10K is applied, the temperature range for PMCs reported by *Petelina et al.* [2005] would be 122–152K with the maximum PMC brightness at ~130K, which is in a good agreement with our findings.

[21] A recently launched Aeronomy of Ice in the Mesosphere (AIM) satellite also has the ability to simultaneously detect mesospheric ice and measure temperature. The latter is retrieved from the 4.324 μm (2312.67 cm^{-1}) CO_2 band data measured by the Solar Occultation For Ice Experiment (SOFIE) on AIM. However, current SOFIE temperature retrievals have a warm bias of more than 7K near the mesopause [*Gordley et al.*, 2009]. This bias is related to non-local thermodynamic equilibrium effects and will be addressed in upcoming data processing.

[22] In the future, we will use the technique for solid phase temperature retrievals described here to analyze the PMC- temperature -water vapor relation from all available ACE-FTS data in 2004–2009 in both hemispheres. Different from some other satellite PMC measurements, the extinction from mesospheric ice particles detected by ACE-FTS in the solar occultation mode is free from effects due to possible small orbit drift, or differences in observation geometry between two hemispheres [e.g., *Robert et al.*, 2009; *Bailey et al.*, 2005]. In addition, the retrievals of solid phase temperature described here reveal the intrinsic temperature of PMCs that is independent of the instrument FOV, or horizontal and vertical patchiness in ice layers. The latter can be significant due to high temporal and spatial variability of physical parameters in the mesosphere. This gives ACE-FTS an advantage in studying the morphology of mesospheric ice particles, including interannual and interhemispheric variability in PMCs, with respect to corresponding temperatures.

5. Conclusion

[23] For the first time, the intrinsic temperature of mesospheric ice particles is retrieved from the satellite data. This is done by analyzing the ACE-FTS ice extinction spectra in the O-H stretch region between 3200 and 3300 cm^{-1}. The retrieval technique is based on the results of *Clapp et al.* [1995] who demonstrated that the position of the O-H stretch band peak depends on temperature and varies from 3230cm^{-1} at T = 120K to 3246cm^{-1} at T = 155K. By fitting the calculated extinction spectra to the measured ones, the temperature of ice (solid phase temperature) is derived with the maximum random uncertainty for an individual retrieval <12K. Different from other satellite measurements, this temperature is completely independent of the temperature in PMC-free regions that often present along the instrument line-of sight.

[24] The assumed ice particle shape affects the retrieved solid phase temperature because the position of the O-H stretch band peak in the calculated spectra is also shape dependent. The use of a compact hexagonal (L/S = 1.1) or rectangular (AR = 1) prisms, as well as spheroids with AR = 2, gives same result for solid phase temperature. The use of spheres or rectangular prisms with the AR $\geq$ 2 changes the retrieved temperature by ~3–4K.

[25] For nearly 400 PMCs measured by ACE-FTS in July 2005, the retrieved solid phase temperature ranges from 120K to 150K with the maximum of ice volume centered at ~135K. This agrees well with other published data [*Lübken et al.*, 2002; *Rapp et al.*, 2002; *Petelina et al.*, 2005; *Gerding et al.*, 2007]. For comparison, the standard gas phase ACE-FTS temperature for the same observations ranges from the minimum of 106K to the maximum of about 230K. For the majority of clouds, such standard temperature at PMC peak is higher than the ice frost point temperature of about 150K. This is likely caused by a wide, about 4 km, FOV of ACE-FTS, as well as high variability of thermodynamic conditions in the mesosphere, including PMC patchiness.

[26] In the future, the technique to retrieve the intrinsic temperature in PMCs presented in this work will be used to analyze the PMC-temperature-water vapor relation from all available ACE-FTS data in 2004–2009 in both hemispheres. This will advance our understanding of the polar upper mesosphere, including the interannual and interhemispheric differences and trends in this region.

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