We would like to sincerely thank reviewer #1 for their insightful comments and suggestions for our manuscript. We have carefully considered each comment and addressed each concern point-by-point below. For ease of readability, each of reviewer #1'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 computational cost is not explicitly presented or analyzed; therefore, it is difficult to judge the cost vs. potential improvements to SOA predictions. For example, overall, a 1% difference in SOA burden is predicted at the surface. While the differences in arid regions and in the free troposphere are more substantial, understanding the computational requirement of this framework would be useful.

AC: We benchmarked and analyzed the computational costs of the new α_{eff} scheme against the base case scheme and included the metrics in Table S3. We added a discussion in L414-L419 with the main point that there is no additional computational cost in GEOS-Chem for our methodology.

L414-L419: From our benchmarking, a one-month simulation has a wall time of roughly 6.6 h from the base scheme compared to the αeff scheme at < 6.5 h (Table S3). The memory utilized by both schemes indicates that the α_{eff} scheme requires the same or less maximum memory usage. Truly significant differences between computational costs may slightly differ depending on run-to-run variability, however it is apparent that the α_{eff} partitioning scheme does not add any computational cost.

Table S3. Computational demand of 1-month benchmarked GEOS-Chem simulations, reported as wall-clock time (actual elapsed runtime), aggregate CPU time (summed CPU usage across cores), and maximum resident memory (MaxRSS).

Scheme	Wall-Clock Time (hh:mm:ss)	CPU Time (dd-hh:mm:ss)	MaxRSS (GB)
True Base [1]	06:37:31	17-16:01:04	5.87
"Base" [2]	06:40:45	17-19:28:00	5.87
α_{eff}	06:24:19	17-02:02:40	5.62

* Simulations were run on a single dedicated node using 64 CPU cores.

[1] The True Base scheme refers to the default complex scheme with no changes to the GEOS-Chem ver. 14.1.1 source code.

[2] The "Base" scheme refers the modified version of the complex scheme that includes volatility-dependent heat of vaporization parameterization which is used to compare to the α_{eff} in the main text.

RC: For the analyses presented in Figure 2, more equilibrium timescales between 1 min and 5 hr could be plotted to help illustrate the transition near Db = 10-15 and the different model treatments above/below this threshold.

AC: We added additional contour lines (5 min and 10 min) to Fig. 2 (and combined with Fig. S2 as per referee 2's suggestion) prior to the 20 min chemical timestep contour for all three aerosol mass loading conditions.

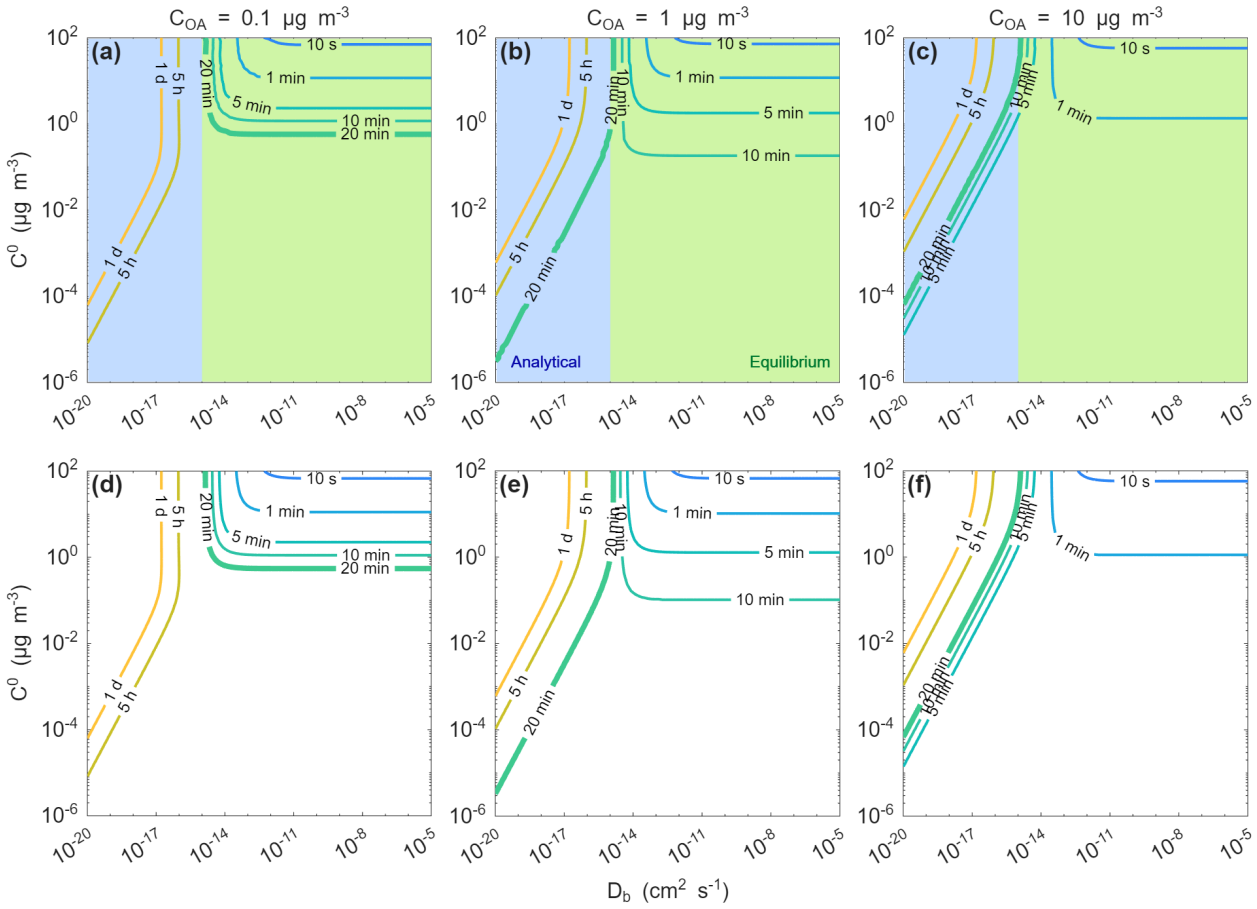


Figure 2. Contour plot of equilibration timescale (τ_{eq}) calculated using the numerical solution as a function of bulk diffusivity (D_b), saturation mass concentration (C^0), and initial pre-existing particle concentrations (C_{OA}) of **(a, d)** 0.1 μg m^{-3}, **(b, e)** 1 μg m^{-3}, and **(c, d)** 10 μg m^{-3}. The τ_{eq} is calculated using the numerical solution **(a-c)** and by the analytical solution **(d-f)**. Two colored regimes are depicted: (1) when $D_b \geq 10^{-15}$ cm^2 s^{-1}, equilibrium partitioning is applied (in green) and (2) if $D_b < 10^{-15}$ cm^2 s^{-1}, then the instantaneous analytical solution is applicable (in blue). The initial gas-phase concentration was 0.1 μg m^{-3} (to simulate condensation), or the initial particle-phase concentration was 0.1 μg m^{-3} (to simulate evaporation), and the temperature was set to 298K.

RC: The manuscript alludes to a follow-up study in which model predictions will be evaluated against observations (Lines 473 – 475); however, there is currently no evaluation of the new modeled SOA, not even qualitatively. Some discussion, e.g., SOA concentrations

are typically underpredicted in the free troposphere, could be added without a full quantitative analysis.

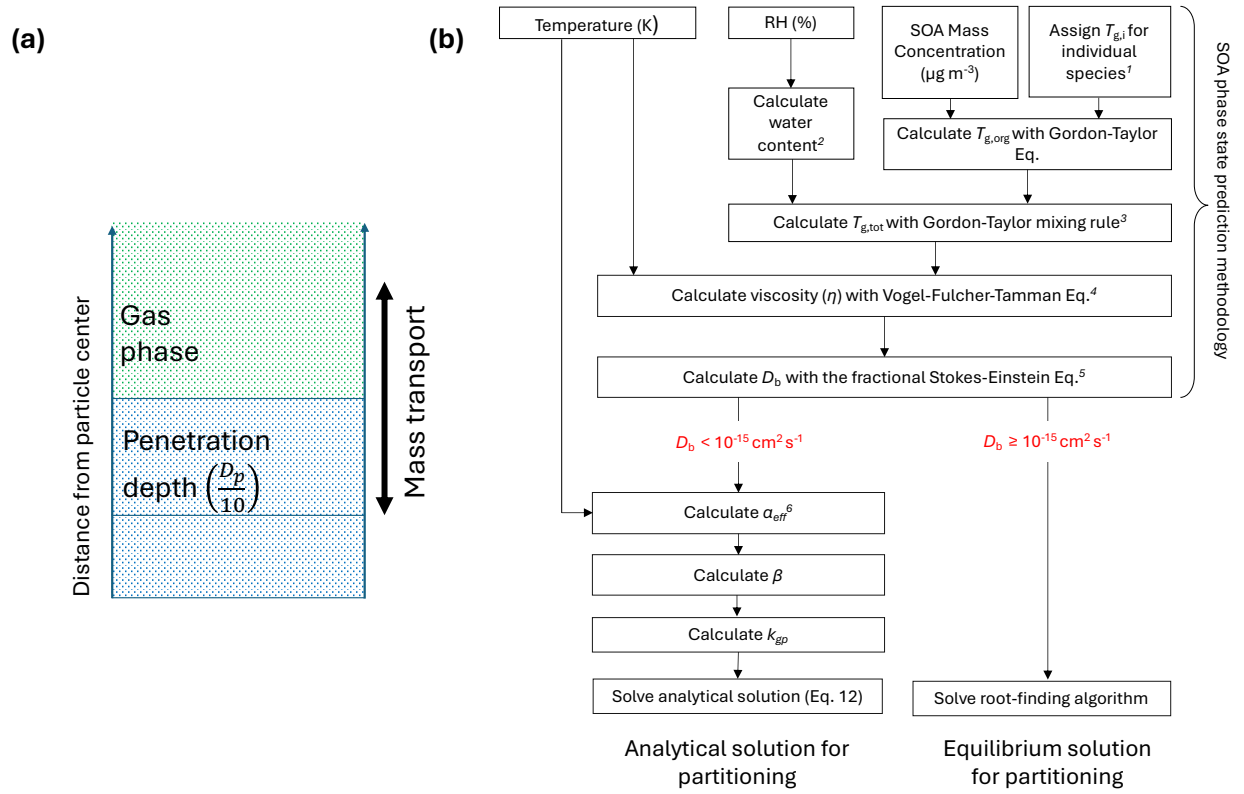
AC: Thanks for this suggestion. We acknowledge that there was a lack of qualitative comparisons of remote or free tropospheric SOA budgets therefore we added the following lines (L491-L495) to discuss the uncertainty of this topic. As POA and SOA are difficult to differentiate, most field campaigns will report total organic aerosol (OA) mass concentration making OA model-measurement comparisons more common (Heald et al., 2011; Hodzic et al., 2020; Pai et al., 2020). Heald et al. 2011 examined and simulated observed field campaign OA found that simulated median OA was underestimated over the total domain. However, in remote areas, it is noted that the model skill was poorer and tends to overestimate aloft compared to source regions(Heald et al., 2011). Analysis of the high-altitude flight observations by Hodzic et al. 2020 showed that GEOS-Chem simulations underpredict the relative fraction of OA observed at free troposphere altitudes. Other ESMs and CTMs exhibited both over- and underestimation of relative OA fraction. Pai et al. 2020 found that OA was typically underestimated in the upper troposphere, however simulated SOA minimally contributes to overall OA. Typically, VBS treatments where POA is non-volatile, SOA (including OPOA) is vastly underestimated compared to POA aloft in the free troposphere (Heald et al., 2011; Hodzic et al., 2020; Pai et al., 2020). Given the campaign-to-campaign variability in both over- and under-estimation of OA (and thus SOA), generalized trends in SOA under remote or free tropospheric conditions remain highly uncertain in the present work. We have added the following sentence in the revised manuscript:

L489-494: As POA and SOA are difficult to differentiate in field measurements, most field campaigns report total organic aerosol (OA) mass concentration making OA model-measurement comparisons more common; typically, OA (including OPOA) mass concentrations were underpredicted at remote free tropospheric altitudes, but in some cases, OA has been overestimated (Heald et al., 2011; Hodzic et al., 2020; Pai et al., 2020).

RC: Suggest adding a simple flowchart or pictorial representation of the modeling approach to the SI.

AC: We created schematics for both the 0-D box modeling and the GEOS-Chem simulations as Fig. S1a and S1b to improve visualization of the two approaches.

L184-L186: Details on the meteorology and emission inventories used are the same as in our previous study, Luu et al., (2025) and Fig. S1b depicts a schematic algorithm for model predictions of viscosity, bulk diffusivity, effective mass accommodation coefficient, and kinetic partitioning, as implemented into GEOS-Chem in this study.



[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: Finally, the writing is generally good, however, there are a number of small grammatical issues or sentence clarifications needed. This is not comprehensive, but some issues are: Line 46; Line 49; Line 67-68; Line 98-99; Line 124; Line 144; Line 183; Line 212; Line 231; Line 253; Line 330; Line 332; Line 338; Line 436; Line 446; Line 456; Line 470-471.

AC: We have corrected and addressed all the noted grammatical issues and added additional clarifications in the lines listed above as well as other lines during proofreading.

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