Reply to Reviewer#1

We thank the referee for the constructive comments on our manuscript. We have made changes to the manuscript based on the recommendations of both referees. The responses to the referee's comments are provided below.

The paper is extremely well written and thorough and addresses a topic of scientific importance. I appreciate the authors' careful analysis and forthright discussion of uncertainties. I strongly recommend publication and have only minor comments and suggestions.

We thank very much the reviewer for the appreciation of our manuscript.

1) The authors discuss inconsistency and uncertainty in the aircraft HCHO measurements used for satellite evaluation, and come up with a defensible approach for dealing with this. It would be helpful somewhere later on to include a brief discussion of the degree to which these uncertainties could impact (or not) their conclusions about OMI and the resulting emissions.

Thanks for the very good point. The aircraft-based validation relies on two instrumental techniques: DFGAS and CAMS. The intercomparison of DFGAS and CAMS suggests a potential overestimation of DFGAS by up to 11% and a potential bias of the adjusted CAMS measurements (see Sect. 2.3) of up to ca. ±6.5%. Assuming that the optimized model columns are proportional to the observed mixing ratios, we evaluated the impact of those potential biases on the regression statistics. The regression slope ranges between 0.6 and 0.7, while the intercept lies in the range $(2.6\text{-}3.3)\times10^{15}$ molec. cm^{-2}. We added this discussion to Sect. 4.2.

2) There is a sensitivity inversion (OPT3) included to assess impacts of the prior error assumptions on the inversion results, which I agree is an important test to include. However, the results and conclusions from this test do not seem to be discussed anywhere.

The results of the comparison with aircraft data with OPT3 were summarized in Table S4 in the supplement, but we agree with the referee that the OPT3 results should have been discussed in the main manuscript. The point is, that the OPT3 run are not very different from those of the OPT2 run. For example, the optimized isoprene emissions of OPT3 are only slightly higher (by ~3%) than in OPT2. The statistics of the comparisons with aircraft data (in 2012 and 2013) are also quite similar. The Sect. 5.1 and 5.3 were updated to convey this information.

3) 281-290, I am confused here b/c the text first says that E is diagonal but then later the text describes a decorrelation length scale which seems to imply the presence of off-diagonal elements. Please clarify.

Thank you for the useful comment. There was a typo on line 287 where the matrix **E** should have been the matrix **B**. The matrix **E** is diagonal, whereas the matrix **B** is not.

4) 543-544, "and comparatively slower increases (< 1%yr^{-1}) in biogenic VOC emissions over many areas due to global warming (e.g. over Amazonia, Southern Africa and Australia) (Fig. 14)."

The phrasing here implies that warming is unequivocally driving a statistically significant, detectable increase in emissions over these regions. Looking at the figure, however, the trend for Amazonia is 0.0%/y and I have a hard time believing that the trends for Southern Africa and Australia are statistically distinguishable from zero. I wonder if what the authors mean to say is that any warming driven isoprene increase is small or undetectable over this period; that is the conclusion I draw from Fig. 14.

We did not mean that warming induces detectable emission increases over all those regions; instead, we meant that warming might cause emission increases that do not exceed 1% yr^{-1}. The uncertainties on the trends are now indicated on Fig. 12 and 14. The increasing emission trend (Fig. 14) is not significant over Amazonia and Southern Africa and it is barely significant over Australia. We therefore changed as follows the sentence pointed out by the referee: "and comparatively slower changes (< 1% yr^{-1}) in biogenic VOC emissions over many areas (e.g. over Amazonia, Southern Africa and Australia) (Fig. 14)."

5) Sections 3.3-3.4, I get the impression that emissions are being optimized on a monthly basis but I don't believe this is explicitly stated. Please specify.

The emissions are indeed optimized on a monthly basis. This is now made clear in Sect. 3.3.

6) In lines 515-520 the authors discuss the difficulty in separating biogenic versus pyrogenic emissions in some regions. I feel that the paper would benefit from a more general discussion of this issue, perhaps earlier on when introducing the inversion methodology. That is, we are solving for 3 separate variables (anthropogenic, biogenic, and pyrogenic VOC emissions) for every grid cell based on a single observed variable (HCHO). To what degree are these terms actually resolved through the inversion, and to what degree does that separation merely rely on the prior and/or only work where (again according to the prior) one source is dominant?

Fair point. We have added the following paragraph in Section 3.4:

The determination of VOC emissions from satellite HCHO data has several limitations. Although the fluxes from three emission categories are inverted simultaneously through the minimization of the cost function (Eq. 1), the distinction between these categories is uncertain, in particular at places and times where more than one category is dominant. The optimization realizes the separation largely based on the a priori magnitude and spatio-temporal patterns of the emissions, through the correlation between a priori errors on the emission parameters. Therefore, errors in the a priori emission distributions might cause errors in the attribution of emissions between different categories. Fortunately, a single emission category is very often dominant over continental areas, e.g. anthropogenic emissions are strongly dominant over northeastern China and biogenic emissions are dominant over Eastern U.S. and most tropical forests. However, biomass burning is a highly episodic source which generally coincides with biogenic source areas, resulting in uncertain top-down emissions for both biogenic and pyrogenic sources. The same is also true in areas (e.g. India) where both anthropogenic and biogenic emissions are significant. In those regions, the total top-down VOC emissions are much better resolved than individual categories.

7) The VOC source optimization is by nature indirect and based on the resulting HCHO abundance. The authors should include some assessment or discussion of the extent to which the VOC emission magnitude updates could in fact be compensating for other factors or model errors that affect HCHO (for example, incorrect VOC speciation, uncertainty in the diel cycle of VOC emissions, errors in NOx emissions or in the HCHO lifetime, uncertainties in the chemistry leading to HCHO, etc.).

We added the following paragraph in Section 3.4:

In addition, the top-down VOC emissions have uncertainties related to the multiple factors that might affect the abundance of HCHO, besides the magnitude of the emissions. This includes, for example, the background HCHO abundance, largely determined by OH radical levels, the incomplete and oversimplified speciation of VOCs in large-scale models, the VOC chemical oxidation mechanisms, the deposition of VOC oxidation intermediates, the diurnal cycle of emissions, especially for biomass *burning, the vertical transport processes control the vertical profile of HCHO, and the NO_x levels, which influence the yields of HCHO from many important VOCs. Although a few of those uncertainties were partially addressed in previous studies (e.g. Oomen et al., 2023), an exhaustive quantitative study of those uncertainties would be a daunting task and is beyond the scope of the present study.*