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Atmospheric Chemistry and Physics Discussions

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

T. Berkemeier et al.

t.berkemeier@mpic.de

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Referee comment 1:

Organic aerosols are prevalent in the atmosphere, but little is known about their impact on ice nucleation. The paper presented here modeled the phase transitions of model organic aerosols representing both anthropogenic and biogenic SOA due to updrafts in clouds. The phase transitions for each organic aerosol were used to predict the ice nucleation regime the particle would be involved in (deposition, immersion, homogenous) and at which temperature and RH. The novel approach and results presented in this paper will greatly help in modeling ice nucleation due to organic aerosols in cirrus and other high altitude clouds. Future studies including other organics will be inter-

esting and are necessary to assess the magnitude of the impact of organic aerosols on IN globally. This paper does a good job of visualizing the competing effects of aging (increased hygroscopicity and increased temp of glass transition) and the concepts presented. The schematics were very helpful and informative. I recommend this paper for publication.

Response:

We thank anonymous referee 2 for her/his positive review of the article and appreciate the suggestions she/he made to further improve the quality of the paper. We will include changes towards all comments in the revised version of the manuscript as detailed below.

Referee comment 2:

General Comments: The stated temperature range where glassy aerosols would be important for ice nucleation is in the range of homogeneous ice nucleation. Therefore, how important globally do you think these organic aerosols are? There is some discussion of impacts at the end of the paper, but more discussion on this temperature range and impacts would make the paper stronger. Do the authors have any suggestions for other types of organic aerosols that should be modeled next? Or predications for which organic aerosol types are most important for ice nucleation globally or in certain regions?

Response:

We see our work as an explorative microphysical modelling study which describes and quantifies the different ice nucleation regimes of organic aerosols for the first time. It is less focused on the global consequences of these mechanisms. While such a study would be very worthwhile, we think quick implications are difficult to draw and an in-depth discussion would be outside the scope of our paper. In Sect. 4, we already point out that aerosol particles originating from aromatic precursors and highly

aged particles derived from monoterpenes have the highest potential to affect ice cloud properties. Despite the fact that aromatic SOA particles might be produced predominantly in the exhaust plumes of urban and industrial areas, it is hard to confine their range of ice nucleation activity to certain regions due to their long lifetime in the atmosphere. The global impact is also hard to estimate, for many different reasons. Not only are the thermodynamic input parameters for our microphysical model (such as diffusivities) still subject to large uncertainties, but also is the actual microphysical process of ice nucleation on glassy particles still little understood, which is reflected in the huge spread of experimentally determined ice nucleation onsets that vary strongly between different investigated substances and experimental techniques. We would like to refer to the discussion of this issue in the revised manuscript, see also author comment to anonymous referee 3.

Referee comment 3:

Specific Comments: Page 16454 Lines 8-9: "low temperature and low humidity" Please specify the range of temperature and humidity or add < XX °C, < XX % RH for more clarity.

Response:

Since glass temperature and glass relative humidity are interconnected, we decided to add exemplary glass transitions temperatures for two values of relative humidity that can be encountered in the atmosphere. We will add the following sentence:

For example, typical α-pinene derived secondary organic aerosol particles are expected to be in a glassy state below about 260 K at 30 % relative humidity, whereas at a higher humidity of 80 %, such glass transition is expected at approximately 215 K (Koop et al., 2011).

Referee comment 4:

Page 16460 Lines 25-26: The definition of RHg is given, but as written it was confusing.

Perhaps write "The quasi-equilibrium glass transition of the aqueous organic, RHg, is shown in grey." or similar.

Response:

We accept the suggestion of the referee and will change the sentence accordingly.

Referee comment 5:

Page 16461: Figure 1 and the discussion of it in the text may benefit from labeling the circles as ABCD or 1234 and referring to them in the text by the number or letter for more clarity and less wordiness in the discussion.

Response:

We agree that the figure would benefit from further labeling. We will assign labels (1234) to the four depicted particles and refer to these from the main text in the following way:

Several morphological stages can be distinguished during the humidification process in Fig. 1. Starting from a homogeneous, glassy particle (1), an increase in RH first leads to liquefaction of a thin outer layer and emergence of a core-shell morphology (2). This liquid outer layer grows in equilibrium with ambient relative humidity and also extends towards the particle centre by diffusion of water into the glassy organic matrix (3), leading to shrinkage of the residual glassy core until the particle is fully deliquesced (4).

Referee comment 6:

Page 16463: A little more discussion of the Baustian 2013 data and how it relates to the model in the text, not only in the appendix, would be beneficial to the reader.

Response:

We agree with the referee and will add the following sentences to Sect. 3.2:

Baustian et al. used optical microscopy in conjunction with a cold stage to detect ice nucleation on glassy sucrose particles (4 μm diameter) during humidification (1 % RH min^{-1}), leading to the nucleation onsets shown in Fig. 2C (brown markers). A range of simulations mimicking the experimental conditions at different starting temperatures leads to a continuous FDRH curve (solid blue line) over the entire temperature range. For details on the calculations see Appendix B.

Referee comment 7:

Figure 1 caption: The specific time scale should be added either on the schematic or at the end of the last sentence in the caption, where it is mentioned.

Response:

We will add the typical time scale for atmospheric updrafts to the caption of Figure 1. The respective sentence now reads:

The speed of the displayed trajectory corresponds to that typical for cloud chamber or environmental cellexperiments (0.1 - 1.5 K min^{-1}, 1 - 15 % RH min^{-1}).

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