Reply to Reviewer#2

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 manuscript title is misleading, as "bias characterization" suggests that individual causes for biases in the OMI HCHO product will be diagnosed and quantified, but it is really a "bias correction" informed by independent observations. The manuscript and Sections 4.1 and 4.2 should be renamed to reflect this.

Thank you for the suggestion. The manuscript has been amended accordingly.

Section 2 should include details of the CrIS isoprene and Bauwens et al. (2016) datasets that the authors compare to in Section 5.3.

We added a new subsection on CrIS isoprene in Section 2. We do not think necessary to do the same for the Bauwens et al. dataset since the nature and main results of that study are already sufficiently described in the manuscript.

Is there any dependence of the sampling coincidence (50 km) on the regression statistics, as was found by Pinardi et al. (2020) for NO_2?

Thanks for this comment. Note that, compared to NO_2, HCHO columns show generally much less spatial heterogeneity around cities due to anthropogenic emissions. We performed tests with different collocation criteria and added the following text in Sect. 4.1:

We tested alternative choices for the collocation distance: a higher value (100 km instead of 50 km) degrades the correlation and yields a slightly lower slope (0.63) than our reference regression. Lower distances (e.g. 20 km) lead to an excessively low number of OMI pixels to be averaged and therefore to poor correlation with FTIR.

The updated / optimized model is evaluated against the same observations that are used to bias correct the OMI HCHO product used to derive emissions. The limitations of this evaluation should be acknowledged, given that it isn't truly independent.

This comment is not correct. The updated model is evaluated against many more aircraft campaign data than the observations used to bias correct the OMI product. See Figure 1 and Table 1.

The description of the OMI HCHO product in Section 2.1 is challenging to follow without prior detailed knowledge of the product. Non-European readers won't necessarily know what "EU-FP7" is. It's not apparent what the "cloud correction" is and what implication this has on the data. Defining what this is might help clarify what is meant by "in lieu of the cloud-corrected AMFs". Is the cloud fraction geometric or effective or something else? It's not clear what's being compensated for with model data over the Equatorial Pacific. Is the background removed in the background correction and the model data used to add back a background? Is the Equatorial Pacific the "reference region"?

The QA4ECV EU-FP7 project is now described as "QA4ECV project of the 7th Framework Programme of the European Union (EU-FP7)". The description of the algorithm has been updated as follows:

(…) *The standard AMF calculations uses the effective cloud fraction and cloud top pressure from the Fresco v7 cloud product (Veefkind et al., 2016), treating clouds as Lambertian reflectors and applying the independent pixel approximation (Martin et al., 2002; Boersma et al., 2004). However, in this work,* *the cloud correction is switched off, except for a strict filtering (effective cloud fraction > 0.2). Clear-sky air mass factors (AMF) are used in lieu of the cloud-corrected AMFs. This choice ensures an optimal consistency with the TROPOMI HCHO dataset (De Smedt et al., 2021). Indeed, the TROPOMI HCHO retrieval is inherited from the QA4ECV algorithm with the aim to generate a consistent time series of early afternoon observations. Finally, to correct for any global offset and for stripes arising between the rows, a background correction is performed on daily basis using the HCHO slant columns over the Pacific Ocean. The TM5 HCHO model columns derived in the same region are finally added to compensate for the background HCHO concentrations.* (…)

It's not stated what fit is used to quantify trends in Section 5.4 and which of the reported trend values in Figures 12 and 14 are statistically significant.

This is a good point. We now make clear that we use a least squares linear regression method. The 1-σ errors of the regression slopes are now given in Figures 12 and 14.

The discussion of emissions trends in Section 5.4 has quite limited discussion of trends in biomass burning and the influence this has on trends in HCHO. For Africa, for example, Andela and van der Werf (2014) reported significant trends in biomass burning activity in Africa and Hickman et al. (2021) reported decline in NO_2 abundances in North Equatorial Africa.

Thank you for this interesting comment. Note that the a priori inventory of biomass burning emissions (GFED4s) takes into account the reported trends in biomass burning activity, e.g. the emission decline in the savanna region of northern Africa (Hickman et al., 2021). The patterns of changes over Africa are complex (see Fig. 13) and a dedicated investigation would be needed to elucidate their causes. Nevertheless, the manuscript now briefly discusses the role of biomass burning as an important driver for long-term trends in HCHO, in particular over Africa.

The Conclusion reads like a mix of concluding statements and as a discussion, as indicated by inclusion of citations.

The referee is correct. We simplified the Conclusion section and removed most citations. The bits of discussion that are now removed from the Conclusions are part of the general text.

Specific Comments:

L. 13: Consider a more informative summary of the comparison to CrIS isoprene than "striking similarities and differences".

For the sake of brevity, we prefer to keep the sentence as is. The details are provided in Sect. 5.3.

L. 50: Should "higher-quality" be "higher spatial resolution"? If not, then perhaps indicate what it is about OMI that makes it higher quality.

Fair point. We changed to "higher-resolution sounders".

L. 180-181: Justification for not using INTEX-B seems to contradict mention in Section 2.1 of the importance of the Pacific Ocean where OMI HCHO data are used to perform background correction.

Our global inversion study addresses continental VOC emissions, and, as explained in the text, HCHO levels over the Pacific are insensitive to emissions over land. Therefore there would be no point in validating the inversion results using such data.

Figure 2: Intercept value and error estimate should be written to account for the scale of the axis (10^{16}).

Although strictly speaking, this comment is of course correct, we keep the figure as is, since it is obvious for the reader that the intercept should not be multiplied by the scale factor of 10^{16}.

Figure 12 caption: Unnecessary to include units in caption, as these are given in the figure.

Corrected as proposed.

L. 537-538: Clarify why a change in the version of the model would cause a change in data for 2017 and 2018? Is the updated TM5 model only applied to those years, rather than the full record being reprocessed to use the updated TM5 model?

The TM5 model changes were not applied to the full record. There are two different versions of TM5 used for the QA4ECV OMI HCHO product. The TM5 version which was run during the QA4ECV project, and the TROPOMI version (TM5-MP) from 2018 onwards. There is no temporal overlap between the two versions. TM5-MP has not been run for the years before 2018. Reasons for change in HCHO profiles are not clear (possibly the time resolution, the definition of the vertical layers, or an update in the convection scheme). They are apparent only in Tropical regions.

L. 543-544: The statement starting "comparatively slower..." is confusing, as isoprene emissions have an exponential dependence on temperature, so should have a large response to warming. Is this statement meant to convey something else?

No, the statement is meant to convey that over the large regions shown on Fig. 12, global warming does not induce fast HCHO trends (>1%/yr). Even with an exponential temperature-dependence of biogenic emissions, a fast warming trend is require to induce a large trend in HCHO columns. Although such warming trends have been observed at many locations (see Stavrakou et al., 2018), spatial averaging over large regions leads to moderate trends, as observed on Fig. 12.

L. 550: Isn't 2012 too early for emissions to level off in India as a result of policies? Emissions controls were only implemented in earnest relatively recently, starting with power plants in 2015 and vehicles in 2018 (see for example Vohra et al., 2021)?

Thank you for this interesting comment. We changed the manuscript and included the following text:

Over India, the apparent stabilization of top-down emissions after 2012 seems contradicted by reports that regulatory measures were not effective in India until the last years (after 2018) (Vohra et al., 2021). More work will be needed to examine the patterns of HCHO changes and the possible causes of the discrepancy.

L. 564: Replace "is believed to be" with "one of"

We changed to "is the dominant source". Isoprene oxidation is not just one of the sources, it is the dominant source in this region.

L. 599: "poor" correlation would be R < 0.4.

It's all relative. But we changed to "low" instead of "poor".

L. 620-621: What's the utility of the sentence starting "This result demonstrates ...". It's an obvious statement. Is this stated because the product used by Bauwens et al. (2016) didn't undergo any validation?

Yes indeed, as implied in the manuscript (e.g. the Introduction), previous inverse modelling studies did not apply bias correction to the HCHO datasets. It might be obvious that bias-corrected is better than not, but it is not obvious that it has such large consequences on top-down emissions.

References:

Andela and van der Werf, 2014, http://www.nature.com/doifinder/10.1038/nclimate2313

Hickman et al., 2021, https://doi.org/10.1073/pnas.2002579118

Pinardi et al., 2020, https://doi.org/10.5194/amt-13-6141-2020

Vohra et al., 2021, https://doi.org/10.5194/acp-21-6275-2021

Boersma et al., 2004, https://doi.org/10.1029/2003JD003962

Martin et al., 2002, https://doi.org/10.1029/2001JD001027

Stavrakou et al., 2018, https://doi.org/10.1029/2018GL078676