

We thank Dr. Krol for his timely review and helpful suggestions. I have incorporated the feedback into the manuscript text. The level of detail of the corrections is appreciated. Below are a few points of clarification.

For ease of reading the text has been formatted like so: Original Text, **Reviewer comment**, *Author response*, and Revised text.

**I miss in the introduction the “satellite” perspective and the total column measurements. In general, also its depletion in the stratosphere requires one or two lines (link to SSA).**

*This is a fair point. Early OCS studies were motivated by stratospheric sulfate questions. We now include two additional lines in the introduction:*

*“For decades, OCS has been a compound of interest as a source of sulfate to the stratosphere.” and “On larger spatial scales, many FTIR stations and 3 satellites currently in operation have recently been used to retrieve spectral signals for OCS in the atmosphere using new retrieval algorithms.” The section on OCS in the Atmosphere has been moved up to better frame the bottom up budget discussion, which includes the latest work on stratospheric OCS.*

**It would help here if you outline the structure of the paper. Since it is a review, this is particularly important. I think the focus is on the “non-closure” of the bottom-up OCS budget, and this should be articulated better in the introduction. If you state: “The ultimate goal of this research is to constrain our estimates of global carbon-climate feedbacks”, this does not cover this paper.**

*We thank the reviewer for pointing out this oversight. We now call out an explicit paper structure in the introduction.*

*“This review seeks to synthesize our collective understanding of atmospheric and biospheric OCS, to highlight the innovative new questions that these data will help answer, and to identify the outstanding knowledge gaps that will need to be addressed moving forward. First, we present what information is known from surface level studies. Then we develop a scaled up global OCS budget that suggests a considerable missing source of OCS to the atmosphere. We examine how the existing data has been applied to estimating GPP and other ecosystem parameters. Finally, we describe where data is available and submit a community research plan. The ultimate goal of this OCS tracer research is to constrain our estimates of global carbon-climate feedbacks.”*

**“...a regional scale modeling approach can avoid many of the within-canopy measurement issues. The carbon uptake by forest ecosystems is large and a crucial component in understanding future climate-carbon feedbacks...” without further elaboration, this remark seems out of scope...what is meant here? References?**

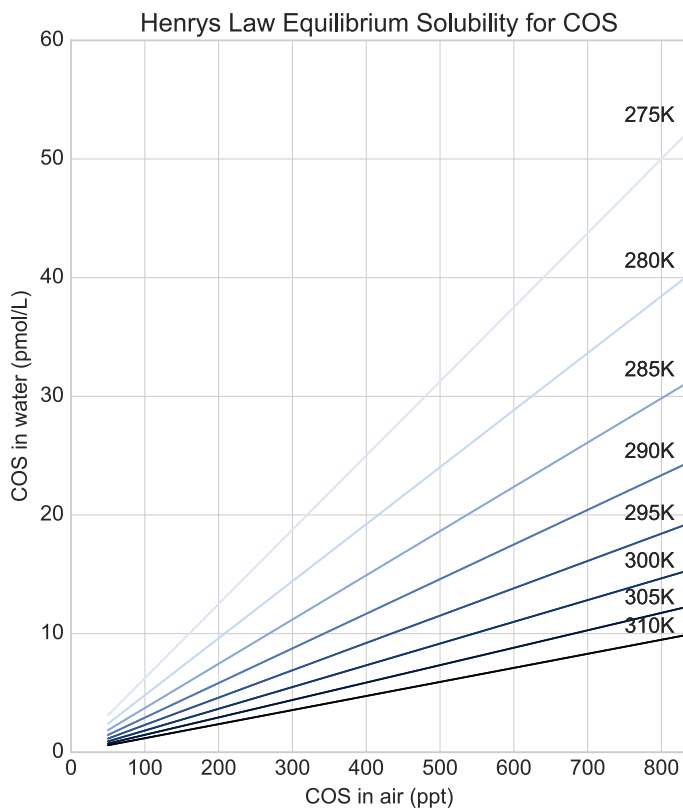
*This is a reference to several ongoing studies, most of which have not made it into the literature yet. However, the strongest evidence to support OCS for regional modeling studies that has been peer reviewed is work by Hilton et al. We’ve now changed the wording to explicitly call on Hilton’s work.*

*This now reads:*

“The application of the OCS tracer in tall canopies poses difficulties because turbulence can be limited in canopies and complicate many traditional methods of trace gas flux measurements (Blonquist et al., 2011; Kooijmans et al., 2017). A possible solution is using the “surface renewal” approach (Paw U. et al., 1995), but it has not been attempted for OCS. Regional scale modeling with OCS appears to sidestep site-level problems, as in Hilton et al., (2017). With more OCS observing towers upstream and downstream of large forested areas, OCS may be able to resolve the daily or weekly carbon uptake by forest ecosystems, a large and a crucial component in understanding future climate-carbon feedbacks.”

### what is the role of the OCS-solubility in water?

*This is mentioned briefly in the Abiotic processes section, however, a figure may be more useful to explain it.*



*Generally, there is less than 1 ppt of OCS dissolved in water. The degradation of OCS by hydrolysis is strongly temperature dependent and becomes relevant at time scales of hours and more. We now include the figures for OCS solubility and hydrolysis in water much earlier in the manuscript.*

*[Re: reported wetland values] so these numbers are substantially higher than uptake fluxes....but are they more wide-spread? I try to integrate to a “global” number.*

*We now include a range of values from the sparse data available. We integrate a global number later in the budget section. This is now included in the text.*

“Much early work on the subject used sulfur-free sweep air leading to biased results, and few soil-only field measurements with ambient air exist, e.g. Whelan et al., (2013) found fluxes ranging from 1 to 27 pmol m<sup>-2</sup> s<sup>-1</sup>. We integrate a “global” estimate of wetland fluxes in Sect. 2.7.”

*[Re: deviation from paragraph structure in soil section with Bunk et al., (2017)]* **here I am lost: I thought emissions were discussed here. I looked at the original manuscript and see that they performed fungi inhibition experiments. Maybe explain this better.**

*We now include discussion of this study both in the discussion of fungal OCS fluxes and OCS exchange by soil, instead of lumping both points into the same location.*

*[Re: upscaling in the Launois et al., (2015) study]:* **maybe explain a bit better what was done in this “upscaling” approach. ..clearly such an approach is very uncertain, so an error-range would be appropriate (101 GgS/yr sounds like an “accurate” estimate. We agree with the reviewer and now have related the Launois et al., (2015) results with more of the language of uncertainty:**

“The recent evidence of soil OCS emissions led Launois et al. (2015b) to include an emissions term in their soil flux estimates by upscaling biome-specific emissions. For global fluxes, Launois et al. (2015b) used ranges typically measured for anoxic soil emissions reported by Whelan et al. (2013), which had dependencies on temperature and flooded state. Using a map of estimated anoxic soils used for methane emission estimates (Wania et al., 2010), OCS production was assessed and allowed to vary by  $\pm 30\%$  in optimization. Typically, peatlands were probable net emitters of OCS, with a mean value of 25 pmol m<sup>-2</sup> s<sup>-1</sup>. Some ecosystems, such as rice paddies, shift from a net source to net sink depending on the flooding state of the soil. Because the reported range of fluxes was centered on 0, Launois et al. (2015b) considered these fields to have net emissions of zero. Peatlands are mainly located in the northernmost regions (above 60° N) and were expected to contribute about 101  $\pm 30\%$  Gg S y<sup>-1</sup> to total emissions, combining the wetland extend from Wania et al., (2010) and the estimated mean peatland flux. Seasonality was also indirectly included in soil fluxes because frozen soil was assumed to have a 0 net OCS flux. In the simulation, emission estimates dominated in some extratropical regions of the Northern Hemisphere, turning them into a net source of OCS in late autumn and winter.”

“Although different processes undoubtedly require further dedicated process studies, the total uncertainty range combining process parameterization and in situ observations remains lower than the current gap in the global OCS budget.”

**I read this as: the uncertainties combined cannot explain the gap in the OCS budget...and other terms have to be found. Is this a correct interpretation?**

*We have clarified the statement:* “Different processes undoubtedly require further dedicated process studies to assess their importance. Despite these unknowns, the current gap in the OCS budget is larger than the estimated ocean emissions, including uncertainties from process parameterization and in situ observations.”

*[Re: the section on volcanic emissions]: I find this section a bit disjunct from the rest of the paper. The relatively small amount of work that has considered volcanic OCS sources has reasonably been almost entirely absent from the discussion of OCS as a tracer for ecosystem processes. I hope this section seems out of place because the terminology and the approach are completely separate from any other component of the OCS budget, and this review will help bring this important body of work into the discussion.*

“It is obvious that the OCS budget needs more observations to support modeling efforts. There is a large missing source, thought to be in the oceans, but the available evidence also supports a larger anthropogenic source. Current leaf-based investigations need to be expanded to include water or nutrient-stressed plants. Despite the large uncertainties of the global OCS budget, many applications of the OCS tracer have been attempted with success.”

**This is strange, this means that the error bar on the anthropogenic OCS emissions is too small ....the same holds for the ocean.**

*This is an excellent point. When examining how uncertainty is estimated, it is difficult to tell how much uncertainty remains considering all of the observations that have not been made. For the oceans, any observations in the proposed OCS source region would be important to have. For the anthropogenic sources, better emissions factors and observations of industrial activities, particularly in Asia, are needed to get a better picture of the budget. Either way, it is not surprising that the measurement uncertainty does not capture the true variability. The text now reads:*

“Examining Table 4, we find a large missing source of at least 1200 Gg S y<sup>-1</sup>, up to 4100 Gg S y<sup>-1</sup>. It has been suggested that ocean OCS production has been underestimated (Berry et al., 2013), but some research points to unaccounted anthropogenic sources closing the budget gap (Zumkehr et al., 2017). This suggests that the uncertainty on our ocean OCS production and/or the industry inventories do not capture their true range of OCS fluxes. Additionally, there is a possibility that the plant sink is overestimated or that there is an unidentified source. More observations in the ocean OCS source region and from industrial processes, particularly in Asia, are needed to further assess their actual magnitude and variation. Current leaf-based investigations need to be expanded to include water or nutrient-stressed plants, perhaps revealing lower OCS plant uptake. Despite the large uncertainties of the global OCS budget, many applications of the OCS tracer have been attempted with success.”

*[Re: Section 3: Applications introduction]: line of the story becomes diffuse here....I would expect the disadvantages of both approaches. However, no distinction is made anymore...are these “general” issues, or specific for the “biosphere model” approach? Or are sections 3.1.1. making this separation? Some more lines outlining the general structure would be helpful.*

*This suggestion will greatly improve this section. We have re-arranged the text in the first and last paragraphs to add more structure to the overall section and address these concerns, e.g.*

“More precisely quantified OCS surface fluxes should improve our knowledge of terrestrial photosynthesis and hence GPP (Sandoval-Soto et al., 2005). Improved knowledge of the global OCS budget will help constrain OCS surface flux components (vegetation, soil, ocean, anthropogenic). Current OCS surface flux budgets are mostly bottom-up estimates derived from leaf-scale or regional-scale experiments (Berry et al., 2013; Campbell et al., 2008; Kettle et

al., 2002; Suntharalingam et al., 2008). In addition to a better global OCS budget, atmospheric OCS measurements can also be used to further constrain bottom-up or mechanistic estimates. Such “top-down” estimates use observed spatial and temporal gradients of OCS in the atmosphere to adjust an independent estimate of surface fluxes (usually called the “prior” estimate).”

*and*

“Among the four satellite OCS products, only TES OCS data have been used for OCS surface flux inversions. As the TES OCS product is limited to over ocean only, the inversion of the OCS terrestrial sinks in Kuai et al. (2015) may be subject to large uncertainties. Thus, for consistency, TES OCS over land may be highly desired. Spectral retrieval over land requires exact details of surface properties, including surface altitude, temperature, emissivity, reflectance, snow cover, etc., which have been considered in the IASI OCS retrieval. A similar retrieval algorithm for TES OCS is currently under development. The accuracy of the surface flux inversion can be further improved by using simultaneously more than one satellite OCS observation, e.g. TES and MIPAS, to provide more constraints on the horizontal and vertical OCS distribution in different parts of atmosphere. Satellite products need to be compared to tower or airborne data, perhaps determining how well the upper troposphere can reflect surface fluxes. This effort is furthered by better estimates of surface fluxes, in particular observations of OCS emissions from the oceans in areas where we assume a large source region might exist (Kuai et al., 2015). Thus better “bottom-up” surface flux estimates constrained by more numerous atmospheric observations can provide powerful constraints on OCS surface fluxes, and thus GPP.”

*[Re: OCS as a tracer for the origins of air masses]: I find the last paragraph a bit (too?) speculative. Why would one use OCS to answer meteorological questions? There are much easier ways.*

*This is referring to a few projects that are currently underway. A drop in OCS concentration will alert us to when a free tropospheric parcel of air has mixed with air from the planetary boundary layer (PBL). This is particularly useful when studying areas of convection, where many atmospheric models disagree. PBL dynamics are notoriously difficult to model, requiring radiosondes on weather balloons for ground truthing. OCS could be an easier and faster way (and less expensive, over the long term) of answering certain meteorological questions.*

We thank Dr. Krol again for a thorough and helpful review. The manuscript is undoubtedly improved and the intent is clearer.

Cheers,

Mary Whelan