We would like to sincerely thank reviewer #2 for their insightful comments and recommendations for our manuscript. We have carefully considered each comment and addressed each concern point-by-point below. For ease of readability, each of reviewer #2's points is denoted with "RC" and is bolded, and our response is denoted with "AC" with detailed discussion and excerpts to changes in the manuscript beneath. We included references to changes in line numbers to our current manuscript.

RC: The authors mention that density is important for the accommodation coefficient; however, the authors never address how density changes with oxidation / production of secondary organic aerosol. However, though other parameter were discussed or set constant for discussion reasons, SOA density was never discussed. What is the impact of this? E.g., the O:C ratio is already being used for Tg so why not use it to calculate density?

AC: We assume that SOA particle density is constant at 1.4 g cm^{-3} based on a previous study showing that the density of various SOA materials ranges from 1.23 – 1.46 g cm^{-3} (Kuwata et al., 2012). This variation is much smaller and almost negligible compared to variations of bulk diffusivity and volatility (both change over orders of magnitudes) for calculations of effective mass accommodation coefficient in Eq. 7. Thus, the potential effect of density change is minimal. We clarify the value of SOA density in the revised manuscript in L119.

RC: The results in Fig. S1 and S2 seem important enough that they should be included in the main paper and not placed into the SI. I would recommend showing both contours in Fig. 2.

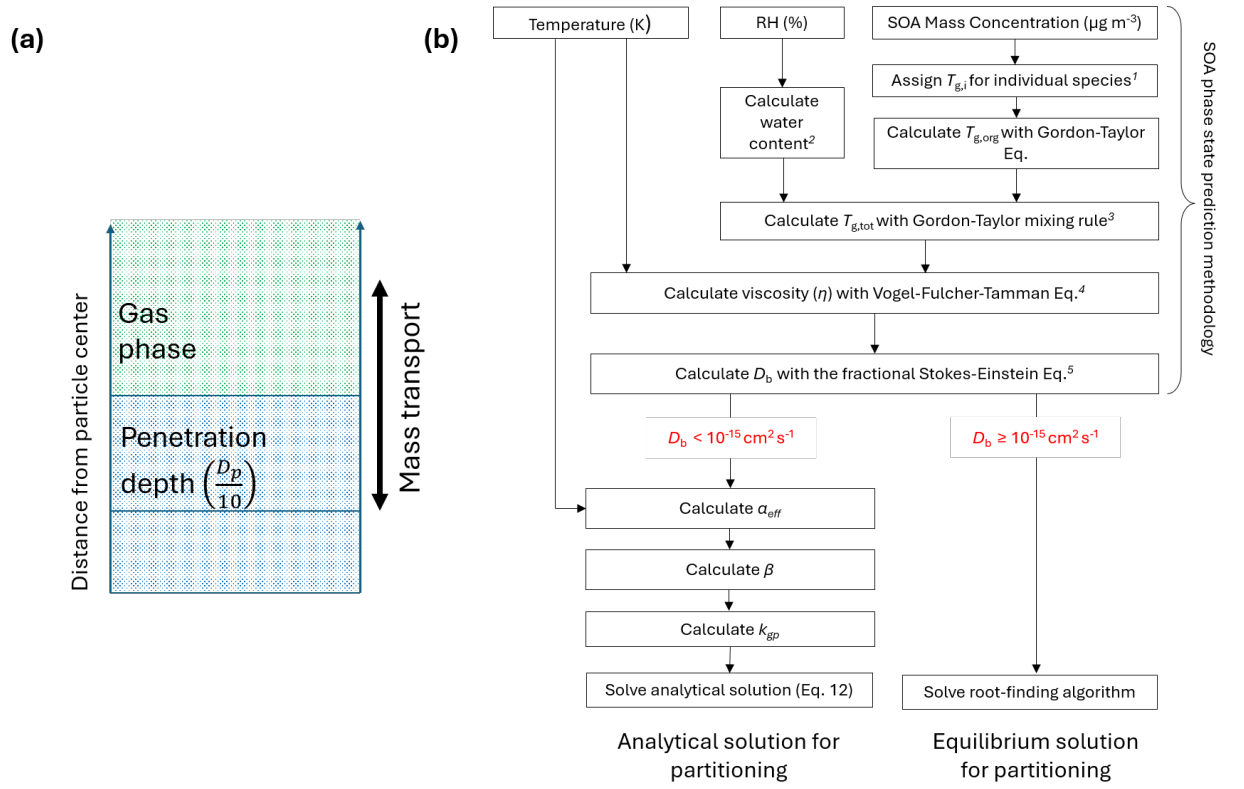
AC: Thanks for your suggestion; we agree to include Fig. S1 in with Fig. 2 (as panels d, e, f) and have made the related changes to the main text and supplemental information. We would like to keep Fig. S2 in the supplemental information as the final particle-phase concentration under specific conditions is not too critical for the equilibration timescale of SOA partitioning.

RC: Line 258-259 -- The authors state that tmix would lead to a significant underestimation in SOA mass for C0 = 10 and 100; however, it would seem that would be true for C0=0.1. Also, since tmix is nearly identical to teq for C0=100, is it really a significant underestimation compared to the other two bins? Also, significant implies statistically analysis; however, with the simplifications and assumptions, how different is a value of 0.04 vs 0.06 for C0=10 and 0.06 and 0.1 for C0=0.1?

AC: Please note that x-axis of Fig. 1 is in log scale; while τ_{eq} and τ_{mix} looks similar for C^0 = 100 μg m^{-3} in Fig. 1f, τ_{mix} is actually by almost a factor of 2 larger than τ_{eq}. Thus, the use of τ_{mix} as τ_{eq} would still underestimate SOA formation. As it is not based on statistical analysis, we removed the word significant from this sentence.

RC: For Fig 3, bulk diffusivities is shown from GEOS-Chem. Is this a normal output or is this something that was specifically done for this study? If done for this study, how was it calculated (online vs offline, assumptions, etc.).

AC: Online calculated SOA bulk diffusivities output from GEOS-Chem are not standard output but was implemented as an archivable diagnostic for the scope of this study, as described in the method section. We detailed the methodology and underlying assumptions in the SOA phase state simulations in our previous paper, Luu et al. (2025). To improve presentation quality of our method and also following the comment by Reviewer 1, we have added new Figure S1 as shown below, which describes the schematic of our method to calculate SOA viscosity and bulk diffusivity.



[1] $T_{g,i}$ for each species was calculated based on Eq. (8) and can be found in Table S1.
[2] The effective hygroscopicity parameter (κ) for SOA is assumed to be 0.1(Lambe et al., 2011; Petters & Kreidenweis, 2007). The density of SOA particles (ρ_{SOA}) is assumed to be 1.4 g cm^{-3}(Cross et al., 2007).
[3] The glass transition temperature of pure water ($T_{g,w}$) is 136 K and the Gordon-Taylor constant for organic-water mixtures (k_{GT}) is assumed to be 2.5(Koop et al., 2011).
[4] The threshold between semi-solid and liquid states depends on the so-called fragility parameter (D) which is assumed to be 10(DeRieux et al., 2018).
[5] We assume that the hydrodynamic radius of a diffusing species is 0.38 nm and x is an empirical fit parameter determined to be 0.93(Evoy et al., 2019).

Figure S1. Modeling schematics of (a) the zero-dimensional box model simulating gas-particle partitioning solving Eq. (5, 6) using α_{eff} to govern reversible mass transport between the gas phase and the particle phase at the set penetration depth. The workflow chart for (b) the revised SOA partitioning scheme implemented in GEOS-Chem shows that environmental conditions and SOA mass concentration are used to calculate bulk diffusivity which is ultimately used to determine the partitioning approach.

RC: For the GEOS-Chem run, the authors discuss differences in regions (e.g., line 367-370); however, these differences are not apparent in the figures shown in the SI.

AC: The referenced differences over the remote Pacific Ocean can be seen best at 10ºN, 135ºW while the most apparent difference for the Indian Ocean is approximately at 0ºN, 90ºE. We added the coordinates to better guide the reader for the supplemental figures.

L376-L377: Over the remote oceans (Indian and Pacific Oceans, 10ºN 135ºW and 0ºN, 90ºE) and central US, we predict up to an order of magnitude SOA budget increase in the upper troposphere (Fig. S4a and S5a).

RC: I appreciate the inclusion of limitations. However, there is no discussion in the impact of inorganics with organics and potential for phase separation and/or inorganic impacting the water enough to make the aerosol potentially less viscous than assumed from a purely organic perspective. Does this have any impact?

AC: Thank you for bringing up this point. Inorganics may either decrease particle viscosity by acting as plasticizers or may increase viscosity due to gel formation (Koop et al., 2011; Richards et al., 2020). These aspects are beyond the scope of this manuscript, but this is an important issue to be addressed in future studies. We revised our statement on phase-separation to include more detailed discussion on the impacts of mixing with inorganics as below:

L442-449: In the current GEOS-Chem configuration, SOA particles are implicitly assumed to be externally mixed from inorganic compounds or, if internally mixed, to be phase-separated, such that the predicted phase state represents only the organic phase. Consequently, the predicted viscosity and bulk diffusivity reflect the organic phase alone. This approach does not account for the influence of inorganic species, such as ammonium sulfate, ammonium nitrate, and sodium chloride on the bulk phase state. If these inorganic compounds are well-mixed with the organic phase, they may decrease viscosity by acting as plasticizers (Koop et al., 2011). If oxygenated organics and divalent inorganic ions are mixed, it might lead to an increase of viscosity by forming highly viscous gels (Richards et al., 2020).

RC: Lines 41 - 45: Only gas-phase oxidation of precursor gases are mentioned for SOA when other processes, including aqueous chemistry and volatilization and subsequent oxidation of gases from primary OA are important and not mentioned.

AC: We revised the statement about SOA formation and included these additional pathways that were missing.

L47-L49: Other pathways of SOA formation include aqueous/heterogeneous chemistry and volatilization and subsequent oxidation of gases from primary organic aerosols (Robinson et al., 2007; Ziemann & Atkinson, 2012).

RC: Line 201, should the first ASOA C0 really be 10^-10 and not 10^-1 (or 0.1)?

AC: Yes, the first ASOA species is supposed to be essentially nonvolatile, and this is hard coded into the GEOS-Chem source code as its reference volatility. To further clarify, we indicated this as below.

L206-L207: The VBS for TSOA covers C^0 of 0.1, 1, 10, and 100 μg m^{-3}, while ASOA uses C^0 of 10^{-10} (nonvolatile), 1, 10, and 100 μg m^{-3}.

RC: Check references formatting. For example, Lines 180 and 183

AC: We have corrected these references formatting.

References

Cross, E. S., Slowik, J. G., Davidovits, P., Allan, J. D., Worsnop, D. R., Jayne, J. T., Lewis †, D. K., Canagaratna, M., & Onasch, T. B. (2007). Laboratory and Ambient Particle Density Determinations using Light Scattering in Conjunction with Aerosol Mass Spectrometry. *Aerosol Science and Technology*, *41*(4), 343–359. https://doi.org/10.1080/02786820701199736

DeRieux, W.-S. W., Li, Y., Lin, P., Laskin, J., Laskin, A., Bertram, A. K., Nizkorodov, S. A., & Shiraiwa, M. (2018). Predicting the glass transition temperature and viscosity of secondary organic material using molecular composition. *Atmospheric Chemistry and Physics*, *18*(9), 6331–6351. https://doi.org/10.5194/acp-18-6331-2018

Evoy, E., Maclean, A. M., Rovelli, G., Li, Y., Tsimpidi, A. P., Karydis, V. A., Kamal, S., Lelieveld, J., Shiraiwa, M., Reid, J. P., & Bertram, A. K. (2019). Predictions of diffusion rates of large organic molecules in secondary organic aerosols using the Stokes–Einstein and fractional Stokes–Einstein relations. *Atmospheric Chemistry and Physics*, *19*(15), 10073–10085. https://doi.org/10.5194/acp-19-10073-2019

Koop, T., Bookhold, J., Shiraiwa, M., & Pöschl, U. (2011). Glass transition and phase state of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere. *Physical Chemistry Chemical Physics*, *13*(43), 19238–19255. https://doi.org/10.1039/C1CP22617G

Kuwata, M., Zorn, S. R., & Martin, S. T. (2012). Using elemental ratios to predict the density of organic material composed of carbon, hydrogen, and oxygen. *Environmental Science & Technology*, *46*(2), 787–794. https://doi.org/10.1021/es202525q

Lambe, A. T., Onasch, T. B., Massoli, P., Croasdale, D. R., Wright, J. P., Ahern, A. T., Williams, L. R., Worsnop, D. R., Brune, W. H., & Davidovits, P. (2011). Laboratory studies of the chemical composition and cloud condensation nuclei (CCN) activity of secondary organic aerosol (SOA) and oxidized primary organic aerosol (OPOA). *Atmospheric Chemistry and Physics*, *11*(17), 8913–8928. https://doi.org/10.5194/acp-11-8913-2011

Petters, M. D., & Kreidenweis, S. M. (2007). A single parameter representation of hygroscopic growth and cloud condensation nucleus activity. *Atmospheric Chemistry and Physics*, *7*(8), 1961–1971. https://doi.org/10.5194/acp-7-1961-2007

Richards, D. S., Trobaugh, K. L., Hajek-Herrera, J., Price, C. L., Sheldon, C. S., Davies, J. F., & Davis, R. D. (2020). Ion-molecule interactions enable unexpected phase transitions in organic-inorganic aerosol. *Science Advances*, *6*(47), eabb5643. https://doi.org/10.1126/sciadv.abb5643

Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop, A. P., Lane, T. E., Pierce, J. R., & Pandis, S. N. (2007). Rethinking organic aerosols: Semivolatile emissions and photochemical aging. *Science*, *315*(5816), 1259–1262. https://doi.org/10.1126/science.1133061

Ziemann, P. J., & Atkinson, R. (2012). Kinetics, products, and mechanisms of secondary organic aerosol formation. *Chemical Society Reviews*, *41*(19), 6582–6605.