Author comment on "Constraining the budget of atmospheric carbonyl sulfide using a 3-D chemical transport model" by Michael P. Cartwright et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-215-AC1, 2022

Firstly, thank you to the anonymous referees for taking the time to evaluate this work.

Here we address all major or general comments made by the two referees.

Please find all responses to minor comments specific to the text in the supplementary document attached.

RC1

- The main concern from RC1 here is the insufficient use of recent references in the introductory material, as well as using primarily out-of-date fluxes and data in the model.

"References to recent studies are missing in the introduction." - We address these concerns by firstly updating the introduction to include a broader review of available literature to better represent recent scientific findings for OCS.

"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the conclusion that TOMCATocs gives better results than TOMCATcon does not seem relevant when TOMCATcon is based on out-of-date estimates (except for the anthropogenic emissions from Zumkehr et al., 2018). " - To address this concern (and others detailed in 1.) we include an additional model run for comparison with TOMCAT_{OCS}. This new model is called TOMCAT_{SOTA} and makes use of flux inventories developed between 2017 - 2022, including work by the following: Stinecipher et al. (2019), Zumkehr et al. (2018), Lennartz et al. (2017, 2020) and Maignan et al. (2021). See the text for more specific detail. This new model run provides an additional comparison for TOMCAT_{OCS}, one that is more relevant to recent findings and strengthens the conclusions made about the quality of TOMCAT_{OCS}.

"In Section 3.3, why choosing to use a single constant LRU value while several studies provide PFT-dependent LRU values?" - On a global scale it seems unlikely that a varying LRU would make a large difference, which is highlighted by Hilton et al. (2017): "the methodological simplification of treating LRU as a constant introduces considerably less error than GPP uncertainty (as estimated by inter-model GPP differences)". While this will be included in the scope of future work, the primary goal was to present a suitable model that compares well with measurements.
"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). " – Firstly, thank you for bringing these works to my attention, the method and discussion sections feature more references to recent literature in line with this request. However, from an actionable perspective this comment poses several challenges. Firstly, the use of CS2 is utilised purely due to its spatial distribution over the tropics from the Kettle et al. (2002) inventory – this is mentioned several times in the text. Secondly, while Kooijmans et al. (2021) does suggest soil uptake of 89 – 146 Gg S/y using SIB4, this paper was published in December 2021 during the final stages of preparing this manuscript. Furthermore, the discussion for Abadie et al. (2022) stared in November 2021, again during final preparations for this work and was actually published nearly 2 months following (11 May 2022) the initial submission of this work (18 March 2022). Finally, it is worth highlighting that the combined biosphere flux presented in this work, used in TOMCAT_{OCS}, is inline with inversion studies by Ma et al. (2021) and Remaud et al. (2022). “

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).” – Remaud et al. (2022) is now also included.

- The main concern from RC1 here is that optimisation of the fluxes would be better achieved using an inversion framework and that balancing the budget is unnecessary.

“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.” – This is a fair statement but does not factor in the accessibility to an inverse framework at the time of undertaking the research. Also, while a balanced budget is indeed under debate (less so in the past 5 or so years), it has been shown for the majority of the period between 2004 and 2018, that the OCS budget is indeed in balance or with a weak trend, depending on latitude.

“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.” – This is indeed a limitation of the method of scaling in this work and is acknowledged by the author(s). The compromise proposed here is to have included the third model run, as discussed in 1. This provides an additional comparison with up-to-date spatial distribution of fluxes, including maps presented in the supplementary material. The quality of the vertical comparison with ACE-FTS suggests the troposphere, surface and atmospheric fluxes are modelled well.

- The main concerns here were regarding the calculation of vegetative uptake of OCS, particularly CO2 and OCS concentration.

“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 modelled by JULES. Considering only the year 2010 does not reflect the yearly increase in atmospheric CO2 concentration and the fertilization effect.” It was assumed that including variability in CO2 concentration is likely to be a smaller source of error or variation in $F_{OCS}$ than other factors, such as GPP product and
OCS concentration. Like LRU this would be included in the future scope of this work.

“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.” – The calculation of vegetative uptake is actually calculated at each time-step in the model (every 6 hours). The text has been updated to make this clearer. But we do establish a first order relationship between vegetative uptake and OCS concentration.

- RC1 main points here are that the paper lacked direction and conclusion, and that some of the comparing TOMCAT\textsubscript{OCS} to TOMCAT\textsubscript{CON} was a challenge and a bit arbitrary.

“It is not clear what the goal of this study is, and the title is confusing.” – The goal of the study was to build a model that simulates OCS well compared to novel satellite observations and surface measurements. In turn to extract information about the implications of the OCS budget estimated here and how this compares with recent literature. TOMCAT\textsubscript{SOTA} helps provide a benchmark for recent work and how a collection of bottom-up estimates does not necessarily yield sensible seasonality at the surface. Hence, highlighting there is still substantial uncertainties in the overall OCS budget. This work agrees with the missing source originating from the tropics but falls short of concluding its exact origin. As mentioned in the text, future work will aim to use an inverse system in combination with data that offers vertical information, such as ACE-FTS, but preferably also in the lower troposphere, like NDACC. Connecting the troposphere to the surface could be the key to improving surface flux estimates.

We have recommended a change in title from:

Modelling atmospheric carbonyl sulfide using gross primary productivity to constrain vegetative uptake

to

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

“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?” – The quality of TOMCAT\textsubscript{OCS} comparison with ACE-FTS is highlighted more so in the revised manuscript, with an underlying message that a good comparison throughout most of the atmosphere suggests that the gradients of OCS are well represented by the model. Therefore, so are the surface and atmospheric fluxes driving it.

RC2

“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.” – The author has improved on this by either removing speculation or better comparing and referencing conclusions, particularly in the results and discussion sections.

“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.” – We include an additional model simulation for 2010 only that reduces atmospheric photolysis. The intention of this simulation is to test if this does resolve the issues in the stratosphere. What we find is while it does reduce negative bias in the Southern Hemisphere, it introduces model bias elsewhere, i.e. positive in the Northern Hemisphere tropics. So we draw the conclusion that the convection scheme or transport is causing the issue. An experiment with 0.75 photolysis rate is presented in the manuscript and an additional one with 0.5 photolysis is shown in the supplement.

“It remains unclear what is taken for the OCS mixing ratios here.” – As described in point 3 of RC1, we utilise the OCS concentration at each time-step in the model to calculate the vegetative flux. The model is initialised at the start of the spin-up period (1994) using 500 ppt however.

“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.” – The introductory material has been updated to include more recent literature. More frequent use of up-to-date references are used throughout the text.

Please also note the supplement to this comment: