

Response to comments of anonymous referees # 1

General comments

The authors presented laboratory results of phase behaviour of secondary organic aerosol (SOA) mixed with ammonium sulphate seed particles using smog-chamber experiments and particle bouncing measurements. The key novelty about this study is the usage of “isoreactive” (see below for a question on this) single- or mixed-precursor volatile organic compounds (VOCs) from both biogenic and anthropogenic sources, while the two hypotheses (RH rules and MR_{org/inorg} dominates) tested are actually well established. The results confirmed the vital role of RH and the dominating effect of the fraction of organics in the aerosol particles in the changes of phase behavior, while also suggested a second-order role of SOA composition (whether it is anthropogenic or biogenic, but not very much related to O:C ratio) when MR_{org/inorg} is close to unity. The experiments of this study were carefully designed and data well interpreted, in support of the conclusions made. The manuscript is also fairly well written. I have a few comments for the authors to clarify, and recommend a Minor Revision before publication.

Many thanks to the reviewer for the comments and suggestions. We have improved the manuscript accordingly. Please find a point-by-point response below.

Specific comments:

Q1. “iso-reactive”: I assume that this term is referring to a similar reactivity towards a certain oxidant for the VOCs or mixtures chosen (Table 1). The question is, what is the dominating oxidant in the chamber experiments? OH radicals or O₃? Or a mixture of them? If it is the mixture, is it a combined reactivity that makes them “iso-reactive”? Please clarify.

Good point. Yes, the term “iso-reactive” in this study means that the initial concentration of VOCs chosen in each system were adjusted towards the OH reactivity, to ensure each VOCs have the equal chance to react with OH radical at the beginning of the experiments. But after the photochemistry starts and the ozone builds up, it is not iso-reactive furthermore. Both OH radical and the formed ozone can oxidize α -pinene and isoprene (OH is the dominant oxidant), but OH radical is the only oxidant for o-cresol oxidation. Here, we take the α -pinene experiment as an example as it forms the maximum ozone concentration (peak: 25 ppb) among all systems. The lifetime of α -pinene towards OH and ozone at the 25-ppb ozone is 5.6 h and 6.4 h. In other systems, the ozone concentration is much lower than 25 ppb so the dominant oxidant for α -pinene and isoprene is OH radical.

To avoid repetition, I added a sentence in Sec. 2.1 clarifying that the experimental design will be described in detail in the Sec. 2.3, and the explanation of the iso-reactive towards the oxidants has been added into the Sec. 2.3 when introduce the rationale behind the choice of precursor and the iso-reactivity.

“The initial VOC concentrations were adjusted toward OH reactivity (dominant oxidant) to achieve the equal chance to compete the OH radical at the beginning of the experiments (referred to as iso-reactivity). When the photochemistry starts and the O₃ builds up, it is not iso-reactive furthermore during the experiments as the formed O₃ during the photochemistry can oxidize α -pinene and isoprene but not for o-cresol.”

Q2. About AMS results: a) the authors used O:C and H:C ratios and specific ion signals of NO⁺ and NO₂⁺ in their analysis, but indicated that only V-mode data were available from AMS measurements (L188). How were those data on elemental ratios and specific ion peaks obtained? Approximated from empirical formulas or high-res fitting on V-mode data? b) L192: how is this ionization efficiency defined here? If it is the number of ammonium nitrate molecules ionized per number of ammonium nitrate molecules introduced, it should be a number much lower than 1. c) 196: please specify on what is “real-time” collection efficiency.

Thanks for your helpful comment. About the AMS, the answers are shown as below:

a) The elemental ratio of O:C used for the proxy of organic oxidation state and the ions of NO⁺ and NO₂⁺ used for the organic nitrate fraction estimation are derived from the high-resolution fitting on V-mode data. This sentence has been added in the method section 2.2 as suggested.

b) Yes, the ionization efficiency means the number of ammonium nitrate molecules ionized per number of ammonium nitrate molecules introduced. The number was wrongly written and has been corrected to 9.38×10^{-8} ions molecule⁻¹.

*c) This “real-time” collection efficiency is not a precise description. Here, we corrected the AMS mass concentrations by comparing to the real-time DMPS unit mass concentration multiplied by the calculated density from AMS chemical composition. The mass ratio of AMS/(DMPS*density) is 0.4 ~ 1.0 in our study, which was used to scale up the mass concentrations of AMS accordingly. This correction only changed the absolute mass but not the ratio of SOA/inorganic or SOA/PM₁. And the ratio is the focus of this study.*

The original sentence (‘In this study, real-time collection efficiency was applied to the data analysis by comparing NR-PM₁ mass with DMPS mass.’) has been replaced by the following sentences:

“In this study, the AMS mass concentrations were corrected by comparing to the real-time DMPS unit mass multiplied by concurrent calculated density from AMS species.

*The mass ratio of AMS/(DMPS*density) is 0.4 ~1.0 in all investigated VOC systems.”*

Minor comments:

Q1. P4/L88: suggest to change “even RH up to 90%” to “even with RH of up to 90%”.

Thanks for your kindness. This has been corrected in the update version.

Q2. P8/L174 and L177: “don’t” to “do not”.

Thanks for your kindness. This has been corrected in the update version.

Q3. P8/L181: “100 Pa s in volatility”? Should it be viscosity?

Yes, correct. This has been revised in the manuscript.

Q4. P9/L184: add “and” before “SOA”. Also in other places including L204, L234, L235, and L244

Thanks for your kindness. These have been corrected in the update version.

Q5. P11/L255: “are” to “is”, and use “>” to denote the decreasing order?

Thanks for your kindness. These have been corrected in the update version.

Q6. P12/L266: “3.5h” to “3.5 h”, and a few other places in the same paragraph.

Thanks for your kindness. These have been corrected throughout the manuscript.

Q7. P12/L270: add “was” before “>0.05”.

Thanks for your kindness. This has been corrected in the update version.

Q8. P14/L319: “BVOC” to “BVOC-“.

Thanks for your kindness. This has been corrected in the update version.

Q9. P14/L341: “need” to “needs”.

Thanks for your kindness. This has been corrected in the update version.

Q10. Figures 1-3: I assume “[-]” in the axis titles means dimensionless. How about just state “dimensionless”?

Thanks for your kindness. [-] have been removed in all figures.