Response to reviewer 4

The review comments and our responses are coloured in blue and black texts, respectively. The changes in the manuscript corresponding to the comments are highlighted in yellow. The line and figure numbers refer to the revised manuscript.

This paper presents the evaluation of HCHO columns from the CHASER model against the TROPOMI, OMI, ground-based MAX-DOAS observations, and the CHASER HCHO vertical profiles against the Atom-4 flight dataset. The authors compare the modelled regional HCHO columns with the TROPOMI and the OMI HCHO columns and analysed the model-observation differences comprehensively. The authors also compare the modelled HCHO columns with the MAX-DOAS columns at three locations in Thailand and Japan respectively. The modelled HCHO profile and the profile from the Atom-4 flights are compared for Amazonia and for the Remote Pacific region, respectively. The authors have also performed sensitivity simulations to assess the impact of anthropogenic, biogenic, and biomass burning VOC emissions, as well as NOx emissions on modelled HCHO. However, I find that one limitation is the lack of discussions on the important role of chemical mechanisms in simulating HCHO in the models, despite that the authors did mention this in the conclusion. There are some previous studies that the authors could cite which addressed inter-model di□refences in modelled HCHO (see below suggestions) Overall, the analysis is thorough and robust. The paper is generally well-written, and the materials are well organised, and is within the scope of GMD. However, the presentation of the paper can be improved. I encourage the authors to make a thoroughly revision of the manuscript.
Below are two relevant papers on model di□erences in modelling HCHO (and CO):

Anderson, D. C., Nicely, J.M., Wolfe, G. M., Hanisco, T. F., Salawitch, R. J., Canty, T. P., … Zeng, G. (2017). Formaldehyde in the tropical western Pacific: Chemical sources and sinks, convective transport, and representation in CAM-Chem and the CCMI models. Journal of Geophysical Research: Atmospheres, 122. https://doi.org/10.1002/2016JD026121 (Figure 13)
Zeng, G., Williams, J. E., Fisher, J. A., Emmons, L. K., Jones, N. B., Morgenstern, O., Robinson, J., Smale, D., Paton-Walsh, C., and Gri□ith, D. W. T.: Multi-model simulation of CO and HCHO in the Southern Hemisphere: comparison with observations and impact of biogenic emissions, Atmos. Chem. Phys., 15, 7217–7245, https://doi.org/10.5194/acp-15-7217-2015, 2015. (Figure 15 and Table 4)

Response: We thank the reviewer for the insightful comments which has helped improving the quality of the manuscript. We have included additional discussion on the role of chemical mechanism on the simulated HCHO.

3.9 Uncertainties in the chemical mechanism

Uncertainties in the chemical mechanisms affect the HCHO simulations. Representation of isoprene chemistry can vary among the gas-phase chemistry mechanisms used in the CTMS. The most commonly used isoprene schemes underestimates observed HCHO by at least 15% (Marvin et al., 2017). Such underestimations are also strongly linked with the errors in the NO_x emission inventories (Anderson et al., 2017). In addition, potential errors in the acetaldehyde emission and chemistry can also lead to underestimated HCHO vmr up to 75 pptv in the lower troposphere (Anderson et al., 2017).

My specific comments are listed below.

Abstract
It feels that the abstract is overly concise and does not reflect fully what are presented in the paper.

Response: We have revised the abstract

L19-20: Please state which comparison this is for, i.e., TROPOMI.

Response : We have revised accordingly

L30: It is the comparison between the CHASER and MAX-DOAS HCHO columns, not mixing ratio. Please also state the disagreement, i.e., CHASER underestimates the HCHO peak in comparison with the MAX-DOAS data at all three locations. You speculate that the model data averaged over a large area might not be able to capture the observed peak at these locations. A mention of this would be useful in the abstract.

Response: We have revised the whole abstract following the reviewer's earlier comment. Such information is included in the revised abstract.

Introduction
L82: How do you evaluate OH?
Response: The OH was validated against Atom observation by Sekiya et al., (2018)

Model, observations, and methods
L93: Is there a reference for this?
Response; Reference has been included

L99-101: A list of the reactions in a table (in supplementary) could be considered if they have not been published before.

Response: The reactions has been published by Sudo et al., 2002 (Reference provided in the reference section of the manuscript)

L110-123: It will be helpful to tabulate these emissions.

Response: We agree with the reviewer's perspective. However, to reduce the number of tables, we decided to describe the emission inventories.

L117: Do you calculate lightning NOx emissions online or prescribe them?

Response: The lightning NO_x emissions were calculated online using the widely-used cloud-top height (CTH) scheme (Price and Rind, 1992).

L124-125: Are there OH observations from OMI and Atom? Please provide details.

Response: OH observations are only available from the Atom campaign, We have revised the sentences as follows:

L146-147 Sekiya et al. (2018) comprehensively assessed CHASER simulated NO_2 abundances using OMI observations. CHASER well reproduced the ATom-observed OH spatiotemporal variation (Sekiya et al., 2018).

Table 1: ANI and OLNE appear first time in Table 1. Please define these simulations in the text.

Response: Table 1 has been revised. All the abbreviations are defined.

L137: What are the TROPOMI grids?

Response: We have removed the sentence

L139: Do you mean that the TROPOMI data are interpolated onto the CHASER horizontal grid?

Response: TROPOMI data has been interpolated onto the CHASER horizontal grid. We have included additional discussion in the revised manuscript as follows:

L196-205 TROPOMI observations are averaged spatially and temporally to the CHASER grid (T42) daily, leading to horizontal representativeness errors. However, the random horizontal representativeness errors are in the order of 5-10%, which is lower than the individual retrieval error of the satellite observations (Boersma et al., 2015). If the model horizontal resolution is increased by 50% (i.e., simulated at a horizontal resolution of 1.4º × 1.4º), the change in HCHO abundances is less than 6% (Fig S1 and Table S1 in supplementary information). The vertical sensitivity of the satellite retrievals is the most relevant source of representativeness error (Boersma et al., 2015). The current study utilizes the TROPOMI AK information to minimize the representativeness error. Therefore, the horizontal representative error will likely affect the results less than other error sources, such as uncertainties in satellite retrieval, emission inventories, and model chemical mechanisms.

L174: Should be “2.3 OMI”

Response : We have revised accordingly

Results and discussion

L231: This section is essentially the comparison of CHASER HCHO with TROPOMI. Maybe “TROPOMI” should be reflected in the section title?

Response: We have revised the section title to : Comparison of CHASER HCHO with TROPOMI observations

L235-239: I am not sure how meaningful these statistics are in terms of the global means as the global HCHO distribution is so inhomogeneous.

Response: Similar statistics were reported in earlier studies on CHASER simulations (Hoque et al., 2022; Sekiya et al., 2018; Ha et al., 2022, He et al., 2023)

L243-245: Would it more suitable to note this in the MAX-DOAS comparison section?

Response: We agree with the reviewer’s perspective. However, we think it is also important to mention here to support the uncertainties in the observations and simulations.

Table 2: These numbers don't have to be in a table. You could include them in the Figure 1 caption. Is the correlation coefficient spatial or temporal?

Response: We agree with the reviewer’s perspective. However, we think the table is important to provide vital information on the global statistics for the readers, without checking the details. We also believe it is important for citation of our work too.
These are spatial correlation and has been described in the caption of table 2.

Figure2: The panels can be larger. Mark the position of the MBE numbers in the panels consistently. Add identifiers to the sub-figure, e.g., (a), (b), … for each region. Then refer to Figure2(a), Figure 2(b), etc., when you discuss them in the following subsections.

Response: Figure 2 has been revised accordingly. We have also ensured the consistency of the figure numbers in the revised manuscript.

L293-: Please refer to the figure(s) and table(s) that your discussions are based on at the beginning of each subsection. Same as the following subsections of (b), (c), etc.

Response: Following the reviewer’s comment we have ensured mentioning the figure numbers in the text.

L300: Do you mean direct HCHO emissions or indirect (degradation of VOCs) HCHO emissions? Please clarify.

Response: We have added the following text to address this comment
L335-338 NMVOC emissions from these sources (i.e., vehicular exhaust, solvent usage, and transport) are considered in the HTAPv2.2 inventory (Crippa et al., 2023). Although CHASER considered HCHO production from the degradation of anthropogenic VOCs, it is likely underestimated, resulting in a lower simulated winter-time HCHO column in this region.

L319: Can you speculate what drives these model-satellite discrepancies in the Europe and W-US in summer and autumn?

Response: We have added the following text in response to this comment

L358-365 In both regions (i.e., Europe and W-USA), the biogenic and anthropogenic contribution to the total HCHO level is equivalent during summer. In autumn, the anthropogenic emission contributions are higher. (Section 3.8). This manifests a potential model underestimation of biogenic HCHO levels in these regions, linked to the uncertainties in the biogenic emission inventory and isoprene mechanism

L328-329: Please refer to the figure you are referring to. Please also note that the C-Africa o□-peak HCHO is overestimated by CHASER compared to TROPOMI (Figures 1 and 2).

Response: We have ensured consistency in the figure numbers throughout the manuscript. We have also revised the discussion on C-Africa.

L333: Figure S4: There is no black curve in Figure S4. Please revise this figure or the caption.

Response: Figure S4 has been revised which is FigS5 in the revised manuscript

L338: Please mention the figure you refer to for these discussions.

Response: The figure number has been included.

L339: Missing ")" in "(De Smedt et al., 2008". Again, please mention the figure here you are referring to over the next few lines.

Response: The figure number has been included and the consistency has been ensured throughout the revised manuscript.

L340-341: "The lower CHASER columns in Southern Africa are likely attributable to underestimated pyrogenic emissions." - Can you confirm this from the following sensitivity simulations?

Response: We have revised the sentence as follows:

L382-385Over South Africa (S-Africa; Fig.2(g)), elevated TROPOMI HCHO columns are consistent with GOME-2 and SCIAMACHY observations (De Smedt et al., 2008). The observed peaks in HCHO columns and FRP values (Fig.S5) are consistent and thus can be attributed to biomass burning. Pyrogenic emissions contribute ~36% to the high HCHO columns in this region (section 3.8).

L355-356: Why particularly mention the biomass burning in N Africa here?

Response: N-Africa is mentioned to compare between two biomass-prone regions. We have revised the sentence.

L358-359: Could the chemical mechanism in the model be at play?

Response: We have revised the sentence as follows:
L41-403 The model overestimates the HCHO columns in S-America, similarly to C-Africa and N-Africa, probably because of the uncertainties in biogenic emission inventories and the isoprene oxidation scheme.

L370: Refer to relevant figure(s) and table(s) earlier in your discussion of the results. Do you have an estimate how the biogenic and biomass burning emissions in India compared to other regions? Are there any specific meteorological conditions in this region that lead to low HCHO and the lack of seasonality?

Response: We have revised this section as follows to address the comment.

L406-419 CHASER well reproduced the observed HCHO spatial distribution in India (Fig.2 (i); r =0.84), with MBE and RMSE of -1.20 × 10^{15} and 1.775 × 10^{15} molecules cm^{-2}. However, the temporal correlation (R=0.18) between the datasets is low. The observed seasonal modulation of ~30% manifests a less-prominent seasonality in HCHO abundances in India. The correlation between temperature variations and isoprene emissions in India is inhomogeneous (Starvakou et al., 2014). India has a diverse landscape, including major forests over the east, northeast, and southwest regions and deserts in northwestern India (Surl et al., 2018). The Indo-Gangetic Plain (IGP) stretches from Eastern Pakistan to Bangladesh and is a major agricultural region in India (Kuttippurath et al., 2022). Thus, averaging the HCHO columns over a diverse landscape can lead to a less prominent seasonality. Moreover, biomass burning compromises 23% of India's total NMVOC (13 Tg/yr) emissions (Stewart et al., 2021). Sensitivity analysis (section 3.8) estimates show biomass burning contribution

to the HCHO levels in India is ~2%., manifesting that the modeled biomass burning emissions for India are underestimated. Considering the diverse Indian landscape, the model satellite comparison over three regions in India (IGP, east India, and South India) is shown in Fig.2 (j-l).

L387: The figure number is missing here.

Response : We have included the figure number.

L422-423: I am not sure what you try to convey here?

Response: The sentence has been removed for clarity.

Figure 5: Could you increase the size of the panels in this figure?

Response: We have revised all the figures

L552: It is important to summarise the NOx emissions in the two inventories you used. What are the di□erences in NOx emissions between these two inventories? It will help to understand the impact of NOx on HCHO and OH.

Response: The differences are mentioned in text. The NOx emissions are shown in Fig.S8. The discussion has been revised.

L605 – 628 The differences between the two NO_x inventories are – (1) HTAP-v3 inventory considers the changes in NO_x emissions from 2000 to 2018, whereas the temporal coverage of HTAP_v2.2 is 2008 – 2010, and (2) Emissions in HTAP-v3 have a higher sectoral disaggregation (Crippa et al., 2023). The comparison-related statistics are given in Table S3. NO_x emissions from both inventories are shown in Fig. S8

On a global scale, HCHO column estimates are mostly unaffected by the changes in the NO_x emission inventories, manifested by the MBE values (Table 6). However, RMSE is 8% lower in the case of standard simulation. OLNE estimates in the higher latitude (>=50^{o}N) are 5% lower than the standard simulations. Such differences do not affect the model–satellite agreement in these regions.

The standard HCHO columns in India, China, and Southeast Asia are approximately 10–20% lower than the OLNE estimates (Fig.5(c)). In fact, those differences are consistent with changes in the regional OH estimates (Fig.6(d)). This finding implies that the changes in the NO_x emissions estimates have affected the OH and HCHO abundances in these regions. Satellite data assimilation results reported by Miyazaki et al. (2017, 2020) indicate that NO_x emissions in India have increased by 30% since 2008, whereas NO_x emissions in China have declined since 2011 (Liu et al., 2016). Over E-China (Fig. 5(a &b)), the standard simulations

reduce the absolute annual mean difference between OLNE and TROPOMI of 3×10^{15} molecules cm^{-2} to 1×10^{15} molecules cm^{-2}, which is consistent with the lower NO_x emissions in this region in the updated inventory (Fig . S8). Over India and SE-Asia, the standard OH concentrations are ~40% lower (Fig.5(d)) than the OLNE estimates, resulting in lower HCHO columns. The lower standard HCHO columns can be linked to the increasing NO_x emissions in these regions (Fig.S8); however, the magnitude of the change in the NO_x emissions for these regions in the updated inventory is likely overestimated.

In E-USA and W-USA (Table S3), the standard simulation reduces the MBE by 26% and 12%, respectively. The reduction in MBE and RMSE values in Africa and South America is less than 10%. Therefore, NO_x emission uncertainties mainly affect the HCHO simulations in India and SE Asia.

L553: you need to define the OLNE simulations before referring to it.

Response: The abbreviations of the simulations have been defined in Table 1 in the revised manuscript.

L565-566: Which figure that you are referring to here?

Response : We have included the figure number.

Figure 6: It will be helpful to understand this figure if the di□erences in NOx emissions are displayed or mentioned.

Response : The differences are mentioned in text. The NOx emissions are shown in Fig.S8. The discussion has been revised.

L595-: This section should be condensed where appropriate. You have compared CHASER and TROPOMI HCHO columns in detail already, so the focus here should be on what those most significant di□erences between OMI and TROPOMI HCHO are and how they compare with the CHASER HCHO.

Response: We removed the redundant information.

L612: Referring to Figure 7 at the beginning of this paragraph.

Response: We have revised the figure numbers in the revised manuscript and ensured consistency throughout the manuscript

L680: Which “observation” do you refer here?

Response: Both OMI and TROPOMI. We have revised the sentence.

L688-689, L703: what are di□erences between OMI SOA and OMI BIRA HCHO products? A brief introduction will be helpful.

Response: We have added the following text to address this comment
L728-731 The most relevant differences between the OMI BIRA and SAO products are related to the underlying CTMs that simulate the apriori profiles and the reference sector correction (Zhu et al., 2016). A comprehensive list of the differences between the two products is available from Zhu et al. (2016).

L762-764: Is this coincidental?
Response: Yes, coincident data were used for the comparison

L808: "In Kasuga, modelled diurnal variations correlate strongly (R=0.85) with the observations". But in Figure 9, the R value is 0.41, not 0.85. Please check.

Response: We have revised the texts.

L837-839: Could you elaborate a bit more on this mechanism?

Response: We have added the following texts.

L904-909 Zhao et al. (2022) reported a similar finding and attributed enhanced CH_4 oxidation in the presence of water vapor to the HCHO mixing ratios above 2 km. At higher altitudes HCHO is produced through the CH_4 oxidation (i.e., CH_4 + OH) initiated CH_3O_2 (methyl peroxy radical) + CH_3O_2 pathway. HCHO production through this pathway is considered in CHASER Therefore, despite the differences in the magnitude, CHASER has shown good skills in reproducing the VOC profiles.

L844: Please check the coordinates for Amazon. You could draw two boxes on the map (Fig. S2) to represent the two studied regions.
Response: We have revised the texts. We agree with the reviewer's perspective of providing the box. However, we adopted the style of earlier studies (i.e., He et al., 2022, Sekiya et al., 2018) using ATom measurements.

L872-873: Have you already defined these sensitivity simulations?

Response: We have defined all the simulations in the Table 1 in the revised manuscript

L932-934: Does the model's course resolution play a role in this case?

Response: We didn't infer the impact of model resolution on this comparison. However, our earlier studies (i.e., Hoque et al., 2022) have demonstrated the impact of horizontal resolution on the model-ground-based comparison.

L944: The last half sentence doesn't read well; do you mean the model underestimates the biogenic contributions?

Response: We have revised the sentence

L1020-1023 Lastly, sensitivity studies were conducted to estimate the contributions of the different emissions sources to the total HCHO columns in different regions. Biogenic emissions were the most significant contributor in most of the regions. In a few cases, biogenic and anthropogenic emission contributions were equivalent. In some regions, only summertime biogenic estimates were found to be reasonable.
