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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heterogeneous ice nucleation of viscous secondary organic aerosol produced from ozonolysis of α-pinene" *by* K. Ignatius et al.

Anonymous Referee #1

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Ignatius present a study in which they study the ice nucleating properties of secondary organic aerosol which was generate and characterised in terms of its physical state. They conclude that up to 20% of SOA particles can nucleate ice in the 'deposition' mode in a narrow temperature range of between -36.5 C and -38.3 C and at saturations between 1.3 and 1.4. Heterogeneous nucleation in this regime is a striking conclusion and of significant potential importance for ice formation in cirrus clouds. I support the publication of the paper once the following comments have been addressed:

1) SPIN is a new instrument and I think this might be the first published ice nucleation data making use of this instrument. Hence, the paper needs to 'validate' the instrument

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as thoroughly as possible. The homogeneous freezing results for ammonium sulphate are valuable in this respect. However, this paper is about heterogeneous nucleation and I would like to see some heterogeneous results for a material which has been studied in the past and a comparison made.

2) P35722 ln 7-8. The Kramer et al. and Cziczo et al. references are for cirrus clouds, not mixed phase clouds. There are plenty of papers out there which discuss mixed phase clouds including two relatively recent review articles:(Hoose and Möhler, 2012; Murray et al., 2012).

3) P35722 ln 20. The reference to Zobrist et al. in the context of 'suggestions that these SOA particles could play a role in ice nucleation' is incorrect. Zobrist et al. (2008) suggested the opposite – they suggested that glassy aerosol would not nucleate ice. This was one of the reasons why it was so surprising that Murray et al. (2010) showed that aqueous glassy aerosol could nucleate ice under upper tropospheric conditions. Also, Virtanen et al. (2010) did not discuss SOA nucleating ice in any detail – it is just mentioned in the abstract.

4) P35723, ln 10-13. The authors refer to modelling studies. It is important to also note the modelling studies performed by Murray et al. (2010) and also Price et al. (2015). These studies are highly relevant here.

5) P35723, ln 25-30. The authors suggest that the SOA in Mohler et al. (2008) liquified and froze homogeneously. The SOA only nucleated well above water saturation. This implied that it was so hydrophobic that it did not take up water until an extreme supersaturation.

6) P35731, I am confused by the discussion of the dependence on size here. It seems to be stated in the text that there is no significant size dependence of ice nucleation, but when I look at fig 4 I see that there is a clear dependence on size. Bigger particles nucleate ice at a lower S.

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7) The authors need to discuss and use the results from Price et al. (2015) throughout their paper. In Price et al. the diffusion coefficient of water in the water soluble fraction of SOA from alpha pinine was quantified over a range of temperatures and RHs. Using these measurements the uptake of water into a solution droplet was modelled for a variety of conditions. They conclude that 'SOM can take hours to equilibrate with water vapour under very cold conditions' and 'for 100 nm particles predicts that under mid- to upper-tropospheric conditions radial inhomogeneities in water content produce a low viscosity surface region and more solid interior, with implications for heterogeneous chemistry and ice nucleation' This is highly relevant and complementary for this paper. For example, when making a judgment concerning the timescale of transformation from a glassy solid to a liquid the pertinent quantity is diffusion. The diffusion coefficient can be used to estimate this timescale. This should be done, for example, on P 35733 (ln 18-30) where the authors note that they observe ice nucleation at an RH well above the RH at which they observe these aerosol to transform to liquid aerosol.

8) P35736, ln 5-10. When commenting that biogenic SOA may be mixed with sulphates and that this may be important for ice nucleation, it would be sensible to bring in the work of Wilson et al. (2012) who showed that glassy aerosol containing a mixture of carboxylic acids and ammonium sulphate also nucleated ice.

9) P35736. A discussion is needed about how their fraction 'frozen' is far higher than previous investigations at the AIDA chamber (Wilson et al., 2012; Murray et al., 2010). Could this be related to the particle size? The studies at the AIDA chamber were with smaller particles than those used here.

10) The authors use the term deposition mode. This should be caveated. Is this true deposition? i.e. deposition of ice directly onto glassy aerosol particles? Or could nucleation occur in the layer of lower viscosity solution which the modelling of Price et al. (2015) (and others) show will form when RH around glassy aerosol is increased? This layer of lower viscosity water has also been experimentally observed in the work from Jonathan Reid's group.

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11) The data for SOA proxies from (Wilson et al., 2012; Murray et al., 2010) must be shown in Figure 6. I suggest the authors focus on the onset RHs where the aerosol was thought to start in a glassy state.

12) I note that the co-authors of the Leinhard et al. ACP 2015 paper have posted a comment, so I won't write much about this. But, I reinforce that comment and state that the Leinhard paper should be discussed in the present manuscript.

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