Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-902-AC3, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Experimental study of H_2SO_4 aerosol nucleation at high ionization levels" *by* Maja Tomicic et al.

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Received and published: 15 December 2017

First of all we would like to thank you for all the comments. This reply provides answers/discussion of the main four comments, to ensure that we understand them correctly and that you accept our ideas for fulfilling them. We appreciate the specific comments as well and intend to answer them in the final version.

(1) "*Currently it is stated that the parametrization from Dunne et al. (2016) is expanded to lower sulfuric acid concentrations and higher ion concentrations. This is an overstatement since Dunne et al. explored new particle formation for a wide range of sulfuric acid, ammonia, temperature and ion concentrations. However, the*

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present study examined NPF at only one temperature (295 K) and one ammonia mixing ratio (1200 pptv). For these conditions, the sulfuric acid and ion concentrations were varied. Given the fact that the authors did not resolve the chemistry of the nucleating clusters, it is also speculative that NH3 is the only possibility of explaining the high NPF rates at low sulfuric acid. In principle, other contaminants (e.g. organics or amines) could probably also explain the data. Therefore, without having identified the chemistry of the nucleating clusters the statements about the chemical parameter space that the current study explores need to be reformulated"

We will make sure to clarify that we only expand the parametrization from Dunne et al. (2016) at a temperature of 295 K and ammonia mixing ratio of 1200 pptv. We will also stress that it is not only NH_3 that explains the high NPF rates since the amount of other contaminants is unknown. We will rephrase some statements about the ternary nucleation rates to clarify that the nucleation pathways in this study are unknown.

(2)"*The results presented in figure 6 are not in agreement with previous studies. At 2e+07 cm^{-3} of sulfuric acid, the contribution from binary neutral nucleation to the total neutral nucleation rate is as high as the contribution from the other channels (binary ion-induced, ternary neutral and ternary ion-induced). For this warm temperature, it is impossible that binary neutral nucleation yields a formation rate of $\sim$0.04 cm-3 s-1 (at sulfuric acid of 2e+07 cm^{-3}) since the clusters evaporate too rapidly (see, e.g., Hanson and Lovejoy, 2006; Ehrhart et al., 2016; Duplissy et al., 2016). At these conditions, binary neutral nucleation should be completely negligible and even the binary ioninduced component should be negligible compared to the ternary channels (Ehrhart et al., 2016; Duplissy et al., 2016). Therefore, a re-evaluation of the different nucleation channels is necessary as well as a more thorough inter-comparison to previous studies. Given the presented results and the results from previous studies it seems very likely that the nucleation rates presented are by far dominated by the*

ternary channel."

We will rewrite the evaluation of the nucleation channels to ensure consistency with results from previous studies. We will rethink the use of figure 6 and possibly leave it out. Also, the fit could be used as a tool in the analysis in order to explain the steep increase in nucleation rates that we see for [H_2SO_4]>$2 \cdot 10^7$ rather than to conclude anything about the nucleation channels. Finally we will emphasize that our results should be seen more like an expansion of the Dunne et al. parametrization to higher ionization levels (for the investigated parameters) than a separate parametrization.

(3) "*Regarding the identification of the relevant nucleation scheme, one possibility would be to use the CI-APi-TOF as an APi-TOF. This should indicate what fraction of sulfuric acid cluster ions contains ammonia molecules (or any other contaminants); based on Schobesberger et al. (2015) it might also be possible to derive an estimate of the ammonia contaminant level. Given the fact that the experiments were made at high ion concentrations, the APi-TOF should yield strong signals, which would shine a light on the nucleation pathway.*"

This is an interesting idea for future studies that we will keep in mind. Especially the idea from Schobesberger et al. of using the the ration of NH_3 to H_2SO_4 in the clusters to determine the corresponding concentration ratio. Unfortunately we cannot redo the experiments at the present.

(4)"*The data evaluation process needs to be explained in more detail. Especially, an additional figure should be added that shows the time development of particle concentration, UV light intensity, H2SO4 concentration, temperature, etc. Based on that figure it should be explained over what period the data for the derivation of J were*

averaged."

We will provide the requested figure and include an explanation of the experimental run sequence and a more in depth description of the data analysis possibly including a correction to the nucleation rate.

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