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Atmospheric Chemistry and Physics Discussions

Interactive comment on "New particle formation in the sulfuric acid-dimethy lamine-water system: Reevaluation of CLOUD chamber measurements and comparison to an aerosol nucleation and growth model" *by* Andreas Kürten et al.

Anonymous Referee #3

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In this study, formation rates published by Almeida et al. (2013) for ternary sulfuric acid (SA) nucleation with dimethylamine (DMA) in the CLOUD chamber are re-analyzed with a method that takes into account self-coagulation. The authors argue that particle formation rates at 1.7 nm are more than a factor of 10 higher than those reported by Almeida et al. (2013), which would imply that SA-DMA new particle formation is significant at lower DMA gas-phase concentrations than previously thought. The revised formation rates agree well with rates calculated by a kinetic aerosol model at different particle diameters. Therefore, the authors conclude that nucleation for the conditions

studied here proceeds at rates that are collision-controlled.

General comments:

I think this manuscript is well written and contains some interesting results and conclusions that makes it suitable for publication in ACP. However, since the manuscript focuses mainly on a re-evaluation of particle formation rates from the paper by Almeida et al. (2013), I think more information needs to be given on the approach used by Almeida et al. for extrapolating their formation rates. I suggest that the authors add a schematic diagram or a table illustrating how 1) Almeida et al. have calculated their formation rates and 2) how the authors of the present study have calculated their formation rates. Such a diagram should also include information on what instruments were used when deriving the particle formation rates, and the necessary corrections. For instance, the authors state on lines 335-338 that Almeida et al. (2013) made an extrapolation from 3 to 1.7 nm when deriving their formation rates at 1.7 nm. How was this extrapolation done? Furthermore, the authors of the present study use data from the smallest SMPS size channel to calculate the formation rate. As the authors admit on lines 344-345, "the smallest SMPS size channels need to be corrected by large factors to account for losses and charging probability, which introduces uncertainty". How were these corrections made, and how large were the corrections relative to the actual measured number concentrations? In addition, the authors assume on line 366 that the growth rate is independent of size which adds more uncertainty. How large are these uncertainties compared to the "error" resulting from the extrapolation method used by Almeida et al. (2013)?

Another general comment I have is related to the fact that there is another recent study focusing on nanoparticle growth for the SA-DMA system in the CLOUD chamber by Ahlm et al. (2016), where most authors of this manuscript were co-authors. In that study, model simulations and measurements with three different instruments indicated an increasing particle-phase DMA/SA molar ratio with increasing particle size due to a decreasing Kelvin-effect of DMA with increasing size, from ∼1.5 to 20 nm. The results

of that study appear, at least to this reviewer, to be inconsistent with the view provided in this manuscript that nucleation and growth up to ∼80 nm are completely collision-controlled. I think there needs to be some explanation, or at least, discussion of this issue.

Specific comments:

1. In the Almeida et al. (2013) paper, the ACDC model reproduced ternary SA-ammonia formation rates almost perfectly, but somewhat overpredicted ternary SA-DMA formation rates compared to observations in the CLOUD chamber. I think it could be worth mentioning that the conclusion within this manuscript, that ternary SA-DMA formation rates in the CLOUD chamber were underestimated by Almeida et al., brings the formation rates much closer to predictions by the ACDC model.

2. Sect. 2.1: Please describe the SMPS measurements including corrections.

3. Line 35: The word “advanced” is not very useful for the reader. It is better to try to explain as clearly as possible the difference between the approach used here and the method used by Almeida et al.

4. Line 40: “modeled and measured size distributions show good agreement”. I think it should be mentioned that this was for one nucleation event that you studied in detail, unless you have analyzed other events as well.

5. Lines 137-138: To what extent was dimethylamine oxidized by OH within the chamber during these events? Were any oxidation products detected and may these have contributed to new particle formation?

6. Line 321: How high is “relatively high”, and how do the authors know there was no sulfuric acid in the chamber? Do the authors think this is a general problem with using a CIMS for measuring sulfuric acid?

References

Ahlm et al. (2016). Aerosol Sci. Technol., 50, 1017-1032, 2016. Almeida et al. (2013). Nature, 502, 359-365.

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