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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heterogeneous freezing of droplets with immersed mineral dust particles – measurements and parameterization" *by* D. Niedermeier et al.

Anonymous Referee #1

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General comments:

The paper shows experimental evidence that Arizona Test Dust (ATD) particles exhibit different ice nucleation abilities depending on the coating of the particles. All experiments address to freezing in the immersion and were performed in the LACIS chamber @ Leipzig. The experiments cover a temperature range between 233.15 K and 240.65 K. A monodisperse particle size of 300 nm was coated with either an organic or different inorganic substances. The results indicate that the uncoated ATD particles and the particles coated with either succinic acid or small amounts of sulphuric acid are the best ice nuclei. On the other, ATD particles coated with ammonium sulphate are

the less effective ice nucleus. Classical nucleation theory was applied to parameterize calculate heterogeneous ice nucleation rates for the differently coated ATD particles. The paper deals with a hot topic since ice particle formation is not yet fully understood, and it is an important process for mixed-phase and cirrus clouds formation.

I have an understanding problem with the temperatures given in the paper. The authors mainly talk about the wall temperature of the LACIS chamber, which leads than to the supercooling temperature, Ts. However, I guess one wants know the real temperature within section 6 and 7 of the chamber, where the ice freezing occurs. Do the particles reach there equilibrium and thus have the same temperature as the ice on the walls? If so, then Ts is the temperature in the chamber, right? If not, then the authors have to give the real temperatures in the chamber. Is it possible that the information is given in the Hartmann et al. 2009 draft (unpublished results)?

In the discussion section, I would be nice to have a comparison with other studies together with the possible atmospheric implications of the results. I believe that such a discussion would increase the depth of the paper.

It is nevertheless a well written manuscript and I recommend it to be published as a full ACP paper, but only after the authors have stated on the general comments as well as on the points below:

Specific comments:

Page 15828 lines 7: Do the authors now the temperature in the chamber so precisely? The investigated temperature range primarily addresses to mixed-phase clouds activation temperatures. Can the same conclusions be drawn also for cirrus temperatures?

Page 15828 lines 13-16: I had a hard time to understand that sentence. All experiments were investigated in dilute solutions, and thus the water activity of the droplets is always close to 1. And now you conclude that water activity is not related to the investigated freezing process? Please rephrase that sentence.

Page 15829 lines 25 -30: This discussion could be more detailed. On what base do Meyers et al. J. Appl. Meteorol 1992 conclude that deposition mode can also occur at water saturation? Why do the authors doubt that condensation freezing takes place at all?

Page 15831 lines 1-2: Which of the mentioned studies do contradict each other and why?

Page 15831 line 25: Why 300 nm? Is that the mean diameter of the ATD size distribution?

Page 15831 line 25: Which additional instrumentation? Since the two cited papers are unpublished papers, I would like to get here some additional information.

Page 15833 Eq.3 How well does the right term of the equation agree with the vapour pressures given by Murphy and Koop Q. Murphy, D. M.; Koop, T. Q. J. R. Meteorol. Soc. 2005? Why did the authors use the expression by e.g., Rogers and Yau 1996, when both vapour pressures are well parameterized? What about the uncertainty and temperature dependence of lf?

Page 15833 line 15: Why did the authors take ΔF and vi as constants? There are parameterizations given in Zobrist et al. JCP 2007 paper.

Page 15833 line 19: ΔF is no longer treated as a function of temperature, as mentioned above. So the T-dependence is redundant.

Page 15834 line 16: How does the temperature gradient within the LACIS influences the results? See also general comments.

Page 15834 lines 20: Why is jhet almost constant?

Page 15835 line 3: The validity of Eq. 8 does not cover the entire investigated temperature range.

Page 15838 lines 26-28: So the simulation can calculate the hygroscopic growth, but

not the ice nucleation. But the goal of this study is to investigate ice nucleation. What does “half quantitative” mean? I don’t see the direct gain of the simulations for this study.

Page 15839 lines 9-10: This means that the particles are completely dry again. What are the thermodynamic conditions within the LACIS for a given wall temperature? See also comment below and general comments.

Page 15839 lines 13-17: What is the temperature within LACIS for a wall temperature of 233.15 K? Since homogeneous ice nucleation should start @ $\sim$235K.

Page 15841 lines 12-14: What is the size of the drops? Could you mark in Fig.4 where the ice starts to occur for the first time? Could the authors add in Fig. 4 also the temperature within LACIS?

Page 15845 line 5: So what is the typical water activity of such a particle?

Page 15845 line 9: What means almost constant temperature. According to Fig. 11, jhet can increase up to one order of magnitude within 1 K (e.g., red curve), so jhet is very sensitive to temperature.

Page 15847 lines 19-21: This conclusions is drawn without showing the water activity data. So either show the data or omit that sentence. Could you investigate more concentrated solutions to prove that conclusion? This would be great data.

Page 15847 lines 27-28 : Do you really believe that the total surface is considerably changing due to a small coating?

Page 15858 Fig.5: Can you give the size of the particles or droplets and not the log(channel)?

Page 15859 Fig.6.: See comment Fig.5.

Page 158639 Fig.10.: How sensitive are the experiments with respect to fice? How many droplets are investigated for one Ts?

Page 15864 Fig. 11: I was wondering about the different slopes of jhet for the differently coated ATD particles as a function of temperature. The results indicate the some IN are better at low temperatures and some at higher temperatures. Do the authors have any idea why? The slopes are clearly smaller than that for homogeneous ice nucleation rate of pure water, which can increase by roughly one order of magnitude decreasing the temperature from 237 K to 236.5 K (e.g., Benz et al. J. Photochem. Photobiol. 2005). Possible reasons?

Technical corrections:

Page 15828 lines 15: change "... before the freezing occurred." to " ... before freezing occurred."

Page 15828 lines 18: replace "...allows us to determine..." by for example " ...suggest ..."

Page 15828 lines 22: Please rephrase that sentence.

Page 15829 lines 5: Either use "ice-forming nucleus" or "ice nucleus" trough out the entire manuscript. I suggest the latter one.

Page 15829 lines 12: Melting point of water or of ice?

Page 15829 lines 12, 15 and 18: I would rather argue with super-saturation or frost point than with the melting point of water (or rather ice).

Page 15831 line 12: add " ice" before "heterogeneous freezing". The author should be consistent within the publication

Page 15832 line 12: Give typical units for jhet, ns ...

Page 15834 line 6: replace "... crystallization velocity of water ..." by "... crystallization velocity of ice ...".

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15827, 2009.