Peer-review of egusphere-2023-1133

We hereby express our sincere gratitude for the two anonymous reviews for their work on reviewing the manuscript: "Combined assimilation of NOAA surface and MIPAS satellite observations to constrain the global budget of carbonyl sulfide" by Ma et al. Below are the comments by the reviewers and our response point by point. Our response is colored in red.

RC1: 'Comment on egusphere-2023-1133', Anonymous Referee #1, 06 Oct 2023

This is an excellent study and important contribution to efforts making use of COS data to study the global carbon cycle. I have a few mostly minor comments:

The authors state the specific goal of exploring inversions using separate and combined datasets (L86-88), which I think is a great contribution. But I don't understand why bias free MIPAS only experiments were not included. It seems that if the combined inversion requires a bias correction to MIPAS, then the MIPAS-only inversion should also be based on corrected data. Please explain why or why not this was not included, and please include bias-corrected MIPAS-only inversion results if you think necessary. I also recommend to remove results based on uncorrected data from the abstract. You could simplify the abstract by mentioning the bias correction is necessary and performed, and limit the reported findings to those based on corrected data.

The bias correction is aimed to tackle the potential systematic bias between the NOAA surface network and MIPAS satellite observations of COS. However, a bias between the two observational streams does not necessarily mean that the MIPAS satellite observations themselves are biased. As we state in the manuscript (L235-237), the derived bias might be due to model errors, e.g. transport and/or stratospheric chemistry. The MIPAS-only inversion is therefore meant as a reference run. As shown in Figure 3, the MIPAS-only inversion overestimates COS at all NOAA surface stations, which implies that the MIPAS observations as sampled in the TM5 model overestimate NOAA COS surface observations. As noted, MIPAS observations are usually not sensitive to the lower troposphere. Since we do not know the "true" MIPAS bias, a bias-corrected MIPAS-only inversion is difficult to perform.

The final message in the abstract "better characterization of biosphere and ocean fluxes by observations is urgently needed" is mixed with respect to recommendations moving forward. There appear to be three messages: (1) better characterization of biosphere prior and uncertainty, (2) better characterization of combined ocean + land fluxes, and (3) better constraint of both by combining surface and satellite observations. It would help if the authors could state more clearly what is the conclusion based on the present study, and what set of steps need to be taken to bring more clarify to tropical COS fluxes. The finding in Section 4.4 (Line 418-419) that "inclusion of MIPAS data in the inversion leads to a better separation of land and ocean fluxes" seems like an important takeaway for the abstract.

We agree with the reviewers three messages, which are concise and clear. The abstract is rephrased to convey the important messages:

"Carbonyl sulfide (COS), a trace gas in our atmosphere that leads to the formation of aerosols in the stratosphere, is taken up by terrestrial ecosystems. Quantifying the biosphere uptake of COS could provide a useful quantity to estimate Gross Primary Productivity. Some COS sources and sinks still contain large uncertainties, and several top down estimates of the COS budget point to an underestimation of sources especially in the tropics. We extended the inverse model TM5-4DVAR to assimilate MIPAS satellite data, in addition to NOAA surface data as used in a previous study. To resolve possible discrepancies among the two observational datasets, a bias correction scheme was implemented. A set of inversions is presented that explores the influence of the different measurement streams and the settings of the prior fluxes. To evaluate the performance of the inverse system, the HIAPER Pole-to-Pole Observations (HIPPO) aircraft observations and NOAA airborne profiles are used. All inversions reduce the COS biosphere uptake from a prior value of 1053 GgS a^{-1} to much smaller values, depending on the inversion settings. These large adjustments of the biosphere uptake often turn parts of the Amazonia into a COS source. Only inversions that exclusively use MIPAS observations, or strongly reduce the prior errors on the biosphere flux maintain the Amazon as a COS sink. Assimilating both NOAA surface data and MIPAS data requires a small bias correction for MIPAS data, mostly at higher latitudes, to correct for inconsistencies in the observational data and/or transport model errors. Inclusion of MIPAS data in the inversion leads to a better separation of land and ocean fluxes. Our inversions with bias correction reduce the global biosphere uptake to respectively 570 and 687 GgS a^{-1}, depending on the prior biosphere error. Over the Amazon, these inversions reduce the biosphere uptake from roughly 300 to 100 GgS a^{-1}, indicating a strongly overestimated prior uptake in this region. Although a recent study also reported reduced COS uptake over the Amazon, we emphasize that a careful construction of prior fluxes and their associated errors remains important. For instance, an inversion that gives large freedom to adjust the anthropogenic and ocean fluxes of CS_2, an important COS precursor, also closes the budget satisfactorily with much smaller adjustments to the biosphere. We thus highlight that better characterization of the biosphere and ocean fluxes are needed, especially over the data-poor tropics."

Section 2.2: I think the choice of the 8th level and high sensitivity AK (> 0.03) MIPAS data is fine. But this begs the question, how do the tradeoffs between data density and surface sensitivity affect inferred fluxes (spatial/seasonal distribution, land/ocean partitioning, etc)? The authors speculate about including higher level data in Section 5 (Line 479) but say nothing about lower levels. I suggest, if it's not too much extra work and the justification is there, for the authors to conduct another run using lower level data, to explore changes in signal and uncertainty. Even at lower levels, the reduced amount of valid data will still overwhelm the NOAA data (e.g., 490 in 2009). At the very least, it's worth discussing and speculating about these tradeoffs.

Agree, and we rerun two inversions on different levels of MIPAS data: the 5th and 8th level. Please note that these two inversions are driven by ERA5 meteorological datasets, instead of ERA-interim due to a recent upgrade of the modeling system. Figures R1 and R2 show inversion S1 using both NOAA and MIPAS observations on the level 8th and 5th, respectively. The major difference is that the MIPAS data on the 5th level has large portion of missing data mainly in tropical regions. Since the information from MIPAS is specifically needed in the tropics, the choice of using 8th level data from MIPAS seems justified. Note that there is a difference between the inversion using ERA5 and ERA-interim (Fig. R1 and Fig. 5), yet this topic is beyond the scope of this paper. The numbering starts from 0 in this context since we used python to program the satellite datasets processing, and we modified the numbers to 6th and 9th by counting from 1 in the revised version.

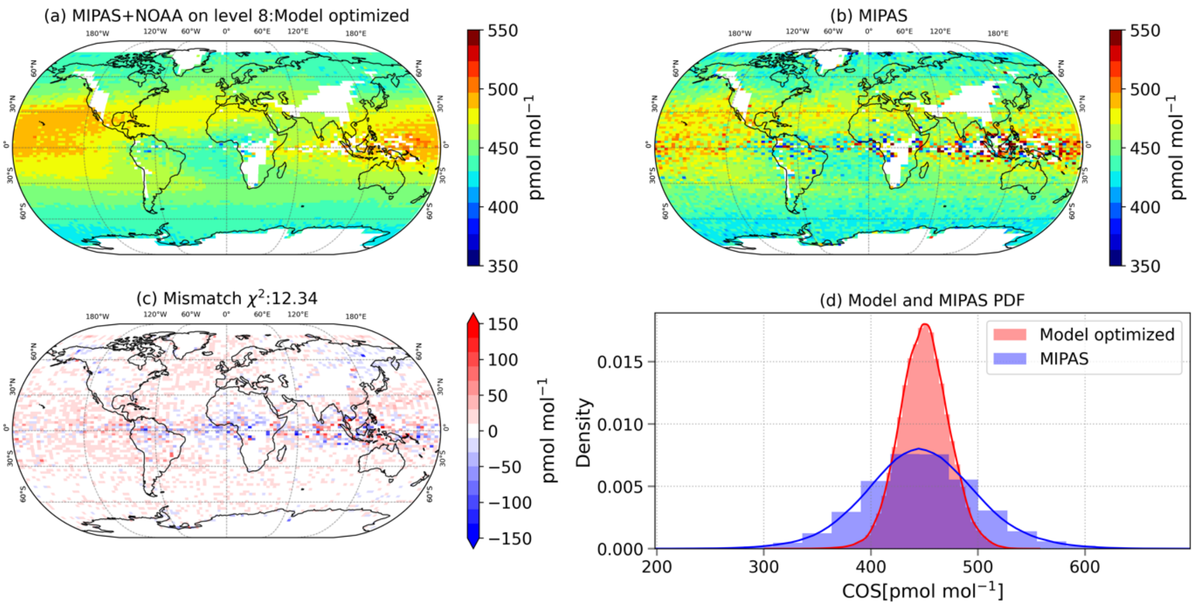

Figure R1. The MIPAS+NOAA S1 inversion using level 8 (numbering starting from 0) of MIPAS.

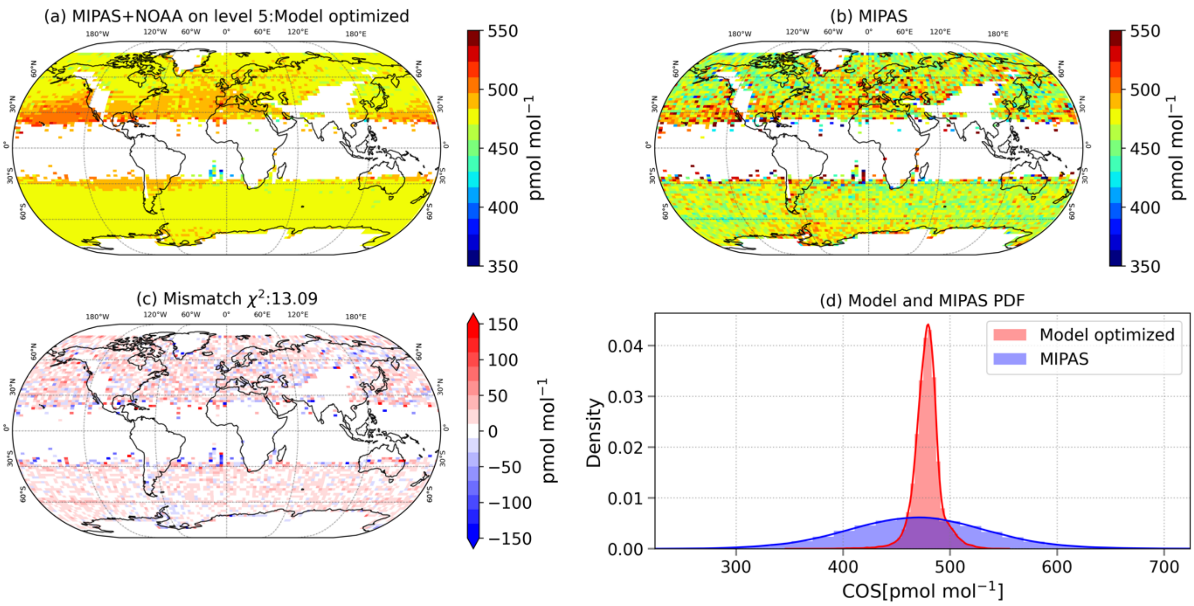

Figure R2. The MIPAS+NOAA S1 inversion using level 5 (numbering starting from 0) of MIPAS.

Line 33: Suggest referring to “atmosphere” somewhere in the description of the transport model (e.g., The transport model describes physical transport by atmospheric winds and chemical loss/production by atmospheric chemistry”)

Agree. In Section 3.3 Line 201, the sentence is inserted:

“The TM5 transport model describes physical transport of tracers by atmospheric winds and their chemical loss and production.”

Line 178: Are SIB4 fluxes year specific or climatology? Does this matter?

The SIB4 fluxes are year specific and this is important since the interannual variability of the biosphere flux is also important to close the gap between sources and sinks of COS using inverse modelling. The SIB4 fluxes are model outputs from Simple Biosphere Model version 4 for the years 2000-2020 and we used these fluxes as prior biosphere flux.

Line 201: Use of "generally the same" is vague. Are there important differences we should care about?

The model settings are identical as in Ma et al. (2021). The only difference is that the MIPAS observations are assimilated into the inverse system TM5-4DVAR.

In Line 201, this changed to:

"The model settings are identical to Ma et al., 2021. The difference is that the MIPAS observations are assimilated into the inverse system and that a bias correction scheme is implemented."

Section 5: I'm curious how much of the Amazon source can be attributed to outliers during a specific year of the inversion. The authors analyze the period 2008-2010, but I don't see any mention of specific years. This could be a more robust finding if the inferred source is persistent over the three-year period.

The inversion period is 2008-2010, and the first 6 months and last 6 months are considered a spin-up and spin-down period. To calculate the statistics of the regional fluxes like the Amazon, the spin-up and spin-down periods are removed, and the year 2009 is used to show the Amazon flux. Fig. R3 shows the COS biosphere flux over the Amazon on a monthly time scale in 2008-2010. When the spin-up and spin-down periods are removed, results seem consistent across the inversion. To further improve the robustness of the source attribution, it would be necessary to extend the time span of the inversions. This would require substantial work that we would like to perform in a future study.

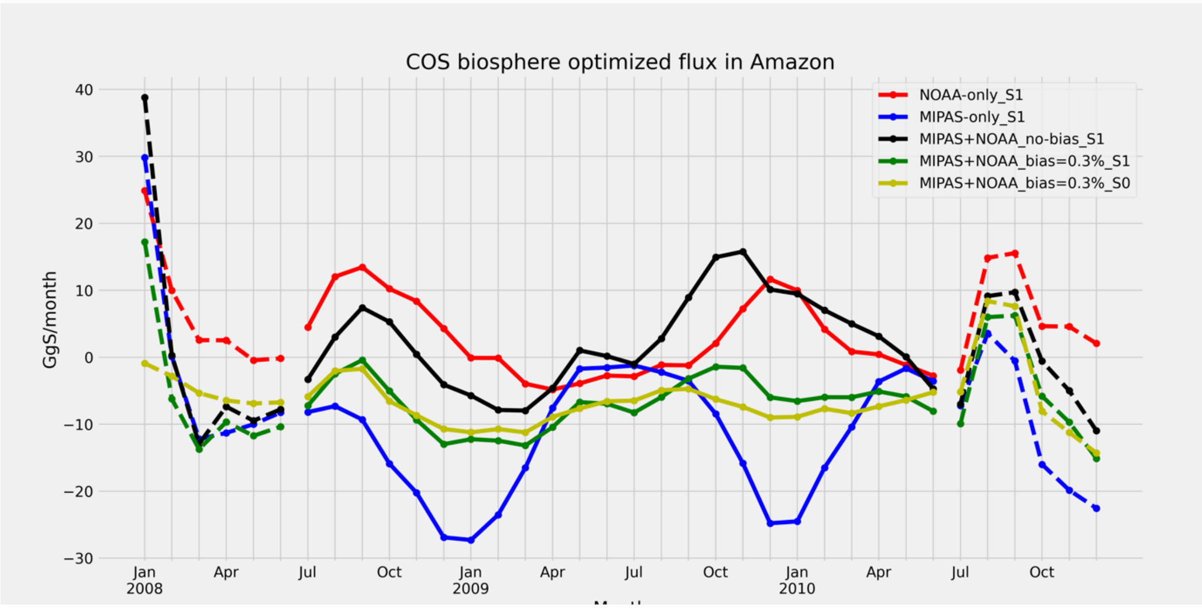

Figure R3. The biosphere COS flux over the Amazon on a monthly time scale after optimization for different inversions in 2008-2010. The dashed lines indicate the spin-up and spin-down periods.

RC2: 'Comment on egusphere-2023-1133', Anonymous Referee #2, 26 Oct 2023

The manuscript by Ma et al. constrained the global budget of COS using a combination of surface and satellite observations. They implemented a bias correction scheme to resolve the discrepancies between these two datasets and evaluated the results using independent aircraft and airborne data. Their results show consistent lower biosphere uptake of COS from the prior, especially in the Amazonia. The observation-constrained COS budget is an important topic and fits the ACP readership. The manuscript is well-written with nice experiment setups. I have minor comments that could potentially improve the clarity of the presentations.

We appreciate the reviewers comment on the manuscript.

My major concern is how robust are the strong reductions of COS sources over the Amazonia. The manuscript also questions the realism of this result. How sensitive are these results to the inversion parameters and observations? How does the result fit in the error bars? Is there any other work or independent data showing a similar conclusion. These additional tests and discussions are needed to better support the conclusion.

We agree with the reviewer that the realism of the result is questionable, but this is mainly due to lack of observations in tropical regions. The uncertainty of the prior fluxes is also poorly quantified. Our result is based on different inversions, and indeed points to large remaining uncertainties in tropical regions. The major challenge of the study is how to improve the separation of ocean and land fluxes. The extra inversions included in the supplementary material show that the inversion scenario SCS2 leads to a better separation of ocean and land fluxes. In SCS2 the prior errors of the ocean fluxes are increased to 150% and the prior errors of the biosphere fluxes are reduced to 10%. On the positive side, we do find a similar result as Stinecipher et al. (2022), who also found a largely reduced GPP flux based on the COS proxy approach using MIPAS satellite data as climatological constraints. The comparison to this reference is already included in the paper and clearly stated in the conclusions.

Stinecipher, James R., et al. "Remotely sensed carbonyl sulfide constrains model estimates of Amazon primary productivity." Geophysical Research Letters 49.9 (2022): e2021GL096802.

Specific comments

L11, is there any support for the claim that Amazonia is a COS source from other data? Otherwise, this would suggest that the posterior results are not realistic.

This is indeed our line of argumentation (e.g. line 392: It is questionable, however, whether positive fluxes from the Amazonia are realistic). Even when MIPAS data are assimilated the tropical biosphere still acts as a “waste bin” for the inversions, meaning that the required global adjustments are projected to the (large) tropical fluxes without much additional costs (unless the errors on the biosphere are substantially reduced, like in the SCS2 scenario).

L15, what data are these errors evaluated against?

The errors are associated with prior or posterior fluxes for the inversions. For example, the prior errors are the errors of the prior fluxes, and the posterior errors are the errors of the posterior fluxes.

L55-72, it would be easier for readers to understand if the budget is shown in a table.

The prior budget is shown in Table 1. The content in L55-72 is not a prior budget used in this study but a summary of literature, so we would like to keep them as text.

Eqn 6, how is “error” defined?

The errors are defined as in Eq. 4. The **B** matrix is the error-covariance of the prior fluxes. The covariance matrix **B** describes the uncertainty statistics associated with the state vector. Off-diagonal elements of **B** are determined by user-prescribed spatial and temporal correlations of the fluxes (details of the used correlation lengths can be found in Ma et al. (2021)).

L231, why is a spin down period needed?

It is a standard procedure to remove the beginning and end of the inversions. In this case, the spin-up and spin-down period is 6 months, and the whole inversion is 3 years. At the beginning of the inversion, the optimized fluxes may be affected by the uncertain initial condition. For instance, COS biosphere flux is turned to positive values during the spin-up period, as shown in Fig. R3. At the end of the inversion, there are less observational data to constrain fluxes. We believe that the spin-up and spin-down approach improves the statistics of the inversion.

Eqn 7, how are the values in the satellite data compared with NOAA measurements? Are their differences consistent with the 0.003 error for beta here?

The values in the MIPAS data are not directly compared with NOAA measurements, but they are directly compared with COS mole fractions sampled in the transport model space, also applying the averaging kernel as discussed in Sect. 2.2. Since MIPAS is a limb sounder, and is affected by weather conditions like clouds, it is impossible to make a direct comparison between MIPAS data and NOAA measurements. Furthermore, it is unknown if the differences are consistent with the 0.003 error. We chose the 0.003 error by trial and error to ensure that the bias correction for the MIPAS data becomes not too large.

L253, how is this balance determined? Does it mean the two terms have similar magnitudes in the cost function? Please clarify.

We chose an arbitrary number of $\sqrt{10}$ as error inflation to balance the cost function in the NOAA data part and MIPAS data part, as shown in Table 4. The two terms do not have similar magnitudes, partly because the number of MIPAS data points is much larger than that of NOAA surface observations.

Fig 3, why do you need a spin up and spin down for the constraint? How did you choose the length? It would be nice to add another line of MIPAS data at the NOAA locations.

The spin-up is needed in this study since the initial condition is not optimized and it will affect the first a few months inversion. The spin-down is needed because at the end of inversion, there are no more observational constraints to improve the fluxes. The length is chosen to be 6 months, because the lifetime of COS is roughly 2 years, and the tropospheric mixing time is roughly one year. The fluxes of the first 6 month are used to make the atmospheric state consistent with observation. Note that the fluxes are also correlated in time. Unfortunately, it is not easy to compare MIPAS data at the NOAA locations since MIPAS does not have sensitivity for the lower troposphere.

Fig 6, it looks like the bias correction parameters are not adjusted to reduce biases in the tropical regions. Why is that?

We think the reasons are: 1) Mixing is fast in the tropics and transport and chemistry errors are much smaller; 2) At higher latitudes MIPAS samples pressure levels in the stratosphere, where the air is older. Thus, transport errors and errors in modelled COS breakdown become more important; 3) The MIPAS data quality is better over the tropics. Note that we use a rather coarse resolution with 25 vertical layers. This might contribute to transport errors in TM5 model, especially in the stratosphere where the model resolution is coarser.

Fig 9, the bars are too thin to be easily identified. Please adjust the width.

Agree. The bars are made wider to be more easily identified as in Fig. R4:

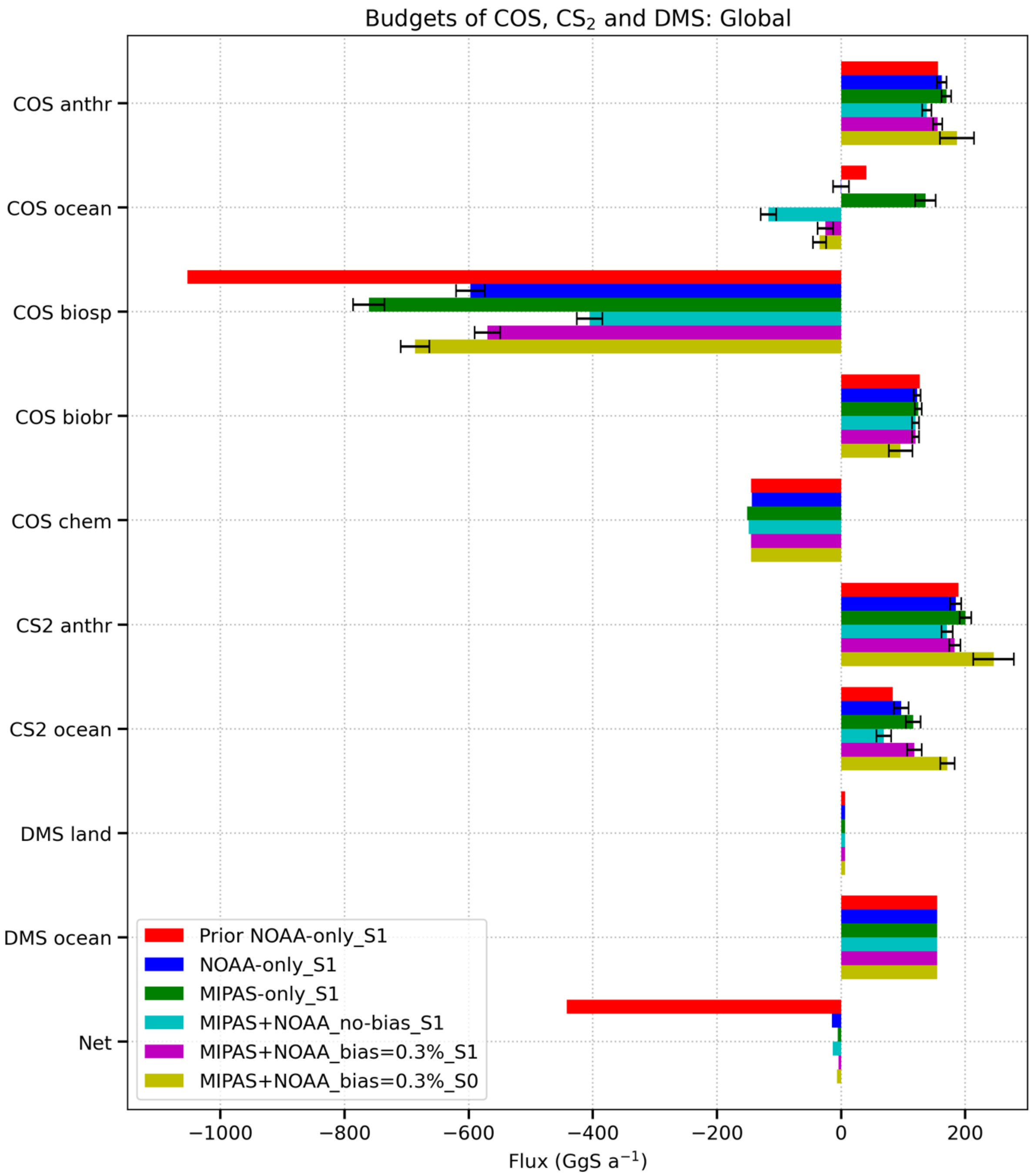

Figure R4. The global budgets of COS from inversions.

491, are these related to the specification of errors? Would the results improve if changing prior error characterizations?

Indeed, we found the results are sensitive to the prior error characterizations. Currently it is hard to conclude which inversion is most realistic. Our result therefore underscores the need to carefully construct the prior error covariance matrix **B** (Eq. 4). About matrix **B**, we need to know more about the errors of the biosphere and ocean fluxes. As an example, inversion SCS2 shows that the ocean fluxes increase when a larger prior error is applied.

Additional modifications:

After discussion with the co-authors, we will replace Fig. 2 with the version below as Fig. R5. The panels (a, c) are modified using altitude grid. Fig. R5 applies the same filtering procedures as described in Sect. 2.2. Also, we changed the numbering of the MIPAS levels by counting from 1 instead of 0.

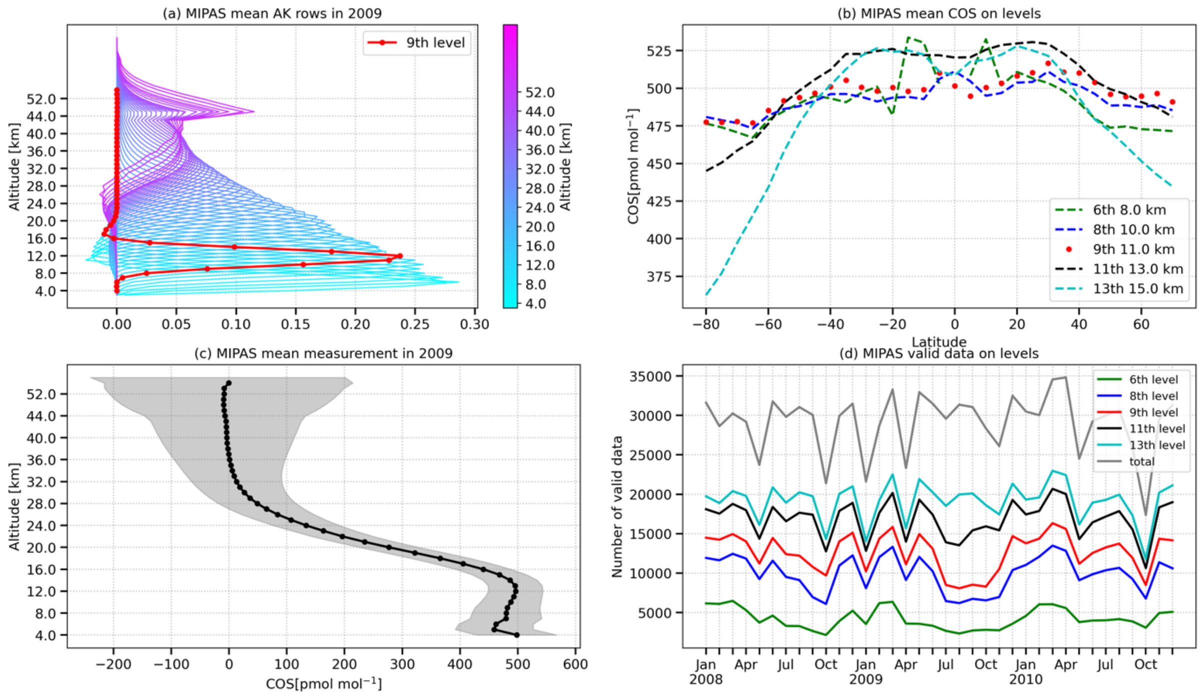

Figure R5. Filtered MIPAS satellite data annual mean in 2009: (a) The selected MIPAS mean averaging kernel rows, colored by their representative altitudes; (b) selected MIPAS COS mean mole fractions on MIPAS levels (6, 8, 9, 11, 13); (c) MIPAS mean COS vertical profile; (d) Number of valid measurements on selected MIPAS levels after data quality control.

In the revised manuscript, the relevant texts have been modified.

We also changed:

"As outlined above, this might imply lack of observations to constrain the model, or a bias in the model transport and chemistry."

into

"As outlined above, this might imply a lack of observations to constrain the model or a bias in the model transport and chemistry. However, a true bias of MIPAS compared to NOAA observations cannot be excluded. Glatthor et al. (2017) also found positive differences of MIPAS retrievals compared to ACE-FTS profiles in the upper troposphere and lower stratosphere."