Atmos. Chem. Phys. Discuss., 6, S2342–S2345, 2006
www.atmos-chem-phys-discuss.net/6/S2342/2006/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Probing ice clouds by broadband mid-infrared extinction spectroscopy: case studies from ice nucleation experiments in the AIDA aerosol and cloud chamber" *by* R. Wagner et al.

Anonymous Referee #2

Received and published: 7 August 2006

Review of: Probing ice clouds by broadband mid-infrared extinction spectroscopy: Case studies from ice nucleation experiments in the AIDA aerosol and cloud chamber

R. Wagner, S. Benz, O. Mohler, H. Saathoff, and U. Schurath

This manuscript details the experimental determination of the infrared spectral properties of crystalline ice particles produced by expansion cooling in the AIDA aerosol and cloud chamber. The authors obtain transmission infrared spectra of ice particles with

diameter equal to 1-15 micrometers, and then compare them to a T-matrix calculation of the transmission of crystalline ice particles with different aspect ratios. The authors state that determination of the aspect ratio from the spectra and measured size distribution alone is difficult. Therefore, retrieval of the particle shape of atmospheric ice particles may be difficult as well. In addition, the refractive index and modality of the size distribution need to be considered in each individual retrieval problem.

Overall, this is a very thorough and well written manuscript. The authors have collected sufficient data to support their claims. The manuscript is certainly useful for the community in general and specifically for remote sensing studies of cloud particles. The paper is acceptable for publication, but I have a few comments listed below. Could you please address these comments?

In general the figures are difficult to read because they are too small. Is it possible to split some of them into multiple figures rather than panels within one figure?

The experiment involves the heterogeneous nucleation of water ice onto mineral dust particles and homogeneous freezing of supercooled H2SO4/H2O droplets. The size of the mineral dust particles may affect the ice particle shape after nucleation, by altering the critical saturation ratio (the relative humidity where nucleation occurs). Did you attempt to model the particle shape and size based on your experimental conditions? What is the effect of using a cylindrical particle shape in your T-matrix calculation when the particles are hexagonal?

There are many physical properties which will affect the shape of ice particles grown under different conditions (pressure, temperature, etc.). I recommend that you search for articles by Kenneth G. Libbrecht at the California Institute of Technology, or see his website here: http://www.its.caltech.edu/~atomic/snowcrystals/ice/ice.htm.

For the homogeneous freezing of H2SO4/H2O droplets, what is the concentration of sulfuric acid in the droplet? Is it possible that you actually freeze a crystalline hydrated form of sulfuric acid, such as sulfuric acid tetrahydrate (SAT), or sulfuric acid dihydrate

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(SAD)? Did you see any evidence of these solid materials in your infrared spectra?

Is the apparatus for measuring the ice particle size on the same horizontal plane as the infrared beam? In Figure 1, it appears that they are not Are you certain that the size measurement is performed on the same particles that the infrared beam is measuring? It is possible due to settling that you are sizing particles that have a different size than those seen by the infrared beam? This is somewhat of a moot point since you retrieve your size based on the infrared spectra, but I am curious.

There are two other recent references of the optical constants of water ice in the near-infrared and far-infrared. They are:

Rajaram B, Glandorf DL, Curtis DB, et al. Temperature-dependent optical constants of water ice in the near infrared: new results and critical review of the available measurements APPLIED OPTICS 40 (25): 4449-4462 SEP 1 2001

Curtis DB, Rajaram B, Toon OB, et al. Measurement of the temperature-dependent optical constants of water ice in the 15-200 mu m range APPLIED OPTICS 44 (19): 4102-4118 JUL 1 2005

On Page 5742 you state “Obviously, this leads to a much poorer agreement between measured and calculated spectra in this atmospheric window region.” This statement is in reference to using the Clapp et al. optical constants instead of the Zasetsky et al. optical constants as plotted in Figure 12, panels 1 and 5 at1500-800 cm-1. It is difficult to tell from Figure 12 how much of a difference is really present. It seems that there is a difference, but it is difficult to quantify, and may not be obvious to the reader. Can you remove the word “obviously” from this statement?

The difference in the calculated spectra using different optical constants is very exciting to me personally and is useful to the community. Could you discuss this difference in more detail, perhaps adding a figure which shows the 1500-800 cm-1 region in more detail? I would like to see a plot of each calculation on the same plot (with the measured

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spectrum) to compare the difference in the calculated spectra, and a quantification of how much they each vary from your measured spectrum (e.g. a percent difference plot).

On Page 5747 you state that a unimodal fit gives a fit which which is “distinctly poorer than the bimodal one”. It is not clear from Figure 15 that the fit is that much poorer. I cannot deduce from the Figure how much poorer the fit is. Could you make the figure more explicit? Perhaps you could magnify the region to which you specifically refer in the text at 1500-1000 cm-1. Perhaps you could include a quantification of how much each size distribution affects the calculated spectrum, e.g. a percent difference plot with respect to the measured spectrum.

Also, on the next page (5748) you make the statement that it is “impossible to distinguish between uni- and bimodal size distributions and to retrieve the correct number concentration of ice crystals” and that the distinction is “extremely difficult”. But you made the statement on page 5747 that you can “distinctly” see a difference between the uni- and bimodal fit in relation to the measured data. This is very confusing. Are they distinct or is it extremely difficult to distinguish any difference? It is not clear from the manuscript which scenario is true.

In the conclusions (page 5749) you emphasize that the input parameters for the retrievals are critical for each retrieval problem due to differences in the quality of the refractive indices, the modeled asphericity of the particles, and the modality of the size distribution. They are listed in this order in the text. Does this imply that the quality of the refractive indices is more important than the modeled asphericity, which is in turn more important than the modality of the size distribution? If so, perhaps you should quantify the differences a little more thoroughly. If not, please include a statement saying this.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 5711, 2006.

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