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Atmospheric Chemistry and Physics Discussions

Interactive comment on "α-pinene secondary organic aerosol at low temperature: Chemical composition and implications for particle viscosity" *by* Wei Huang et al.

Anonymous Referee #2

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General comment:

The authors discuss the formation of α-pinene secondary organic aerosol within two environmental chambers and its chemical-physical characterisation. Two different kinds of experiments are described and they are meant to mimic different possible formation and evolution conditions of SOA in the atmosphere, at different temperatures and relative humidities. The formed aerosol was characterised with a series of different analytical techniques, that allow the measurement of size distribution of particles, chemical composition and degree of oligomerisation, desorption temperatures. Generally speaking, the formation of SOA at low temperature has been very little studied in the laboratory, because of evident experimental challenges that such measurements can present. I think that the potentiality of the authors' experimental setup in this respect is really interesting; the coupling of the two different types of experiments that they performed with the extensive chemical-physical characterisation of the formed α-pinene SOA provides some interesting and valuable insights in this relatively little investigated field of atmospheric aerosols research. That said, I find there are a few points within the manuscript needs to be clarified, especially in the final discussion that the authors present at the end of the paper, where the inferred viscosity of the formed SOA is discussed. If the authors can address my points and questions below, I would recommend the publication of the manuscript in Atmospheric Chemistry and Physics.

Specific comments:

Line 59-60: "The fraction of total SOA mass from monoterpene oxidation products is estimated to be ~15 % globally, and can be higher in 60 some regions (e.g. in the boreal forest) (Heald et al., 2008)". I think it would be worth mentioning here up to what kind of values the SOA from the oxidation of monoterpenes is estimated.

Line 123: "Here, we discuss a subset of the SOA15 dataset". This is not clear, do the authors mean that in this paper they are discussing a subset of 15 dataset, or a subset of a total of 15 experiments? I think it would be worth adding some information in the manuscript on how many experiments of each kind are being discussed, possibly in Table 1 where all the different conditions for all the different types of experiments are summarised.

Line 131: "was used to prepare SOA particles in a reproducible manner". Does this statement refer to the previous paper by Möhler et al. (2008) or is there data from this work that support this? Please add either the appropriate literature reference or report some data in support of this statement in the Supplement.

Line 144: "a fraction of the SOA particles was then transferred to the dark AIDA chamber". Could please the authors provide a little bit more detail on how this transfer is done?

Line 147: “Figure 2”, and Line 119. In panel A, which refers to the type 1 experiments, α-pinene was added over two separate steps. Could please the authors mention why? I think this is a quite important point, since in the discussion of the results the concentration of α-pinene and of SOA particles in the different kinds of experiments is often mentioned as a variable that influences the resulting chemical-physical properties of the produced aerosol (for example at Lines 283-291).

Lines 155-157: Could the authors please mention how this 5% uncertainty in the quantification of the water vapours and the uncertainty of the dew point hygrometer reflects on the final uncertainty on the determined RH value?

Line 176-177: “An AMS collection efficiency (CE) of $0.4-0.5$ was used, except for the CH experiment where CE was 0.7, likely due to higher particle water content”. What was this decision based on? Could the authors please explain this a little more in detail or provide a literature reference to support it?

Line 193: “For each experiment, backgrounds were determined by sampling from the AIDA chamber before adding any precursor gases”. If the authors want to mention this aspect of their experimental procedure, which I think is quite important, could they please indicate what they measured on average in the background or give a literature reference if such measurements were previously published? Or provide some data about this in the Supplement?

Lines 283-291: I agree with the general conclusion of this paragraph, the authors state that from these results the importance of the experimental conditions when interpreting laboratory data is fundamental. As I mentioned in the comment on Line 147, I believe this discussion would benefit from the authors’ explanation of why the concentration α-pinene was kept that much higher for type 1 experiments. If the two sets of experiments were performed at the same (or at a more comparable) VOC concentration this would not be a variable anymore and they would be able to completely discriminate the effects of α-pinene concentration from the effects of temperature on the chemical-physical properties of the formed SOA. Are the authors thinking of working in this direction?

Line 299 and following discussion: “Here we show that thermograms may also be used for qualitative information on particle viscosity - My biggest concern with the author’s claim that they can get some information on the particles’ viscosity from thermograms regards what is actually the effect of viscosity at the high temperatures at which the thermograms are taken. The authors say: “at the higher temperatures where adducts desorb, particle matrix effects may become less important” (Lines 324-325), including particles viscosity. By heating up the deposited particles, the viscosity of the particles is going to change (decrease); even at the relatively low Tmax they measured at about 45 °C, is the viscosity of the particles still going to play a role in the rate of diffusion of the desorbed compounds? And what about when the temperature is even further increased? Is there a way the authors can support these claims more strongly? - Line 365: “Calculated particle water content derived from AMS measurements is prone to large uncertainties (Engelhart et al., 2011); we observed a qualitative positive correlation with RH (data not shown)”. If the authors want to mention this positive correlation I think it would be appropriate to show it in the Supplement, even if the water content quantification is characterised by a large uncertainty as they mention. - Line 376: “The dependency of Tmax on filter mass loading was not linear, and for our FIGAERO reached a plateau at mass loadings of 2$-$4 μg.”. Looking at the data reported in the Supplement in Figure S6 panels B and D, I do not think it is true that Tmax plateaus for CD experiments. Both the shape of the thermograms and the Tmax value change with different mass loading. This could mean that all the previous discussion on the CD experiments could be affected by the choice of sampling times (Table S1). Could please the authors comment on this aspect and rephrase this statement?

Line 380: “In this study, α-pinene SOA physicochemical properties such as chemical composition, phase state, and viscosity were investigated”. I would tone this down and rephrase this because of the very uncertain link between what is observed in the thermograms and the phase state/viscosity of the particles. Some aspects were actually investigated (size distributions, chemical composition, degree of oligomerisation, etc.) but the phase state/viscosity of the particles can just be supposed.

Technical comments:

Line 57: change “24.8 % mass contribution to global monoterpene emissions” to “24.8 % mass contribution to global monoterpenes emissions”.

Line 183, 269 and 307: “Table 2”. There is no Table 2 in the main manuscript, could please the authors double check this reference?

Figures 4,5, S2, S5 and 7: It is a little confusing for the reader having the results for type 2 experiments often displayed before type 1 experiment. I think it would help the reader if the sequence of the data displayed in the figures reflected the sequence with which the different kinds of experiments are presented in the abstract and in section 2.1. Possibly adding a label “Type 1 (or 2)” would help the reader, too.

Figures S3 and S4: I don’t think these figures are referred to at any point in the manuscript. If this is the case, could please the authors either add references to these figures in the main manuscript or add some more context to them in the Supplement?

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