Constraining the budget of atmospheric carbonyl sulfide using a 3-D chemical transport model

Final Response to Referee Comments

Due to the extensive corrections required, large portions of the text are now different. Hence some referee comments are now unnecessary. We have tried to address every comment as best we can, including a line number where it best relates in the current text. Author responses can be found in italics.

Commenter Summaries

First

Michael P. Cartwright and co-authors present an evaluation of two OCS flux inventories using OCS concentration observations from 14 NOAA towers and from the ACE-FTS satellite instrument. The first inventory is mainly based on Kettle et al. (2002) and is used as a control for the second inventory. The authors define this second inventory using the LRU approach to compute vegetation OCS fluxes, then scaling the other OCS flux components to obtain a balanced global OCS budget. The OCS fluxes from both inventories are transported with the TOMCAT atmospheric transport model. Finally, the surface OCS flux components are evaluated by comparing the seasonal cycles of the simulated OCS concentrations to NOAA flaks measurements. The vertical profiles of the simulated OCS concentrations are also evaluated against ACE-FTS OCS concentration profiles between 5 and 30 km. The authors conclude on a better performance of the new balanced OCS flux inventory compared to the control simulation in terms of seasonal cycle representation. The strength of this work is to make use of ACE-FTS observations to evaluate the simulated OCS concentration vertical profiles. However, major revisions should be considered before publication.

Second

This paper presents forward simulation of OCS using the TOMCAT model. Two main simulations are presented: a control simulation and a simulation in which GPP from the JULES model is converted into an OCS flux using the LRU approach. Results generally show an improvement with the surface observation network, and a favourable comparison with the ACE-FTS observations. The paper is well written, with a clear structure. However, the results are rather thin in the sense that the field of OCS research is moving rapidly, and formal inversion systems are now in place. In that sense, the "hand-adjusted-flux" approach in this paper may be a bit outdated. The comparison with ACE-FTS and the use of a new model (TOMCAT) and biosphere model (JULES GPP) provides sufficient new information. The paper ~~can~~ might provide a valuable addition to the existing literature, after addressing some major issues.

Major Comments

First

1. This work makes insufficient use of OCS state-of-the-art. References to recent studies are missing in the introduction. For example, when describing atmospheric OCS trend, Hannigan et al. (2022) should be mentioned as they found positive trends in the troposphere and in the stratosphere

between 2008 and 2016 at most of the studied sites. The reference of the study by Glatthor et al. (2017) should also be contrasted with the increasing trend found by Hannigan et al. (2022) in the free troposphere at the Jungfraujoch site between 2008 and 2016, followed by a decreasing trend since 2016–2017. Ma et al. (2021) should be presented as an inversion study and completed with Remaud et al. (2022) that also conclude to a missing source in the tropics. The underestimated OCS anthropogenic source suggested by Zumkehr et al. (2018) was also supported by Aydin et al. (2020).

Using the inventory of Kettle et al. (2002) is also a major weakness as many studies have provided new OCS flux estimates since. Therefore, the conclusion that TOMCATocs gives better results than TOMCATcon does not seem relevant when TOMCATcon is based on out-of-date estimates (except for the anthropogenic emissions from Zumkehr et al., 2018). Many limitations that were highlighted in OCS literature arise from using the inventory from Kettle et al. (2002). For example, oxic soil contribution considers a constant atmospheric OCS concentration while recent studies have shown the importance of considering variable atmospheric concentrations for both soil and vegetation OCS fluxes (Kooijmans et al., 2021, Maignan et al., 2021, Abadie et al., 2022). Important processes are also not included in the control inventory. Indeed, oxic soils can not only take up OCS but also produce OCS, and it has recently been shown that anoxic soils cannot be neglected at the global scale (Abadie et al., 2022).

In Section 3.3, why choosing to use a single constant LRU value while several studies provide PFT-dependent LRU values? For example, these sets of LRU per PFT can be found in Seibt et al. (2010), Whelan et al. (2018), Maignan et al. (2021). Moreover, as JULES land surface model distinguishes several PFT categories, it would be possible to use PFT-dependent LRU values.

In Section 3.4, the OCS fluxes that were adjusted to obtain a balanced budget should be contrasted with more recent estimates. For example, choosing to scale CS2 oceanic emissions to 439 GgS/y is not supported by Lennartz et al. (2020) who estimated a total source of 70 GgS/y from CS2. An oxic soil budget of 322 GgS/y is also not in line with the recent estimates of Kooijmans et al. (2021) and Abadie et al. (2022) based on the mechanistic soil model of Ogée et al. (2016).

More recent studies should be added in Table 2 to compare to the OCS budget from this work, such as Maignan et al. (2021), Kooijmans et al. (2021), Remaud et al. (2022).

2. The scaling of OCS fluxes to better match estimates made after Kettle et al. (2002) and to obtain a balanced OCS budget seems quite arbitrary. Such adjustments should be made using an inversion framework as done in Ma et al. (2021) or in Remaud et al. (2022). Without an analytical inverse system that optimizes the fluxes, why aiming at a balanced COS budget? A balanced OCS budget is also not required if analyzing the detrended OCS concentrations.

Moreover, this scaling assumes that the OCS flux spatial distribution of each component is not modified compared to the control inventory, which might not agree with flux distribution obtained in more recent studies.

3. Not considering interannual variations is a strong assumption that should at least be better justified. The study from Chen et al. (2017) does not conclude that interannual variability in GPP amplitude can be neglected. GPP interannual variability could easily be included in this work as GPP is modeled by JULES. Considering only the year 2010 does not reflect the yearly increase in atmospheric CO2 concentration and the fertilization effect.

Otherwise, could the impact of not considering OCS flux interannual variability be quantified? For example, OCS vegetation uptake could be defined as a first order relationship with OCS mixing ratio. Therefore, inter-annual variations in OCS vegetation flux might have a strong impact on the simulated atmospheric OCS concentrations.

4. It is not clear what the goal of this study is, and the title is confusing. It is mentioned that this work evaluates the suitability of gross primary productivity to estimate the OCS vegetative uptake. However, doing so by comparing a vegetation OCS uptake based on GPP to a vegetation OCS uptake based on NPP and NDVI seems outdated (Sandoval-Soto and Stanimirov, 2005). Moreover, the modification of several OCS flux components with the rescaling makes it difficult to compare seasonal cycles of TOMCATcon and TOMCATocs regarding vegetation OCS fluxes. Please specify clearly what are the main goals of this study.

Should this work focus more on the advantage of using ACE-FTS compared to other available OCS concentration observations? Or on the information that can be retrieved from ACE-FTS about the modelling of OCS atmospheric sinks?

Second

This paper presents forward simulation of OCS using the TOMCAT model. Two main simulations are presented: a control simulation and a simulation in which GPP from the JULES model is converted into an OCS flux using the LRU approach.

Results generally show an improvement with the surface observation network, and a favorable comparison with the ACE-FTS observations.

The paper is well written, with a clear structure. However, the results are rather thin in the sense that the field of OCS research is moving rapidly, and formal inversion systems are now in place. In that sense, the “hand-adjusted-flux” approach in this paper may be a bit outdated. The comparison with ACE-FTP and the use of a new model (TOMCAT) and biosphere model (JULES GPP) provides sufficient new information. The paper can might provide a valuable addition to the existing literature, after addressing some major issues.

First, when the results are described in the paper, often hand-waving argumentation is used to explain the deviations between model and observations. “Likely caused”, “could be attributed”. Here, we have to believe the judgement of the authors, since rarely additional arguments are presented. Likewise, the underestimation of modelled OCS in the tropical stratosphere is explained by too fast removal. These observations call for additional simulations to verify whether speculations hold true. Two suggestions here: (1) a simulation with tagged tracers to be used in a more detailed analysis of e.g. seasonal cycles (2) a simulation with reduced photochemical removal in the tropical stratosphere. Presentation of the results would give the paper more body.

Second, the LRU approach is defendable, and uses seasonal CO2 mixing ratios to convert GPP into a OCS flux. It remains unclear what is taken for the OCS mixing ratios here. In the recent papers of Ma et al. (quoted), and Kooijmans et al. (Biogeosciences, 2021, not quoted) it is clearly shown that OCS fluxes become substantially smaller in regions with low OCS abundance (F_OCS = -vd*OCS). I am a bit surprised that nothing is mentioned on how OCS mixing ratios are used to convert GPP to a OCS flux. If a constant value of e.g. 500 ppt is taken, the final OCS flux may become substantially smaller. Kooijmans et al., (2021) report a drop in SIB4 from 922 Gg S yr^{-1} in the original SiB4 to 753 Gg S yr^{-1} when accounting for varying OCS mixing ratios.

Finally, the field of OCS research is moving fast. The paper therefore misses quite some recent references that are relevant for the work. The authors should update the reference list (and discussions) with more recent papers.

Further comments are in the accompanying annotated pdf file.

Minor Comments

RC1

Abstract:

L25: “At the surface, the model captures background concentrations at most of the surface sites to within the maximum and minimum of the seasonal measurements”. It does not seem to be a strong condition to satisfy. It might be better to highlight results on the seasonal cycle amplitude or the phase. – *The error bars represented in Figure 1 are now standard deviation, rather than maximum and minimum. Results of SCA are highlighted in the new abstract.*

1.Introduction:

L49: “The main source of atmospheric OCS is oceanic emission”. This could be replaced by "one of the main sources" as anthropogenic emissions are also a major source of OCS. For example, Zumkher et al. (2018) estimated an anthropogenic OCS source of about 400 GgS/y. Aydin et al. (2020) suggested that this estimate could be underestimated with an anthropogenic OCS source of about 600 GgS/y. – *Updated. Line 64.*

L57: “with estimates ranging from 210 to 2400 Gg S yr-1 (Kettle et al., 2002; Sandoval-Soto and Stanimirov, 2005; Suntharalingam et al., 2008; Berry et al., 2013; Glatthor et al., 2015; Kuai et al., 2015; Launois et al., 2015b; Ma et al., 2021)”. Add references to more recent studies such as Maignan et al. (2021), Kooijmans et al. (2021), Remaud et al. (2022) for vegetation OCS uptake estimates. – *Updated. Line 75-77.*

L61: “OCS hydrolysis also occurs in soil, again catalysed by carbonic anhydrase”. Note that OCS can also be consumed by other enzymes in soils, such as nitrogenase, CO dehydrogenase, or CS2 hydrolase (Smith and Ferry, 2000; Masaki et al., 2021). – *Updated. Line 81-82.*

L63: “with an estimated annual loss of 127-355 Gg S (Kettle et al., 2002; Montzka et al., 2007; Berry et al., 2013; Glatthor et al., 2015; Kuai et al., 2015)”. More recent studies should be mentioned, such as Kooijmans et al. (2021) and Abadie et al. (2022) that lead to smaller soil OCS budgets. – *Updated. Line 83.*

L65: “Soil has also been observed to act as an emitter of OCS in warm conditions (Maseyk et al., 2014)”. This was not observed only for warm conditions. OCS emissions have also been related to soil types (Whelan et al., 2013), nitrogen content, light radiations reaching the soil surface (Spielmann et al., 2019; Kitz et al., 2020). – *Updated. Line 85-88.*

L69: “the latter of which has been used as a benchmark for more recent studies”. Please add the references of the studies. – *Updated. Line 92-93.*

2. Observations:

Section 2.1: Please provide and detail the uncertainties associated with ACE-FTS retrievals. – *Updated. Line 145-147.*

3. Chemical transport modelling of OCS:

Section 3.1: What is the timestep used to run the TOMCAT model? – *6 hours. Updated. Line 172-173.*

L139 to L224: Please make it clearer which fluxes have been used for TOMCATcon and which one have been used for TOMCATocs. – *Updated. $TOMCAT_{CON}$ uses inventory in Section 3.2 and $TOMCAT_{OCS}$ uses inventory in Section 3.3.*

L153: "The three sink terms are an oceanic sink, soil uptake and a vegetative sink". OCS photolysis in the stratosphere and OCS oxidation by OH radical in the troposphere should also be included in OCS sinks, as atmospheric OCS reactions are not explained before in section 3.1. – *We include a summary of OH and Photolysis loss in Section 3.1, before describing all the fluxes. It is clear the same scheme is used for all model runs. Line 166-170.*

Equation 1: Precise the unit for each term of this equation. What is used for OCS background concentration? – *Updated. Line 222-226.*

L173: Please replace "LRU is the normalised ratio of OCS assimilation rates to CO2 at the leaf-scale. This is then normalized by background concentrations of the two gases" by "LRU is the ratio of OCS assimilation rates to CO2 at the leaf-scale, both normalized by their respective concentration". – *Updated. Line 223-224.*

L180: "but is slightly under half that of the largest estimation of 1115 Gg S in Table 2 from Montzka et al. (2007)". Launois et al. (2015) estimated a larger plant OCS uptake than Montzka et al. (2007) for the ORCHIDEE land surface model. – *Updated. Line 242.*

L204: "at Northern Hemisphere (NH) NOAA-ESRL sites". Please precise which sites and whether they receive air masses mainly coming from the ocean. – *Updated. Line 277-278.*

Table 2: Why were more recent studies not included in this table for comparison? Such as Maignan et al. (2021), Kooijmans et al. (2021), Remaud et al. (2022). – *Updated to include Remaud et al. (2022).*

4. Results:

L243 to L245: "TOMCATCON was initialised using OCS values in each grid box from TOMCATOCS, after 10 years (1994 – 2003) spin-up. Only 2004 monthly mean mixing ratios from TOMCATCON have been included, as this flux inventory has a 245 net negative budget and therefore a negative trend over longer periods". Should this be in Section 3 as it is related to the method? – *Moved to section 3. Line 187.*

L244: "Only 2004 monthly mean mixing ratios from TOMCATCON have been included, as this flux inventory has a net negative budget and therefore a negative trend over longer periods". Please precise the net negative budget. The trend in atmospheric COS concentrations should not be an issue if you remove the trend and compare the detrended atmospheric concentrations. – *Updated. Line 188. Figure 2 now presents monthly anomalies.*

L249: "Error bars associated with the observations represent the maximum and minimum values for each month at every site". Representing the standard deviation would be a better indication of the uncertainty of the mean value. – *Updated. Figure 1 error bars are now represented by standard deviation.*

L253: "Comparisons between TOMCATocs and TOMCATcon are shown here to emphasise the improvements made by the flux inventory developed in this study". How could the improvements obtained with TOMCATocs on atmospheric OCS concentrations be compared to the improvements made when using inversion systems such as in Ma et al. (2021) and Remaud et al. (2022)? – *We compare TOMCATOCS to a new model run TOMCATSOTA, which utilises fluxes that are more up-to-date in the literature and similar or the same as those used in as prior fluxes by Ma et al. (2021) and Remaud et al. (2022). See discussion and supplement.*

L254: “The root mean square error (RMSE) for the entire period is shown for each site, alongside the seasonal cycle amplitude (SCA)”. Precise that in the following you also compare the phases of observed and simulated seasonal cycles. – *SCA is now displayed in Figure 2.*

L255: “Generally, there is an improvement in RMSE across all the sites, but in some cases, there is a degradation, which is mostly attributed to background concentration, rather than the model’s ability to capture a suitable seasonal cycle, hence both are shown.” By “background concentration”, do you mean the average concentration? If so, could you please show that the degradation in RMSE is due to the average concentration? – *I did mean average concentration. Figure 1 shows $TOMCAT_{OCS}$ and $TOMCAT_{CON}$ monthly mean model values, but Figure 2 shows monthly anomalies. So we are able to distinguish the impact from poor estimation of average concentration and of the seasonality.*

L264: “This seasonal cycle resembles that of CO2, hence GPP is a suitable proxy for calculating OCS uptake”. Please rephrase as similar seasonal cycle is not reason enough to use GPP as a proxy of vegetation OCS uptake. – *I agree. This was rephrased. Line 377.*

L275: “Here we show realistic amplitudes in the seasonal cycle from TOMCATOCS, 76 ppt at LEF and 71 ppt at HFM, compared to observed values of 123 ppt and 128 ppt, respectively”. Please rephrase as the SCA at these two sites are still largely underestimated. – *Rephrased. Line 399-401.*

L279 to 282: The constant LRU value used in this study could be compared to other LRU estimates for the same vegetation types found at LEF and HFM to see if it could be underestimated. If a constant OCS mixing ratio was used to compute OCS vegetation uptake, this could also affect the SCA. – *I have updated the text in places to make it clear we are using OCS concentration from online within the model. A brief discussion of LRU in the literature is made in Section 5: Line 565-570.*

L329: “OCS values decline above and below the UTLS due to removal by photosynthesis at the surface”. Soils can also absorb OCS at the surface. – *Updated. Line 456-457.*

L345: “potentially attributed to slower surface OCS uptake”. It could also be due to an underestimated surface OCS uptake. – *Updated. Line 472-473.*

Figure 2: What could explain the net distinction between higher mixing ratios in NH compared to SH found in TOMCAT in JJA and SON? – *Weaker removal of OCS in the SH.*

L364: “this suggests the upper atmospheric sinks are modelled well by TOMCATocs”. Isn't it in contradiction with the steeper gradient of TOMCATocs in the stratosphere mentioned above? – *Slightly rephrased to play down the performance. Line 492-493.*

Section 4.2 attributes model-observation mismatches to OCS sources or sinks, what about the potential mismatches from TOMCAT transport? – *This is mentioned in Section 4.3. Line 516.*

5. Discussion:

L376: “the TOMCATocs simulations of atmospheric OCS concentrations and the vegetative flux, which are dependent on one another in the model”. The simulated OCS concentrations are dependent on vegetation OCS fluxes that were transported, but it is not said which OCS concentrations are used to compute the vegetation OCS fluxes. – *OCS concentration from the model is used in the calculation of vegetative uptake. See Line 228.*

L385: “inverse modelling of OCS fluxes shows that some combination of a larger tropical oceanic source and vegetative sink resolves the budget”. Kooijmans et al. (2021) also show that considering a variable atmospheric OCS concentration reduces the vegetation sink in the tropics, meaning that a smaller tropical OCS source would be needed to close the budget. – *See above.*

L392: “such as calculating OCS uptake using a constant LRU value of 1.6 is not representative of reality”. It could be interesting to give the range of values proposed for LRU in the literature, to illustrate how LRU values can vary. – *Updated. Line 565-575.*

L394: “Our estimation of vegetative uptake in this work does not replicate OCS uptake universally and it is unclear if this is due to localised differences in LRU or on the GPP fields themselves”. Could you provide a global map of vegetation OCS uptake obtained with your approach using a constant LRU value and compare it to similar maps found in the literature (Berry et al., 2013; Kooijmans et al., 2021; Maignan et al., 2021) to analyse the spatial distribution of the fluxes? Could you also provide a map of TOMCAT simulated atmospheric CO2 concentrations used to compute vegetation OCS uptake? – *See the supplement for these additions.*

L396: “the distribution is based on work by Kettle et al. (2002) and has since been updated, for example by Ogée et al. (2016)”. It has also been updated by Sun et al. (2015). It could be interesting to compare the spatial distribution of your soil OCS fluxes to other maps based on the mechanistic approach from Ogée et al. (2016) (Kooijmans et al., 2021; Abadie et al., 2022). – *See the supplement for these additions.*

6. Conclusion:

L453: “Therefore, further study following on from this work will be to derive an a posteriori set of fluxes using an inversion scheme based on an up-to-date prior, and surface observations and a dataset containing vertical information near the surface”. Important drawbacks are acknowledged by the authors as this work does not rely on an optimization framework and uses out-of-date OCS fluxes. These drawbacks should explicitly appear in the abstract. – *Abstract has been mostly rewritten.*

Minor comments:

L19: “To compensate for this larger vegetative sink”. I would not use "larger" here as it has not been explained yet that it is larger than the vegetation OCS uptake from Kettle et al. (2022). – *Updated.*

L30: Is this really “Hawaiin” and not “Hawaian” (replace everywhere if needed)? – *Updated to Hawaiian everywhere.*

L56: Please replace “Vegetative uptake is the most important atmospheric sink of OCS” by “Vegetative uptake is the most important sink of atmospheric OCS”. – *Updated. Line 74.*

L107: Please develop the abbreviation HITRAN. – *Updated. Line 144-145.*

Table 1: Please replace “Barrow” by “Utqiagvik (formerly Barrow)” and “Cape Grim” by “Kennaook / Cape Grim”. For PSA and SPO stations, please precise “Antarctica (United States)”. – *Updated.*

L135: Should “surface emission fields” be replaced by "surface flux fields" as surface OCS fluxes are not only sources? – *Updated. Line 175.*

L136: Please replace “six sources and three sinks” by "six net sources and three net sinks" as soils can be both a sink or a source of OCS for example. Please also name the sinks and sources here. – *Updated*

L147: Remove "an" in this sentence “Eleven anthropogenic sources of OCS were an quantified by Zumkehr et al. (2018)”. – *Updated*

L175: Please develop the abbreviation WATCH. – *Updated*

L235: “so we only compare the main simulations”. Please precise that it is TOMCATocs. – *Updated*

L301: Please remove “Gg” in the following “from 17.7 Gg ppt to 3.4 ppt”. – *Updated*

Figure1: Please improve the resolution of the figure to be able to read the RMSE scores. – *Updated*

RC2 (from attached pdf document)

Section 1

L19: confusing, since you say "towards the lower end", so expect "smaller". – *Abstract has been mostly rewritten.*

L39: define lifetime: global burden/stratospheric loss or stratospheric burden/loss? – Updated. Line 48-50.

L55: confusing. "DMS accounts for OCS oceanic emissions". DMS is emitted and an uncertain fraction is oxidized to OCS. refer recent findings: Jernigan, C. M., Fite, C. H., Vereecken, L., Berkelhammer, M. B., Rollins, A. W., Rickly, P. S., Novelli, A., Taraborrelli, D., Holmes, C. D., & Bertram, T. H. (2022). Efficient production of carbonyl sulfide in the low-NO x oxidation of dimethyl sulfide . Geophysical Research Letters, x, 1–11. https://doi.org/10.1029/2021gl096838 - *Updated. Line 71-73.*

L66: recent update: @article{abadie2022global, title={Global modelling of soil carbonyl sulfide exchanges}, Kooijmans, L. M. J., Cho, A., Ma, J., Kaushik, A., Haynes, K. D., Baker, I., Luijkx, I. T., Groenink, M., Peters, W., Miller, J. B., Berry, J. A., Ogée, J., Meredith, L. K., Sun, W., Kohonen, K. M., Vesala, T., Mammarella, I., Chen, H., Spielmann, F. M., … Krol, M. (2021). Evaluation of carbonyl sulfide biosphere exchange in the Simple Biosphere Model (SiB4). Biogeosciences, 18(24), 6547–6565. https://doi.org/10.5194/bg-18-6547-2021- *Updated. Line 85-88.*

L84: I find this sentence strange. Normally one would use OCS vegetative uptake to estimate GPP.....so this becomes rather confusing. – *Removed this line. While we do clarify that the LRU approach does suitably estimate vegetative uptake, this had been shown previously.*

L92: are – *Updated.*

Section 2

Section 3

L153: do I miss the chemistry terms here (OH, and photolysis)? – *Line 166-170.*

L169: I think you have to be clear about units here. GPP and Fcos differ by orders of magnitude... – *Updated to be more clear on units. All appropriate scaling is performed in the calculation. Line 223-227.*

L178: I think Ma et al. showed that is even more important to account for OCS mixing ratios. Now it remains unclear how [OCS] is used to convert GPP to F_OCS – *We use OCS from the model at each timestep. The text has been updated to make this more clear. For example on Line 228.*

L194: fluxes?

L195: uptake?

L197: I understand, but should be something like: due to the resulting improvements ....

L206: ocean emissions

L219: I think OH loss is mostly tropospheric. – updated.

Table 2: I read the Ma et al. table well, this is the imbalance in their budget.... – *This has been updated in Table 2.*

L236: did not read that in section 2.1, nor in section 3.1 – updated.

Section 4

L237: This does not make sense. Why not compare both simulations? – As the budgets of the other model runs are negative, it would make correction throughout the entire atmosphere very challenging.

L243: mm, this was not stated in the method section. Actually, this belongs in the method section, and the 10 year initialization is mentioned in the TOMCAT_CON description. – Updated.

L248: please do not repeat the method section here. – Updated.

L250: What I no not understand why monthly means are compared, while ACE-FTS is co-sampled. I think co-sampling is important at the surface. Also, I find the metric max-min to estimate the monthly error in observations misleading. – Figure 1 updated to use standard deviation.

L272: This improvement is rather disappointing. This implies that budget terms and their seasonality are not OK yet? – *It is not an excellent improvement. However, when compared also to TOMCATSOTA, we see that it is a fairly good improvement. While TOMCAT$_{SOTA}$ does well in the NH in terms of SCA, it performs poorly at LEF and HFM.*

L279: Here it is really vital to investigate the modeled diurnal cycle. And during strong uptake OCS goes down, and the LRU formulation might break down. This is very handwaving argumentation. – *This sentence was removed. But our work does indeed suggest the LRU approach still underestimates at heavily vegetated regions.*

L282: Well, also handwaving: was JULES NEE validated with flux measurements at this location (of at Harvard Forest)? – *JULES GPP was validated against FLUXNET. The text has been updated accordingly. Line 405-407.*

L287: did you check this? Or is this speculation. Coudl anthropogenic emissions influence MHD is some month?

L289: The big question hanging here is: is this due to model shortcomings, or due to the applied COS fluxes. Given the fact that inverse models capture the seasonal cycle at MHD, you would be tempted to say that fluxes are not yet optimal, e.g. too high ocean emissions over the Atlantic... – *This is likely the cause and text has been updated. Line 415-419.*

L298: did you check (e.g. with tagged tracer run), or can you provide a reference. – *Updated as above.*

L301: ??

Figure 1: increase font size... AND What are the Blue error bars? CAPTION: is. – Figure 1 has been updated and the blue bars now represent standard deviation.

L337: Again: here you speculate. It would be good to perform tagged tracer runs to backup these statements. – This was not considered in the scope of corrections.

L342: I assume you mean something like the maximum concentration in the vertical.

L343: ?? If ACE connot measure, this does not make sense. – *Removed.*

L347: There is nothing between these two seaons? – *Updated Line 473.*

L359: There is abundant speculation in the paper. Some things really need to be checked better ....One thing is this "faster removal". What about reducing the photolysis rates to see whether the disagreement disappears? – *The author improves on argumentation and discussion. We include an additional model run to test the comments on photolysis loss in the stratosphere.*

L364: Not sure if this is the ACE uncertainty, since this is the variability in observations. Does ACE provide an observational error? – *Amended to be standard deviation.*

L366: see comment above

Section 5

L386: I think you overstate the quality of your simulations here. There are significant remaining deviations that apparently are resolved when emissions are optimized. Now the seasonal cycles are sifted and mostly underestimated (Figure 1). – *Amended the text to suggest that our flux also points towards a missing tropical source, like the inversion studies referenced. Less so that our model compares as well as theirs.*

L388: ???? – Removed.

L394: and sensitive to the unknown OCS mixing ratio at the uptake locations... – *Updated. Line 565-566.*

L397: see also new paper Abadie (2022) – *Updated. Line 570-571.*

L410: Again,, OH mostly acts in the troposphere. – *Updated this sentence. Line 539-541.*

L413: Again, a speculation that can be easily tested.. – *Removed this sentence.*

Section 6

L448: d – *Updated. Line 613.*