Response to reviews

We thank the reviewers for their constructive comments. In the following section, reviewer comments are in black. Point-by-point responses are in blue labeled with [R]. Line numbers in the responses correspond to those in the track-change version of the revised manuscript. Specific modifications to the manuscript are in red.

Reviewer 1

This paper investigates the impact of full-volatility-range organic emissions on the global budgets of organic aerosols using the GEOS-Chem atmospheric chemistry transport model. The authors highlight the importance of incorporating a wide volatility spectrum of organic precursor emissions from anthropogenic sources and open biomass burning on global SOA production. This approach leads to greater POA and SOA burdens compared to the previous model configuration. Overall, the proposed developments appear to improve the model's agreement with a range of observational datasets used for the present study, compared to the previous organic aerosol representation.

General comments:
The manuscript provides a detailed description of the emissions used for this study. I acknowledge the authors' effort in presenting all of the different sensitivity simulations; however, the aim of this strategy is not clearly explained until the end of the manuscript, which makes it challenging for the reader. The discussion of the results is well-organized, though some repetitions exist at the beginning of certain sections. Aside from these minor concerns, the discussion of the modeling methods could have been more explicit in some cases to better justify the conclusions of this study, e.g., regarding whether the assumptions used are driven by observational/laboratory constraints, computational/modeling limitations, or both. Therefore, I recommend a revision to address such issues before the submission is accepted.
[R0] We sincerely appreciate the time and effort that Reviewer 1 dedicated to providing insightful feedback on our manuscript. We have provided more information on the default schemes and the revised scheme, especially for the SOA yield, vapor wall loss correction, and gas-particle partitioning. We have also added more description in the model-observation comparison and discussion in the global OA budget. We provide point-by-point responses below and have revised the manuscript and supporting information (SI) accordingly.

Major comments:
The authors present their new scheme as a "full-volatility-range" representation. However, it appears to be more of a hybrid volatility-based scheme that combines traditional VBS theory and empirical SOA yield parameterizations. The latter is also supported by the authors' statement in the Conclusions that "the SOA scheme remains a volatility-lumped and yield-based approach." Stating this explicitly in the Methods,

rather than in the Conclusion of the manuscript, would improve the transparency of the methodology section and help the reader better understand these developments.

[R1] We agree with the reviewer and have renamed our revised scheme as "full-range volatility-binned (FV) OA scheme" and clarified the scheme in the Methods in Lines 167-178: "Unlike the default complex OA scheme, which represents organic species using a limited number of discrete volatility bins, the FV OA scheme represents all OA precursors across continuous volatility bins. For S/LVOCs, we adopt four lumped volatility bins in the gas phase with decadal C^* ranging from 10^{-1} to 10^{2} μg m^{-3}, and five lumped species in the particle phase with decadal C^* of $10^{\leq -2}$ to 10^{2} μg m^{-3}. IVOCs are represented by four lumped gas-phase species that have decadal C* ranging from 10^{3} to 10^{6} μg m^{-3}.

The simulation of gas-phase S/LVOC oxidation follows the default complex OA scheme, in which the oxidation products with C^* below 10^{-2} μg m^{-3} are lumped into one nonvolatile species. The SOA formation from VOCs and IVOCs is simulated by using the VBS approach, but with updated SOA yields and vapor wall-loss corrections to better represent low-loading gas-particle partitioning (Zhang et al., 2014; Chen et al., 2024)."

The authors also state in the Methods that "Under low-NO_x conditions, the IVOC-SOA is treated as nonvolatile mass with a constant SOA yield of 73%." As far as I understand, this suggests that a constant yield is applied for every IVOC category in this case, which is a strong assumption. IVOC oxidation is parameterized completely differently between high- and low-NO_x conditions; i.e., under high-NO_x conditions, oxidation products are distributed among separate volatility bins, while under low-NO_x conditions, SOA-IVOCs are parameterized as nonvolatile with a fixed yield. It would be very useful for the reader if the authors discussed more explicitly why they use different parameterizations under high- and low-NO_x conditions and what the added value of this assumption is compared to other modeling studies. I would also propose further discussing the implications for SOA partitioning/evaporation under different NO_x conditions in the model (and what thresholds are used).

[R2] The NO_x-dependent SOA yields from the photooxidation/ozonolysis of VOCs and IVOCs are commonly adopted in CTMs (Carlton et al., 2010; Pye et al., 2010). This treatment is consistent with observed differences in laboratory experiments (Ng et al., 2007; Chan et al., 2009). In GEOS-Chem, the amounts of precursors consumed under high- and low-NO_x conditions are calculated online through the loss of peroxy radicals via NO and HO_2 pathways, respectively. This branching ratio is determined by temperature and the concentrations of HO_2 and NO (Henze et al., 2008).

In this study, we update the high-NO_x SOA yields based on the experiments for the photooxidation of *n*-alkanes, while the low-NO_x SOA yields remain those of the default scheme due to the lack of low-NO_x experiments for *n*-alkanes. We agree with the reviewer that the fixed low-NO_x SOA yield for IVOCs is highly uncertain, as recent studies suggest a lower SOA yield of naphthalene under low-NO_x conditions (Manavi and Pandis, 2022). Most IVOCs and NO_x are co-emitted from combustion sources, so a large fraction of IVOCs is oxidized under high-NO_x conditions near source regions,

although some IVOCs can be transported to low-NO_x regions. The uncertainty in the low-NO_x SOA yield for IVOCs may have limited impacts on the global SOA budget.
We have modified the descriptions in Lines 142-146: "Moreover, this scheme simulates SOA formation from the oxidation of VOCs (isoprene, terpenes, and aromatics) and IVOCs (treated as naphthalene) via a commonly used volatility-basis-set (VBS) approach with NO_x-dependent SOA yields (Pye et al., 2010). Precursor consumption under high- and low-NO_x conditions is calculated from peroxy radical loss via NO and HO_2 pathways, respectively (Henze et al., 2008)", and Lines 197-199: "Under low-NOx conditions, following the default complex scheme, a constant SOA yield of 73% is assumed for the IVOC oxidation to form SOA (Chan et al., 2009; Pye and Seinfeld, 2010)".

Minor comments:
Abstract: The transition to "population exposure" and "air quality improvements" seems quite abrupt, as this is not the focus of this study. I suggest moving this sentence to the conclusion/discussion section, rather than the abstract, which should focus on the actual developments the paper presents.
[R3] We agree with the reviewer and have deleted that sentence from the abstract.

Lines 61-62: It would be helpful to briefly refer to why an overestimation appears for the nonvolatile POA assumption.
[R4] We thank the reviewer for the suggestion. This sentence is revised in Lines 63-66 as "Some CTMs assume that POA is nonvolatile. This may lead to overestimation of POA concentrations by more than 100% in outflow and remote regions, because most POA emissions are semivolatile and therefore partially evaporate under atmospheric dilution (Jo et al., 2013; Murphy et al., 2017; Brewer et al., 2023)."

Section 1: An outlook paragraph presenting what each of the following sections discusses will be helpful to the reader.
[R5] We thank the reviewer for the suggestion. We have revised Lines 96-103 in the Introduction section as follows: "In this study, we develop a new OA simulation framework in GEOS-Chem chemical based on volatility-binned anthropogenic and biomass burning FVOC emissions. Model performance is evaluated against a comprehensive observation dataset that includes surface, shipborne, and airborne measurements, with POA and SOA further assessed against aerosol mass spectrometer source apportionment. Building on this evaluation, we provide new constraints on the global distributions, source contributions, and population exposures of POA and SOA. The results are compared among studies, and the sensitivity of the simulations to wet-deposition parameterizations is discussed."

Section 2.2: A table presenting the global annual emissions of S/LVOCs and IVOCs from anthropogenic sources and biomass burning sources used for each scheme (i.e., the simple and complex OA schemes, etc.) would be useful.
[R6] Thank the reviewer for this suggestion. We have added a new table (Table S5 in

the Supplement) that lists the global annual emissions of S/LVOCs and IVOCs for all three schemes used in GEOS-Chem and their caveats.

Lines 129-133: Here, the authors refer to heterogeneous reactions for IEPOX and glyoxal/methylglyoxal, but they also include a sentence about neglecting isoprene SOA formation under low-NO_x conditions. Please explain briefly why low-NO_x isoprene SOA formation is neglected and whether this impacts heterogeneous SOA production. If this is not the case, it would be better to move this sentence earlier to improve the flow to avoid confusion.

[R7] Since both the traditional low-NO_x isoprene SOA pathway and the heterogeneous isoprene SOA pathway in the model may double-count isoprene SOA formation under low-NO_x conditions. We therefore followed the model settings of Pai et al. (2020) adopted by the default complex OA scheme, in which the traditional low-NO_x pathway is turned off to avoid double-counting.

Turning off this pathway does not affect the heterogeneous IEPOX SOA production, because the IEPOX uptake pathway is a separate formation channel that is explicitly represented in the model.

We have modified Lines 159-161 as follows: "Because IEPOX uptake is the dominant pathway for isoprene SOA under low-NO_x conditions (Surratt et al., 2010), the traditional low-NO_x VBS pathway for isoprene is disabled to avoid double-counting, following Pai et al. (2020)."

Line 137: Please provide information about the enthalpy of vaporization used for all components.

[R8] We have added the description of the applied enthalpy of vaporization (ΔH_{vap}) in Lines 154-155: "A constant enthalpy of vaporization (ΔH_{vap}) of 41.6 kJ mol^{-1} is adopted to calculate temperature-dependent partitioning (Chung and Seinfeld, 2002)", and discussion in Lines 206-209: "For ΔH_{vap}, we still use 41.6 kJ mol^{-1} in the FV OA scheme. This treatment may underestimate the temperature sensitivity of SOA formation. Low C^* species may have higher ΔH_{vap}, but specific ΔH_{vap} values are not well constrained in the literature (Epstein et al., 2010; Saha and Grieshop, 2016)."

Lines 148-149: Please clarify whether POA and SOA are allowed to re-evaporate, particularly under low-NO_x conditions.

[R9] POA and SOA species can undergo reversible gas-particle partitioning in response to changes in temperature and OA mass loading, unless those with $C^* \leq 10^{-2}$ ug m^{-3} or form from heterogeneous pathways. We have added the following sentence to clarify the re-evaporation of POA and SOA in the FV OA scheme in Lines 154-155: "A constant enthalpy of vaporization (ΔH_{vap}) of 41.6 kJ mol^{-1} is adopted to calculate temperature-dependent partitioning (Chung and Seinfeld, 2002)" and in Lines 161-162: "The SOA heterogeneous pathways are considered as irreversible processes in the model".

Lines 204-212: The authors appear to attempt to justify why they compare observations

from 2011–2022 with a single model simulation as the reference year. Although this approach includes several assumptions, it is common practice in such modeling studies, focusing mostly on the "climatological" evaluation of the model rather than interannual variability. However, it is unclear from the discussion whether only one year of observation (that of the model simulation) is used, or if the model is run with "average-year" emissions for comparison with observations from several nearby years. Note, however, that since the authors state that biomass burning contributes 46% to total SOA formation, this can vary significantly between years due to climate variability (e.g., El Niño). The earlier statement, therefore, appears more valid for anthropogenic emissions than for biomass burning emissions. Moreover, in Sect. 4, lines 403-404, the authors mention: "Significant changes in SOA production are expected in the future because of the reduction of anthropogenic emissions driven by tightened regional air quality standards (e.g., in China) and the intensified wildfires." This appears somewhat inconsistent with the earlier claim that using a single year would counteract potential biases. Overall, I recommend explicitly stating your methodology for the model evaluation and simply discussing the limitations of using a single reference year.

[R10] In this study, ground-based in situ measurements from 2015–2022 are used to evaluate the FV OA scheme against the reference simulation of 2018 to reduce computational cost. The reference year of 2018 is chosen as it is the middle year of the observation period, so that the potential biases caused by the rapid decrease in anthropogenic emissions in China can be counteracted. The interannual variations of anthropogenic emissions in other regions are relatively small, with limited influence on OA concentrations. For open biomass burning, the global emissions generally vary by less than ±30% during the observation period, potentially biasing simulated concentrations at wildfire-affected sites. We have clarified the statement in Lines 274-285: "To reduce computational cost, all of the observation-derived POA and SOA concentrations are compared to the simulated campaign-mean or season-mean concentrations for the reference year of 2018. The interannual variations of OA concentrations are expected to be small because of minor changes in anthropogenic emissions of organic precursors during this period in most regions except for China (Xu et al., 2026). Anthropogenic emissions in China had a rapid decline from 2013 to 2020, followed by a slow decline afterwards (Xu et al., 2026; Geng et al., 2024). By using the middle year of observations as the simulation reference, the potential biases in model–observation comparisons may be counteracted. Moreover, global open biomass burning emissions vary by ±30% during the evaluation period (van der Werf et al., 2025; Wiedinmyer et al., 2023). This variability is partly buffered in total OA by relatively stable anthropogenic and biogenic sources, but still affects model-observation comparisons on individual sites that are strongly influenced by wildfire. Additionally, recent studies show that the interannual variations of meteorology affect annual mean OA concentrations by within ±10% and have a minor influence on the trends (Ridley et al., 2018; Chen et al., 2024)".

We have further discussed how the interannual variations of open biomass burning affect the global OA burden estimate in this study in Lines 445-448: "Moreover, global anthropogenic emissions of S/LVOCs and IVOCs are relatively stable between 2010

and 2020 (Xu et al., 2026). The interannual variation of the SOA burden may be dominated by changes in the open biomass burning emissions (Wiedinmyer et al., 2023; van der Werf et al., 2025)".

Lines 355-356: Since this is not a climate study, it is better to move this sentence to the discussion section.

[R11] We have modified and moved the sentence to Lines 437-438 as follows: "The higher OA burden suggests that the climate impacts of OA may have been overlooked in previous studies."

Lines 356-357: The authors imply that roughly 31% of the total SOA burden in the model comes from anthropogenic SOA. How does this finding compare with other studies? Please discuss.

[R12] Our study suggests a higher anthropogenic SOA than previous studies, mainly due to the higher burden of SOA from S/LVOCs and IVOCs (Pye and Seinfeld, 2010; Tsimpidi et al., 2017; Hodzic et al., 2016; Jo et al., 2023; Pai et al., 2020). We have modified the corresponding discussion in Lines 440-443 as follows: "Open biomass burning, anthropogenic sources, and biogenic sources account for 48%, 32%, and 20% of the SOA burden, respectively. Our simulations suggest S/LVOCs and IVOCs from open biomass burning and anthropogenic sources together account for 1.2 Tg of the global SOA burden, much greater than previous estimates of 0.2-1.0 Tg (Pye and Seinfeld, 2010; Hodzic et al., 2016; Tsimpidi et al., 2017; Pai et al., 2020; Jo et al., 2023)."

Lines 360-361: Such a sentence should also be placed in the introduction to justify the later choice of an additional sensitivity simulation focusing on the wet removal efficiency. Otherwise, this comes very abruptly in the text.

[R13] We agree that the motivation for the wet deposition sensitivity simulation should be introduced earlier. We have added the following sentence at Lines 93-95: "Large uncertainties remain in the global budgets of POA and SOA, which limit our understanding of the climate and health effects of OA. Besides, the new parameterization for wet deposition may alter the OA budgets substantially (Luo et al., 2020; Lou et al., 2020)."

Section 2.4: Does the model separately track the different species calculated by VBS? How many (additional) species does the model use for the different sensitivity simulations? How much does the computational cost increase depending on the simulation setup?

[R14] We added 25 tracers and removed 1 tracer compared to the default complex OA scheme. The increase in computational cost is low, as the wall time of the revised scheme on our server increased by only 0.97% compared to the default complex scheme. When compared to the default simple scheme, our revised scheme increases the wall time by 15.43%.

Section 4: Please briefly discuss which of the assumptions used in the model have the greatest impact on the reported increases in POA and SOA burdens.
[R15] We have added corresponding discussion in Lines 433-435 as follows: "Compared to the default complex OA scheme, the POA burden increases 9.2 times, and the SOA burden increases by 56%. The former is attributed to the lower-volatility distribution of S/LVOC emissions, and the latter mainly to the increases in IVOC emissions and SOA yields."

Technical Comments:
Line 33: validation → evaluation
[R16] This correction has been made accordingly.

Line 45: "Immediately?" Under what ambient atmospheric conditions? Please remove or rewrite by explaining in more detail.
[R17] We have removed the word "immediately".

Line 400: Better split into a new paragraph.
[R18] Yes, it is now split into two paragraphs.

Table S1: What do OPOAs stand for? Also add some explanation for other species' names.
[R19] The definitions of the species listed in Table S1 are added in the table caption as follows: "POA and OPOA stand for primary organic aerosol and oxidized primary organic aerosol, respectively. IVSOA, ASOA, TSOA, and ISOA represent SOA species formed from gas-particle partitioning of oxidation products of IVOCs, aromatics, terpenes, and isoprene, respectively. SOAGX is formed from heterogeneous uptake of glyoxal and methylglyoxal. SOAIE is formed from heterogeneous uptake of isoprene epoxydiols (IEPOX), and LVOCOA represents heterogeneous isoprene SOA from non-IEPOX products. IONITA, MONITA, and AONITA represent organic nitrate from isoprene, monoterpenes, and aromatics, respectively."

Manuscript: The names of the main simulations, i.e., "Default" and "Base," are somewhat confusing since they both imply "reference" simulations. You can consider adopting more descriptive names/abbreviations or explicitly note their definitions throughout the manuscript (esp. figures/tables) to avoid confusion.
[R20] We have added "FV_" to the names of the simulations with the revised model scheme (e.g., "Base" to "FV_Base", Zero_Biof to "FV_Zero_Biof", etc.) to avoid confusion.

References:
Carlton, A. G., Bhave, P. V., Napelenok, S. L., Edney, E. O., Sarwar, G., Pinder, R. W., Pouliot, G. A., and Houyoux, M.: Model Representation of Secondary Organic Aerosol in CMAQv4.7, Environ. Sci. Technol., 44, 8553-8560,

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Hodzic, A., Kasibhatla, P. S., Jo, D. S., Cappa, C. D., Jimenez, J. L., Madronich, S., and Park, R. J.: Rethinking the global secondary organic aerosol (SOA) budget: stronger production, faster removal, shorter lifetime, Atmos. Chem. Phys., 16, 7917-7941, https://doi.org/10.5194/acp-16-7917-2016, 2016.
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Reviewer 2

This manuscript describes the implementation and evaluation of a full-volatility range OA scheme in the GEOS-Chem model. This represents an important effort to expand the representation of OA sources, especially from anthropogenic and biomass burning S/LVOCs. The paper is relatively straight-forward and clear. The main deficiency is a lack of detail in the text that make it difficult to understand how the results compare to the previous scheme in GEOS-Chem and what contributes to these changes. The authors should be able to easily expand the text and figures to provide these details.

[R0] We are grateful for the constructive comments from Reviewer 2. In the revised manuscript, we updated the citation of the MEIC-global-FVOC emission inventory paper, provided more details on the settings of the default schemes and the full-range volatility-binned OA scheme in individual sections, and added more information on the model-observation comparison. We have provided the model evaluation for the default schemes to compare with our revised scheme. We also have added more discussion in the global OA budget, especially the impact of open biomass burning and fragment processes. We provide point-by-point responses below and have revised the manuscript and supporting information (SI) accordingly.

I also note that the paper is heavily dependent on a new anthropogenic emission inventory (cited on p5, line 150-151) – until that paper has been peer-reviewed, this current manuscript should not be accepted for final publication.

[R1] We would like to inform the reviewer that the MEIC-global-FVOC emission inventory paper (Xu et al., 2026) has now been published in *National Science Review* (doi:10.1093/nsr/nwag281). We have modified the following sentence in Lines 210-211: "Anthropogenic emissions of the 9-bin lumped organic species are provided by the newly developed MEIC-global-FVOC inventory (Xu et al., 2026)." The reference in the manuscript has also been updated accordingly.

Major comments:

1. Section 2.2: This section is a bit confusing. Given that the authors are comparing their simulation with the default "complex OA" scheme in GEOS-Chem, it seems they should start with a complete description of this scheme (i.e. biogenic SOA formation is not mentioned in the first paragraph which seems to focus on the default scheme).

[R2] We agree with the reviewer that Section 2.2 would benefit from starting with a description of the default complex OA scheme. We have moved the description of the FV OA scheme to Section 2.3 and restructured Section 2.2 to present the default scheme more clearly: "The complex OA scheme, first described by Pye and Seinfeld (2010), represents S/LVOCs with two surrogates that have C^* of 1646 and 20 μg m^{-3} (Figure 1a and Figure S1a). This scheme applies a simplified treatment of gas-phase S/LVOC oxidation, with a rate constant of 2×10^{-11} cm^3 $molec^{-1}$ s^{-1} and 100 times lower C^* for their oxidation products (Shrivastava et al., 2006; Pye and Seinfeld, 2010). The oxidation products undergo gas-particle partitioning to form SOA, while S/LVOCs that remain in or condense into the particle phase during atmospheric processes without

chemical processing are classified as POA. S/LVOCs emissions are estimated as 1.27 and 1 times the OC emissions for anthropogenic sources and open biomass burning, provided by CEDS and FINNv2.5, respectively. The complex OA scheme has an option to treat POA as non-volatile, which is no longer recommended because of the large SOA underestimation (Schroder et al., 2018). Moreover, this scheme simulates SOA formation from the oxidation of VOCs (isoprene, terpenes, and aromatics) and IVOCs (treated as naphthalene) via a commonly used volatility-basis-set (VBS) approach with NO_x-dependent SOA yields (Pye et al., 2010). Precursor consumption under high- and low-NO_x conditions is calculated from peroxy radical loss via NO and HO_2 pathways, respectively (Henze et al., 2008). IVOC emissions are estimated as 66 times naphthalene emissions (Pye and Seinfeld, 2010). The OM:OC ratios of POA and SOA are assumed to be 1.4 and 2.1, respectively (Turpin and Lim, 2001). A constant enthalpy of vaporization (ΔH_{vap}) of 41.6 kJ mol^{-1} is adopted to calculate temperature-dependent partitioning (Chung and Seinfeld, 2002).

In addition, the complex OA scheme includes the SOA formation pathway of heterogeneous uptake of oxidation products from isoprene and monoterpenes, including isoprene epoxydiols (IEPOX), glyoxal, methylglyoxal, and nitrophenols on aerosols (Marais et al., 2016; Fisher et al., 2016; Bates et al., 2021). The heterogeneous uptake coefficient of IEPOX is sensitive to aerosol acidity and nucleophile concentration, while the other species use fixed reactive uptake coefficients. Because IEPOX uptake is the dominant pathway for isoprene SOA under low-NO_x conditions (Surratt et al., 2010), the traditional low-NO_x VBS pathway for isoprene is disabled to avoid double-counting, following Pai et al. (2020). The SOA heterogeneous pathways are considered as irreversible processes in the model."

2. Section 2.3: The section is missing details on the measurement techniques used (other than AMS), the particle size sampling, and the uncertainties. On P7, line 199: was the PMF performed consistently across all AMS/ACSM datasets? What is the uncertainty on these assigned factors? It is also a bit puzzling that the authors made such a comprehensive effort to assemble surface observations, but then selected only two aircraft campaigns. Why not more? Why these two campaigns?

[R3] We thank the reviewer for this suggestion. We have added the description for the methods and techniques of OC measurements in Lines 261-263 as follows: "The OC data are based on offline quartz-filter measurements following the IMPROVE_A, EUSAAR_2, or NIOSH heating protocols, and online measurements using OC-EC analyzers". For AMS/ACSM datasets, we have also added more description for the uncertainties in Lines 270-274: "We use 165 worldwide campaign-average surface concentrations (2015-2022) (Tables S8-S10 in the Supplement) and the season-mean concentrations from 21 sites (2015-2019) in the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS) in the model evaluation. For most campaigns, PMF was performed separately in each study, whereas the ACTRIS data were analyzed by a standardized PMF protocol across all sites. The uncertainties of the PMF-resolved POA and SOA mass loadings are generally within ±40% (Zheng et al., 2020)". The reason for using the measurements from WINTER and KORUS-AQ campaigns has been

added in Lines 295-296: "The two campaigns are chosen for model evaluation because they sampled anthropogenically influenced atmospheric backgrounds and the PMF results are publicly accessible".

3. It is strange to present model comparisons with observations prior to describing the new model budget. I would suggest that the authors invert their ordering in Section 3 (i.e. start with what is now Section 3.3).

[R4] We understand the rationale for presenting the new budget earlier to help readers see the main result sooner. We have chosen to retain the current structure for the following reasons: (1) the evaluation establishes the credibility of the revised scheme before the new budget is presented; (2) readers can first understand the separate contributions of the emission update and the scheme update, which makes the subsequent budget changes easier to interpret. To address the reviewer's concern and improve readability, we have revised the last paragraph of the Introduction in Lines 96-104 as follows: "In this study, we develop a new OA simulation framework in GEOS-Chem chemical based on volatility-binned anthropogenic and biomass burning FVOC emissions. Model performance is evaluated against a comprehensive observation dataset that includes surface, shipborne, and airborne measurements, with POA and SOA further assessed against aerosol mass spectrometer source apportionment. Building on this evaluation, we provide new constraints on the global distributions, source contributions, and population exposures of POA and SOA. The results are compared among studies, and the sensitivity of the simulations to wet-deposition parameterizations is discussed."

4. The plots do not provide sufficient comparison with the default scheme (for the model version, year and emissions that the authors have simulated). For example, the statements on p10 line 294-295 and p11 line 346 are not currently supported. Observational comparison should show default and new (Figures 3, 4, 5, 6). As should all budget plots (Figure 9 and 10).

[R5] We have added new figures in the SI (Figure S7, S9, and S10) to present the model performance of the default complex OA scheme and show the improvements of our updated simulation. We have also modified the discussion in Lines 379-384: "As shown in Figure S10, the default simple OA scheme with a nonvolatile POA assumption overestimates POA, while the default complex OA scheme underestimates POA. Both of the default schemes overestimate SOA during the WINTER campaign, but underestimate it during the KORUS-AQ campaign. In contrast, the FV OA scheme in this study shows more robust performance for POA and SOA vertical profiles across different source regions and chemical regimes" and Lines 433-434: "Compared to the default complex OA scheme, the POA burden increases by 9.2 times, and the SOA burden increases by 56%".

5. The plots do not sufficiently detail which sectors contribute to the simulated changes. The manuscript should include plots comparing the default POA and SOA from biogenic, fire, and anthropogenic sources, respectively. In particular, while the primary

advance of the scheme is the focus on anthropogenic and biomass burning full-volatility sources, the authors seem to have made substantial changes to the biogenic SOA formation. These are not quantified or discussed in the text (or even mentioned in the abstract).

[R6] Compared to the default complex OA scheme, the POA increase in our updated framework is attributed to the lower-volatility distribution of S/LVOC emissions, and the SOA increase mainly to the increases in IVOC emissions and SOA yields. Overall, both anthropogenic sources and open biomass burning together drive the increases in POA and SOA. In contrast, the change in biogenic SOA is limited.

We have added the discussion in Lines 441-445: "Our simulations suggest S/LVOCs and IVOCs from open biomass burning and anthropogenic sources together account for 1.2 Tg of the global SOA burden, much greater than previous estimates of 0.2-1.0 Tg (Pye and Seinfeld, 2010; Hodzic et al., 2016; Tsimpidi et al., 2017; Pai et al., 2020; Jo et al., 2023). The estimated biogenic SOA burden is 0.4 Tg, which falls within the range of previous estimates of 0.3-0.7 Tg. The relative fraction of biogenic SOA in SOA is lower because of the increased contribution from S/LVOCs and IVOCs."

6. How much of the fire source is POA vs SOA? P6 suggests that the emission of IVOC and S/LVOC from fires are large (exceeding the anthropogenic emissions). Can the authors comment on how this much SOA from fires is consistent with previous field studies? (e.g. Cubison et al., 2011)

[R7] In our simulation, the open biomass burning-derived OA burden is 1.4 Tg, of which about 22% is POA, and 78% is SOA. Our simulated SOA production from open biomass burning (67.3 Tg yr^{-1}) is much higher than the global net OA enhancement from BB aging of 8 ± 7 Tg OA yr^{-1} estimated by Cubison et al. (2011), and is at the high end of the smog chamber-based estimate from Bruns et al. (2016). Cubison et al. (2011) may not fully account for the SOA formation from the aging and conversion of POA, which leads to a relatively low estimate compared to other observation-based estimates (Hodzic et al., 2016). Our simulation, however, may overestimate SOA production from open biomass burning, as the global emissions from FINNv2.5 are generally higher than other emission inventories (Wiedinmyer et al., 2023).

We have added the following discussion in Lines 456-463: "The estimated SOA production from open biomass burning is 67.3 Tg yr^{-1}, which falls within the range of other model estimates (Shrivastava et al., 2017), lies at the high end of the chamber-based estimates from Bruns et al. (2016), and is much higher than the observation-based estimates reported by Hodzic et al. (2016). This may be explained by possibly higher fire emissions in FINNv2.5 and the contribution of IVOCs and SVOCs that are not fully captured by observation-based approaches (Cubison et al., 2011; Wiedinmyer et al., 2023). It is worth noting that the emissions from open biomass burning remain highly uncertain. Their OC emissions differ by over 100% in global annual estimates among emission inventories, and the difference is even larger regionally (Carter et al., 2020; Wiedinmyer et al., 2023), which is a key limitation of current SOA budget estimation."

7. The fire source would be highly variable year-to-year (and thus statement on p7, line

207 is false). Given that the study appears to find a large fire source, it's not clear that one year of simulation is sufficient to characterize a contribution from fires. The authors could either simulate additional years (5-10 years?) to provide some variability estimate, or clearly state this uncertainty up-front on their single-year values (in abstract and throughout).

[R8] We agree with the reviewer that the interannual variations of open biomass burning can affect the OA concentration and burden. This in turn affects the model-observation comparisons and the global OA burden estimate in this study. We have added the description in Lines 281-283 in the Methods section: "Moreover, global open biomass burning emissions vary by ±30% during the evaluation period (van der Werf et al., 2025; Wiedinmyer et al., 2023). This variability is partly buffered in total OA by relatively stable anthropogenic and biogenic sources, but still affects model-observation comparisons on individual sites that are strongly influenced by wildfire" and the discussion in Lines 445-448 in Section 3.3: "Moreover, global anthropogenic emissions of S/LVOCs and IVOCs are relatively stable between 2010 and 2020 (Xu et al., 2026). The interannual variation of the SOA burden may be dominated by changes in the open biomass burning emissions (Wiedinmyer et al., 2023; van der Werf et al., 2025)".

Additional comments:

1. P2, line 56: define C^* upon first usage

[R9] We have added the definition of C^* in Line 60 as follows: "saturation concentration (C^*)".

2. P4, line 127: provide reference for all loss correction factors

[R10] We have added a reference for the vapor wall-loss correction factor in Lines 178-184: "Organic vapor wall loss can lead to significant underestimation of SOA yields for the high-NO_x oxidation of aromatics, the low-NO_x oxidation of aromatics, and the high-NO_x pathway of isoprene SOA formation, respectively. We therefore adopt vapor wall-loss correction factors of 1.2, 1.9, and 2.2 that were obtained by Zhang et al. (2014) in chamber experiments for the three categories."

3. P5, line 146-147: is fragmentation not considered? If not, might the scheme overestimate the oxidation products of S/LVOCs?

[R11] Fragmentation reactions are not explicitly considered in our current scheme because their parameterization remains highly uncertain. Omitting fragmentation processes may lead to OA overestimation in remote regions, while our model-observation comparisons do not show large model bias in remote environments and at high altitude. We have added the following discussion in Lines 509-515: "Moreover, there is a lack of a better representation of multi-day aging of OA, which may further improve the spatial and temporal model skills. Chemical loss pathways (e.g., via fragmentation and photolysis) may affect the OA simulation significantly. The fragmentation loss is not explicitly represented in our FV OA scheme because the model-observation comparisons do not suggest large model bias in remote environments and at high altitude. In previous studies, the SOA photolysis losses were

implemented to balance the large increase in SOA burden after adding uncertain SOA production, which is a historical problem of underestimated SOA concentrations due to the limited capability to represent IVOC and SVOC emissions and their SOA formation (Hodzic et al., 2016; Lou et al., 2020)."

4. P6, lines 173, 190: These figures (Figure S3 and S4) are new results of this scheme and should be shown in the main text, not the SI.
[R12] We have moved Figures S3 and S4 to Figures 2 and 3.

5. Figure 1: missing a description of circles vs triangles. Also unclear if oxidation arrows are pointing to specific circles or groups (e.g. the kOH, SVOCs arrow is placed next to the gas phase C^*=1 bin). Where do emissions of BVOCs fit on this diagram?
[R13] We have revised the figure caption of Figure 1 as follows: "Colored circles and triangles represent gaseous and particle-phase tracers in the schemes, respectively". All VOC precursors, including BVOCs, follow the pathway of VOC oxidation on this diagram, except for those that form SOA from heterogeneous reactions.

6. Figure 2: only shows surface. Perhaps the authors could also show a cross-section like in Figure 7d-f in the SI?
[R14] We have added a new figure in the SI (Figure S5) showing the zonal concentration differences between "FV_Base" and "Default" for OA, POA, and SOA.

7. P8, line 232: The figures are shown in absolute concentrations, so these percentages do not match the information provided. I suggest if the authors want to cite percentages that they provide percent difference plots in the SI.
[R15] We have revised the discussion in Lines 308-310 as follows: "By contrast, the simulated SOA mass concentrations increase by 1-5 μg m^{-3} in East Asia, Europe, southern America, and Africa compared to the default complex OA scheme, mainly due to the increased IVOC emissions (Table S5 in the Supplement)".

8. Figure 5e-h: missing stats on panels
[R16] We have added statistical metrics (NMB and r) to each panel of Figure 7e-h (former Figure 5e-h).

9. Figure 8: missing units on the cumulative probability distribution plots
[R17] We have added a description for the units to both axes of the cumulative probability distribution plots in the figure caption of Figure 10 (former Figure 8) as follows: "cumulative distributions represent the population percentages (in the unit of %) exposed to annual mean POA and SOA concentrations (in the unit of μg m^{-3}) in different regions and countries".

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