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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Immersion freezing of water and aqueous ammonium sulphate droplets initiated by Humic Like Substances as a function of water activity" *by* Y. J. Rigg et al.

Anonymous Referee #3

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The manuscript by Rigg et al. describes a study of heterogeneous immersion ice nucleation in aqueous ammonium sulfate solution droplets. The authors present ice nucleation experiments using ice nuclei (IN) composed of natural organic matter – Leonardite and Pahokee peat – as surrogates for organic constituents in atmospheric aerosol particles. The experiments were performed using micron-sized aqueous droplets with different ammonium sulfate concentrations employing an experimental setup that has been characterized thoroughly in previous studies. The experimental data were then analyzed using various descriptions (both time-dependent and deterministic) of heterogeneous ice nucleation that are currently being used in the literature. From fitting these descriptions to the experimental data it was possible

to extract a parameterization of immersion ice nucleation induced by Leonardite and Pahokee peat IN. One important result is that the data are in agreement with the so-called water-activity approach thus allowing for the application of the derived parameterizations also for other types of solute.
The topic of this manuscript is of interest to experimentalists and modelers working in the area of ice nucleation and cloud microphysics and, hence, fits well into the scope of ACP. Both the experiments as well as the data analysis appear to be carefully performed and executed. The manuscript is well written and the length of the text, the figures, and the supplement are all appropriate. In conclusion, I consider the paper being publishable in ACP, after the following minor comments have been taken into account.

Minor comments:

1. P. 4921, L.9-11: I think it would be appropriate to reference the paper by Karcher and Lohmann, JGR 108(D14), 4402 (2003) in this context.

2. P. 4922, L.13-15, 23-26, and Table 1: It was not clear to me whether the concentration of LEO and PP given in Table 1 is that after filtration or before (it is mentioned in the text that $\sim$75% of the PP and LEO is lost during filtration).

3. P. 4924, L.10-13: I do not understand why a partial solubility of PP may explain the difference in surface area measurements using SEM and BET.

4. P. 4925, L.16-19: I assume that the agreement of the measured ice melting points to the ice melting curve (from AIM?) implies a negligible solubility of LEO and PP. Is this correct? This would imply that the dissolved fraction of LEO and PP does not change aw significantly.

5. P. 4926, L.1-2: I think it would be appropriate to reference the papers by Young and Leboef ES&T (2000) and by Koop et al. PCCP (2011) in this context.

6. P. 4926, L.3-7: I think it would be appropriate to reference the paper by Karcher and Lohmann, JGR 108(D14), 4402 (2003) in this context, see above.

7. P. 4928, L.17: In Figs.2a and 3a, isn't Jhet(exp) increasing stronger than exponentially with decreasing T? Is it possible that the very steep increase of the last points at low temperature is already affected by homogeneous ice nucleation? May this contribution result in a potential bias in terms of the derived Jhet(exp)? Please proof that you can exclude such contributions from homogeneous ice nucleation.

8. P. 4930, L.25-26: Regarding the increase in alpha with decreasing temperature seen here (and also in previous studies): Can you speculate about any rational explanation/reason for this observation which implies that the compatibility between the IN and ice in the immersion mode becomes worse at lower temperature?

9. Fig. 4. and 5: The Jhet(exp) model seems to be significantly off the data in the panels for the lowest aw. Can you provide an explanation?

10. P. 4938, L.10-11: I am puzzled by the statement that 'the alpha(T)-model yields alpha and Jhet values directly from the experimental data, thus, no fitting is involved.' Isn't alpha fitted such that Jhet matches the observed Jhet values?

Technical comments:

11. P. 4927, L.20: Typo: 'Jhet *is* calculated for all. . .'

12. P. 4928, L.17: I assume 'a temperature decrease by *about* 10 K. . .' is meant?

13. P. 4931, L.12-14: I assume with 'that can *lie* outside of the . . . probed in the laboratory' you rather mean to say 'that can *be predicted also* outside of the . . . probed in the laboratory'?

14. Fig. 4. and 5: The frozen fraction f is without unit, hence I suggest to remove 'a.u.'

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 4917, 2013.