

Dear Cedric,

Thank you for your detailed comment to our manuscript. I will address your points in the following and am pleased to answer any remaining questions:

Dear Sinikka,

I read your paper with great interest and I have a few comments that relate primarily to the photoproduction part of your work. Note I generally support the reviewers' comments on better acknowledging and including uncertainties associated with your estimates. This is especially important because the paper's conclusions contradict the main findings of other published work.

Thank you for bringing up the point of better acknowledging the uncertainty in the photoproduction again, which has been addressed in the reply of the reviewers in the revised manuscript. While we contradict the conclusion of Launois et al. (2015), i.e. that direct emissions could account for the missing source, we do not contradict the many oceanic measurements that had been performed in various open ocean regions in different ocean basins and across different seasons. We hope that this point is clarified in the revised manuscript where we added a table (Tab. 5) to compare to >4000 previous measurements.

In particular:

1. The photoproduction rate calculations deserve to be explained in more details, especially when it comes to the global calculations. In particular, are the rates reported integrated over the mixed layer (as is suggested by equation 2) or are they an average for the mixed layer, or at a discrete depth (as would be suggested by the pmol L⁻¹ h⁻¹ units reported on figure S5)? The caption mentions production rates but that's it. If the rates are integrated over the mixed layer then the units of photoproduction rates should be in units of pmol per unit time and unit AREA (not per unit VOLUME). It would be good to clarify this because it would help facilitate comparisons with other approaches.

Note I attached (see supplement) a number of figures of calculated rates using the Fichot and Miller (2010) model implemented with the single wavelength-resolved apparent quantum yield of Weiss et al. (1995) for comparison purposes and to illustrate the differences in photochemical rates calculated at different depths or for different depth ranges (mixed layer, and sunlit layer).

Thank you very much for providing the plots to illustrate the point of the differences in photochemical rates for different depths. The model in our study treats the mixed layer as one single box and integrates the photoproduction rate constant over this respective depth, as described in equation 2. Thanks for pointing us to the mistake in the caption, the photoproduction rates illustrated in S-Fig. 5 is the average for the mixed layer, which will be corrected in the revised manuscript.

2. The attempt to constrain the variability in the photochemical rate constant p is welcomed, but it should also be recognized in the paper that using wavelength-independent p for the global calculations can also lead to significant and potentially large uncertainties in the calculated rate. In the von Hobe (2013) paper, it was suggested this had a little impact, but I believe the comparison using wavelength-independent and wavelength-resolved constants was made at the same location where there should be minor differences in the spectral characteristics of the UV downwelling irradiance spectrum. For global calculations, this simplification can be much more problematic because the spectral characteristics of the downwelling irradiance can vary quite dramatically with latitude or with atmospheric conditions. This is potentially a large source of uncertainty that would be good to discuss in the paper.

We will highlight the difference between using a wavelength integrated p and a wavelength resolved p in the revised manuscript in the section about uncertainties. A dependence of the photoproduction rate constant on CDOM makes sense from a chemical point of view as described in von Hobe et al. (2003), but the exact shape of this relationship remained to be defined. Our

attempt to constrain a global relationship includes photoproduction constants that account for these differences as they are optimized in an inverse set-up of the model. With data from different oceanic regions across three basins, we suggest this relationship as a first step to account for this variability. As is shown by our comparison to measurements from previous cruises (new table 5), this simplification simulates OCS concentrations in a range similar to previous measurements despite the uncertainties mentioned.

To test the sensitivity for the photoproduction rate constant in relation to the missing source, we performed a simulation with an arbitrarily increased photoproduction by a factor of 5 in the tropics. This factor is much larger than the uncertainty in our p-CDOM-relationship. This simulation yields tropical (30°N-30°S) concentrations of 35.1 pmol/L and an integrated flux of 160 Gg S yr⁻¹ from this area. The strong hydrolysis in warm tropical waters prevents the OCS from accumulating and results in tropical emissions too low to account for the missing source.

We added the following paragraph to the revised manuscript, p. 12, l. 21ff.:

“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.”

3. It should also be clearly acknowledged that the modeled downwelling (or downward) irradiance (as described in the paper) are crude approximations and do not even include the effects of clouds for example. This is another important source of uncertainty.

For the case studies during the cruises, we used measured radiation which includes the effect of clouds and the absorption by gases/aerosols in the atmosphere, as it was measured at the surface directly on the ship. For the global simulation, the effect of clouds and other absorption is indirectly considered. We use Era Interim data, where measured data is assimilated (Dee et al., 2011). Monthly climatologies of sunshine duration and daily incoming radiation, which include the effects of clouds and atmospheric absorption of aerosols and gases (Dee et al., 2011), are then used to fit mean monthly diurnal cycles of radiation. While the fitted diel cycle is always parabolic, the integral of the daily incoming radiation includes the effect of any attenuation in the atmosphere. The main uncertainty lies in the conversion to the UV light, which is counterbalanced by the photoproduction rate constant p that is optimized to this specific way of treating radiation in the model, across several regions and latitudes (tropical Indian Ocean, tropical Peruvian upwelling, Northern Atlantic ocean from the von Hobe et al. (2003) study). The fact that the model simulates concentrations of OCS in line with previous observations supports the validity of these simplifications.

4. It should also be better mentioned that there are uncertainties in the a_{350} retrieved from satellite data and that uncertainty will be compounded when p is derived from a_{350} . There is also uncertainty due to the fact that the GSM model used to retrieve the absorption by detritus+CDOM at 443 nm and not just CDOM. Finally, another uncertainty is associated with the use of a single spectral slope for CDOM to derive a_{350} from a_{443} . All these combined uncertainties can amount to a large error in the estimates of OCS photoproduction alone, and I think there is a need to better acknowledge all sources of uncertainty and their implications on the conclusions, and there is a need for a more cautious language with regards to the conclusions of this work.

We agree that pointing out these uncertainties is important and add a more detailed discussion on this in the new section “3.2.2. Uncertainties”. However, assuming $a_{443\text{nm}}$ is only CDOM would lead to an overestimation and thus a conservative estimate in our case. As our comparison of the

resulting global simulation with measurements shows, our simulated concentrations agree very well with previous and independent observations. A large error through these combined uncertainties thus seems unlikely. Given that the direct emissions are a factor of ~5 lower than what would be needed to account for the missing source, and that the previous measurements support our simulated concentrations, these uncertainties do not interfere with the main conclusion.

I also would like to take the opportunity to use the attached figures of COS photoproduction rates integrated over the mixed layer to highlight that the regions studied in your paper are generally not as photochemically active (generally lower photoproduction rates) as the region studied in Mihalopoulos et al. (1992). This is in relation to the comment posted by Dr. Belviso.

Thank you for providing these plots of your model calculations which shows the photoproduction rates if a globally constant, i.e. not CDOM dependent, photoproduction rate constant (or quantum yield), is assumed. This is different to our approach, where the amount of CDOM has a double effect on the photoproduction rate and, thus, shows a different spatial pattern, with hot spots in regions of high CDOM, i.e. in the Peruvian upwelling where we carried out our second study. The chemical background for a CDOM dependent photoproduction rate constant is described in detail in von Hobe et al. (2003). Still, including these two regions together with the photoproduction rate constants from the Atlantic region into one global p-CDOM-relationship yielded simulated oceanic concentrations of OCS that are in line with observations across different ocean basins and latitudes (Tab. 6 in revised manuscript).