

Referee #1:

We thank referee 1 for the helpful and constructive comments and have implemented the following changes in the revised version of the manuscript (our answers in bold font).

Page 2, line 10: There is good reason to think atmospheric OCS has not been stable for the past four decades. The available firn air measurements suggest an OCS decline in the atmosphere through the 1980's and 1990's (Montzka et al., 2004). Many of the data sets in this compilation are from the 80's and 90's. The impacts will likely be limited to the interpretation of the atmospheric data and the atmospheric change is on the order of 10%. This is not a science paper and the possible standardization and calibration issues between labs are probably on the same order, so this is not a major. However, a brief cautionary note to the potential end users of the data product is necessary.

We agree with the reviewer that we should mention reported decadal trends in the introduction. We have thus changed the following:

p. 2, line 10:

Atmospheric mixing ratios of OCS show larger annual than interannual variations (Montzka et al., 2007). Small negative trends between 10-16% decrease derived from firn air and flask measurements have been reported for the 1980 to 2000 period (Montzka et al., 2004). Since 2001, small positive trends <10% per decade were derived from OCS observations in the Southern hemisphere (Kremser et al., 2016).

And we added to the discussion about atmospheric OCS measurements:

It should also be noted that small decadal trends as reported in the introduction could influence the reported differences, as the measurements reported here span a period of 1982-2018. Also, possible standardization and calibration issues could potentially be larger than the range of reported trends, so using the dataset in new trend studies should only be done with caution.

Page 3, line5: Would be nice to add a column to Table 2 or 3 indicating the presence/or not of the met or other physical data.

We agree that this is helpful and have added a new Table 4, also in combination with the comment by referee #2 about additional trace gas measurements.

Page 4, line 8: In the Excel and text files that I downloaded, I see only NaN notation. Are these for missing or N/A data? Please include explanation in the manuscript.

Both -999 and NaN values are reported, but have different meanings. The -999 are present only for some rare occasions where "below detection limit" has been reported for the COS/CS2 concentration data (we double checked the submitted data file). However, we agree that a clarification for the NaN values (which means no reported data in the additional data section) is required. We have thus added:

Missing additional data (physicochemical parameters, meteorological parameters etc.) were filled with NaN (not a number), to facilitate readability in data handling software.

Page 4, line 21: It should be "e.g. with liquid N2" because the cryogen is not always liq. N2.

We changed this as suggested.

Page 6, line 23: From what I see in Fig. 3e, about half the data have longer than 4 hr resolution and therefore not appropriate for diurnal work based on the criteria used in this manuscript.

We have double checked the data, and come to the same result. This is also the reason why we highlighted the need for higher resolved data in the recommendations (section 4).

Page 6: Carbonyl sulfide in seawater: It is interesting that concentrations are higher in the Southern Hemisphere (Fig. 3a). Is this expected from a mechanistic perspective? Can it be because a larger fraction of the SH measurements is from regions of high OCS production? I'm also puzzled by Fig. 3c, in terms of two NH mins and when they appear.

The higher concentrations in the Southern Hemisphere are most likely a result of the sampling locations, as the reviewer already suspected. In the Southern Ocean, high concentrations of CDOM and low temperatures lead to high OCS concentrations in the water. Measurements in a comparable region in the Northern hemisphere show a similar trend (e.g. Cutter et al., 1999), and high concentrations are also expected in the Arctic Ocean. From a mechanistic point of view, there is also less ozone in the southern hemisphere's stratosphere (McKenzie et al., 2003), which would mean a higher UV concentration at the surface, which in turn would lead to a higher OCS production and concentration in seawater.

Concerning the seasonal variation: The Northern Hemisphere dataset contains data from a large variety of regions and biogeochemical regimes; ranging from shelf (North Sea, Atlantic shelf) to open ocean locations. The spatial variation seems to dominate this dataset, and we expect that the seasonal variation will become more pronounced if more data across different biogeochemical regimes become available to average out the effect of location.

Page 7, line 4: The figure reference should be to Fig. 3c.

We changed this as suggested.

Page 7, line 9: Better to be more specific and say "seasonal variability" instead of "temporal variability" here, I think. There are a few more instances in the paper where the same change would work better.

We changed this as suggested in several places in the manuscript.

Page 9: Carbon disulfide in the marine boundary layer: There are data from the north Pacific too (Fig. 2). Anyway, a 40+ ppt average in the boundary layer sounds pretty extreme for CS₂. The global coverage is pretty poor as pointed out in the manuscript. It would be useful to put this number in context with data from flask networks that see marine influence if possible.

Indeed, atmospheric mixing ratios of CS₂ of more than 40 ppt are very high compared to free tropospheric values as e.g. reported from flights during the GTE/CITE3 campaigns (Cooper and Saltzman, 1993), where maximum values reached 3 ppt in the boundary layer (Fig. 8 in Cooper and Saltzman, 1993). However, continental air seems to carry higher mixing ratios of CS₂, as e.g. summarized for several locations in Khan et al. (2017), Fig. 5 therein. We cannot completely rule out a problem of local contamination in the measurement systems as the source for high CS₂ values (which would have to be occurring independently in different measurement systems), but we suspect that the influence of land air masses may have had an influence on the measured air sample over the ocean, which can carry up to 1200 ppt CS₂. We have added a reference to these studies as suggested in order to put the numbers into perspective:

The reported CS₂ concentrations are generally higher than those reported from airborne measurements in previous studies, where values <10 ppt in the boundary layer have been reported (Cooper and Saltzman, 1993). An influence of continental air carrying a higher concentration of CS₂ might be a possible explanation for elevated values (see e.g. compilation in Khan et al. (2017) of up to 1200 ppt). The short atmospheric lifetime of CS₂ sets a limit to long range transport, so this explanation would only hold for coastal and shelf regions. The data reported here have undergone

calibration procedures as reported in the original manuscripts and elevated values are consistent across different labs and locations, so contamination problem of the local measurement systems are unlikely but cannot be ruled out completely to be responsible for the elevated mixing ratios.

Recommendations: It would be nice if the authors also comment on priorities (location wise) going forward with the ocean going OCS and CS₂ measurements. Where do we need the measurements most in terms of improving the ocean OCS source? For example, there are no data from the Arctic Ocean, but there is also precious little from the vast southern Pacific.

We agree with the reviewer and have added the following to the section 4 Recommendations:

In order enable the identification of large scale patterns and the quantification of the oceanic source strength, we identify locations for future measurements. For OCS seawater concentration, large gaps exist in the open Pacific Ocean and the Arctic Ocean. The Arctic Ocean would be especially interesting due to the unique composition of dissolved organic matter derived from river input, which could influence OCS production in the water. Marine boundary layer OCS is required especially from the Arctic Ocean. The data coverage for CS₂ is very scarce, but especially measurements in water and marine boundary layer from high latitudes (Southern Ocean and Arctic Ocean) as well as Indian Ocean and Southern Pacific would be helpful. Generally, for both gases, water concentration profiles would be helpful to understand their processes in the subsurface. This is important for CS₂ which has a long lifetime in water, so that mixing processes could bring subsurface CS₂ in contact with the atmosphere. Similarly, repeated measurements from the same locations would be helpful to decipher any trends.

Table 2: Cruise ID#27 should include CS₂.

We have added the information as suggested.

Fig. 1: Hard to match the lines representing the cruises to what is in the legend because a lot of the detail gets lost. The points are too small to be seen easily.

We have added labels to the lines in the plot and adjusted the caption accordingly.

Fig. 3: Check b) and d) labels.

We have adjusted the labels.

Fig. 6: When I look at the cruise ID#7 that has both ocean and air side CS₂ measurements in Fig. 1, I see a cruise that starts in Europe and ends in South America. However, I do not see boundary layer data in Fig. 2d from this cruise, although the cruise is listed in Fig. 6b

This is because the data were not fully georeferenced (no longitude given), as it is digitalised from a pdf document where no longitude was provided. The cruise track was provided as a separate figure in the original publication, and was digitalised to show the rough locations of the measurements in Fig. 1. The measurements (digitalised from a measurement vs. latitude plot) is thus included in the calculations, but not in the map plot (note that caption indicates that only georeferenced data were shown).

Referee #2

We thank Róisín Commane for her helpful comments that help to improve the manuscript. Please find our answers below.

For the atmospheric MBL data, how often are there other trace gases available to characterize the air mass type? This is something I think could be a suggestion for future data collection requests (along with CDOM - rather than Chlorophyll a). Newer/Laser-based measurement systems often also measure CO₂ and CO, which could be used to identify MBL data with recent continental influence/pollution vs cleaner air (CO lifetime about a month in the remote MBL). I know most of this information is not available for previously obtained data but it would be great to include in the wish list and include as a potential category within the database. This would allow for proper interpretation of data from top-down atmospheric inversion studies (rather than just the marine/oceanic community).

We have added a new table 4 (also for any additional data available) and have indicated additional data as reported in the publications. Including this data in the database would be beyond the scope of this collection, but we agree that it is useful to provide the information that additional data exists. We have also included the following in section 4 Recommendations, in order to raise awareness for the advantage of additional trace gas measurements:

“In order to decipher the history of the air mass and identify potential continental influence, it would also be helpful to measure additional trace gases such as CO or other anthropogenic tracers simultaneously.”

Fig 2: Only a few points (potentially in anthropogenic outflow in Europe) mean that it is hard to see more subtle changes in the true MBL observations. Could you replot with the OCS MBL limit as 450-600 ppt (which would still be a reasonable range)?

We have replotted the figure as suggested.

Table 2: Adding a column to indicate when ancillary data is available would be quite useful here. I would also recommend changing the D = details designation when you do A-O in the grouping column. Another option would be to skip A, D and O in the grouping column since you use it for digitized/original and A: analysis.

We have added the suggested column as a separate table (Table 4). In addition, we have changed the abbreviations as to avoid any confusion: digitalized: di instead of d, original: or instead of o, analysis: An instead of A, details: Det instead of D.

Fig 3: Labels are a bit off in the pdf I have here. (b) not there, (d) up high. Check it for the final version. Please add a color bar for the red/green in the middle panel. It will have a better impact than looking through the text for it. What do the plots look like if you separate the tropics from the NH/SW? Is the seasonal cycle of the OCS in NH and SW outside the tropics the same or offset?

We have adjusted the labels in Figure 3 and have also added a legend (also for Figure 5 for CS₂). The first row of panels in Fig. 3 and 5 now shows the latitudinal bands separated for the Northern and Southern hemisphere. Concerning the temporal resolution it is difficult to compare the seasonal variation in the temperate latitudes to the tropics, because splitting up the dataset in even smaller parts leads to even less data in each bin, so that it is difficult to decipher reliable trends. We have plotted a separate seasonal cycle for the Northern hemisphere >23° and the tropics -23° to 23° to compare the variation (see below). However, we believe that there is too little data available, so that the influence of the location (shelf/open ocean) for the individual datasets might obscure any trend. We will thus not include this figure in the main paper.

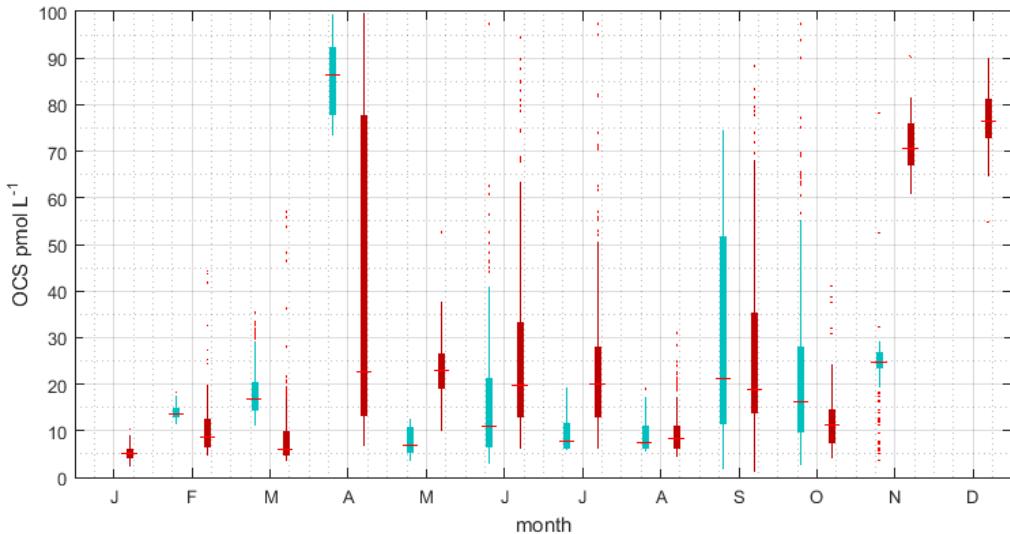


Fig. 1: Seasonal variation of OCS seawater concentration in northern hemisphere temperate latitudes (red, $>23^{\circ}\text{N}$) and tropics (blue, 23°S - 23°N).

Seasonal vs temporal variability: Usually temporal variability is used to described days to weeks and anything more uses seasonal. I agree with the other reviewer on this point and recommend changing the various instances of that.

We have made adjustments as suggested in the text.

References:

Cooper, D. J., and Saltzman, E. S.: Measurements of atmospheric dimethylsulfide, hydrogen sulfide, and carbon disulfide during gte/cite 3, *Journal of Geophysical Research: Atmospheres*, 98, 23397-23409, 1993.

Cutter, G. A., Walsh, R. S., & de Echols, C. S. (1999). Production and speciation of hydrogen sulfide in surface waters of the high latitude North Atlantic Ocean. *Deep Sea Research Part II: Topical Studies in Oceanography*, 46(5), 991-1010.

Khan, A., Razis, B., Gillespie, S., Percival, C., and Shallcross, D.: Global analysis of carbon disulfide (cs 2) using the 3-d chemistry transport model stochem, *AIMS ENVIRONMENTAL SCIENCE*, 4, 484-501, 2017.

Kremser, S., Jones, N. B., Palm, M., Lejeune, B., Wang, Y., Smale, D., and Deutscher, N. M.: Positive trends in southern hemisphere carbonyl sulfide, *Geophysical Research Letters*, 42, 9473-9480, 10.1002/2015gl065879, 2015.

McKenzie, R. L., Björn, L. O., Bais, A., & Ilyasd, M. (2003). Changes in biologically active ultraviolet radiation reaching the Earth's surface. *Photochemical & Photobiological Sciences*, 2(1), 5-15.

Montzka, S. A.: A 350-year atmospheric history for carbonyl sulfide inferred from antarctic firn air and air trapped in ice, *Journal of Geophysical Research*, 109, 10.1029/2004jd004686, 2004.

Montzka, S. A., Calvert, P., Hall, B. D., Elkins, J. W., Conway, T. J., Tans, P. P., and Sweeney, C.: On the global distribution, seasonality, and budget of atmospheric carbonyl sulfide (cos) and some similarities to co2, *Journal of Geophysical Research*, 112, 10.1029/2006jd007665, 2007.