This study established a mass-balance inversion framework that integrates CrIS-retrieved isoprene columns into the LMDZ-INCA chemistry-transport model to constrain global, monthly biogenic isoprene emission maps spanning 2013–2020. This represents a significant scientific contribution. Overall, I find the methodology generally sound, and particularly acknowledge the author' thorough justification of key assumptions and parameter selections. The manuscript is well-organized and clearly written. However, there are major concerns that must be addressed before I can recommend publication.

Response:

We sincerely thank the referee for their constructive and thoughtful comments on our manuscript. Below, we offer detailed responses addressing each point raised.

Major Concern

The validity of inversion critically depends on demonstrating whether posterior results converge when using different prior emissions inventories. The manuscript exclusively employs LMDZ-INCA land module simulations as the prior inventory. It is imperative to conduct additional inversions using an alternative emissions inventory—ideally one mechanistically distinct from LMDZ-INCA, with differing total magnitude, spatial distribution, and seasonal variation in isoprene emissions. This test would reveal whether the inferred total isoprene emissions align with those derived using LMDZ-INCA as prior, or quantify the extent to which discrepancies between the two prior inventories are reduced.

This validation using independent prior emissions is particularly crucial because the study finds that the posterior isoprene emission seasonal cycle using LMDZ−INCA as prior is entirely reversed compared to inventories such as MEGAN (Figure3). This is a new and important conclusion but has to be addressed carefully. What seasonal cycle does the prior LMDZ-INCA itself exhibit? Would using MEGAN as prior similarly yield a reversed seasonal pattern in the inversion? Without addressing these points, the scientific robustness of the posterior emissions—especially their seasonal cycle—remains limited. Given computational constraints, this sensitivity test could be restricted to a single representative year.

Response:

We have conducted sensitivity inversions using MEGAN-MACC and MEGAN-ERA5 as isoprene prior in 2019, respectively, which shows a minimal difference (<3.5%) in global annual posterior totals. Regionally, the largest difference occurs in Oceania, attributed to the substantial divergence in Oceania priors. For the seasonality, both MEGAN-MACC and MEGAN-ERA5 derived posteriors show a peak in JAS but reach minimum in DJF period, consistent with our findings. Besides, the satellite observed isoprene and HCHO column concentrations also exhibit a similar seasonal pattern as posteriors, which further demonstrates the reliability of isoprene posterior peak in JAS while minimum in DJF. Detailed discussion on sensitivity inversion on prior has been added as Section 2.6 The impact of prior choice on inferred isoprene emissions in the manuscript, and the aligned seasonality of the recent HCHO-based isoprene inversion, different prior tests, and satellite observations have been added in Lines 363-366, Lines 374-378, Figure 3, and S20.

2.6 The impact of prior choice on inferred isoprene emissions

To evaluate the sensitivity of the inversion to the choice of prior emissions, two additional sensitivity experiments are conducted using MEGAN-MACC (Sindelarova et al., 2014) and MEGAN-ERA5 (also known as CAMS-GLOB-BIOv3.1) (Sindelarova, 2021; Sindelarova et al., 2022) isoprene inventories, both of which are mechanistically distinct from the ORCHIDEE-based prior employed in the main analysis. The inversions are performed for the year 2019 following the same setup and observational constraints. Results show that the inferred global total isoprene emissions differ by less than 3.5% among the three prior configurations: deviations between the MEGAN-MACC-based inversion (500 Tg yr^{-1}) and our posterior global total (485 Tg yr^{-1}) are 3.1%, while those between the MEGAN-ERA5-based inversion (495 Tg yr^{-1}) and our posterior are 2.1%, suggesting that the inversion framework remains robust to the choice of prior in global annual totals (Fig. S15). From a regional perspective, the largest differences occur in Oceania, where posterior emissions derived from MEGAN-MACC and MEGAN-ERA5 differ from our reference posterior by 60.6% and 17.4%, respectively (Fig. S16). Although Oceania shows the largest posterior discrepancies globally, these differences are substantially smaller than those in their priors (19 Tg yr^{-1} in ORCHIDEE, 108 Tg yr^{-1} in MEGAN-MACC, and 61 Tg yr^{-1} in MEGAN-ERA5 in 2019), indicating that the inversion effectively reconciles regional inconsistencies and converges toward observational constraints even where prior emissions diverge markedly. Overall, these tests demonstrate that the optimized emissions are primarily driven by observational constraints rather than by the characteristics of the prior inventory.

Lines 363-366:

"This seasonal cycle agrees with recent HCHO-based inversion results (Müller et al., 2024) but differs markedly from that in current bottom-up inventories: MEGAN-MACC (Sindelarova et al., 2014) and MEGAN-ERA5 (also known as CAMS-GLOB-BIOv3.1) (Sindelarova, 2021; Sindelarova et al., 2022) (Figs. 3 and S20)."

Lines 374-378:

"Besides, sensitivity inversions using MEGAN-MACC and MEGAN-ERA5 as priors also reproduce a JAS maximum and DJF minimum, reversing the original prior seasonality. The posterior seasonality derived from all three priors aligns with that observed in CrIS isoprene and OMPS HCHO concentrations (Fig. S20), indicating that the retrieved temporal variability reflects the observed atmospheric signals and demonstrating the robustness of the inferred seasonal cycle."

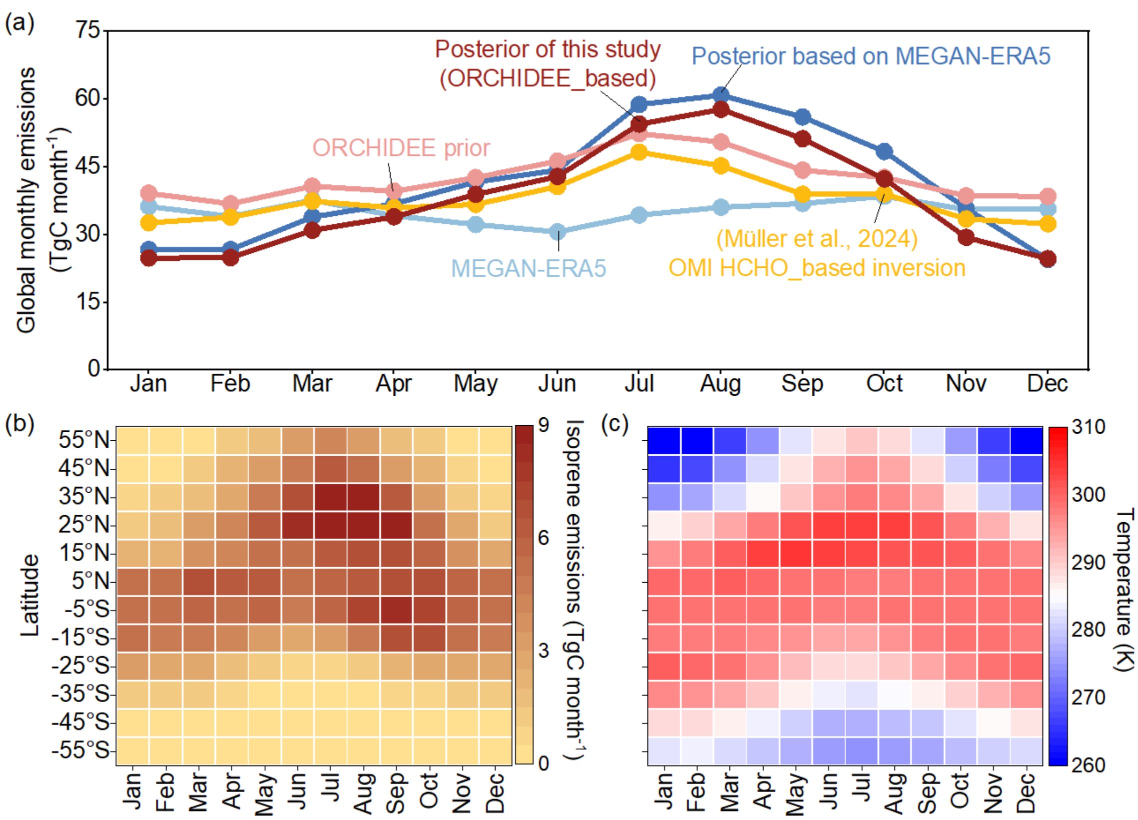

Figure 3. Monthly mean isoprene emissions from 2013 to 2020. (a) shows the global monthly pattern of ORCHIDEE prior and our posterior in this study, MEGAN-ERA5 (also known as CAMS-GLOB-BIOv3.1) inventory (Sindelarova, 2021) and posterior based on MEGAN-ERA5, as well as OMI HCHO-based isoprene inversion result (Müller et al., 2024). MEGAN-ERA5 is based on MEGAN v2.1, updated with ERA5 meteorology and CLM4 land cover (Sindelarova et al., 2022). (b)-(c) display monthly distributions of our estimated isoprene emissions (TgC) and temperature (K) by every 10° latitude band, respectively. We here only present the latitude range from 60°S to 60°N where emissions dominate (~99%). Temperature is acquired from ERA5. The monthly distributions of two MEGAN inventories (MEGAN-MACC and MEGAN-ERA5), precipitation from ERA5, and the Leaf area index (LAI) from Pu et al. (2024) are presented in Fig. S25.

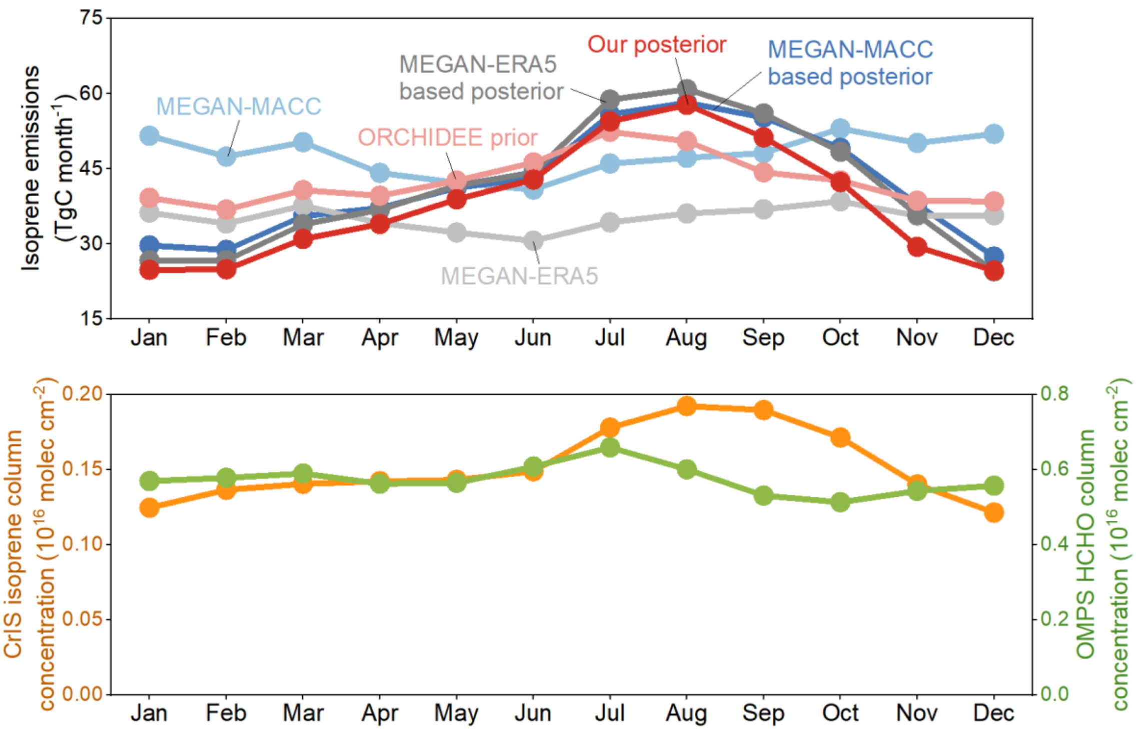

Figure S20. Monthly variation of isoprene emissions (ORCHIDEE prior and posterior, MEGAN-MACC prior and posterior, MEGAN-ERA5 and posterior) and CrIS observed isoprene column and OMPS HCHO column concentrations.

Other Comments

Sensitivity of perturbation magnitude (Lines 180–193):

The authors analyze the β+25%/β-40% value, with a global mean value of 0.9, to argue that the method is not sensitive to the perturbation magnitude and that the relationship between isoprene emissions and concentrations can be assumed linear. However, as seen in Figure S2, β+25%/β-40% ratios reveal substantial spatial heterogeneity. The manuscript should:

Present the grid-scale variance of β(+25%)/β(−40%) in the main text.

Further investigate how the nonlinearity reflected by β(+25%)/β(−40%) correlates with spatial patterns of OH, NO_2 pollution, or other meteorological drivers.

Response:

We have added discussions on the grid-scale $\beta_{+25\%}/\beta_{-40\%}$ ratio and its correlation with potential influential factors (isoprene, OH, NO_2, and radiation) in Lines 219-224 in manuscript. Overall, the statistics of grid-scale $\beta_{+25\%}/\beta_{-40\%}$ shows that the average ratio falls within 0.86-0.90 across months, suggesting a localized linear-response exist in most grids. Grids with large-deviated $\beta_{+25\%}/\beta_{-40\%}$ ratio occur in low-isoprene environments, where the linearity between isoprene emissions and concentrations weakens.

Lines 219-224:

"The grid-scale statistics of $\beta_{+25\%}/\beta_{-40\%}$ shows that the average ratio falls within 0.86-0.90, and median value within 0.85-89 each month (Fig. S7). The remaining deviations, primarily located in low-isoprene environments (Fig. S8), point to localized nonlinear responses, yet the overall relationship between isoprene emissions and its concentrations can be considered approximately linear at the grid scale within the range of perturbations and corrections of the inversions."

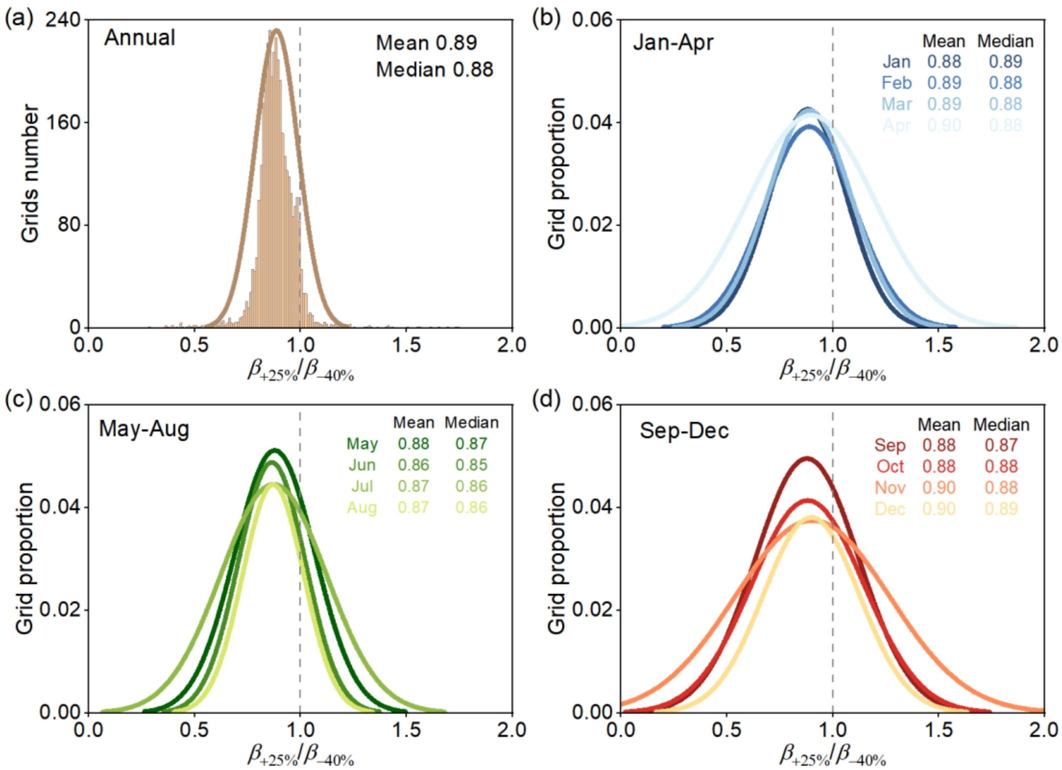

Figure S7. Statistical summary of the grid-scale β ratio ($\beta_{+25\%}/\beta_{-40\%}$) derived from the regression-based inversion in 2019. (a) shows the annual mean statistics, while (b)–(d) present the monthly distributions for Jan–Apr, May–Aug, and Sep–Dec, respectively. The tables inside (b)-(d) correspond to mean and median value of $\beta_{+25\%}/\beta_{-40\%}$ for each month.

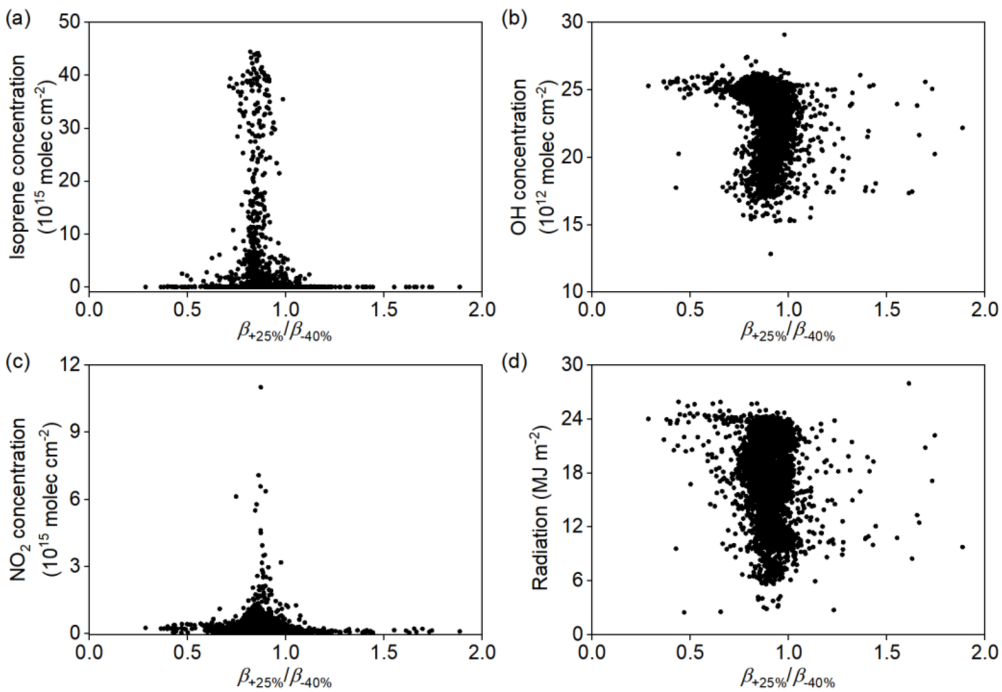

Figure S8. Correlation between annual mean grid-scale β ratio ($\beta_{+25\%}/\beta_{-40\%}$) and (a) simulated isoprene column concentrations, (b) simulated OH column concentrations, (c) simulated NO_2 column concentrations, and (d) radiation from ERA5.

Model chemistry representation:

The chemical mechanisms governing VOCs—especially isoprene oxidation pathways—vary significantly across models and directly influence simulated isoprene concentrations and their response to emissions. The model description section should explicitly detail the VOCs chemical mechanisms included in LMDZ-INCA.

The limitations section should explicitly address the potential impacts of using a single model framework on the results.

Response:

We have added a detailed description of the VOC chemical mechanisms, particularly those directly related to isoprene and HCHO in Lines 153-160 in the manuscript. In brief, LMDZ-INCA includes a comprehensive reactive VOC chemistry scheme with 14 isoprene reactions and 80 HCHO reactions, using up-to-date reaction rates.

In addition, we have included a discussion of the potential impacts of using a single model framework on the results in Lines 560–564 in the manuscript.

Lines 153-160 in Section 2.2:

"LMDZ-INCA contains a state-of-the-art CH_4–NO_x–CO–NMHC–O_3 tropospheric photochemistry scheme with a total of 174 tracers, including the chemical degradation scheme of 10 non-methane hydrocarbons (NMHCs): C_2H_6, C_3H_8, C_2H_4, C_3H_6, C_2H_2, a lumped C>4 alkane, a lumped C>4 alkene, a lumped aromatic, isoprene and α-pinene. The mechanism comprises 398 homogeneous, 84 photolytic, and 33 heterogeneous reactions, and is continuously updated to integrate newly identified chemical processes and reaction pathways, thereby improving the representation of atmospheric composition and oxidation capacity (Hauglustaine et al., 2004; Folberth et al., 2006;

Pletzer et al., 2022; Sand et al., 2023; Terrenoire et al., 2022; Novelli et al., 2020; Wennberg et al., 2018). Reactions directly related to isoprene and HCHO are listed in Tables S1-S2."

Lines 560-564 in Section 4:

"Moreover, relying on a single model framework introduces structural uncertainty that cannot be fully quantified here. Model-specific formulations of boundary layer mixing, photochemistry, and deposition can affect the simulated column–emission relationships. These issues could be mitigated through retrievals that include vertical sensitivity information, continued model development, and cross-model ensemble evaluations to better represent atmospheric isoprene processes."