

## Response to the comments of Anonymous Referee #2

### Referee Specific Comment:

1. The authors emphasized several times that "This is the first time that spatial distributions of the SOA phase state over China are investigated by a regional CTM." However, I don't feel this add any merits to this study. Is it because it's hard to apply such parameterizations in regional CTM, or people think it's meaningless to investigate this issue with regional CTM? The true value of the study should be stated here.

Response: We thank the positive review and further very helpful suggestions from the Referee #2. The phase state of SOA particles plays an important role in many aerosol related processes, e.g., gas-particle partitioning, heterogeneous and multiphase reactions, particle size distribution evolution and ice nucleation pathways. However, current CTMs usually assume that SOA particles are homogeneous and well-mixed liquids, thus rapid establishment of gas-particle equilibrium is often applied in simulations of SOA formation and partitioning.

Recent measurements show that SOA can exist in liquid, semi-solid or solid states. The semi-solid or solid phase state can prolong the equilibration timescales in the gas-particle partitioning, indicating a need of considering kinetic limitations in simulating the SOA partitioning into highly viscous particles. Therefore, it is important to know the spatial distributions of the SOA phase state and viscosity, the information of which is usually missing in current CTMs. The above aspects have been clearly stated in the Introduction section. In the revised abstract, we have deleted the sentence that "This is the first time that spatial distributions of the SOA phase state over China are investigated by a regional CTM". We revised the first sentence of the abstract as below to show the significance of this study. We also adjusted the Conclusion Section (refer to the reply to your comment 3).

Line 14 – 19: “Chemical transport models (CTMs), however, usually assume that SOA particles are homogeneous and well-mixed liquids, with rapid establishment of gas-particle equilibrium for simulations of SOA formation and partitioning. Missing the information of SOA phase state and viscosity in CTMs impedes accurate representation of SOA formation and evolution, affecting the predictions of aerosol effects on air quality and climate”.

2. Also, I didn't understand why the authors choose summer 2018 as the simulation period in the abstract. In the main text, I can see that it's for comparison with observations. If so, the observation-simulation comparisons should be a major point, as should be highlighted in the abstract.

Response: We add the following sentence in the revised abstract to highlight the result of the observation-simulation comparison.

Line 23 – 24: “The simulated  $T_g$  of dry SOA ( $T_{g,org}$ ) agrees well with the value estimated from ambient volatility measurements at an urban site in Beijing”.

3. I feel the implications are not fully investigated. After reading the whole article, it's still not clear why one should care so much about the viscosity, and how the improved simulation of viscosity would refresh our understanding on SOA and its atmospheric processes. These points should be more clearly stated and described.

Response: Please refer to the reply to your comment 1 that the importance of the phase state and viscosity in many aerosol related processes has been clearly stated in the Introduction section (particularly paragraph 2 of the Introduction). In the main text of the manuscript, we also calculated the diffusion coefficients and mixing timescales of organic molecules within SOA, which has implications in how to properly treat the SOA partitioning (instantaneous equilibrium vs. kinetic partitioning) in CTMs. For example, Line 242 – 248: “We highlight the mixing timescale of 1 hour as the time step adopted in CTMs is often  $\sim 0.5 - 1$  hour (Maclean et al., 2021). Current CTMs usually assume that the gas-particle partitioning of SVOCs reaches equilibrium quickly within the time step (Pankow, 1994; Donahue et al., 2006). When the mixing timescales of organics within SOA particles are  $\leq \sim 1$  hour, the instantaneous equilibrium is a reasonable assumption. However, when the mixing timescales of organics are longer than  $\sim 1$  hour, non-equilibrium between the gas phase and the particle phase, i.e., the kinetic partitioning may need to be considered in simulating the SOA formation in CTMs (Shiraiwa and Seinfeld, 2012; Zaveri et al., 2018; Li and Shiraiwa, 2019; Zaveri et al., 2020; He et al., 2021; Jathar et al., 2021; Maclean et al., 2021; Shiraiwa and Pöschl, 2021; Shrivastava et al., 2022).”

Following your suggestion, we revised the abstract moving the sentence with implications of the mixing timescale that is calculated from SOA viscosity to the last sentence of the abstract, to highlight the implication of this study.

Line 40 – 45: “We also calculate the characteristic mixing timescale of organic molecules in 200 nm SOA particles to evaluate kinetic limitations in SOA partitioning. Calculations show that during the simulated period the percent time of the mixing timescale longer than 1 h is  $> 70\%$  at the surface and at 500 hPa in most areas of the northern China, indicating that kinetic partitioning considering the bulk diffusion in viscous particles may be required for more accurate prediction of SOA mass concentrations and size distributions over these areas”.

We also adjusted the Conclusion Section deleting the sentence that “In summary, our simulations demonstrate the spatial distributions of the glass transition temperature and viscosity of SOA particles over China on a regional scale for the first time”. We moved the implication of the mixing timescales to be the last paragraph:

Line 425 – 431: “(5) The mixing timescale of organic molecules in 200 nm SOA particles is calculated based on the simulated particle viscosity and the bulk diffusion coefficients of organic molecules. Calculations show that at the surface and at 500 hPa, the percent time of  $\tau_{\text{mix}}$  longer than 1 h is  $> \sim 70\%$  in the northwest of the “Hu

Huanyong Line”. The implication of this result is that when the  $\tau_{\text{mix}}$  values are greater than roughly 1 h, which is longer than the typical time step in CTMs, the instantaneous equilibrium partitioning usually assumed in SOA formation simulations is subject to be re-evaluated. We recommend to test the effects of kinetic partitioning considering the bulk diffusion in viscous particles on the prediction of SOA mass concentrations and size distributions over the areas with long mixing timescale of organic molecules.