Response to Referee #1

Review of Improved representation of isoprene-derived secondary organic aerosol in CAM6-Chem reveals regional contrasts in its long-term changes over China by Wenxin Zhang et al., This manuscript presents an updated representation of isoprene-derived secondary organic aerosol (ISOA) in CAM6-Chem, incorporating both the IEPOX (low-NOx) pathway and the MAE/HMML (high-NOx) pathway, along with an explicit treatment of aerosol liquid water in multiphase chemistry. The authors evaluate the updated mechanism against multiple observational datasets and use it to investigate long-term (2000-2019) ISOA trends across China. The study finds that while national-mean ISOA trends are weak, they mask strong regional contrasts, with increasing ISOA mass in Southwest China (SWC) driven primarily by enhanced biogenic isoprene emissions and decreasing ISOA in the Shaanxi-Gansu-Ningxia region (SGN) driven by declining sulfate and changing NOx emissions. Overall, this is a careful and well-executed modeling study that addresses a known gap in global models. The manuscript is suitable for ACP, but several aspects require clarification and strengthening before publication.

We are very grateful to the reviewer for the constructive evaluation and helpful suggestions, which have allowed us to clarify and improve the manuscript. Before addressing the reviewer's comments point by point, we briefly summarize the major revisions made in response to Referee #1. The reviewer's comments are shown in **bold blue text**, while our responses and the corresponding revised manuscript text are shown in black text. Line numbers refer to the revised manuscript.

In response to Referee #1, we made several major revisions to strengthen the manuscript. We developed a clearer conceptual framework by interpreting regional ISOA changes as the balance between precursor supply and heterogeneous reaction capacity, and added a new synthesis schematic (Fig. 9) to summarize the contrasting controls in SWC and SGN. We also revised the multiple linear regression interpretation to emphasize statistical associations rather than strict causal attribution, added a predictor covariance/collinearity discussion and a new Pearson correlation analysis (Fig. S9), and included a brief summary of the regression approach in the main text. In addition, we expanded the discussion of key model assumptions and uncertainties, including HMML/MAE uptake parameters, 100% yield and non-volatility assumptions, phase separation, viscosity, and gas–particle partitioning, and revised the related discussions of model bias and broader implications.

Major Comments

Comment#1: The manuscript contains an important underlying idea that ISOA formation reflects a balance between precursor supply (e.g., isoprene, NOx) and heterogeneous reaction capacity (e.g., sulfate, aerosol liquid water, acidity). However, this is not fully developed into a clear conceptual framework. For example, on lines 369-373, the authors note that increases in isoprene emissions coincide with decreases in sulfate and aerosol liquid water, implying competing controls on ISOA formation. Similarly, the regional contrast between SWC and SGN is interpreted in terms of precursor supply versus reaction medium limitations (e.g., lines 410-417). These are strong and insightful points, but they need to be synthesized into a broader framework. Consider explicitly framing the results in terms of regimes (e.g., precursor-limited vs. reaction-medium-limited ISOA formation) and summarizing this in a conceptual schematic or synthesis figure.

Response: We thank the reviewer for this insightful suggestion. We have revised the discussion to explicitly frame the regional ISOA trends in terms of the balance between precursor supply and heterogeneous reaction-medium capacity. In this framework, SWC represents a precursor-driven regime, where enhanced isoprene emissions dominate the ISOA increase, whereas SGN represents a reaction-medium-limited regime, where reductions in sulfate and aerosol liquid water suppress heterogeneous ISOA formation. We also added a conceptual schematic to summarize these contrasting regional mechanisms. These revisions are provided in lines 613–620, and the new schematic is shown in Fig. 9.

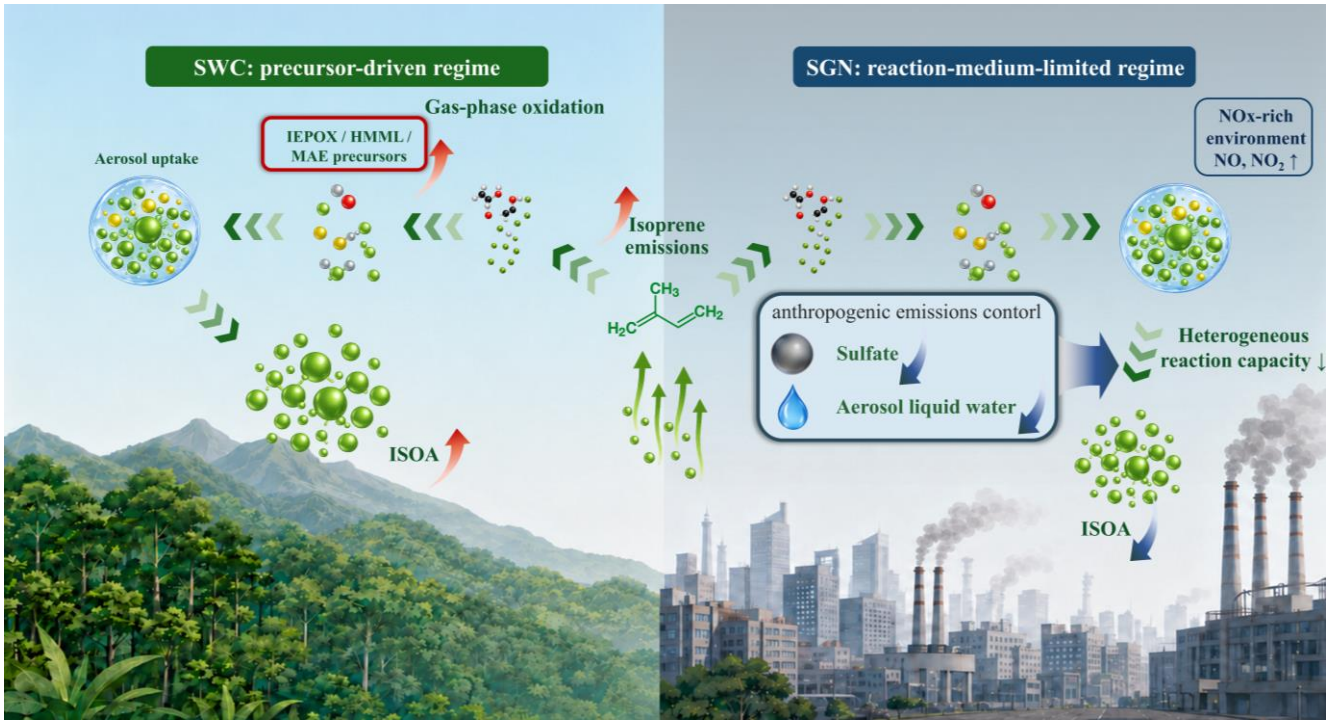

Figure 9. Schematic illustration of contrasting regional controls on surface isoprene-derived secondary organic aerosol (ISOA) formation in Southwest China (SWC; left) and the Shaanxi–Gansu–Ningxia region (SGN; right). The background scenes and selected graphical elements were created with the assistance of OpenAI's image-generation tools in ChatGPT (OpenAI, 2026).

Lines 613–620:

Taken together, the contrasting ISOA trends in SWC and SGN highlight the need for a coupled precursor-multiphase reaction framework. The SWC case represents a more precursor-driven regime, in which enhanced biogenic isoprene emissions and NOx-dependent oxidation promote epoxide precursor formation and enhance ISOA production despite less favorable aerosol multiphase reaction conditions. In contrast, the SGN case represents a more reaction-medium-limited regime, in which declining sulfate and aerosol liquid water suppress heterogeneous uptake and particle-phase product formation, even when precursor supply remains relatively high. This regime-based interpretation emphasizes the balance between precursor supply, NOx-dependent oxidation chemistry, and aerosol multiphase reaction conditions, as summarized schematically in Fig. 9.

Comment#2: The manuscript relies heavily on multiple linear regression to quantify the relative importance of drivers. For example, in the supplement (Text S1), the authors state that isoprene emissions account for ~49.9% of the relative importance of ISOA variability in SWC based on standardized regression coefficients. Similarly, in the main text (Section 3.3), regression-based contributions are used to attribute long-term trends. While these diagnostics are useful, the predictors (e.g., sulfate, aerosol water, emissions) are likely to be strongly covarying over time, which complicates interpretation. Because aerosol liquid water is calculated thermodynamically within MOSAIC from inorganic aerosol composition, including sulfate, sulfate and aerosol liquid water are not fully independent predictors. This should be acknowledged in the regression interpretation, particularly where sulfate and aerosol liquid water are both treated as separate explanatory variables. Please clarify that regression-based importance reflects statistical association rather than strict causality and discuss potential collinearity among predictors. I suggest tempering the language when assigning quantitative contributions. For example, replace "accounts for X%" with "is strongly associated with" or "emerges as a leading statistical predictor" while also discussing the potential impact of predictor covariance on the interpretation.

Response: We thank the reviewer for this helpful suggestion. We have revised both Sect. 3.3 and Supplementary Text S1 to clarify the interpretation of the multiple linear regression results. In the revised manuscript, the regression-based relative importance and trend contributions are now described as diagnostic indicators of statistical associations, rather than as strict causal attribution or fully independent contributions. We have also tempered the language when discussing quantitative contributions, replacing causal-sounding expressions with wording such as "is strongly associated with" and "emerges as a leading statistical predictor". In addition, we added a specific discussion of predictor covariance and potential collinearity, particularly among sulfate, aerosol liquid water, and aerosol acidity, which are physically coupled within the aerosol chemical environment and therefore should not be interpreted as fully independent predictors. This new discussion has been added in Sect. 3.3, lines 557–568. To further support this interpretation, we also added a pairwise Pearson correlation analysis among predictors in Supplementary, with the results presented in the new Fig. S9.

Lines 557–568:

We further evaluated potential predictor correlations and collinearity by calculating pairwise Pearson correlation coefficients among the predictors used in the regression analysis (Fig. S9). The results indicate that some predictors are not fully independent. In both SWC and SGN, sulfate, aerosol liquid water, and $\log_{10}(H^+)$ show moderate to strong positive correlations, with correlation coefficients of 0.70–0.81 in SWC and 0.75–0.88 in SGN. These correlations are physically expected because aerosol liquid water is thermodynamically calculated in MOSAIC based on inorganic aerosol composition, including sulfate, as well as meteorological conditions. Sulfate also contributes to aerosol acidity and aerosol liquid water formation, while aerosol liquid water can further influence aqueous-phase uptake and processing. Therefore, sulfate, aerosol liquid water, and aerosol acidity are treated here as physically connected components of the aerosol chemical environment. Accordingly, the regression-based relative contributions associated with these covarying predictors should be interpreted as diagnostic indicators of statistical associations with ISOA variability, rather than as a strict decomposition of independent causal effects, because they may partly reflect coupled variations among sulfate, aerosol liquid water, and aerosol acidity.

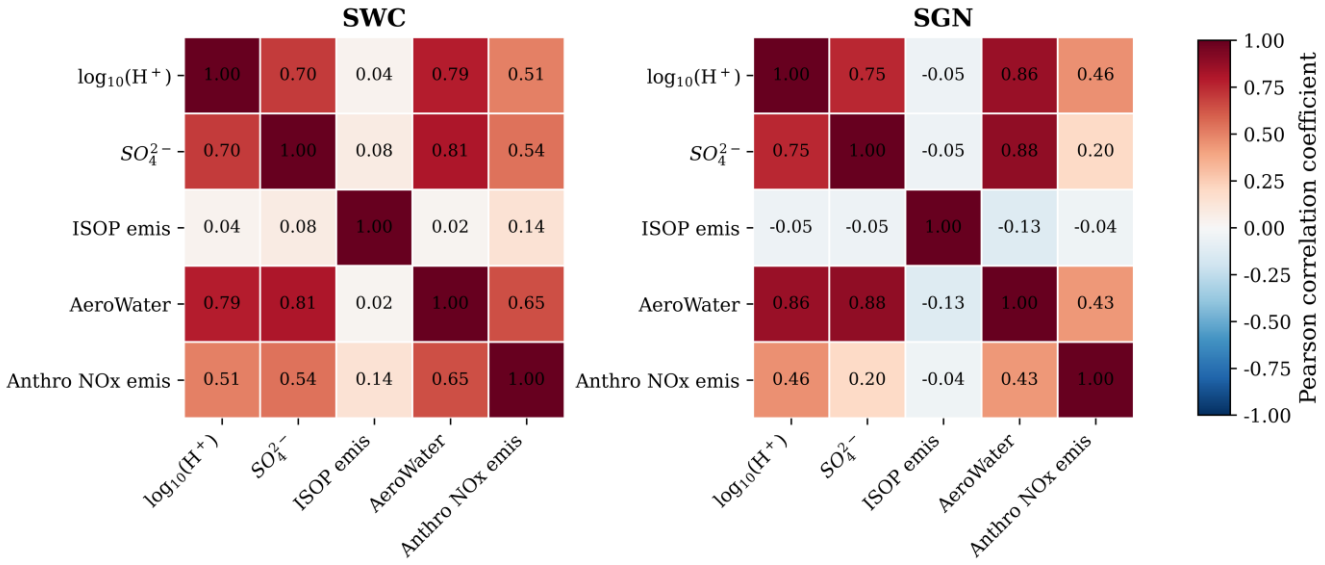

Figure S9: Pairwise Pearson correlation coefficients among predictors used in the multiple linear regression analysis for Southwest China (SWC) and the Shaanxi–Gansu–Ningxia region (SGN). The predictors include aerosol acidity represented by $log_{10}(H^+)$, sulfate (SO_4^{2-}), biogenic isoprene emissions (ISOP emis), aerosol liquid water (AeroWater), and anthropogenic NOx emissions (Anthro NOx emis). Warm colors indicate positive correlations, whereas cool colors indicate negative correlations.

Comment#3: Several key assumptions and uncertainties are central to the modeling framework and should be more clearly discussed. For example, lines 151-153, HMML and MAE are assumed to have the same solubility and diffusivity as IEPOX and lines 196-198, SOA yields from epoxide uptake are assumed to be 100% and treated as non-volatile. These assumptions are consistent with prior studies but introduce uncertainty in both magnitude and trends of ISOA that shape interpretation. How do these assumptions influence the reported findings concerning the relative importance of high-NOx vs. low-NOx pathways, sensitivity to sulfate and aerosol liquid water and long-term trend attributions?

Response: We thank the reviewer for this important comment. We agree that these assumptions represent key uncertainties in the current modeling framework and that their implications should be more clearly discussed. We have revised Sect. 2.1.2 to explicitly acknowledge that, due to limited laboratory constraints on the multiphase kinetics of HMML and MAE, both species are represented using the same solubility and diffusivity values as IEPOX. We now clarify that this treatment allows a consistent implementation of reactive uptake for the three epoxides, but introduces uncertainty in the absolute uptake rates of HMML and MAE and therefore in the simulated quantitative importance of the high-NOx pathway relative to the low-NOx pathway.

In addition, we expanded the limitation discussion in Sect. 4 to address the broader implications of these assumptions. Specifically, we now discuss that the assumptions of 100% SOA yield from epoxide reactive uptake and non-volatile epoxide-derived products may affect the absolute ISOA burden and may amplify the apparent sensitivity of ISOA to sulfate, aerosol liquid water, and acidity, because stronger reactive uptake is directly translated into non-volatile SOA formation. We also clarify that these assumptions may influence the quantitative interpretation of long-term trend contributions. Therefore,

the regression-based trend attributions are interpreted as diagnostic statistical associations under the adopted chemical framework, rather than as exact causal contributions independent of these model assumptions. These revisions have been incorporated in Sect. 2.1.2 (lines 179–190) and Sect. 4 (lines 664–672 and lines 675–677).

Lines 179–190:

Due to limited laboratory constraints on the multiphase kinetics of HMML and MAE, HMML and MAE are treated as IEPOX-like epoxide precursors in the current implementation. This treatment is consistent with previous modeling practice. For example, Pye et al. (2013) treated HMML like MAE in terms of heterogeneous uptake and assumed the particle-phase reaction rate constants of MAE to be the same as those of IEPOX due to the lack of kinetic data. Zhang et al. (2022) also noted that most HMML-related reaction parameters in models are commonly assumed to be the same as those for IEPOX. Following these studies, HMML and MAE are assigned the same solubility, diffusivity, and condensed-phase reaction constants as IEPOX in our implementation. Although a recent field-based study has estimated effective uptake parameters for IEPOX and lumped HMML/MAE, these constraints are not directly applicable here because they do not provide a complete set of standalone, compound-specific solubility, diffusivity, and branch-resolved condensed-phase reaction constants for HMML and MAE (Zhu et al., 2025). As a result, this IEPOX-like treatment introduces uncertainty in the absolute uptake rates of HMML and MAE and may affect the modeled quantitative partitioning between high-NOx and low-NOx ISOA formation pathways.

Lines 664–672:

HMML and MAE are also represented using the same solubility, diffusivity, and condensed-phase reaction parameters as IEPOX because compound-specific laboratory constraints remain limited. This IEPOX-like treatment introduces uncertainty in the absolute uptake rates of high-NOx epoxides and in the quantitative $ISOA_{HMML+MAE}/ISOA_{IEPOX}$ ratio. The assumptions of 100% ISOA yield from epoxide reactive uptake and non-volatile epoxide-derived ISOA introduce additional uncertainty, although they are consistent with previous global ISOA modeling studies (Marais et al., 2016; Jo et al., 2019, 2021). Lower effective yields or partial volatility, especially for products such as 2-MT and 2-MGA, could reduce simulated ISOA concentrations and their sensitivity to sulfate, aerosol liquid water, and acidity. Consistent with this need, gas–particle partitioning treatments for 2-MT in regional models suggest a useful direction for future improvement in global models (Pye et al., 2018; Shrivastava et al., 2022).

Lines 675–677:

Taken together, these assumptions mean that the long-term trends and regression-based attributions diagnosed here should be interpreted within the adopted epoxide-focused chemical framework, rather than as exact causal contributions independent of model structure and parameterization.

Comment#4: The updated model improves SOA representation but still exhibits substantial positive bias for key tracers (e.g., lines 232-233, normalized mean biases of 47.4% (AIETET) and 81.6% (AHMGA) are reported). While the manuscript notes that similar biases exist in previous studies, the causes of these discrepancies are not explored in detail. Provide additional discussion of possible sources of bias such as: missing competing loss pathways, uncertainties in heterogeneous uptake, representation of precursor chemistry.

Response: We thank the reviewer for this helpful suggestion. We have expanded the discussion of the remaining positive biases in the model evaluation section. In the revised manuscript, we now clarify that the positive biases in AIETET and AHMGA may arise from several sources. First, missing or simplified competing product pathways may cause an over-allocation of epoxide precursors to the explicitly represented ISOA products. For example, IEPOX-derived products may be partly diverted to C5-alkene triols or other particle-phase products that are not explicitly represented here. Second, uncertainties in heterogeneous uptake parameterization, including mass transfer, aerosol phase state, organic-shell viscosity, and phase separation, may affect the simulated product distribution and the abundance of ISOA subspecies. We now also clarify that the current core-shell uptake treatment assumes the same reaction rate coefficient in the organic shell and aqueous core; if phase separation and viscous organic coatings impose stronger diffusion limitations than represented in the model, effective epoxide uptake may be lower in the real atmosphere, leading to overestimation of modelled epoxide-derived ISOA. Third, uncertainties in the multiphase uptake and reaction parameters of HMML/MAE may contribute to the AHMGA bias, because compound-specific constraints for these high-NOx epoxides remain limited. We also note that similar positive biases have been reported in previous modeling studies, but that the exact sources of bias may differ across models and configurations. These revisions have been incorporated in Sect. 3.1 (lines 299–314).

Lines 299–314:

Several factors may contribute to these positive biases. First, missing or simplified competing loss pathways may cause an over-allocation of epoxide precursors to the explicitly represented ISOA products. For example, a fraction of IEPOX-derived products in the real atmosphere may be diverted to C5-alkene triols or other particle-phase products that are not explicitly represented here, thereby reducing the effective yield of AIETET (Pye et al., 2013). In addition, previous studies have shown that AIETET and AHMGA are sensitive to factors such as pathway branching, mass transfer, reaction kinetics, and aerosol phase state. For example, uncertainties in the relative rates of water addition versus organosulfate formation can affect the predicted product distribution between 2-MT and organosulfate products (Budisulistiorini et al., 2017). Similarly, assumptions related to aerosol phase state, organic-shell viscosity, and phase separation can influence IEPOX reactive uptake and may affect the simulated abundance of IEPOX-derived tracers (Zhang et al., 2018, 2019; Chen et al., 2024b). In particular, the current core-shell uptake treatment assumes the same reaction rate coefficient in the organic shell and aqueous core. If phase separation and viscous organic coatings impose stronger diffusion limitations than represented here, effective epoxide uptake may be reduced in the real atmosphere, and the model may overestimate ISOA formation, especially under conditions with high organic coatings or strong phase separation. For AHMGA, uncertainties in the multiphase uptake and

reaction parameters of HMML/MAE may also contribute to the model bias, because compound-specific constraints for these high-NOx epoxides remain limited (Zhang et al., 2022; Zhu et al., 2025).

Comment#5: The manuscript focuses on China, which is appropriate given the emissions context. However, the findings likely have broader implications. For example, on lines 80-85, the authors discuss prior work linking sulfate reductions to declines in ISOA in the United States. The present results extend this by showing that inclusion of high-NOx pathways can alter both magnitude and attribution of ISOA trends. What may be inferred for the US or other regions based on the findings reported here?

Response: We thank the reviewer for this insightful comment. We agree that although the present study focuses quantitatively on China, the findings have broader mechanistic implications for other regions where biogenic isoprene emissions interact with anthropogenic emissions and aerosol chemical conditions.

Previous studies in the United States, especially in the southeastern United States, have shown that $ISOA_{IEPOX}$ formation is strongly influenced by sulfate and aerosol acidity, and that reductions in sulfate can lead to decreases in $ISOA_{IEPOX}$ (e.g., Marais et al., 2016; Budisulistiorini et al., 2017; Liu et al., 2023). Our results are consistent with this general understanding, but further suggest that sulfate reduction alone may not fully explain ISOA trends in regions where high biogenic isoprene emissions overlap with elevated NOx conditions. By explicitly including high-NOx epoxide pathways, the model can represent additional HMML/MAE-derived products, which could potentially alter both the magnitude and composition of ISOA and, therefore, could have implications for interpreting long-term ISOA changes.

For the United States, this implies that sulfate reductions probably remain an important driver of declining $ISOA_{IEPOX}$, especially in the southeastern United States. However, in typical regions where biogenic isoprene emissions increase substantially or anthropogenic NOx emissions continue to rise, NOx-dependent epoxide formation might become more relevant, and the simulated response of total ISOA could differ from that inferred from sulfate reductions alone. Therefore, the net ISOA response would likely reflect the combined effects of isoprene emissions and related precursor supply, as well as the heterogeneous reaction environment. More broadly, in other high-isoprene regions such as the Amazon and Central Africa, abundant biogenic isoprene emissions provide sufficient precursor supply and could potentially drive substantial ISOA variability, which may not necessarily follow the influence of sulfate changes alone. In regions strongly affected by urban pollution or increasing anthropogenic NOx emissions, NOx-dependent chemistry could also become more relevant for interpreting ISOA changes.

Thus, the broader implication of this work is that ISOA trends in different regions may be better interpreted as the combined outcome of isoprene emissions, NOx-dependent gas-phase precursor supply, and the heterogeneous reaction environment, with the dominant factors varying among typical regions rather than being universally driven by sulfate changes alone. Given that the present study focuses on China, we have limited this discussion to a qualitative outlook rather than extending

the analysis to other regions. We have added a brief discussion in the outlook section to emphasize that future global-scale quantification of these effects would be important for assessing their broader implications lines 684–687.

Marais, E. A., Jacob, D. J., Jimenez, J. L., Campuzano-Jost, P., Day, D. A., Hu, W., Krechmer, J., Zhu, L., Kim, P. S., Miller, C. C., Fisher, J. A., Travis, K., Yu, K., Hanisco, T. F., Wolfe, G. M., Arkinson, H. L., Pye, H. O. T., Froyd, K. D., Liao, J., and McNeill, V. F.: Aqueous-phase mechanism for secondary organic aerosol formation from isoprene: application to the southeast United States and co-benefit of SO_2 emission controls, Atmos. Chem. Phys., 16, 1603–1618, https://doi.org/10.5194/acp-16-1603-2016, 2016.

Budisulistiorini, S. H., Nenes, A., Carlton, A. G., Surratt, J. D., McNeill, V. F., and Pye, H. O. T.: Simulating aqueous-phase isoprene-epoxydiol (IEPOX) secondary organic aerosol production during the 2013 Southern Oxidant and Aerosol Study (SOAS), Environ. Sci. Technol., 51, 5026–5034, https://doi.org/10.1021/acs.est.6b05750, 2017.

Liu, Y., Dong, X., Emmons, L. K., Jo, D. S., Liu, Y., Shrivastava, M., Yue, M., Liang, Y., Song, Z., He, X., and Wang, M.: Exploring the factors controlling the long-term trend (1988–2019) of surface organic aerosols in the continental United States by simulations, J. Geophys. Res.-Atmos., 128, e2022JD037935, https://doi.org/10.1029/2022JD037935, 2023.

Lines 684–687:

Future global-scale simulations and attribution analyses would also be useful for evaluating whether the updated ISOA mechanism has broader implications for other high-isoprene regions, where climate change and human activities may shift the relative roles of biogenic precursor supply, anthropogenic emissions, and the heterogeneous aerosol reaction environment in shaping future ISOA responses.

Minor Comments

Comment#6: Some sections could be streamlined for clarity (e.g., repeated discussion of pathway sensitivities).

Response: We thank the reviewer for this helpful suggestion. We have reviewed the manuscript and streamlined several sections to improve clarity and reduce repetition. In particular, we revised the repeated discussion of pathway sensitivities and consolidated overlapping statements regarding the sensitivities of ISOA formation to precursor supply and heterogeneous reaction conditions. Where appropriate, we shortened redundant explanations and kept the more detailed discussion in the sections most directly related to the corresponding results. These revisions improve the readability of the manuscript without changing the scientific interpretation.

Comment#7: Consider adding a synthesis figure summarizing regional drivers of ISOA trends.

Response: We thank the reviewer for this helpful suggestion. This comment is closely related to the Comment#1. In response, we have added a new synthesis schematic, now shown as Fig. 9, to summarize the contrasting regional drivers of ISOA trends in Southwest China (SWC) and the Shaanxi–Gansu–Ningxia region (SGN). The figure explicitly frames regional ISOA formation as the balance between precursor supply, including isoprene and NOx, and heterogeneous reaction capacity, including sulfate, aerosol liquid water, and aerosol acidity. Specifically, SWC is characterized as a precursor-driven regime, where enhanced biogenic isoprene emissions and gas-phase oxidation products favor ISOA formation, whereas SGN is

characterized as a reaction-medium-limited regime, where reductions in sulfate and aerosol liquid water weaken heterogeneous uptake and aqueous-phase processing. We have also added a synthesis paragraph in lines 613–620 to clarify this framework and link the figure to the preceding trend and attribution results.

Comment#8: A brief summary of the regression approach in the main text (currently primarily in the supplement) would improve accessibility.

Response: We thank the reviewer for this helpful suggestion. We have added a brief summary of the multiple linear regression approach in the main text to improve accessibility. The new text describes the construction of the regression framework linking simulated monthly ISOA concentrations to key controlling factors and clarifies its two complementary applications: standardized monthly anomalies were used to diagnose the relative statistical importance of predictors on a common scale, whereas unstandardized monthly anomalies in physical units were used for the trend attribution diagnostic presented in Table 1. This revision has been incorporated in Section 3.3 (lines 528–541).

Lines 528–541:

To quantitatively diagnose the contributions of influential factors and better understand why ISOA trends differ between SWC and SGN, we constructed a multiple linear regression framework linking simulated monthly ISOA concentrations to key controlling factors. This regression framework was used in two complementary ways. Both analyses were based on regional monthly anomalies derived by first spatially averaging the original monthly gridded data over SWC and SGN to obtain regional-mean monthly time series and then removing the mean seasonal cycle. First, these monthly anomalies were standardized to zero mean and unit standard deviation to compare the relative statistical importance of different predictors on a common scale, as summarized in Text S1 and Table S1. This standardized analysis primarily diagnoses the leading predictors of month-to-month ISOA variability. Second, for the trend attribution diagnostic presented in Table 1, we used the same monthly anomalies without standardization, retaining the original physical units of each predictor. Multiple linear regression was subsequently applied to these unstandardized anomalies to obtain sensitivity coefficients in physical units. These sensitivity coefficients (α) describe how regional ISOA responds to changes in each predictor under the regression framework. For each predictor, its contribution to the simulated 20-year ISOA trend ($C_{/20yr}$) was then estimated by multiplying its regression sensitivity by its own 20-year trend ($dX_{/20yr}$). The relative contribution of each factor was further expressed as C/dY (%), where dY denotes the simulated 20-year ISOA trend in the corresponding region.

Comment#9: Line 230: "We validated the simulated AIETET against ….". Validated should be evaluated.

Response: We thank the reviewer for this suggestion. We have replaced "validated" with "evaluated" to use more appropriate wording for the model-observation comparison.

Comment#10: Line 308 / Figure 4b imprecise wording: The authors state that "Aerosol pH also varies substantially through the year and is relatively higher in late spring and early summer." However, Fig. 4b presents standardized

seasonal cycles (z-scores), not aerosol pH values. Therefore, the figure does not show the actual pH magnitude or allow the reader to assess whether pH is chemically "high" in an absolute sense. Please either revise the wording to indicate that standardized pH anomalies are higher in late spring/early summer, or provide actual monthly pH values, perhaps in the supplement.

Response: We thank the reviewer for pointing out this imprecise wording. We agree that Fig. 4(b) presents standardized seasonal cycles rather than absolute aerosol pH values. Therefore, the original wording could be misinterpreted as referring to the absolute pH magnitude. We have revised the sentence to clarify that the standardized aerosol pH anomaly, rather than the absolute aerosol pH value, is relatively higher in late spring and early summer. This revision has been incorporated in Sect. 3.2 (lines 401–403).

Lines 401–403:

The standardized sulfate and aerosol liquid water anomalies are positive in winter and become negative in summer, while the standardized aerosol pH shows positive anomalies in late spring and early summer (Fig. 4(b)).