Atmos. Chem. Phys. Discuss., 6, S577–S582, 2006
www.atmos-chem-phys.org/acpd/6/S577/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Efficiency of the deposition mode ice nucleation on mineral dust particles" *by* O. Möhler et al.

O. Möhler et al.

Received and published: 17 April 2006

Reply to Paul DeMott

We acknowledge the comments and questions from all three referees of our paper. The comments are indeed helpful to improve the manuscript. Some important points have been addressed by all reviewers. Nevertheless, we think that all referee comments deserve separate answers. This is our reply to the comments of Paul DeMott.

Answer to general comments:

When we started a few years ago to run the AIDA aerosol chamber as an expansion chamber at cirrus cloud temperatures, we decided to use Arizona Test Dust (ATD). This sample was available in large enough quantities to develop and improve methods

of dust dispersion and characterisation in our laboratory. During the first experiments we mainly measured, in series of experiments at different temperatures, the threshold relative humidities for the onset of ice nucleation. Only recently, reliable measurements of ice crystal number concentration became available which we then related to the respective ice saturation ratio to obtain the activation spectra discussed in the present paper. Because AIDA cloud expansion experiments are time consuming, and because we obtained only a limited amount of appropriate dust samples with a large enough mass fraction of particles with diameters less than 3 μm, we were able to conduct only a limited number of experiments with the two desert dust samples. We want to acknowledge again here that the Asian dust sample AD1 was provided by Lothar Schütz from the University of Mainz, Germnany, and the Saharan dust sample was collected by Kahled Megahed in Egypt.

The present paper is restricted to experiments were reliable data sets of both ice number concentration and ice saturation ratio were available, and to temperatures well below the homogeneous freezing temperature of about -37 °C where significant deposition nucleation occurred. Some more characteristics of heterogeneous ice nucleation on mineral dust at higher supersaturation, close to the homogeneous freezing threshold, and higher temperatures, where other heterogeneous freezing modes like immersion or condensation freezing may become important, are discussed in the companion paper by Paul Field et al. This appeared tu us as a reasonable separation to write up the results in two companion papers. Process modelling will be the subject of further papers which are in preparation, as indicated in the manuscript and pointed out correctly by Paul DeMott. We believe that any of these papers contain a variety of new methods, aspects, and possible applications to be published as separate papers.

We already mentioned in our replies to referees #1 and #2 that the present paper should be considered as a first case study to demonstrate the capabilities of the AIDA chamber for investigating heterogeneous ice nucleation on dust aerosols. We think that from the naturally limited number of experiments there is already clear evidence

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that the deposition ice nucleation on the surface of mineral particles may be described with only a limited number of parameters as function of the temperature and the ice saturation ratio. This is in agreement to previous work, which also formulated heterogeneous ice nucleation as a freezing spectrum rather than a stochastic process like the homogeneous freezing rate equation (e.g. DeMott et al., 1998, Geophys. Res. Lett. 25, 1387-1390). We agree that more investigations, including process modelling, are certainly needed to get a better idea of the variability of the ice activation spectra with temperature, ice saturation ratio, and origin or properties of the mineral particles. More expansion runs with freshly dispersed dust samples at different cooling rates and the aspect of ageing in repeated expansion runs with natural dust samples, as suggested by Paul DeMott, are certainly on top of our own list of priorities.

We are open to any cooperations and further discussions of the topic of heterogeneous ice nucleation of mineral particles which we believe is important to properly assess the impact of dust aerosols on cirrus cloud formation. We are happy to share our own dust samples with other groups and appreciate receiving any further samples which are thought to be of atmospheric relevance and which are available in large enough quantities for laboratory studies.

Answers to specific comments:

1. We will mention the phenomenon of preactivation noted by Roberts and Hallett (1968) in the discussion section. The point that deposition ice nucleation on the mineral dust particles may not be treated in the same stochastic sense as homogeneous freezing will also be more clearly mentioned in the manuscript. We agree that this is an important issue that should be addressed in future laboratory and modelling studies.

2. The introduction will be modified according to comments by Paul DeMott. We apologize for the incorrect and incomplete reference to the work by DeMott et al. (1998). A respective discussion of the heterogeneous freezing nuclei spectra inferred from the wave cloud investigations will be added to the manuscript.

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3. We used a bulk density of 2.5 g cm^{-3} and a dynamic shape factor of 1.5 to convert the aerodynamic diameter measured with the APS into the volume equivalent sphere diameter shown in the figures. The charge correction has been made for the data shown in the figures. Obviously, the correction is incomplete which may partly be explained by the fact that the contribution from multiple-charged particles larger than 0.8 μm are not considered in the correction algorithm of the SMPS instrument. It cannot completely be ruled out from the present measurements but appears likely that some more uncertainties of the charge correction remain unresolved. However, for the conclusions of the present paper, these uncertainties appear to be of minor importance. Concerning the combination of the mobility and aerodynamic measurement techniques to obtain the full aerosol size distribution, we think that the volume equivalent sphere diameter is well defined in relation to the mobility and aerodynamic diameter. Nebulous may be the concept of dynamic shape factors to account for aspherical particle shape. We think that the volume equivalent sphere diameter in fact underestimates the surface of highly asperical and structured particles like minerals.

4. Yes, we implied in the statement that there are no smaller particles to be lost by diffusion. We will explicitly mention that.

5. Please note the logarithmic scale in Fig. 2. Because the (minor) fraction of dust particles larger than 0.5 μm in diameter actually interferes with the detection of small ice crystals, we have determined the number concentration of ice particles larger than 0.8 μm from the relative increase of the OPC count rate (see Equation 2). From the fact that the dust particles are not expected to significantly grow by water uptake under the conditions of our experiments (see our replies to referees #1 and #2), and from a comparison with the retrievals from in situ FTIR extinction spectra, we concluded that any increase of the OPC count rate can be explained by growing ice crystals. There is no significant detection delay due to settling of ice crystals to the sampling tube of the OPC in our experiments because of the strong mixing of the chamber volume. This will be mentioned in the updated manuscript. So far we have not yet systematically

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investigated the impact of mixing on the crystal growth rates. We expect this to be only a minor issue for the small and almost spherical crystals regarded here. Mixing and advection aspects may be more important in experiments were ice crystals larger than 50 to 100 μm are grown and characterised at well controlled temperatures and supersaturations in the AIDA chamber.

6. From a number of 20 individual temperature sensors we know that the whole interior of the AIDA chamber is well mixed as discussed in our paper. The water data from the chilled mirror instrument sampling from a fixed location is in excellent agreement with the in situ TDL method that measures an average water concentration along a light path through the chamber. Good agreement was also achieved between the SID and OPC-Welas data sets. The OPC-Welas used a vertical sampling tube at the bottom of the chamber, the SID instrument a horizontal sampling tube at a location about 2 m above. Furthermore, excellent agreement is achieved between the ice crystal number concentration measured with the optical probes and retrieved from the FTIR extinction spectra (another paper on this subject will soon be submitted to ACPD) . The latter are measured along an infrared light path across the whole chamber at a hight of about 4 m above the bottom of the chamber. These are only part of the arguments and findings which may demonstrate that the AIDA expansion runs discussed in the present paper are conducted under well controlled and defined homogeneous conditions regarding temperature, water vapour, relative humidity, and ice crystal number concentration. Uncertainty and variability of temperature and relative humidity have also been discussed in our reply to referee #1. Again, settling and deposition effects may become more important in experiments at warmer temperatures with larger ice crystals.

7. Resonant vibrations means that the TDL signal showed an increased noise due to mechanical vibrations of the multipath cell probably induced by the vacuum pumps.

8. One of the major conclusions is that new ice particles nucleate in an expansion run as long as the ice saturation ratio S_i increases. The formation of new ice crystals almost stops as soon as S_i reaches its peak value. Furthermore, there is experimental

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evidence that the ice activated fraction of the mineral dust particles as function of S_i is independent of the rate of change of S_i. For clarity, we will replace the cooling rate by dS_i/dt in the discussion sections.

9. and 10. See our answer to the general comments above and our replies to referees #1 and #2.

Technical corrections:

Will be considered.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 1539, 2006.