Reply to the comments of Reviewer#1

We sincerely thank the reviewer for the positive evaluation of our manuscript and the constructive comments. The reviewer's comments are in black, our responses in blue.

This manuscript presents an important study that uses TROPOMI HCHO and CHOCHO data to quantify global VOC emissions from biogenic, pyrogenic and anthropogenic sources. The results indicate a large proportion of unidentified VOCs over the tropics, which is an important finding that will motivate a lot of future studies. The manuscript is very well done, and the results are interesting and convincing. I only have a few minor comments.

1. The inversion framework relies on TROPOMI HCHO and CHOCHO retrievals, but these products are themselves subject to uncertainties. As the authors show, CTMs substantially underestimate CHOCHO, yet the TROPOMI retrievals use CTM a priori profiles to compute air mass factors. It is unclear how uncertainties in these a priori profiles propagate into the inversion. It would be informative to assess how the retrieved HCHO and CHOCHO column densities would change if air mass factors were recalculated using CTM fields updated with the optimized VOC emissions.

The reviewer is right to point out that the vertical profiles assumed in TROPOMI HCHO and CHOCHO retrievals are a large source of uncertainty. However, the error caused by vertical profile shape errors (the so-called smoothing error) is taken care of through the application of the satellite averaging kernels to the model profiles (see for example, Lerot et al., 2021). This process removes errors due to vertical profile inconsistencies in the comparison of satellite columns with "smoothed" model columns. We changed the last sentence of Sect. 3.1 as follows: "These kernels are applied to the modeled vertical profiles to account for the instrument's altitude-dependent sensitivity and remove errors due to vertical profile inconsistencies in the comparison of satellite columns with the model (Oomen et al., 2024)."

2. The inversion framework appears to attribute model-satellite discrepancies in HCHO and CHOCHO solely to emission errors. However, both species are secondary products, and their yields depend on chemical mechanisms and NOx levels. It is therefore not clear how much of the discrepancy arises from uncertainties in HCHO and CHOCHO production pathways rather than emission errors. It would be helpful for the authors to comment on how the inversion accounts for, or is affected by, these chemical uncertainties.

Agreed. We added the following text discussing areas of uncertainty in the derivation of top-down emissions (Sect. 3.2). "The inverted emissions have uncertainties due to several factors affecting the HCHO columns, besides the magnitude of the emissions, such as the background HCHO levels due to methane oxidation, incomplete or incorrect information regarding VOC speciation in emission inventories, the VOC oxidation

mechanisms, the deposition of oxidation intermediates, the transport processes influencing the vertical profile of chemical compounds, and the NOx concentrations, known to influence the OH levels as well as the yields of HCHO and CHOCHO from key VOCs including isoprene."

3. Because satellite retrievals are available only under clear-sky conditions, sampling biases in HCHO and CHOCHO are likely. The authors note that CHOCHO sinks may differ under cloudy conditions, but it is unclear how such sampling biases are treated within the inversion framework and how they might influence the emission estimates. A brief discussion of this issue would strengthen the manuscript.

Indeed, the abundances of both HCHO and CHOCHO are affected by cloudiness. As explained in the manuscript, only observations with a cloud fraction less than 20% for CHOCHO and less than 40% for HCHO were retained for processing. However, we compare the satellite monthly averages to corresponding MAGRITTE monthly averages calculated from daily values accounting for the number of measurements (and averaging kernels) for each day. This has been now clarified in the manuscript. The last paragraph of Sect. 3.1 now reads "(...) the modelled monthly averaged columns are based on daily values at the satellite overpass time (~13:30 local time), while accounting for the number of observations and averaging kernels provided with the TROPOMI retrievals. These kernels..."

4. TROPOMI overpasses occur around 2 PM local time, when biogenic VOC emissions typically peak. It is not clear whether the inferred biogenic VOC emissions represent instantaneous emissions at overpass time or whether they are scaled to a daily mean. Clarification on this point would help interpret the emission magnitudes.

No, the top-down biogenic VOC emissions do not represent instantaneous emissions at the overpass time. The temporal variability of emissions is kept identical in the optimised and prior models (see Equation 4 in the manuscript).

5. Figure 4: Add figure legend.

Done as requested.

References

Lerot, C. et al., Glyoxal tropospheric column retrievals from TROPOMI - multi-satellite intercomparison and ground-based validation, Atmos. Meas. Tech., 14, 7775-7807, https://doi.org/10.5194/amt-14-7775-2021, 2021.

Oomen, G.-M. et al., Weekly-derived top-down volatile-organic-compound fluxes over Europe from TROPOMI HCHO data from 2018 to 2021, Atmos. Chem. Phys., 24, 449-474, https://doi.org/10.5194/acp-24-449-2024, 2024.