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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Studies of heterogeneous freezing by three different desert dust samples" *by* P. J. Connolly et al.

Anonymous Referee #3

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General comments:

This paper presents freezing experiments with three different desert dust samples in the AIDA chamber in a temperature range of -12° C to -33°C. Ice crystal shapes, sizes, and concentrations have been measured with various instruments. For data analysis, a new quantity, the ice-active surface site density (IASSD) has been introduced. The data is divided into two different scenarios, depending on whether droplets or ice crystals form first. Simulations with the ACPIM model have been used to interpret the experimental data with this new assumption. These data end the new approach to parameterize ice formation may be useful for the atmospheric community. I therefore recommend this paper for publication after minor revisions have been made which are discussed below:

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Introduction: In recent years, many groups have worked with mineral and desert dusts to study ice nucleation and possibly our theoretica understanding of the underlying mechanism. I suggest to cite and also discuss briefly the more recent works in the introduction as well (e.g. Eastwood et al. 2008, Zimmermann et al. 2008, Vali et al. 2008). In the results section, the authors frequently refer to ice crystal habit measurements e.g. from Bailey and Hallett. It would be good to mention these and their importance in the introduction already because it seems a major topic in the discussion, especially because the CPI images have a prominent position there.

IASSD: I think that new approaches to describe ice nucleation and freezing phenomena are needed, especially those which incorporate active sites. However, I have some comments and question regarding the IASSD approach in this paper: First, it seems that the method described here is rather similar to the one in Marcolli et al (2007). I would appreciate a reference to this work and a discussion of the similarities and differences between both methods. I also see a link to the work presented in Vali (2008) which should be discussed. For the model simulations: Is the IASSD calculated for each time step, independently of previous time steps? How is the problem treated that the distribution of active sites in reality is fixed and does not change with time, e.g. an active site of a given quality sits on one particle and stays there. A density function implies to me that each time a number of active sites for a given particle surface is used to calculate if a given droplet freezes, the nucleating properties of the particle in this droplet may be different. In other words: Does the model conserve the properties of the ice nuclei and their specific active sites for a simulation? How is the situation treated, that an IN may have many active sites? When only one causes the droplet to freeze, all remaining active sites need to be removed from the population. Related to this: Is the number of active sites similar to the number of ice crystals? This may all be covered by the poisson statistical approach but it is not entirely clear to me. I suggest to clarify this a bit more. Since there are many recent attempts to improve heterogenous nucleation theory (e.g. Marcolli et al. 2007) with new data this paper would be much stronger by tying in to these attempts rather than introducing a new scheme which is

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more empirical.

Freezing vs. deposition nucleation: In the discussion it seems to be unquestionable, that immersion freezing of CCN-activated droplets is the source for the ice crystals. However, there is in principle the possibility that deposition nucleation may be a competition to immersion freezing above water saturation. For a discussion of this it would be helpful to have at least one figure of an experiment of scenario 1 where ice activated earlier than the droplets. It would be interesting to know if all dust particles are activated as CCN at t1 or if there are remaining dry particles which may act as deposition nuclei in parallel.

Modeling: In sections 4.3.1 – 4.3.3 in many model runs a tuning of parameters like total water was needed to match the experimental data best. The authors should discuss briefly what the cause for these initial discrepancies might be and what the implication is for the quality of the simulations.

ESEM-EDX (section 4.4): The second sentence in this section promises a discussion of chemical and morphological differences as the source for the different behaviors of the dusts in the freezing experiments. However, the whole section is rather descriptive and the expected discussion is missing, even in the discussion and conclusions section. As it is now, section 4.4 is rather unconnected to the topic of rest of the paper and might be even removed.

Specific comments:

page 475, line 12: Wouldn't be “heterogeneous deposition nucleation” be more correct? page 475, line 18: did you mean: does not require THE presence of... page 479, line 11: Liquid DROPLETS formed... The rest of the sentence should be re-written, it sounds a bit strange to me. Page 479, line 26: over-predicts? Page 479, line 28: ...is the fact THAT the modeled.... Page 480, line 2: ...prediction of THE ICE crystal concentration(s)? Page 480, lines 12/13: add commas after ice concentration and drop concentration. Page 480, line 18: Same comments as for p. 479, l. 11.

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Page 481, line 26: Same comments as for p. 479, l. 11 and p. 480, l.18. Page 487, line 1: ...nucleation OF three different... Page 487 lines 25- 2(next page): I suggest to re-formulate this sentence and divide it into two. E.g. start a new sentence at: Whether this is....

Figures: 2, 4, 5, 6, 8, 9, 10, 11 are pretty hard to read in the current size. At least the fonts should be bigger. However, this may be due to the smaller page size in ACPD compared to ACP and may be better in the final print. From the set of figures 5, 6 ,8 ,9 ,10, and 11 I suggest to use only a smaller set of representative figures which highlight the features which are important to the discussion. Furthermore: It seems to me that all figures represent scenario 2 in which droplets form before ice. I would appreciate if at least one figure for scenario 1 could be included, since the trend for this scenario is already highlighted in Figure 2.

Eastwood, M. L., Cremel, S., Gehrke, C., Girard, E., and Bertram, A.ăK. (2008). Ice nucleation on mineral dust particles: Onset conditions, nucleation rates and contact angles. J. Geophys. Res., 113.

Marcolli, C., Gedamke, S., Peter, T., and Zobrist, B., (2007). Efficiency of immersion mode ice nucleation on surrogates of mineral dust. Atmospheric Chemistry And Physics, 7(19), 5081–5091.

Vali, G, (2008). Repeatability and randomness in heterogeneous freezing nucleation. Atmospheric Chemistry And Physics, 8(16), 5017-5031.

Zimmermann, F., S. Weinbruch, L. Schutz, H. Hofmann, M. Ebert, K. Kandler, A. Worringen, Ice nucleation properties of the most abundant mineral dust phases. J. Geophys. Res., 113, D23204.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 463, 2009.

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