Response to the comments of Anonymous Referee #2
Referee General Comment:

I am interested in the content of this paper and very concerned about the research results. From the perspective of scientific significance, I support the publication of this paper. However, reading this paper is very difficult, and I suggest adjusting the paper to highlight the main theme.

Response: We thank the positive review and very helpful suggestions from the Referee #2. We have carefully revised the manuscript as below.

Referee Specific Comment:

1. Firstly, I hope to see a clear conclusion. It feels like the paper ends in the midst of discussion. Please clarify the conclusion of the paper in the revised manuscript. If there is more to discuss, please use a separate subsection for that discussion.

Response: Thanks. We have inserted the discussion into the Result Section and revised the conclusion as follows.

Line 383 – 419: "We previously developed a new parameterization predicting the glass transition temperature of an organic compound as a function of its volatility (Li et al., 2020). Based on this new parameterization, we use the WRF-Chem model and simulate the T_g and viscosity of SOA particles over China in June of 2018. This is the first time that spatial variations in the SOA phase state over China are investigated on a regional scale. The main conclusions are summarized below.

(1) Simulations show that T_g values of dry SOA ($T_{g,org}$) range from ~287 K to 305 K over most areas of China at the surface, consistent with our previous simulated results based on a global transport model (Shiraiwa et al., 2017). The $T_{g,org}$ is higher in the northwestern China than the southeastern China. This geospatial variation in $T_{g,org}$ is related to the simulated SOA volatility distributions that SOA particles in northwestern China have relatively low volatilities.

(2) Considering the water uptake by SOA particles, the SOA viscosity also shows a prominent geospatial gradient that highly viscous or solid SOA particles are mainly found in the northwestern China. The frequency of liquid SOA particles in most areas with the latitude higher than 30 °N is less than 20 %. A very large spatial variation in SOA phase state over the Qinghai-Tibet Plateau was found and we recommend measurements in ambient particle phase state to be conducted over this area, one of the most sensitive regions to climate change.

(3) 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 τ_{mix} longer than 1 h is > ~ 70 % in the northwest of the "Hu Huanyong Line". The implication of this result is that when the τ_{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.

(4) The average volatility (C^*) and $T_{g,org}$ of the simulated SOA agree well with the values estimated from ambient measurements of OOA volatilities at the IAP site in Beijing, where ELVOCs were not determined in the observed OOA (Xu et al., 2019). The sensitivity simulation considering the formation of ELVOCs shows that compared to the base simulation, the frequency of a liquid phase state does not change in most areas of the northern China. In some areas of the southeastern China the SOA particles become more viscous with the percent time that a SOA particle is in the liquid phase state decreases by up to 12 %. It needs more field volatility measurements to evaluate the effects of ELVOCs as well as the enthalpy of vaporization on OA phase state estimations over China.

(5) Differed from the base simulation that SOA components are assumed to be phase separated from inorganic compounds in particles, we conduct a sensitivity simulation assuming that the organic and inorganic compounds are internally mixed in one phase. We show that the water absorbed by inorganic species has a significant impact lowering the simulated viscosity over the southeastern China, with the liquid frequency increased by 15 – 45 %. Future work should consider the effects of the mixing state of organic and inorganic compounds on the simulations of phase state of multicomponent particles in ambient air.

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. The further calculations of the mixing timescale of organic molecules in SOA particles have an implication of the need to evaluate the effects of the phase state on predictions of SOA gas-particle partitioning, and thus the SOA mass concentrations and size distributions in CTMs."

2. The theme of the paper is not sufficiently clear, and there is a discrepancy between the focus of the conclusions in the conclusion section and those in the abstract. It is unclear what your main focus is. Please clarify your focus and then revise the paper accordingly, making choices about what to include. For instance, you might consider relocating some figures from the main body of the paper to the appendix, and vice versa.
Response: In response to this comment, we have re-written the conclusion section (see the response to your comment 1) making it more consistent with the focus of the abstract. As the glass transition temperature, the viscosity of SOA particles, the bulk diffusion coefficients, and the mixing timescale in SOA particles have not been shown in previous studies on a regional scale, we kept all the figures in the main text file for comparison by future studies. We have revised the manuscript to make the structure more clear and the main theme highlighted. The revision is included below.

Line 26 – 27: "We also calculate the characteristic mixing timescale of organic molecules in 200 nm SOA particles to evaluate kinetic limitations in SOA partitioning."

Line 59 – 61: "Accurate predictions of the viscosity need the information of molecular structures and functional groups (Song et al., 2016; Rothfuss and Petters, 2017; Gervasi et al., 2020; Galeazzo and Shiraiwa, 2022); however, molecular specificity is often unavailable in ambient measurements, leading to the prediction of the phase state of ambient SOA particles difficult."

Line 82 – 84: "It is needed to conduct simulations to investigate the SOA phase state varied with locations and the time. Simulations of the SOA phase state in China on a regional scale have not been available."

Line 232 – 239: "We highlight the mixing timescale of 1 hour as the time step adopted in CTMs is often ~ 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 $\leqslant$ ~ 1 hour, the instantaneous equilibrium is a reasonable assumption. However, when the mixing timescales of organics are longer than ~ 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)."

Line 288 – 291: "The phase state of SOA particles is affected by ambient conditions and the particle chemical composition (Koop et al., 2011). Figure 6 shows the median values of viscosity as a function of RH, T and the mass fraction of low-volatility compound (SOAX with C^* of 0.1 μg m^{-3} at 298 K) calculated for selected regions in the northern China, southern China, northern Qinghai-Tibet Plateau, and southern Qinghai-Tibet Plateau as specified by white boxes in Fig. 5a."

Line 365 – 381: "Besides RH, the mixing state of the organic and inorganic species in atmospheric particles also plays an important role in the phase state of ambient particles. The SOA components are assumed to be phase separated from inorganic compounds in particles in our base simulation, which is consistent to recent ambient observations showing that the phase separation with an organic-rich shell and an inorganic core was a frequent phenomenon in individual particles (diameters > 100 nm) collected over the North China Plain (Li et al., 2021a). To assess the potential effects of inorganic compounds on the phase state of ambient particles, we perform a sensitivity calculation (case C, Table 1) assuming that the organic and inorganic compounds are internally mixed in one phase. In this case the water absorbed by inorganic species can lower the particle viscosity relative to the organic fraction alone. Figure 10b shows that the water associated with inorganic species can significantly

lower the viscosity over most areas of China, with the liquid frequency increased by 15 – 45 % in the southeast, and 5 – 15 % in some areas of the northeast. The effects over dry lands in the northwestern China are relatively small. Previous studies showed that at such mixing condition with one phase, on one hand, it is expected that the inorganic salts that often have lower T_g compared to SOA compounds would further lower the particle viscosity (Dette and Koop, 2015). On the other hand, the presence of divalent inorganic ions could increase the viscosity of mixed organic-inorganic particles, enabling a humidity-dependent gel phase transition through cooperative ion-molecule interactions (Richards et al., 2020). For complex mixtures of primary OA, SOA and inorganics, it was found that three distinct phases could occur (Huang et al., 2021). The impacts of the mixing state of organic and inorganic compounds on the phase state of multicomponent particles in ambient air warrant further investigations in future studies (Lilek and Zuend, 2022; Schervish and Shiraiwa, 2023)."

We have moved a scatter plot in the original supplement file to the main text file as below. This new figure shows that the factors including RH, T and the SOA volatility affect the viscosity predictions.
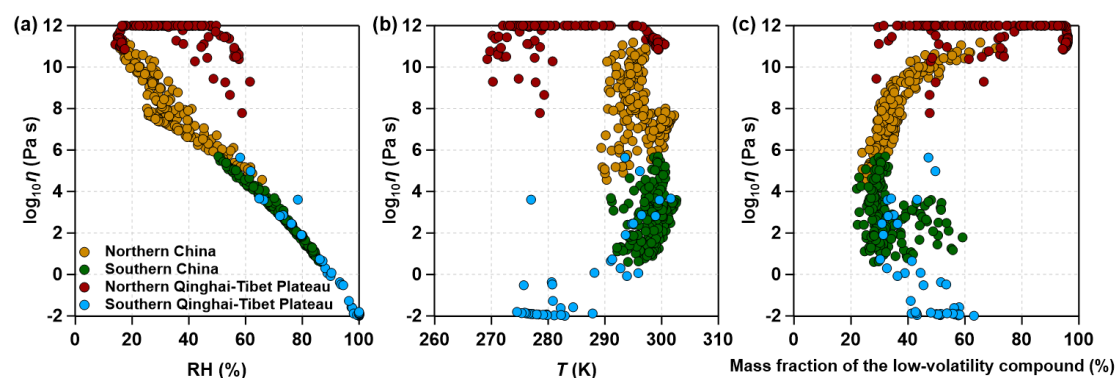

Figure 6. The median values of viscosity as a function of (a) RH, (b) T and (c) the mass fraction of SOAX ($C^* = 0.1$ µg m^{-3} at 298 K) calculated for selected regions in the northern China, southern China, northern Qinghai-Tibet Plateau, and southern Qinghai-Tibet Plateau as specified by white boxes in Fig. 5a during May 20 – June 23 in 2018.