Reviewer 1

We thank the reviewer for the warm comments and appreciation of our work, and for the interesting suggestions which contribute to improve 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.

L95-97. The cited work by Clarisse et al. (2023) was based on NH3 retrievals. What are the specific parameters and corrections that ensure HRI consistency for methanol during the regularization process?

The spectral range for the CH_3OH HRI was kept to 960 to 1080 cm^{-1} as in the previous versions of the product. Regularization was achieved by removing the vector space associated with the 10 lowest eigenvalues of the covariance matrix in the calculation of the inverse of the covariance matrix. Two remaining small corrections were applied to remove residual biases between the different IASI instruments that were observed in the average CH_3OH HRI timeseries. Between 13/04/2015 and 7/10/2015, a correction of +0.02 was applied to the CH_3OH HRIs derived from IASI/Metop-A measurements. In addition, a correction of -0.02 was applied to the CH_3OH HRIs from IASI/Metop-C measurements. This information has now been added to the manuscript.

L106 onwards: How is the methanol retrieval impacted by using a single vertical a-priori profile for land? Methanol is a longer-lived species (~5 days) and its vertical profile can change based on injection height due to fire plume injection (large wildfires) or deep convection processes (like in the Tropics). How has this been evaluated in your retrievals. What is the uncertainty that this bring into the columns estimates and the global inventory? This is very briefly discussed in ~L400-405.

The impact of using a unique profile is important on the retrieved columns, and this is why the application of averaging kernels is essential to harmonize the vertical profiles in the column comparisons (Sec. 2.1). We now provide a better explanation of the role of averaging kernels (Sect. 2.7): "*Since the total column averaging kernel (AVK) increases steeply with altitude (Fig. 1), application of the AVK to the model profiles increases the columns wherever the model profile shape shows higher values in the mid- and upper troposphere, compared to the methanol profile used as a priori in the retrievals*." The vertical and spatial distribution of the AVK is also further described (see next comment).

Fig 1. Out of curiosity, how much variability is there in the IASI AVK in land and the ocean? Are there regions or seasons that show large sensitivity biases from the average profile? What would that mean for the retrievals in those regions?

Thanks for the very good remark. We've added the following figure in the Supplement (Fig. S1). It displays the distribution of the seasonally-averaged total column AVK at a layer right above the surface (0-140m altitude). It is highest (0.3-0.5) above deserts and semideserts, where observation conditions are most favourable due to high thermal contrast. It is lowest above tropical oceans and tropical forests (~0.1). We added the following text in Sect. 2.1: "*The sensitivity of IASI is lowest near the surface (AVK around 0.1–0.2) and highest in the upper troposphere and stratosphere (around 2–3). The spatial and seasonal distribution of the AVK is displayed on Fig. S1.*"

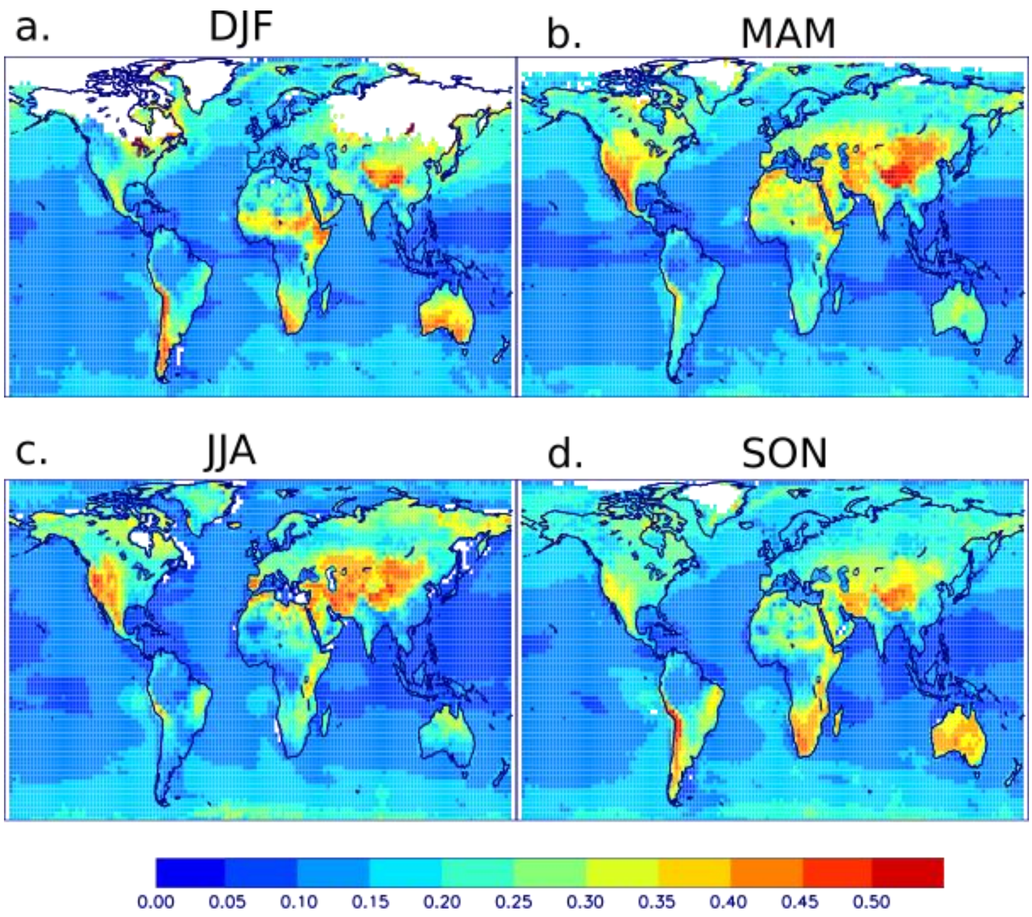


Figure. Seasonally-averaged distribution of the IASI total column averaging kernel at the first IASI grid layer (0-0.14 km altitude). Average over 2008--2019.

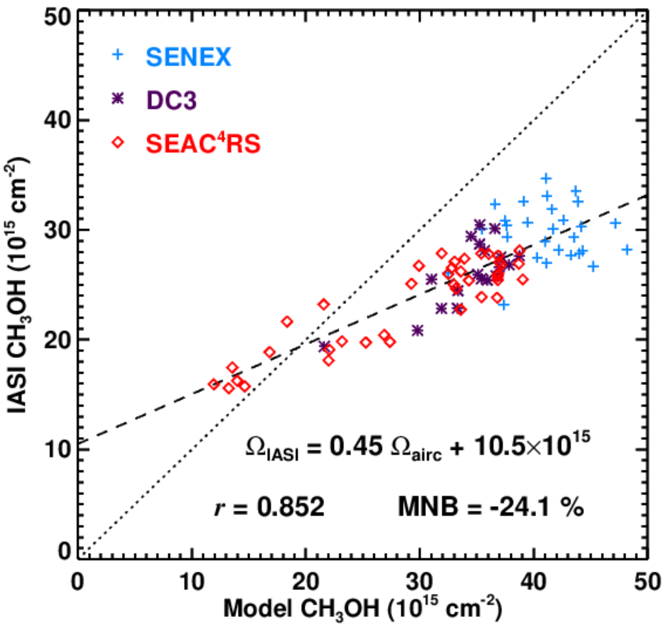
Section 2.2. Is there a reason why the aircraft datasets selected for the bias correction are all in the U.S.? Would it be helpful to have a more diverse representation of the global regions (i.e., Amazon, Asia?) to perform a bias correction that would be applied to global data? – this is discussed in the conclusions!

Yes, it could be helpful to have datasets over source regions outside the U.S. We agree that the observational dataset used to characterize the IASI bias has its limitations, which motivated the extensive model comparisons with additional datasets (aircraft, surface in situ, FTIR). As discussed in the manuscript, the IASI bias determined from three U.S. campaigns overestimates the bias over boreal forests and (to a lesser extent) over tropical ecosystems. Future work should consider a more extensive set of observations for bias correction.

L148: rationale for excluding urban and fire plumes: but even low-resolution models (2x2.5) show evidence of large urban or wildfire plumes unless those are also excluded from the model configuration (not the case as described in Sec. 2.5.1). Please explain how the removal of those observations impacts the results *sp. bias-correction.

Thank you for this comment. As stated in the manuscript, the filters concern only a few percent of the data for the campaigns used as constraint. We performed a sensitivity inversion constrained by aircraft data, without application of the urban and biomass burning filters. We added the following text in Sect. 3: "*We verified that the above results are only minimally affected by the filtering of urban and pyrogenic plumes mentioned in Sect. 2.2. Without these filters, the slope (0.45) and intercept ($10.6 \cdot 10^{15}$) of the above relationship are essentially unchanged.*"

The figure below replicates Fig. 8b of the manuscript, using optimised model values based on the sensitivity inversion without data filtering.



Same as Fig 8b of main manuscript. The modelled columns are constrained by the aircraft in situ measurements, without urban and pyrogenic data filtering. The regression and statistics are similar to the main manuscript results.

About overestimations after bias-correction – throughout the paper, the authors note that for large columns, the bias correction leads to an overestimation of the columns. What does Figure 8 look if including data from the Arctas campaign? Is a linear regression enough for this bias correction? If this exercise is not possible due to model output availability, it would be good to have a discuss about future directions where more datasets on higher latitudes can be included for the bias corrections.

We disagree – we stated clearly that the overestimation mostly concerned boreal latitudes. Comparison with other aircraft campaigns and with surface in situ data show improved agreement after inversion based on bias-corrected data. Including ARCTAS on this plot would require an additional inversion specifically for the year 2008, which was not conducted. Nevertheless, we agree that adding more data to the derivation of the bias correction would be useful and will be considered in future research.

Minor:

L257: is Gamma_SM a typo?

Indeed. Corrected.

Figure 4. Minor: would it be helpful to have the details from L345-350 as insets in Figure3?

Done.

L397: I thought regularized IASI data was used for the study.

True, but in order to avoid jumps in the monthly number of data in the course of the time series, we decided to use only Metop-A.

Figure 5: I believe that more context and/or explanation of what this figure shows would improve the manuscript. See the question above about using a single vertical profile and sensitivity dependencies.

We have added the following text in Sect. 2.7: "*Since the total column averaging kernel (AVK) increases steeply with altitude (Fig. 1), application of the AVK to the model profiles increases the columns wherever the model profile shape shows higher values in the mid- and upper troposphere, compared to the methanol profile used as a priori in the retrievals.*"

L419-420: I am curious what source can exist there that have not traditionally be accounted for (thus the need to increase emissions by such large factors)

Broadly similar increases in biogenic emissions were found in previous work (Stavrakou et al. 2011, Wells et al. 2014) and in our own IASI-based inversions (see Figure 9). As discussed in Sect. 4, the underestimation might partly result from the neglect of soil emissions in MEGAN.

Fig 6 and Fig 9: This is minor, but the direction of the panels is flipped between the two figures: Figure 6 shows the obs, a priori and optimized vertically, while Fig 9 shows it horizontally.

This is correct, but we choose to keep the figures as they are.

L505. Isn't panel i. SH Africa? Should this instead be (h and j).

Thanks, this is corrected now (panels f, h and j).

L523-524: Can you say more about how the estimates of methanol pyrogenic emissions presented here might be impacted by the earlier IASI overpass time (9:30 am) when most fires have not started or re-activated yet?

Due to the lifetime of methanol (several days), the diurnal cycle of its release by fires does not play a crucial role, as it does for shorter-lived species (e.g. NOx). Furthermore, as explained above in the manuscript (Sect.2.7), the dominance of the biogenic flux makes the top-down results uncertain for biomass burning emissions.

L564-565: This sentence is a bit confusing. I understand the gist of it, but re-writing is needed. One suggestion would be to add "Using" at the beginning of the sentence.

We changed the sentence as follows: "*Since the emission optimisations are constrained by IASI columns that are bias-corrected using aircraft data, …*".

L568 and L643: What about the for GoAmazon? Is that really a negligible bias? The model completely misses the vertical profile of methanol (in both the a priori and optimized), so is there really an improvement?

The sentence states that there is an improvement in the comparison statistics, which is indeed the case. Regarding the vertical profile, as indicated on Fig. 12g, the number of measurements per altitude bin is much lower above 2km altitude than below 2km. The discrepancy at the higher levels is therefore of marginal importance and could be due to unresolved gradients at the model resolution.

L575: Does this mean that this top-down emission estimate overestimate biogenic emissions in this region (and others that do not have stringent controls on VCP?)?

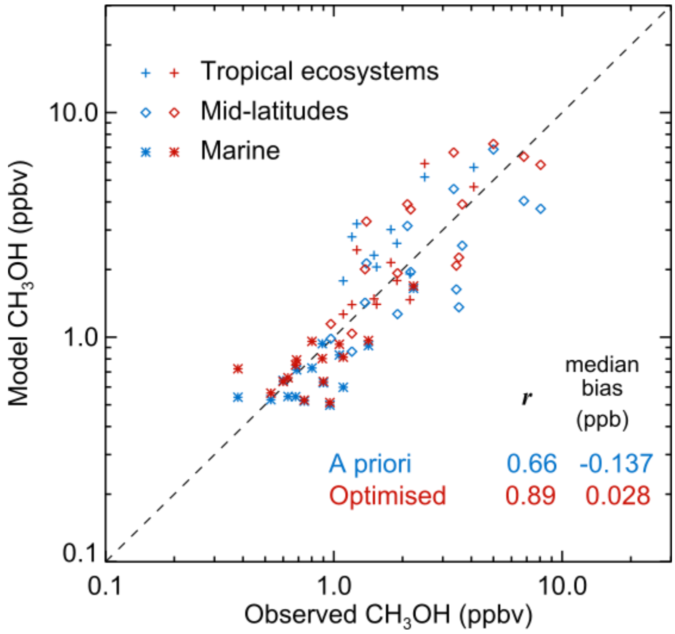
This point is already discussed in this paragraph: "*Part of the methanol emission increase inferred by our inversion might therefore be wrongly attributed to biogenic emissions. In the free troposphere during KORUS-AQ, the strong correlation of methanol with acetone suggested an important biogenic contribution, however. Furthermore, the seasonal variation of top-down methanol emissions over northern China (Fig. 11d) shows a much stronger enhancement in spring than in fall, similar to other regions at mid-latitudes and consistent with a predominantly biogenic source. Incorporation of VCP emissions in methanol emission inventories will be needed to improve the assessment of biogenic emissions over East Asia*."

L595: About ARCTAS-July and the exercise where you added fire observations (removed the CH3CN filter): if the injection heights for those plumes are underestimated (as they usually are), then the model columns would be overestimated if the AVK is applied - since the instrument is more sensitive to CH3OH when its lofted higher in the atmosphere (Fig 1). Could this be a cause for the overestimation?

Sure, the injection heights could be incorrect, and in fact, the pyrogenic emission magnitude could be biased, thereby affecting the comparison. But the important point here is that the conclusion of the comparison remains unchanged against either filtered or unfiltered data.

Figure 13: This figure might convey better information if datapoints were colored or grouped by location, since that's where most of the variability is observed (as discusses by the text).

Excellent suggestion. We now use different symbols for tropical ecosystems, mid-latitudes and marine. The updated Figure 13 is shown below.



Updated Figure 13.

L680: Since its known that the IASI columns that are biased corrected are too high for boreal regions, and that anthropogenic emission might be underestimated in Asia, this number should come with that warning (i.e., an upper bound due to x, y and z) or a margin of certainty [137 – 160] Tg yr-1

The lower bound suggested by the referee (137 Tg yr-1) appears difficult to justify. A precise range is difficult to provide. It would require carefully-designed additional sensitivity inversions and improved bottom-up anthropogenic methanol emissions (incorporating VCP emissions), which are not available yet.

L690: Could this be the reason for the overestimation in other Boreal forest?

The performance of the deposition model is good at other sites dominated by coniferous trees, but it has not been tested at other boreal forest sites (e.g. in Russia and Canada). A potential overestimation of dry deposition over boreal forests e.g. over Canada would imply lower biogenic emissions but similar top-down methanol columns, since the optimisation is constrained by satellite data. Therefore, the model overestimation would remain against ARCTAS-July methanol data.

Reviewer 2

We thank the reviewer for the warm comments and appreciation of our work, and for the interesting suggestions which contribute to improve 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.

Section 2.5.2: While secondary production accounts for ~15-30% of the total emissions, I wonder how accurate this estimate may be. Nicely, a brief discussion is presented in lines 554-556, showing that changing the overall yield of CH_3O_2+OH (from 11.4% to 13%) does not change the results. However, it is stated (line 240) that many other uncertainties are present in the chemical production.

There are indeed many uncertainties in the chemical reactions, but the 13% overall yield was constrained by model comparisons with methanol observations.

Would the consideration of these uncertainties considerably change any of the results obtained in the inversion? Additionally, I wonder how well RO_2 is represented in the MAGRITTE model. I assume that most of it is coming from the oxidation of CH_4, and therefore I would appreciate some additional discussion on the simulation of the OH fields in the model.

Thanks for this interesting point. We added the following text in Sect. 2.5.2: "*The model-calculated OH levels are a significant source of uncertainty for both the secondary production and the photochemical sink of methanol. For example, the representation of halogen chemistry (Sherwen et al., 2016), lightning NOx (Ghosh et al., 2025), biogenic VOC emissions (Williams et al., 2013) and their degradation mechanisms (Novelli et al., 2020) are potential causes of biases in the calculated OH concentrations. On the global scale, the MAGRITTE-calculated, mass-weighted tropospheric OH concentration average is $11.8\cdot10^5$ molec.cm^{-3}, very similar to a recently reported multi-model average (($11.1\pm1.6)\cdot10^5$ molec.cm^{-3})(Naik et al.,2013).* "

Section 3: It is very interesting to note that the MAGRITTE model inversion is able to reproduce the observations in an exceptional way. Furthermore, I appreciate the investigation of the possible bias present in the IASIv4 data, which I consider the real main strength of this manuscript. However, I wonder if the authors could elaborate on the risk of using only an observational dataset over North America. Is this region really representative of the IASI bias? Based on the work of Bates et al. (2021), North America is mostly influenced by biogenic emissions, while biomass burning as well as chemical formation are comparatively much lower. Could the IASI bias be different in other areas of the globe, for example in regions strongly influenced by biomass burning or over the tropics, due to the much stronger influence of methanol chemical production?

We agree with the reviewer that the observational dataset used to characterize the IASI bias has its limitations, which motivated the extensive model comparisons with additional datasets (aircraft, surface in situ, FTIR). As discussed in the manuscript, the IASI bias determined from three U.S. campaigns overestimates the bias over boreal forests and (to a lesser extent) over tropical ecosystems. Future work should consider a more extensive set of observations for bias correction.

Figure 9: Based on what is presented (see, for example, line 385), the focus of the inversion is on terrestrial emissions, and therefore the IASI data over oceans were excluded. I would very much appreciate it if all figures including IASI data could be masked over the ocean, to avoid confusion during reading.

Thanks for this suggestion. Done.

Line 600: It would be great if the Spearman correlation could also be listed, to provide a more complete overview of the comparison.

Done. The following text was inserted: "*while Spearman's rank coefficient is increased from 0.86 to 0.89*".

REFERENCES

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