

Interactive comment on “Properties of polar stratospheric clouds obtained by combined ACE-FTS and ACE-Imager extinction measurements” by A. Y. Zasetsky et al.

A. Y. Zasetsky et al.

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Authors' replies to comments on Atmos. Chem. Phys. Discuss., 7, S5440–S5443, 2007, “Properties of polar stratospheric clouds obtained by combined ACE-FTS and ACE-Imager extinction measurements” by A. Y. Zasetsky et al.

Comments by M. Höpfner:

The authors appreciate Dr. Höpfner's helpful comments.

(1) -> *The composition of the liquid ternary solution PSCs retrieved is only mentioned in the abstract. It would be helpful if this result could be described in more detail in the body of the paper*

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Omission of this information was an oversight. A paragraph has been added at the end of Section 4 to add this information.

(2) p13272, l24: 'Hopfner et al., 2002':

-> *This reference for MIPAS/Envisat PSC observations is not appropriate here since it only describes balloon-borne data, better use one of the following: [Spang et al.(2005),Höpfner et al.(2006a)].*

The references have been corrected.

(3) p13276, l16: *'for solid hydrates and ice particles, the discrete dipole technique (Draine et al., 1994) was used.'*

-> *Could you tell which particle shape has been assumed?*

The statement in the manuscript is too brief and therefore is misleading. In fact, DD calculations were tested for a few particle shapes, but in the final analysis published here, these were not used. The sentence in question has been modified as follows to clarify this:

For solid hydrates and ice particles, the discrete dipole technique [Draine, B.T. and P. J. Flatau, 1994] was tested for a variety of aspect ratios up to 10, but due to the relatively small size of the particles, the spectra did not depend significantly (within the noise) on particle shape, so the Mie approximation was used throughout this work.

(4) p13277, l18: *'The total error is computed as a combination of the statistical error of the least squares fitting and the uncertainty associated with the particle shape and the finite length of the spectral region.'*

-> *Could you also provide an estimate on the error due to the used refractive index data? E.g. the data of STS by Biermann et al. seems to have some problems as shown in [Wagner et al.(2003)] (this is especially of interest for the composition retrieval of ternary solution particles mentioned in the abstract).*

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It is very difficult for us to assess the effects on our results of the errors in the *Biermann et al* refractive indices. Unfortunately, the analysis of Wagner *et al* contains approximations (the assumption of lognormal distributions, for example) that will also have associated uncertainties. More important in the present context, however, are the errors associated with the atmospheric (gas phase) interference in the satellite measurements, which are considerably larger than the errors introduced by the differences in the indices of refraction. Thus, we have chosen to use the (self-consistent) Biermann indices of refraction.

(5) p13279, l.22: *'The band near 820 cm⁻¹ (see inset) is characteristic of NAT (Hopfner et al., 2002).'*

-> *The reference to Höpfner et al., 2002 is not adequate here. The band has first been spotted by [Spang and Remedios(2003)] in CRISTA observations and has spectroscopically been identified as a NAT-feature by [Höpfner et al.(2006b)] in MIPAS/Envisat measurements.*

The references have been corrected

(6) p13287, Figure 2:

-> *Could you describe in the caption the shaded regions?*

Done.

(7) p13288, Figure 3 and p13290, Figure 5:

-> *Since the volume density of PSCs is in the order of $\mu\text{m}^3/\text{cm}^3$ i.e. of 10-12 either units or the factor might be missing in the caption of the y-axis.*

The units are $\mu\text{m}^3/\text{cm}^3$ and they were omitted from the caption by mistake. This has been corrected.

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Comments of anonymous referee #1:

1. page 13274, line 16. *The ATMOS and mkIV measurements have provided a valuable body of work on interpretation aerosol signatures in solar occultation data. In particular, Steele et al (applied optics 2003; 2006) has provided a through discussions of the retrieval of aerosol parameters of interest and the errors associated with these retrievals. Although Steele's work focused on sulfuric acid aerosols, the principles are completely relevant to Zatesky's work. I suggest the authors review those papers and incorporate relevant references into their paper.*

While the publications mentioned above are important, we do not feel that they are sufficiently relevant to our work to warrant reviewing them in our paper. The work reported in Applied Optics (2003) was based on observations from an aircraft using the JPL MkIV interferometer, while that in Applied Optics (2006) combined simulated measurements with an error analysis based on the ATMOS instrument, which was carried on the space shuttle during the 1980s. Both of these instruments are much larger and more sophisticated than the ACE instrument and the platforms in which they were mounted were much more stable and better serviced than the small satellite on which the ACE instrument is mounted. The errors, therefore would not be expected to be comparable to those of the ACE instrument and we feel it would be misleading for us to imply similarity. Instead, we have included an error analysis specific to our conditions. This gave rise to the error bars on figures 3 and 5. We feel that this is a more appropriate approach.

2. pg 13274 line 24 *The authors provide lat and lon boundaries for the 20 cases that they identified with a volume of more than 0.5 #956;m3/cm3. Are these boundaries or the spatial distributions of the ACE PSC observations consistent with other PSC climatologies? Would a map of the location of the cases be valuable to the reader?*

The lat/lon ranges given are those in which the observations contained PSCs. The satellite made on the order of 500 sunrise occultations during the time in question, but

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the purpose of this paper is not to compile a climatology of PSC observations, but to report the extension of the optical range, permitting us to quantify smaller particles. We agree that publication of PSC climatology is important and we hope to compile such a climatology for comparison with other similar data in a future publication, but that was not our intention in the present publication.

3. page 13278 lines 7-14 this is essential information - The authors write: "As a result, if the reference spectra of a given component reproduce one or more of the major features in the measured spectrum, then the presence of that component in the observed sample can be considered to be proven" What if there are many solutions that are equally good - how do we see any of that information in what is presented here: The reports only show a single result but don't talk about how this compared to other fits - are there a number that are within noise, or only one

Apparently there seems to have been a misunderstanding about our procedure. We have revised this part of the manuscript, beginning at the sentence quoted. We have also added a paragraph and an additional reference at the same place to help clarify the explanation. We hope that this makes it clear that our procedure returns only one single result, which is the one that is best in the least squares sense.

4. page 13278 line 20: The authors write: "The relative intensities othe characteristic features, of course, give the amount of that component present in the sample" Yet, no information about amount is presented in the results. Earlier in the paragraph the authors argue that there is residual gas phase information. How does that impact the amount and the best fit of the aerosols?

In this case as well, our description was incomplete; we relied on the information provided in the references to our previous work on this subject. The quoted comment refers to the fact that the coefficients returned by the least squares procedure depend on the intensities of the characteristic features (the extinction bands) of the respective components in the spectrum. This aspect of the procedure is described extensively in

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the two references we provided (Zasetsky *et al.* (2004) and Zasetsky *et al.* (2007)) so it is not appropriate to repeat the description here.

5. pg 13278 line 23 Earlier in the paper, 20 cases of significant cloud volume were cited, yet at this point the table has 6 cases with composition identification. What happened to the other 14 cases? I would be very interested in statistics of the composition of the 20 cases.

The cases in the Table are selected because they are representative of the compositions of the twenty cases analysed. It is not our intention to report a comprehensive climatology of PSCs in this paper; that will be done in later publications as appropriate. Here, we report the successful application of a retrieval method that uses an extended optical frequency range and we demonstrate the capability of this method to derive information on a wider range of particle sizes than has been possible before.

6. page 13279 and following. This section leads into discussion of the two cases shown with detail spectra. Are these representative of the only 2 distinct types of cases that were seen in the 20 first identified? Are they the best looking cases?

These are not the only two distinct cases, but they represent compositions and size distributions that were observed frequently, as stated in the manuscript. They were chosen also because they illustrate the important capabilities of the retrieval method that we wish to emphasize.

7. page 13280 line 17 and following Figure 4 illustrates the 'best fit' aerosol model to the observations, yet it appears that there are NAT like features in the spectrum between 1200 and 2000 cm^{-1} . This is a case where it would help to see all the fits that are within the uncertainty of the spectra. Rather than only identifying the minimum residual, it would be valuable to know what range fit the spectra. Alternatively, the fitting procedure finds the weighting of all of the aerosol models that minimizes the difference, yet the labeling only identifies on aerosol species. We all of the other aerosols weighted as zero?

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As pointed out in the description of our retrieval procedure, it does not do a series of fits. The procedure produces only one result –the one that is the “best” in the least squares sense. Also, there is no preselection of the components except the selection of all possible components of PSCs: Ice, NAT and the different STS components. All of the spectra were analysed by considering all of these possible components and the results are the coefficients returned by the fitting procedure, as described in the references on the basic method. Where there is a negligible amount of a component (as, for example the NAT in occultation sr7818), it is because the coefficient returned by the least squares procedure is negligibly small.

It is worth noting that this is a good illustration of a case where reliance on visual inspection of the spectra can lead to errors. In our laboratory work on cryogenic spectra of nitric acid aerosols, we have shown that it is difficult to distinguish between the spectrum of NAT and that of supercooled concentrated HNO_3 , if the latter has roughly the same mole fraction as the NAT. The nitrate bands look very similar on visual inspection but those of the crystalline material are sharper and shifted very slightly in frequency. These small differences, however, lead to significant differences in the goodness of fit parameters when the spectra are analysed in our procedure. In summary, we did not weight any of the possible components to zero; the distributions shown are simply the coefficients returned by the procedure, as described in this and previous reports of our retrieval procedure.

8. page 13272 The abstract and conclusions are written much more broadly than the paper. The abstract suggest that there is analysis of clouds for a month, and then some typical characteristics are provided. All we see in the body of the paper are a table with 6 of 20 cases, and spectral fits for two cases. There is also a disconnect where the high HNO_3 content - this is not discussed fully in the paper, nor do we get a sense of what fraction of the 20 cases had these characteristics.

We have rewritten the abstract to emphasize that the point of the paper is to report on the extended wavenumber range of the measurements.

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9. *There is a large body of theory about the conditions that result in NAT, ice, or STS PSCs. In the caption figures, temperature are mentioned. I think this paper would be much stronger if the authors addressed the composition of the clouds in light of the temperature and chemical information that ACE and others measure. This is hinted at in the concluding statements, but should be thoroughly analyzed and discussed in the context of the 20 cases first identified*

We have done calculations using the equilibrium models and these show that all of the compositions we report here are generally consistent with the temperatures and gas phase compositions in the regions where the observations were made. Unfortunately, such considerations are of very limited value in the actual retrievals, because the occultation method samples a very long horizontal path, which often includes a wide range of conditions that could be used in the equilibrium calculations. In general, gas phase profiles that are obtained from occultation measurements are reasonably accurate due to the large scale horizontal mixing in the stratosphere. PSCs, however, are limited in horizontal extent and might be located anywhere along the observation path. Since the temperatures along the observation path might vary considerably (depending on the location of the vortex), it is very difficult to compare PSC properties measured by occultations with those obtained in equilibrium calculations.

10. *Although the size distributions do have reported errors, Table 1, which reports aerosol volume, has no uncertainties on the quantities. It is important to also include that information*

To report uncertainties in the compositions would imply that the samples are homogeneous ensembles of internally mixed particles, whereas (see the discussion above) they might be externally mixed and in quite different locations along the line of sight. We report error bars on the particle sizes because these are independent of internal vs. external mixing and also because the emphasis of the paper is on the ability of the extended wave number range to give particle sizes in a wider range than is possible with the FTIR alone.

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Comments of anonymous referee #2:

Regarding the focus of the abstract: we agree that this was not correct and we have revised the abstract to remove any perception that we intended to report a climatology of PSCs. We have discussed this point in our replies to anonymous referee #1 so we will not belabour it here.

Regarding the number of dense clouds observed (twenty) and the number reported in detail (six), we have inserted a paragraph at the end of Section 2 (the top of page 5 in the submitted manuscript) to explain the situation further. The development of the methods we have used for this work was time consuming and the analyses were done manually. It is inappropriate to use such a method for the analysis of a large number of observations but we felt its successful development and application to be worth reporting. Although only twenty clouds were analysed, the six reported in detail are representative of the entire set and in the interest of saving journal pages, we did not feel it worth reporting the details of the other results that, to a large extent, simply duplicate the ones reported.

Regarding the composition of the STS solutions, we have added a paragraph at the end of Section 4 (bottom page 10 of the submitted manuscript) to clarify this situation.

Regarding the statement about the variability of physical and chemical conditions in the stratosphere, we agree with the referee's point that similar conditions can be found at different times and have removed the statement from the manuscript.

Regarding the explanation of the retrieval procedure, the referee is correct that it is not a standard method, but one that we developed over a period of about ten years in our laboratory – originally to measure aerosols in cryogenic flow tubes and later for application to satellite measurements. We have revised this section as requested, in an attempt to make the description clearer. In particular, we have dropped the “min” superscript and “P” subscript, since these mean simply “a minimisation on P”, which is already stated in the text. We have also added an additional verbal explanation of

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the procedure after the equation. This information is in the references to our previous work, but we feel its repetition here is appropriate to save the general reader the effort of looking up this literature. The weight matrix, W , must be applied to the whole quantity $(M - KP)$ because it essentially reduces the contribution of certain wavelength ranges (in the measurement and hence also in the computed spectra) to the computation of the chi squared. The double vertical bars, as the referee correctly points out, indicate that the terms are matrices.

We have tested our retrieval method on ice and water (as well as on commercial particles, as reported in Zsazsky *et al.* (2007)), rather than on ternary solutions of acids because the proof of the method's ability does not depend on the complexity of the system, but rather on the correctness of its retrievals. In order to know that the retrieval is correct, the properties (composition and size distributions, etc.) of the observed sample must be known. It is very difficult to prepare samples of chemically complex aerosols in which all of these properties are known accurately. The ice/water system and the commercial silica spheres on which we have tested the method are systems in which these properties are known accurately.

Regarding the extended wavenumber range obtained by including the imager measurements, the referee is correct that each of the two imagers gives only one frequency, in contrast to the FTS, which gives a high resolution spectrum containing thousands of frequencies. The FTS, however, covers the range of frequencies containing the chemical information in which there are absorptions (*i.e.* the chemical information) and the wavelengths recorded by the imagers contain only scattering information. Thus it is not necessary for the imagers to record broadband spectra as the FTS does. It is only necessary for them to provide values of the scattering intensity. The FTS and the imagers do look at the same region of space. It is correct that the field of view of the FTS is only approximately 4 km across, while the imagers look at the entire solar disk, but only those imager pixels that correspond to the FTS field of view are used in the retrieval analysis. Both instruments, of course, are aligned coaxially.

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The remaining minor comments of referee #2 have been addressed and the requested modifications have been made, with the following two exceptions:

- Page 13273, line 20: we believe the Danilin *et al.* reference is the one to which the referee refers. Professor Tabazadeh is an author of that paper.

- Page 13280 line 13: the text of the comment that is shown in the website has some places where it shows character codes instead of characters and as a result it is difficult to understand the request, but we have rewritten the sentence at the indicated line to try to make it clearer.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 13271, 2007.

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