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Interactive comment on “Deposition freezing on mineral dust particles: a case against classical nucleation theory with the assumption of a single contact angle” by M. J. Wheeler and A. K. Bertram

Anonymous Referee #1

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General comments

This is an interesting paper that analyzes deposition freezing of two clay minerals, namely kaolinite and illite, using recently developed nucleation models. The main finding is that the data cannot be described using classical nucleation theory and the assumption of a single contact angle. This is in accordance with other recent ice nucleation studies. There are some issues that should be resolved prior to publication in ACP.

Particle size distributions should be analyzed and discussed in more detail. Welti et al. (2009) have published a size distribution of kaolinite from Fluka that peaks at ca. 500

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nm with only a minor fraction of the particles larger than 1000 nm. This is in strong contrast to the determination performed in this study by static laser light scattering yielding an average particle size of 8000 nm for the same kaolinite. One reason for this discrepancy might be aggregation of kaolinite particles in the water suspension. Particles that are smaller than 1000 nm cannot be discerned by optical microscopy. If a large fraction of such particles were present on the hydrophobic glass slide, the nucleated fraction would be much smaller. To see whether this was the case, a sample loaded with the clay minerals using the ultrasonic bath technique should be investigated by electron microscopy. It would also be interesting to know whether nucleation always occurred on large (i.e. optically discernable) clay particles. Also, the assumption of spherical particles leads to a considerable underestimation of surface area of supermicron clay mineral particles and should be refined using measured surface areas of kaolinites and illites.

Heterogeneous ice nucleation on kaolinite (Fluka) has recently been measured by Lüönd et al. (2010) in immersion mode and analyzed using the same nucleation models. They obtained contact angle values of ca. 90° compared to values between $3\text{--}14^\circ$ in this study. Does this imply that kaolinite is a much better ice nucleus in deposition than in immersion mode? The authors might comment on this.

Deposition freezing rates depend on supersaturation with respect to ice (S_{ice}) and on temperature. In the present study, only trajectories reaching $S_{ice} = 1$ at 242 K have been investigated and the dependence of nucleation onsets on temperature is not discussed. Eastwood et al. (2008) have shown for kaolinite (Fluka) that there is little temperature dependence of S_{ice} in the temperature range from 236 – 246 K. However, the temperature dependence might become more important for the smaller particle loadings used in this study. The authors should discuss this issue and state for what temperature range their parameterization is valid.

The Bertram group presented immersion mode ice nucleation results of kaolinite (Fluka) already in Eastwood et al. (2008) yielding values that are at the upper end

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of the error bars of the values presented here. Some values in this study are at $S_{ice} < 1$ (including error bars). Do the results presented in this study have a bias to low S_{ice} ?

Welti et al. (2009) also used kaolinite (Fluka) for their ice nucleation studies in immersion mode. The comparison with this study should therefore be more detailed.

Specific comments

Page 21176, line 1: The authors should specify what exact quality of illite they purchased from Clay Minerals Society.

Page 21181: how was equation 8 applied? By numerical integration or analytically?

Page 21182: exp is missing in equation 10.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 21171, 2011.

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