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

O. Möhler et al.

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We acknowledge the comments and questions from all three referees of our paper. These 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 from referee #1.

Answers to general remarks:

Concerning the different modes of ice nucleation we followed the nucleation terminology by Gabor Vali (Bull. Am. Met. Soc. 66, 1426, 1985) who defined the deposition nucleation as the formation of ice in a supersaturated vapour environment and the immersion freezing as the ice nucleation on a nucleus suspended in a supercooled water droplet. It is also common to refer to immersion freezing if the nucleus is immersed in a supercooled solution droplet instead of pure liquid water.

From our experimental data it is difficult to determine the amount of liquid water adsorbed to the particle surface before ice crystals start to grow. Some evidence that liquid water uptake is limited to at least a minor volume fraction of the particles can be derived from the forward scattering channel of an in situ scattering and depolarisation instrument (see Möhler et al., Atmos. Chem. Phys. 3, 211-223, 2003). This channel is very sensitive to particles growing by water uptake, either liquid or ice. Unfortunately, reliable data from this instrument was obtained only in a few experiments. In all cases, significant scattering increase was always correlated to the detection of growing ice crystals by an optical particle counter and by FTIR extinction spectroscopy. Unfortunately, the depolarisation channel of the same instrument which selectively detects the formation of a-spherical ice particles, was not available. The data from the scattering instrument is not included in the present paper because the data sets are incomplete and difficult to interpret without having available all channels. Recently, a new version of the same instrument proved to more reliably and sensitively measure both intensity and depolarisation from in situ aerosol and ice particle scattering and may therefore be more capable to differentiate between liquid water uptake and ice growth.

In the AIDA experiments, ice nucleation mainly occurred at a relative humidity with respect to ice between 100 and 120 %. At the temperatures between 223 and 196 K, this corresponds to relative humidities with respect to liquid water between about 50 and 70 %. Furthermore, Arizona Test Dust showed almost no hygroscopic growth in laboratory studies at room temperature (see e.g. Vlasenko et al., Aerosol Sci. Technol. 39, 452-460, 2005; Gustafsson et al., Atmos. Chem. Phys. 5, 3415-3421, 2005). From this and from the experimental evidence discussed above we conclude that the ice nucleation mode discussed in the present paper can be interpreted as deposition nucleation without significant liquid water uptake by the mineral particles prior to the nucleation and growth of ice crystals. From the experimental data it is not possible to infer direct information about the mode of ice nucleation.

Answers to specific comments:

A short explanation of deposition and immersion mode ice nucleation according to the definition by Vali (1985) is added to the introduction (*page 1541, line 25*). On *Page 1541, line 10* we will add references to the work by Kärcher and Lohmann (J. Geophys. Res. 108, 4402, doi:10.1029/2002JD003220, 2003), Lohmann et al. (J. Geophys. Res. 109, D16204, doi:10.1029/2003JD004443, 2004), and Haag et al. (Atmos. Chem. Phys. 3, 1791-1806, 2003).

As already mentioned in the manuscript, it is difficult to compare the elemental compositions of different dust samples because they may markedly depend on the size distribution of the individual particles in the sample. It is certainly beyond the scope of this paper to discuss and compare the natural variety of elemental and mineralogical composition of desert dust collected from the ground or sampled on aerosol filters for later analysis. In general, only limited information about the mineral composition can be derived from the bulk elemental analysis of dust samples. The presence of large amounts of SiO_2 and Al_2O_3 indicate the presence of layer silicates, but also of SiO_2 modifications, most likely quartz. All dust samples used in the present studies showed larger fractions of SiO_2 which indicates the presence of quartz particles. Minor abundance of kaolinite is also likely. Recent FTIR analysis of the SD2 sample showed signatures of calcite. Recent EDX analysis of individual particles (Martin Gallagher, private communication) showed larger particles in the Saharan dust sample to mainly contain Si. Other elements like Al, Fe, and Ca were more prominent in the smaller particles. In the ATD sample, the smaller particles also showed major Si peaks which may be a result of the grinding process. This gives some evidence that more quartz particles may be present in the ATD aerosol compared to AD1 and SD2. The XRF analysis of ATD agrees with the composition given by the manufacturer except for SiO_2 mass fraction which was about 5 % below the range given by the manufacturer. It should be noted that the mass fractions do not sum up to 100 % because some of the less stable

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fractions like carbonate are lost when the sample is melted into a borax matrix at high temperature prior to the analysis. We will additionally indicate this mass loss in Table 1 of the manuscript.

The above discussion will be included in the manuscript. The mineralogy of the dust samples used in the AIDA experiments will be discussed in more detail in a separate paper.

On *page 1550, line 19* we suggest to add: The small optical sensor can be operated at the same temperature as the aerosol chamber without additional heaters and with only a very short sampling tube. Therefore, the risk of ice particle sampling losses and evaporation is markedly reduced.

The whole interior of the chamber volume, except for a few centimetre thick transition layer to the chamber walls, is well mixed with a ventilator located in the bottom part of the volume. Therefore, all aerosol and ice particles experience during the course of an experiment the same change of mean temperature and relative humidity within a fluctuation and variability range of about ± 0.3 K and ± 4 to ± 5 $\%$, respectively (see also discussion of uncertainties below). Therefore, the relation between f_i and S_i is well defined in each experiment. Only the peak S_i depends on the rates of ice particle formation and S_i increase as a consequence of the mass transfer of water vapour to the growing ice particles. This should however not affect the relation between f_i and S_i.

The statement that f_i only depends on S_i is valid for any individual AIDA expansion experiment where ice nucleation occurs in a narrow temperature range of 1 to 2 K. The parameter a actually varies with temperature as already discussed in the manuscript. The temperature dependence seems only week for ATD but is more obvious for the desert dust samples. We will point out the temperature dependence of the parameter a more clearly in the manuscript. To the conclusions we will also add a warning to modellers that rigorous application of the parameterisation for atmospheric dust particles and for temperatures beyond the range of our experiments may involve significant uncertainties (see also comments by Paul DeMott to which we will respond in a separate author comment).

We agree that the discussion of the uncertainty of S_i is somewhat misleading and incomplete. S_i is calculated from the ratio of the actual water vapour e_w measured by TDL and $e_{s,i}$ calculated from the mean gas temperature T_g. The TDL result is scaled to the MBW chilled mirror instrument that measures the frost point temperature with an accuracy of ± 0.1 K. Therefore, the uncertainty of e_w from TDL, after scaling, is only ± 1.3 % at 220 K and ± 1.7 % at 190 K. The mean gas temperature is measured with an accuracy of ± 0.3 K, resulting in an uncertainty for $e_{s,i}$ of ± 3.9 % at 220 K and ± 5.2 % at 190 K. Therefore, the mean S_i related to the mean T_g is measured with an accuracy of about ± 4 % at 220 K and ± 6 % at 190 K.

The above discussion assumed that the ice nucleation observed in the chamber is initiated and controlled by the mean gas temperature T_g. An additional systematic uncertainty of the measured ice nucleation onset may arise from the fact that the ice nucleation is initiated by a temperature somewhat lower than the mean temperature (it is unlikely that the ice formation is initiated by the warmer fraction of the chamber volume). The temporal and spatial variability around the mean temperature is typically less than ± 0.3 K. If the observed ice formation would be initiated at $T_g - 0.3$ K instead of T_g, $S_i(T_g)$ discussed in the present paper would systematically underestimate the ice nucleation thresholds by about 4 % at 220 K and 5 % at 190 K.

The manuscript will be changed in order to more clearly point out the different sources of error from measurement uncertainty and variability in the aerosol chamber. The measurement uncertainty of S_i will be added to the data shown in Figures 7 to 9.

For clarity, let us summarize here that the typical uncertainties of measured temperatures are ± 0.1 K for the MBW chilled mirror frost point temperature and ± 0.3 K for the AIDA gas and wall temperature sensors. The variability of gas temperatures is typically

± 0.3 K inside the the cold box, ± 0.2 K inside the aerosol vessel under static conditions (no pumping), and ± 0.3 K inside the aerosol vessel during pumping expansion experiments.

It is already mentioned in the abstract that for our experiments the heterogeneous ice nucleation of dust was approximated by an exponential function of S_i.

Technical corrections:

All technical corrections suggested by the referee will be included in the revised version of the manuscript.

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

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