Atmos. Chem. Phys. Discuss., 6, S2489–S2495, 2006
www.atmos-chem-phys-discuss.net/6/S2489/2006/


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.

**R. Wagner et al.**

Received and published: 17 August 2006

The authors would like to thank referee 1 for his thoughtful annotations. Please find below our answers to the individual comments.

1) Dependence of the ice particle shapes on variations in temperature and ice saturation ratio.

We completely agree that it would be worthwhile to analyse the AIDA ice nucleation experiments with respect to the sensitivity of the ice particle shape to the temperature and the time-dependent profile of the ice saturation ratio during ice formation and ice

crystal growth. Concerning the data sets which were analysed for the present work, however, we feel that there is insufficient information about the actual particle shape to perform such an analysis. As described in the manuscript, the information obtained from the infrared spectra is quite limited due to the size/shape ambiguity of the retrieval problem. In the past, an imaging instrument like the cloud particle imager CPI did not belong to the basis instrumentation of the AIDA chamber and was only available for specific measurement campaigns, usually dedicated to experiments at higher temperatures (T > 237 K) where there is also enough water vapour available for growing ice crystals to sizes which can easily be resolved by the CPI instrument. In the meantime, an optical particle imager has been constructed in our own laboratory which at least equals the resolving power of the commercially available CPI system. But still, at lower temperatures (237 < T/K < 195) the ice crystals might not grow to sizes to be resolved by the instrument. One could try to deliberately reduce the seed aerosol number concentration, or, more precisely, the number concentration of ice-active seed aerosol particles, so that fewer ice crystals are formed which then could grow to larger sizes. However, such a strategy would also significantly reduce the count rate of our optical particle counters for the ice particle detection. Thereby, the quantitative analysis of the expansion experiments with respect to the determination of nucleation rates and ice activation spectra, so far one of the primary goals of the expansion experiments, would be made more difficult.

A promising instrument development might be the second generation probe SID2 (http://strc.herts.ac.uk/pi/over.html). The device should be capable of classifying the shape of cloud particles down to micrometer sizes, relying on the analysis of the recorded angular scattering patterns. In a recent AIDA measurement campaign, SID2 was already attached to the chamber for a series of expansion experiments at 210 K involving different seed aerosol particles with varying critical ice saturation ratios, thus leading to different time profiles of Sice during the activation. The data analysis is still in progress - but it is certainly an interesting point to look at the predominant ice particle habits which are formed at this temperature and whether there is a pronounced



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habit change for the different employed seed aerosols. Note that the information obtained from the AIDA expansion experiments would rather resemble an atmospheric in situ cloud observation (the ice crystals experience variable growth conditions due to the changing water vapour concentration during the expansion) than be comparable to laboratory studies where ice crystals where grown at nearly constant temperature and humidity conditions to infer habit diagrams as function of temperature and ice supersaturation, e.g.:

Bailey, M., and Hallett, J., Growth rates and habits of ice crystals between -20° and -70° C, J. Atmos. Sci., 61, 514-544, 2004.

Libbrecht, K. G., and Yu, H., Crystal growth in the presence of surface melting: supersaturation dependence of the growth of columnar ice crystals, J. Cryst. Growth, 222, 822-831, 2001.

Coming back to our present work, there is indeed only one limited conclusion regarding the ice crystal habits which can be inferred from the FTIR spectra analysis. As already noted in the paper, we observed for the vast majority of expansion experiments that our infrared spectra of smaller crystals (equal-volume sphere diameters less than 6 micrometers) could be accurately fitted with compact shape parameters (aspect ratios from 0.5 to 3.0) and that there was a nice agreement between the deduced ice particle number concentrations and the independent OPC measurements. This implicates that within the broad range of different ice cloud formation conditions which were covered by our experiments (temperature range from 238 to 195 K, critical ice supersaturations ranging from a few percent for very ice-active mineral dust samples like Arizona test dust up to 50-70 % for homogeneously nucleating sulphuric acid droplets) rather compact ice crystal habits were predominantly formed. But due to the solution ambiguity of the FTIR retrieval, we are unable to further distinguish between e.g. thick plates (aspect ratio 2-3) or short columns (aspect ratio 0.5).

We propose to include some aspects of this discussion into the revised manuscript.



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We will change the paragraph beginning with line 24 on page 5738 as follows:

"In summary, our results demonstrate that the approach of modelling aspherical ice crystals by circular cylinders with moderate aspect ratios of $\phi = 0.5 - 3.0$ is appropriate for retrieving number densities and diameters from the FTIR extinction spectra of smaller ice crystals over a wide range of experimental conditions. It is well known that the ice crystal growth, and thereby the ice crystal habit, strongly varies with temperature and ice supersaturation (e.g. Bailey and Hallett, 2004; Libbrecht and Yu, 2001). To deduce the ice crystal habit diagrams from laboratory measurements, the ice crystals are usually grown at nearly constant temperature and humidity conditions. In contrast, the ice crystals which are formed during an AIDA expansion experiment experience variable growth conditions due to the changing water vapour concentration and temperature in the course of the expansion. Nevertheless, we can state that within the broad range of different ice cloud formation conditions which were covered by our experiments (temperature range from 238 to 195 K, critical ice supersaturations ranging from a few percent for very ice-active mineral dust samples like Arizona test dust up to 70 % for homogeneously nucleating sulphuric acid droplets at 195 K) rather compact ice crystal habits were predominantly formed within growth times of the order of a few minutes. But due to the solution ambiguity of the FTIR retrieval, we are unable to further distinguish between the formation of e.g. thick plates ($\phi = 2 - 3$) or short columns ($\phi = 0.5$). Further information about the actual shape of small-sized ice crystals may be obtained in future chamber studies employing the second generation probe SID2 (http://strc.herts.ac.uk/pi/over.html). This new device should be capable of classifying the shape of cloud particles down to micrometer sizes."

2) Determination of ice nucleation rates from AIDA expansion experiments.

Yes, from the WELAS recordings shown in Fig. 2 one can deduce the rates of new ice particle formation. If also the time-dependent aerosol volumes are known, one can derive the volume-dominated nucleation rates. An example for the quantitative analysis of the WELAS measurements is given in the paper by Benz et al. (2005), addressing



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homogeneous ice nucleation in supercooled cloud droplets:

Benz, S., Megahed, K., Möhler, O., Saathoff, H., Wagner, R., and Schurath, U., T-dependent rate measurements of homogeneous ice nucleation in cloud droplets using a large atmospheric simulation chamber, J. Photochem. Photobiol. A, 176, 208-217, 2005.

As for the heterogeneous ice nucleation experiments, the paper by Möhler et al. (2006) - in the meantime published as ACP article - suggests a formulation for the formation rate of ice crystals generated by heterogeneous ice nucleation on mineral dust particles:

Möhler, O., Field, P. R., Connolly, P., Benz, S., Saathoff, H., Schnaiter, M., Wagner, R., Cotton, R., Krämer, M., Mangold, A., and Heymsfield, A. J., Efficiency of the deposition mode ice nucleation on mineral dust particles, Atmos. Chem. Phys., 6, 3007-3021, 2006.

Both papers are already cited at the end of the first paragraph in the introduction.

3) Are the efforts to obtain the absolute size distribution worth doing it?

Yes, we definitely think it is worth the effort - in the context of the major scientific goals which we pursue with our chamber studies. Besides the determination of freezing thresholds (critical ice saturation ratios) for various aerosol types, the quantitative analysis of the expansion experiments with respect to nucleation rates and ice-activated fractions of seed aerosol particles makes it necessary to precisely infer the absolute, time-dependent ice particle number concentrations. Fortunately, we can rely on different instruments for the ice particle detection. In many scenarios, the data from the optical particle counters are indeed reliable. But we have also indicated in our discussion (page 5740) that there are cases where the operation of the OPCs to deduce Nice(t) might fail, either because the ice crystal mode cannot be properly separated from the remaining seed aerosol particles or because very small ice crystals are not



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detected by the instruments. In these cases we have to rely on the FTIR retrieval results for Nice. So we considered it worth doing a crucial test of how reliable the FTIR retrievals with respect to the absolute number size distributions really are - just by comparing the FTIR retrieval results to the OPC measurements in selected scenarios where the OPCs provide precise Nice(t) data. As stated in our summary, we also consider our results as a potentially valuable "guideline for other laboratory studies of ice nucleation, which frequently have to rely on FTIR extinction spectroscopy without independent information on the accuracy of the retrieval results."

Although we have already indicated in the introductory part of our manuscript that the actual ice particle number concentrations are of prime importance for the quantitative analysis of our expansion experiments, we have missed to do so in the abstract and therefore propose to add the following statement (page 5712, line 9):

"The measured infrared spectra were fitted with reference spectra from T-matrix calculations to retrieve the number concentration as well as the number size distribution of the generated ice clouds. The precise evaluation of the time-dependent ice particle number concentrations, i.e., the rates of new ice particle formation, is of particular importance to quantitatively analyse the ice nucleation experiments in terms of nucleation rates and ice activation spectra."

4) Number size distributions or volume concentrations as a measure of accuracy?

With the additional statement in the abstract it should now become clear from the beginning that the absolute number size distributions are of particular concern for the analysis of our chamber experiments. But it is of course important to realise that in spite of the sometimes severe size/shape ambiguities in the number size distribution retrievals the overall ice volume concentrations show much less variation; see also e.g. the discussion of Fig. 9 on page 5739: the difference in the retrieved ice volume concentrations for the unimodal and multimodal number size distribution fit scenarios is well below 10%. Hence, the ice water contents of the clouds are much more accurately



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defined by the FTIR retrieval and we really should emphasise this in our manuscript. We propose to add a paragraph in the Summary and outlook section on page 5750, line 15:

"Given that the determination of nucleation rates and ice activation spectra is a main objective of the AIDA expansion experiments, our work has focussed on the performance of the FTIR retrieval for deriving accurate number size distributions of ice crystals. In spite of the frequently encountered severe size/shape ambiguities in these number size distribution retrievals, we want to emphasise that the simultaneously retrieved ice water content of the clouds shows much less variation. In our case studies, the different size/shape representations of the ice crystals usually gave rise to a less than 10% difference in the retrieved ice water content."

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