

Dear reviewer,

We appreciate your effort and time on the reviewing work, especially during the epidemic time. We have copied the remarks from reviewer #2 as below, and put our response to the comments point by point. The text in blue color is from reviewer #2 and the black text is our response.

Anonymous Referee #2

We would like to thank reviewer #2 for their time in reviewing this manuscript.

Comments

The uptake of OCS is tied to the OCS concentration within the canopy. There are large variations in OCS uptake as OCS depleted air flows through vegetation, e.g. Berkelhammer et al. 2020 (<https://doi.org/10.1029/2019GL085652>). Most canopies do not see free troposphere concentrations of OCS. This will affect the plant uptake flux component substantially and should be addressed.

This comment was also made by reviewer #1. Because we use an inversion framework, we employ a zero-order flux approach, which is technically easy to implement. Indeed, we find adjustments that point to a large drawdown of COS in the canopy. Like we mention in the discussion, in analyzing the flux adjustment, we should be aware that part of the adjustment is due to the “concentration” effect. Based on the comments of both reviewers, we will make this issue clearer in the revised manuscript.

DMS should not be considered a major source of OCS. Many researchers still refer to the 7% yield figure from the Barnes 1996 paper, but note that chamber studies proceeded without NO_x and at high DMS concentrations. Subsequent studies demonstrated that an alternative chemical pathway is typically taken, and that changes in NO_x affect OCS formation greatly (e.g. [https://doi.org/10.1016/S1352-2310\(98\)00120-4](https://doi.org/10.1016/S1352-2310(98)00120-4)). In other words, if you would like to include DMS, it is important to also model NO_x. This is most likely such a small contribution that in the Whelan et al., 2018 synthesis, it was concluded that DMS should only be included as a source of uncertainty in the ocean flux rather than a source itself.

Currently we do not simulate NO_x chemistry in the COS inverse modelling, since this largely increases the complexity of the modelling system. However, DMS is normally emitted over remote oceans, where low NO_x concentrations prevail. We do not fully agree with the reviewer that the COS yield of 0.7% should not be included as long as the COS budget is not closed. Nevertheless, we will perform a NO-DMS inversion in which we place the DMS-related COS emissions to the “unknown” category.

If the lifetime of CS₂ was 12 days, it might make sense to model CS₂ separately, since the associated OCS will not show up in the air parcel until it has traveled nearly around the globe. However, more recent evidence suggested that the lifetime is much shorter than that. For example, see the 3D atmospheric transport study performed by Anwar Khan which focusses only on CS₂ and estimates a lifetime of less than 4 days at maximum: [10.3934/environsci.2017.3.484](https://doi.org/10.3934/environsci.2017.3.484).

We thank the reviewer to point out this publication. We have checked the rate constant used in our paper for CS₂+OH, which is $2 \times 10^{12} \text{ cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$ (line 214). Assuming that the OH average concentration is about $10^6 \text{ molecules cm}^{-3}$, we recalculated the lifetime as about 5.79 days. We cited the reference Khalil & Rasmussen (1984), but the rate constant is from another paper (Jones et al., 1983). The lifetime quoted in the paper will be corrected to ~6 days accordingly. Khan et al. (2017) presented a global modelling study of CS₂ and quote a lifetime of 2.8-3.4 days, based on a different evaluation of the CS₂+OH rate (Sander et al., 2006). This lifetime is still half of our lifetime, and because reviewer #1 also pointed to the novelty of including the CS₂ precursor, we will present a sensitivity study for the CS₂ lifetime in the revised manuscript.

The SiB3 model had a known phenology problem, where the growing season starts too soon (by a couple of weeks, perhaps) and ends too early. Does SiB4 have this issue? It would be good to check the seasonal timing by comparing to SiB3 or even another slightly complicating proxy, e.g. SIF.

Our biosphere fluxes are based on simulations with the Simple Biosphere model, version 4 (SiB4) (Haynes et al., 2019). New in SiB4 compared to SiB3 (used by Berry et al. (2013)) are capabilities to simulate i.e. carbon pools, land cover heterogeneity, and leaf phenology. Further, COS uptake formulations in SiB4 are the same as in Berry et al. (2013). One of the new features in SiB4 compared to SiB3 is that it includes prognostic phenology, and this phenology is no longer depending on satellite products. So, if SiB3 had a problem with the phenology this does not automatically mean that SiB4 has the same problem. In fact, from an ongoing comparison with field observations the phenology looks good in SiB4. Results will be presented in a manuscript that is currently in preparation by co-author Linda Kooijmans. Therefore, we consider further discussion of the prior SiB4 fluxes beyond the scope of this study.

The abstract should be revisited to better reflect the conclusions of the study and to tidy up the language, e.g. sources of OCS are obviously included in current budgets.

The abstract will be revised to better show the conclusions and the cohesion of language.

In short, revising the inclusion of DMS and addressing the first order plant uptake issue will certainly create a different budget overall, and may affect the conclusions.

We will discuss these issues in the revised manuscript. We argue, however, that a zero-order approach is sufficient for a first inverse modelling study, since the fluxes are allowed to adjust to the concentration effect. Concerning DMS, we consider this issue currently unresolved, and we will include a NO-DMS inversion.

References:

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