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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Time dependence of immersion freezing" *by* A. Welti et al.

Anonymous Referee #2

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Review of "Time dependence of immersion freezing" by Welti et al.

The authors have investigated the time dependence of immersion freezing of kaolinite particles at several temperatures. This dataset provides an excellent test for different parameterizations used to describe ice nucleation. I congratulate the authors for carrying out elegant experiments that appear to be carefully done. Although the results are excellent, the writing and discussion should be improved before publication in Atmospheric Chemistry and Physics. Below are specific comments.

1. The experiments look at freezing from approximately 5% frozen to 95% frozen. Although useful, this doesn't necessarily cover all atmospheric conditions. For example, under some conditions only a small percentage of the dust particles in the atmosphere may be activated as ice nuclei (IN). Do the results for these measurements at high fraction frozen extrapolate to low fractions frozen? This should be discussed.

2. Some of the references don't seem appropriate. For example, on page 12625, lines 25-29, the authors have the following sentence: "In summary, many major mixed-phase cloud characteristics (light scattering, precipitation formation, chemistry) depend on the size of the cloud droplets, which in turn is linked by the Bergeron-Findeisen process to the rate at which ice nucleates in the interior of the supercooled cloud droplets (Tabazadeh et al., 2002)." Tabazadeh et al. 2002 focuses on surface nucleation of water droplets, so this reference doesn't seem appropriate. Please carefully check all references to ensure only appropriate references.

3. In the introduction the authors do mention briefly that others have experimentally investigated the stochastic and singular component in immersion freezing. However, the authors could do a better job of describing what has been done previously and how their experiments are different than previous work. For example have others looked at time dependent freezing of mineral dust, and do the conclusions from the current study differ from previous conclusions?

4. Abstract, line 17-20. Should this sentence read ". . .yields an equivalent effect of -1K temperature shift for an increase in times scale by a factor of 10"?

5. Is it possible that in your experiments some of the kaolinite particles are not activated as CCN? This is probably discussed in Luond et al. 2010, but it is worth repeating this discussion here.

6. Figure 4 shows a curve for homogeneous freezing. How was this calculated and what are the uncertainties of this calculation? The homogeneous curve overlaps slightly with the heterogeneous freezing data at 236 K. Has this been taken into account when fitting the heterogeneous data? If not please explain why it is valid to ignore this overlap.

7. Page 12630, line 19-21. Please quantify what you mean by minor. Less than 1%?

8. Page 12630, line 24. Please quantify what you mean by small. Less than 1%?

9. Page 12632, line 22-24. “Compared to homogeneous freezing however, immersion freezing has a gentler slope in the frozen fraction. This indicates a temperature dependent increase in the reduction of the energy barrier to ice nucleation.” I struggled to understand what the authors are trying to say in the second sentence. Please restate to make this point more clear.

10. Page 12633, line 1-3. Please state here what the error bars represent in the figure and how they were determined.

11. Page 12638. The authors should also state that although the alpha-pdf model gave the best fits, this model was not able to fit all the data within experimental error. This probably should also be mentioned in the conclusions and the abstract.

12. Page 12640, lines 219-25. “The rapid change in the frozen fraction for increasing residence time, calculated for the active site model is unexpected, as this model, although also based on CNT, is conceptually closer to the singular model than the alpha-pdf or the stochastic model.” To me this is not surprizing. Both the alpha-pdf and the active site model can approach the stochastic model depending on the parameters used in the model. In your case, the fits have given parameters that make the active site model closer to the stochastic model compared to the alpha-pdf model.

13. Please include references for Equations 13 and 14.

14. Page 12640, lines 23. “Probably the assumption of a 6nmˆ2 active site area and the resulting number of active sites generated per particle is too high.” I don’t think this discussion is necessary and could be removed especially since it is speculative.

15. Page 12642, line 9. Do the authors have a reference to show that mineral dust is partly hydrophobic.

16. Page 12642, line 15-16. “Also the observable time dependence of the frozen fraction would be distinctly different for different IN, as can be already seen in the variance between the two tested particle sizes.” I don’t see how the variance between

the two tested particle sizes necessarily tells you anything about the possible difference between particles with different chemical compositions. The authors have not proven that the differences they observed between the two sizes is due to a difference in chemical composition.

17. Page 12642, line 18-19. “The IN activity could also be an impermanent feature, as from the moment the aerosols are released into the atmosphere until they become active in clouds they are exposed to sunlight and chemical compounds in the air that might lead to loss or increase of their nucleation activity. . .” The authors could add references here.

18. Page 12642, line 25-26. “Therefore we recommend to include a time-dependence in numerical calculations of the evolution of mixed-phase clouds.” It should be mentioned that this recommendation is based on only the current study, which utilized one mineral from one vendor, whereas mineral dust in the atmosphere can contain many different minerals. At this point the authors could also discuss if all other studies carried out with mineral dust are consistent with this recommendation.

19. Page 12643, lines 23-26 and page 12644, lines 1-12. This section was confusing to me. On one hand, this paper shows that a single contact angle and classical nucleation theory cannot describe the experimental data well, but on the other hand, in this section the authors are using a single contact angle and classical nucleation theory to derive a surface tension. It is not clear to me that the surface tensions from this analysis have any physical meaning.

20. Page 12644, lines 13-17. It would be interesting to see a comparison between predictions with the average active contact angle of the population and the time dependent data, similar to the comparisons shown in Figure 7. Otherwise it is hard to judge the accuracy of the average active contact angle of the population.

21. Section 6.2. The authors do not refer to any figures in this section. Does the discussion refer to figure 10?

22. Figure 2. I assume the data shown in this figure is based on calculations. This should be stated and references to the calculations included. Do the calculations for droplet radius assume monodispersed droplets?

23. Figure 3. I don't see a black line in this figure.

24. Figure 4, 5, and 7. Please clarify what the error bars represent in these figures. Do the error bars represent the standard deviations of the measurements (one sigma or two sigma) or do they represent confidence intervals.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 12623, 2012.