

Interactive comment on “Competition between water uptake and ice nucleation by glassy organic aerosol particles” by T. Berkemeier et al.

Anonymous Referee #1

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Recently researchers have suggested that glassy secondary organic aerosols can cause ice nucleation. However, it is still not clear if these particles act as ice nuclei under atmospheric conditions because the processes involved are numerous and complex. In this manuscript the authors have come up with an elegant way to model these processes. Confidence in the modelling approach was gained by comparing model results with recent laboratory results. Then, model results were used to predict atmospheric conditions that may lead to ice nucleation by glassy secondary organic aerosols. The paper represents a substantial contribution to scientific progress on the topic. I expect this study will motivate future laboratory studies to constrain better some of the physical parameters needed for modelling ice nucleation on glassy secondary organic aerosols. I highly recommend this paper for publication in ACP after the authors

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have adequately addressed the following comments.

General comments regarding uncertainties in the calculations:

Figure S4 shows FDRH of four different SOA precursors as well as their quasi-equilibrium glass transition, with uncertainties represented by shaded bands. For clarity and ease of reading, it would be helpful to state in the figure caption the source of the uncertainties (i.e. what uncertainties are considered in the figure). I assume the uncertainties come from uncertainties in T_g, org , kGT and κ_{org} .

Two sets of uncertainties are reported in the manuscript. The first set (shown in Figure S4) uses $kGT = 2.5 \pm 1$. The second set of uncertainties (shown in Figure S6, orange shaded) includes a wider range of kGT . From the document I could not tell which set of uncertainties is most applicable to the present modelling studies. Please clearly discuss which set of uncertainties is most applicable to the present modelling study and justify. This may have been discussed in Koop (2011), but additional discussion in the current manuscript would be very helpful.

There are large uncertainties in calculated FDRH when all the parameters are taking into account (see Figure S6). These uncertainties should be mentioned in the conclusions and perhaps the abstract. In addition, it may be helpful to the community if the authors gave some directions on measurements that are needed to help reduce the uncertainties.

Page 16473, line 15-20. If the uncertainties in the model predictions are considered, are the freezing results above 230 K still inconsistent with the model predictions?

Other general comments:

Page 16461, line 20-25. Full deliquescence relative humidity (FDRH) is defined as the point where the entire particle is homogeneously mixed and its water activity corresponds to that of a liquid (i.e. it is larger than that of the quasi-equilibrium glass transition). Why is the constraint of “entirely homogeneously mixed” needed? I can

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image a case where the water activity corresponds to a liquid everywhere in the particle, but the particle is still not homogeneously mixed due to slow diffusion in the liquid. Does this situation ever arise?

Specific comments:

Page 16461, line 5: I believe it should be "from" instead of "form"

For alpha-pinene are the authors using proxies corresponding to ozonolysis or OH oxidation. Please specify for clarity.

Page 16462, "Ice nucleation regimes": Through the section, please reference the up-draft velocity and the corresponding type of clouds, when possible (e.g. at page 16462, line 19).

Page 16467, line 14: Should "latitude" be replaced with "altitude".

Figures:

All figures. Please indicate what size of droplets the homogeneous freezing line corresponds to.

Figure 1 caption: I believe there is a typo for "typical".

Supplement:

Table S1, column 1. References are not properly formatted. For example see "Kautzman 122/". Also there is some nomenclature in this column that is not defined. For example what does "CARB" represent.

I can't see Table S3 mentioned anywhere in the text (either main text or supplemental). Please discuss somewhere how these parameters are used.

Line 52: ("cf figure") give a number to the figure if possible.

The caption of Table S2 does not appear to be consistent with the data in the table.

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