We thank the reviewer#2 for the review that helps to improve our manuscript. In the following, we address the raised points directly, with the *review in italics* and *our reply in bold* font.

*I find it striking that such broad generalizations are made from data that cover a fairly small portion of the global ocean. The authors have a great deal of new observations to address the important issue of quantifying OCS fluxes from the ocean. But to draw broad conclusions without considering the previous data more carefully is inappropriate. Their data add to the picture in useful and interesting ways. To "conclusively answer the question of whether the missing OCS source... can really be ascribed to the direct OCS emissions from tropical oceans" would seem to require another level of effort that isn’t yet part of this manuscript.*

We agree that we need to add information on a comparison of our model simulation with previous cruises, which we now added in a new Table (Table 5) where we compare previous studies from various seasons and regions with our box model simulation (>4000 measurements). We discuss this in a new section “3.2.1 Comparison to previous ship-based measurements” (p.11). We find overall good agreement even though we compare monthly climatological means from the box model simulation with cruise data. This further increases the confidence in our model simulation.

In addition, we emphasize again that the box model had already been established previously and tested with data from several cruises and seasons (p. 6, l.22ff). We further improve the model by consolidating the aCDOM dependence of the photoproduction rate constant $p$ and constrain the model by important new data from tropical regions which had been missing previously.

See also our response to a similar comment by reviewer #1.

**New Tab. 5**

<table>
<thead>
<tr>
<th>References</th>
<th>Season</th>
<th>Region</th>
<th>Mean OCS±std. [pmol L⁻¹]</th>
<th>n</th>
<th>L2016 mean [pmol L⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mihalopoulos et al. (1992)</td>
<td>Mar/May 1986</td>
<td>open Indian Ocean (20°N-37°S)</td>
<td>19.9±0.5*</td>
<td>20</td>
<td>11.2±6.3</td>
</tr>
<tr>
<td></td>
<td>Jul 1987</td>
<td>OCEAT III</td>
<td>19.9±1.0*</td>
<td>14</td>
<td>17.7±13.1</td>
</tr>
<tr>
<td>Utheofer et al. (1995)</td>
<td>Apr/May 1992</td>
<td>North Atlantic Ocean (47°N 20°W)</td>
<td>14.9±6.9</td>
<td>118</td>
<td>42.8±11.3</td>
</tr>
<tr>
<td></td>
<td>Jan 1994</td>
<td>48.50°N, 10-17°W</td>
<td>5.3±1.6</td>
<td>120</td>
<td>8.9±3.2</td>
</tr>
<tr>
<td></td>
<td>Sep 1994</td>
<td>48.50°N, 10-17°W</td>
<td>19.0±8.3</td>
<td>33</td>
<td>33.4±3.5</td>
</tr>
<tr>
<td>Utheofer and Andreac (1998)</td>
<td>Mar 1995</td>
<td>West Atlantic (32°N, 64°W)</td>
<td>8.1±7.0</td>
<td>323</td>
<td>15.8</td>
</tr>
<tr>
<td>von Hobe et al. (2001)</td>
<td>Aug 1999</td>
<td>Sargasso Sea/BATS (32°N, 64°W)</td>
<td>8.6±2.8</td>
<td>518</td>
<td>8.1</td>
</tr>
<tr>
<td></td>
<td>May/June 1998</td>
<td>Atlantic meridional transect (53°N-34°S,25°W-20°E)</td>
<td>18.1±16.1</td>
<td>440</td>
<td>27.8±47.9</td>
</tr>
</tbody>
</table>
New section 3.2.1:

“The global simulation of OCS surface water concentrations generally reproduced the lower picomolar range of concentrations (Tab. 5), the seasonal pattern of higher concentrations during summer compared to winter (as e.g. in (Ulshöfer et al., 1995)) and the spatial pattern of higher concentrations in higher latitudes (e.g. Southern Ocean, (Staubes and Geogrii, 1993)). Given that monthly means of a model simulation driven by climatological data of the input parameters is compared to cruise measurements, the absolute mean error of 6.9 pmol L$^{-1}$ and the mean error of 3.7 pmol L$^{-1}$ indicate an overall good reproduction of observations (differences between observation and model output were weighted to number of observations in Tab. 5). It has to be noted that on average, the model overestimates OCS concentrations as indicated by the positive mean error, suggesting our emission estimate to be an upper limit to direct oceanic OCS emissions in most regions. Largest deviation from observations are found in the Southern Ocean (vgl. (Staubes and Geogrii, 1993) in Tab. 5), where the model underestimated observations by ~40%. While this can have several reasons, i.e. a possible violation of the underlying assumption of a constant OCS production in regions with deep mixed layers such as the Southern Ocean, or the missing satellite data for CDOM during polar nights, it is a clear indication of the need of more observations from high latitudes. However, this underestimation does not infer with our conclusion drawn for the tropical oceans, where the location of the missing source is derived from top-down approaches.”

New paragraph with detailed description of the evolution of the box model:

“... A box model to simulate surface concentration of OCS is further developed from the latest version from (von Hobe et al., 2003, termed vH2003), where concentrations along the cruistacks of 5 Atlantic cruises have been simulated and compared. The vH2003 model results from successful tests and validation to observations on several cruises to the Altantic Ocean covering all seasons (i.e. (Flöck and Andreae, 1996)in January 1994, (Uher and Andreae, 1997a)in April/May 1992, (Von Hobe et al., 1999)in June/July 1997, (Kettle et al., 2001)in September/October 1998). By comparing photoproduction rate constants of the 5 cruises to CDOM absorption, (von Hobe, 2003)suggests a second order process for photoproduction with the photoproduction rate constant being dependent on the absorption of CDOM in seawater.

In our approach, we test vH2003 along the cruise track of two cruises, include a new way of determining the photoproduction rate constant (see below) and apply it with global climatological input (termed L2016).

(Kettle, 2000; Kettle, 2002) applied a similar version of vH2003 globally, which included an optimized photoproduction constant from Atlantic transect cruise data, an optimized constant light-independant production and a linear regression to obtain CDOM from chlorophyll a. In comparison to K2000, we use (i) a new way of determining the photoproduction rate constant incorporating information from three ocean basins, (ii) the most recent parameterization of light-independent production available, and (iii) satellite observations for sea surface CDOM instead of an empiric relationship based on chlorophyll a.

Launois et al. (2015) implemented parameterizations for light-independant production, hydrolysis and air-sea exchange similar to vH2003 in the 3D global ocean model NEMO-PISCES. The main differences to the approach used here is the lack of accounting for mixing in L2016 (discussed in section 3.2.2., which will theoretically lead to higher simulated concentrations in our case) and the application of a photoproduction rate constant in our model that incorporates information from three open ocean basins in contrast to one from a study in the North Sea (Launois et al., 2015).”
I realize that the model helps the authors extrapolate their results to broader scales, but the results derived are only as good as the data considered by the model. If the authors really hope to be able to “constrain the variability of OCS emissions in the tropic(al ocean)” then I would think they would have to consider the details of previous ocean-going measurements (ocean basin, ocean regime, season, etc.) together with their new data to determine if, in fact, that most of the major global ocean regimes have been adequately sampled to allow such a conclusion.

Additional to considering previous measurements (see above), we also added a comparison of the water properties of our cruises and the assumed source region (Pacific Warm Pool, new Tab. 4). The properties encountered during our 2 cruises which we use to further improve and validate the box model (OASIS, ASTRA-OMZ) comprise the range of properties found in the Pacific Warm Pool except for SST and windspeed. We discuss this in p. 10, l. 20ff in the revised manuscript.

New Tab. 4:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Parameter</th>
<th>Parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>SST [°C]</td>
<td>27.0±1.0</td>
<td>19.6±2.6</td>
</tr>
<tr>
<td>SSS [g kg⁻¹]</td>
<td>35.0±0.3</td>
<td>35.1±0.3</td>
</tr>
<tr>
<td>wind speed [m s⁻¹]</td>
<td>8.2±1.7</td>
<td>7.5±1.8</td>
</tr>
<tr>
<td>aso [m⁻¹]</td>
<td>0.039±0.02</td>
<td>0.146±0.02</td>
</tr>
<tr>
<td>H [W m⁻²]</td>
<td>226.5±303.0</td>
<td>196.4±283.1</td>
</tr>
<tr>
<td>SR [J m⁻²]</td>
<td>1.9±0.1±1.7</td>
<td>1.6±0.4±5.1E⁶</td>
</tr>
<tr>
<td>pH [-]</td>
<td>8.0±0.01</td>
<td>---</td>
</tr>
<tr>
<td>MLD [m]</td>
<td>43.3±15.8</td>
<td>18.9±7.5</td>
</tr>
</tbody>
</table>

Discussion of Tab. 4 in the text:

p.10, l.20: “The water masses encountered during the cruises to the Indian Ocean (OASIS) and eastern Pacific (ASTRA-OMZ), which are used to constrain the global box model, differ considerably with respect to the properties relevant for OCS cycling and thus span a large range of possible OCS variability. The properties encountered during these two cruises comprise or exceed the ones of the Pacific warm pool (climatological averages, Tab. 4), which is where the missing source has been located (Glatthor et al., 2015; Kuai et al., 2015). Both higher SST and lower wind speeds (Tab. 5) would decrease the OCS sea surface concentrations in the ocean and thus decrease emissions to the atmosphere: higher SSTs favor a stronger degradation by hydrolysis (Elliott et al., 1989), and lower wind speeds decrease the transfer velocity k and thus the emissions to the atmosphere. Lower integrated daily radiation (SR in Tab. 4) in the Pacific warm pool also points to lower OCS production. Hence, our new OCS observations presented here likely span the range of emission variability in the tropics.”

Note that the box model has been used in a number of studies comparing observed and simulated OCS for individual cruises from different parts of the oceans and always yielded reasonable results.

For example, it is informative and important that their results are consistent with an upper flux limit from Kettle et al., 2002, but no mention is made of how that consistency actually increases our understanding of total OCS flux from the ocean. If the sampled regions were similar, then this is a confirmation of that original estimate, but potentially not much progress in understanding the broader-scale contribution of the ocean to atmospheric OCS abundances.

Kettle et al. 2002 built his global model on data from a transect through various regimes in the Atlantic, so our new data from the Indian, Atlantic (comparable) and East Pacific increase the data bases and model test cases for two new ocean basins (discussed on p.6 l.32ff in the revised manuscript).

“(Kettle, 2000; Kettle, 2002) applied a similar version of vH2003 globally, which included an optimized photoproduction constant from Atlantic transect cruise data, an optimized constant light-independent
production and a linear regression to obtain CDOM from chlorophyll a. In comparison to K2000, we use (i) a new way of determining the photoproduction rate constant incorporating information from three ocean basins, (ii) the most recent parameterization of light-independent production available, and (iii) satellite observations for sea surface CDOM instead of an empiric relationship based on chlorophyll a."

Suggestion: scale back the broad conclusions and focus on your results and how they compare with others, or consider the broader literature on OCS flux measurements (observation-based and model-derived) in a more detailed fashion to determine if the available data allow an accurate quantification of the global and, most importantly, tropical OCS flux from the ocean.

As described above, we considered the broader literature by (i) comparing our model simulations to 9 studies and (ii) describing more detailed in which regions the box model had been tested previously. With changing the title to “Direct Oceanic OCS emissions are unlikely to account for the missing source of atmospheric carbonyl sulfide” we acknowledge the higher uncertainty associated with the indirect emission estimates.

On uncertainties. Any comparison of derived oceanic fluxes with a shortfall needs to fully consider uncertainties. Many uncertainties are discussed (air-sea exchange, mixed layer depth, parameterization of production, etc.,) but aren’t explicitly included in the derivation of the direct flux of 130 GgS/yr and in the discussion of the global budget discrepancy (no uncertainty is provided on this number).

We restructured the results part and discuss uncertainties now in a new section “3.2.2 Uncertainties” (p. 12, l. 1ff). From our comparison to measurements from section 3.2.1, we use the mean error to derive an uncertainty for the global emission estimate, which results in an uncertainty of 50 Gg S per year.

p. 12, l.1: “Simulated concentrations and fluxes carry uncertainties from input parameters and process parameterizations. One major uncertainty associated with the mixed-layer box model approach arises from the fact that it does not adequately account for downward mixing and vertical concentration gradients within the mixed layer. Under most circumstances, and especially in the tropical open ocean, where hydrolysis greatly exceeds surface outgassing and low a350 makes photoproduction extend further down in the water column, the model tends to overestimate the real OCS concentrations, as was shown for our two cruises above. Therefore, we deem the fluxes from our global simulation to represent an upper limit of the true fluxes. Only at high latitudes would we expect more complex uncertainties, because hydrolysis at low temperatures is slow and only photoproduction and loss by outgassing are directly competing at the very surface. Other uncertainties are associated with the calculation of the photoproduction rate. The wavelength of 443nm combines the absorption of detritus and CDOM, which could have an impact especially in river plumes, where terrestrial material is advected into the ocean. As it is the CDOM that is important for photochemistry, assuming the 443nm is purely CDOM would lead to an overestimation of photoproduct and thus is a conservative estimate. It also has to be noted that a single spectral slope from 443nm to 350nm in the global simulation is a simplification. Furthermore, using a wavelength integrated photoproduction rate constant instead of a wavelength resolved approach, which would take global variations in the CDOM and light spectra into account, is an additional simplification. It has been shown that this does not lead to large differences regionally (von Hobe, 2003), but potentially could lead to variations globally. Our p-CDOM-relationship is a first step for constraining this variability globally in one parameterization, as it incorporates optimized photoproduction rate constants optimized to observations and thus accounting for differences in the light and CDOM spectra. More data from different regions can help to further constrain this relationship in future studies. However, despite these simplifications, the simulated concentrations agree very well with previous observations (n>4000, Tab. 5).
To test the sensitivity of our box model to the photoproduction rate constant, we performed a sensitivity test with a photoproduction increased by a factor of 5 in the tropical region 30°N-30°S, note that this factor is considerably larger than the uncertainty in the p-CDOM-relationship. This leads to an annual mean concentration of 35.1 pmol L⁻¹ in the tropics (30°N-30°S), resulting in tropical direct emissions of 160 Gg S as OCS per year. The efficient hydrolysis in warm tropical waters prevents OCS concentrations from accumulating despite the high photoproduction, and still results in emissions too low to account for the missing source.

With an mean error of 3.7 pmol L⁻¹ in the OCS surface water concentrations added to (subtracted from) the modelled concentration and subsequent calculation of fluxes using annual mean climatologies for wind, pressure and SST (same data sources as global simulation forcing data), we calculate an uncertainty of 60%, which translates into a total uncertainty of the integrated global flux of 80 Gg S yr⁻¹.

Similarly, the origin of stated uncertainties in the derivation of indirect fluxes from DMS are not discussed, but I would imagine are substantially larger then estimated. Without a fair treatment of these uncertainties, it isn't possible to gauge the true magnitude of the budget shortfall, which is a main point of the manuscript.

We agree that the uncertainty of the DMS conversion is large, which is why we took the highest published estimate of 0.7% OCS yield from DMS oxidation as a conservative estimate for remote, pristine atmospheric conditions. The emission pattern of DMS does not point to a specific tropical source. This implies that if emissions and subsequent oxidation of DMS was to account for the missing source, a conversion process specifically favored in the tropical troposphere is required. We now discuss the conversion factor of OCS to DMS in greater detail (p. 12, l. 31ff). Any additional investigation on the conversion factor is beyond the scope of this paper, as the spatial pattern of the missing source would then require an atmospheric, not an oceanic driver to produce a pattern with a hot spot of OCS production in the Pacific warm pool.

The revised section on OCS emission factors p. 12, l. 31:

“A significant contribution to the OCS budget in the atmosphere results from oceanic emissions of DMS and CS₂ that are partially converted to OCS on time scales of hours to days (Chin and Davis, 1993; Watts, 2000; Kettle, 2002). A yield of 0.7 % for OCS is used for the reaction of DMS with OH (Barnes et al., 1994), which results in a global oceanic source of DMS from OCS of 80 (65 - 110) Gg S yr⁻¹ based on the procedure described in section 2.5. The uncertainty range of 65-110 Gg S yr⁻¹ originated from the uncertainty in oceanic emissions, not the conversion factor. This conversion factor is much more uncertain, as the formation of OCS from DMS involves a complex multi-step reaction mechanism that is not fully understood. It has been shown in laboratory experiments, that the presence of NOx reduces the OCS yield considerably (Arsene et al., 2001), which would make our indirect emission estimate an upper limit. However, the yield was measured under laboratory conditions, and may be different and more variable under natural conditions. DMS emissions do not show a pronounced hot spot in the Pacific warm pool region, but as DMS transports much more sulfur across the air-sea interface than OCS, even low changes in the OCS yield could affect the atmospheric budget of OCS. As the spatial oceanic emission pattern of DMS does not reflect the spatial pattern of the assumed missing source, a locally specific tropospheric change in the conversion yield would be one potential way of bringing the patterns in agreement. While it is possible that the OCS yield could vary under certain conditions, e.g. it cannot be excluded that the low OH concentrations in the broader Pacific warm pool area as suggested by Rex et al. (2014) influence the yield, the (local) increase of the conversion factor would need to be in the order of a factor of 10-100.”

Details: P1, line 7-8. It only has implications for GPP derived from OCS observations on certain scales, not all.

Now changed
p.2, line 25-26. It also makes much more chemical sense given our understanding of how COS interacts with carbonic anhydrase in leaf waters.

We added this point on p. 2, l. 25ff: “This revision of vegetation uptake was suggested to (i) take into account the different deposition velocities of CO₂ and OCS within the leaf and base it on GPP instead of net primary production (Sandoval-Soto et al., 2005) as well as (ii) to better reproduce observed seasonality of OCS mixing ratios in several atmospheric models (Berry et al., 2013; Suntharalingam et al., 2008; Glatthor et al., 2015; Kuai et al., 2015).”

Table 1 and p. 9 line 27-30. The paper from Suntharalingam does not state or suggest that the missing source is oceanic.

This is correct, so we took the citation out of the text on p. 9. We still leave it in Tab. 1, as it is one of the missing source estimates. The caption of Tab. 1 specifies that if the missing source is assigned to the tropical oceans (which other studies do), then this would result in the total ocean flux listed.

p.3 line 13. Underrepresentation of global flux is also true for the measurements made here, despite the use of a box model for extrapolation.

This is true theoretically, but the difference in photoproduction constants between the open Atlantic ocean and the North Sea has been compared specifically in Uher and Andreae (1997a). We thus adjusted the sentence to: p. 3, l. 25: “However, the oceanic OCS photoproduction in the ocean model included a parameterization for OCS photoproduction derived from an experiment in the North Sea (Uher and Andreae, 1997b), which might not be representative for the global ocean, as indicated by an order of magnitude lower photoproduction constants in the Atlantic ocean compared to the German Bight (Uher and Andreae, 1997a).”

Figure 3, and line 10, p. 9. Is R = 0.7 or R² = 0.7 in this relationship? These different values seem given for one relationship. Also, given the non-normal distribution of these results this value is not as significant as one might presume.

Because the data is not normally distributed, we used Spearman’s rank correlation coefficient r, which is correctly displayed in the figure, and now also corrected in the text. By relating the photoproduction rate constant to a350, we establish a semi-empirical relationship, which is a reasonable and globally available, but not perfect proxy (i.e. CDOM is only an approximation for the precursor).

The conclusion section is a bit unusual in that it includes speculations not supported by the work. It also reads like a research planning document. On what basis do the authors conclude (p. 11, line 22) that observations could be reproduced “without increasing the vegetation sink”? An extensive body of recent literature has suggested that the interaction between OCS and vegetation is best explained by a substantially larger sink than discussed in earlier papers; to discount those studies without substantial evidence is inappropriate, making this conclusion one that does not follow from the evidence presented in the paper.

In the last chapter we meant to conclude our results and give a systematic outlook on further research. We changed the section title to “Conclusion and Outlook”. Nevertheless, we still want to discuss possible ways of reducing uncertainties in the emission estimates and closing the budget. Suggestions that could be misinterpreted as discounting previous research have been deleted.
References


Uher, G., and Andreae, M. O.: The diel cycle of carbonyl sulfide in marine surface waters: Field study results and a simple model, Atmospheric Geochemistry, 2, 313-344, 1997a.


