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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Oceanic emissions unlikely to account for the missing source of atmospheric carbonyl sulfide" *by* Sinikka T. Lennartz et al.

Anonymous Referee #1

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Overview:

The authors present new bottom-up measurements and analysis of COS and CS2 from 3 ocean cruises. The ocean source is a dominant source of uncertainty in global COS budgets so the authors should be commended for presenting new, high quality data. However, the central conclusion in the manuscript title and text is not supported by the measurements. Nevertheless, the measurements and analysis provide a very important contribution to understanding COS budgets and I suggest only simple, but critical, revisions to the title and text.

Specific Comments:

The title and several statements in the text should be changed so that the conclusions become consistent with the data. In particular, the measurements are not a representative sample for extrapolating to the global source and thus conclusions on the global source should not be made. There are of course many other exciting conclusions that are possible. The measurements are not representative of the global source for following reason. Global satellite observations show global hot spot for the source in the Pacific Warm Pool for most of the year and in June/July/Aug more broadly across the tropical and mid-latitude Pacific. The cruise measurements from this study that are used in the global extrapolation do not cover this critical region. If the author's were looking to uncover information on the missing source they should target locations/times that top-down data points to for the missing source. However, the cruise data presented here are in times and locations were the top-down data suggest that the ocean source should be small or even a sink. I still think the global analysis is useful to include because it is already done and likely points to the problem with scaling up from non-representative data.

Robust conclusions for this study could instead focus on describing the ocean source for the times/locations of the three cruises shown in Figure 1. A qualitative comparison could also be made with previous top-down analysis. This seems to be good ground for an exciting conclusion of consistency between top-down and bottom-up estimates. In this case there appear to be some strong similarities between the bottom -up and top-down estimates. The TranPEGASO cruise covers a section of the Atlantic in Oct/Nov, showing a small source. This is qualitatively consistent with the MIPAS data along the same path in Sept/Oct/Nov. The Oasis cruise covers a small area in the central Indian Ocean in July/Aug showing a sink. This is roughly consistent with a MIPAS Jun/Jul/Aug map and a TES June map that show this region to be on the border between a source and sink. ASTRA-OMZ show a strong source in October for the Peru-Chile upwelling region. MIPAS Sept/Oct/Nov doe not show this. However, MIPAS is an upper troposphere estimate so it is not expected to provide a close relationship to surface fluxes in regions without strong atmospheric convection such as the Peru-Chile upwelling re-

gion. TES provides a lower altitude sensitivity and could provide a better top-down on small regions of sources such as the Peru-Chile upwelling regions. While TES data have only been published for June, TES retrievals for other months are in preparation.

Several revisions are needed in the introduction. Page 2 indicates that top-down studies were not consistent with the Kettle bottom-up estimates for sources and sinks. This should be corrected to say that the bottom-up and top-down info does not agree with Kettle. Kettle was a misinterpretation of the bottomup information from plant studies which was first pointed out in the bottom-up study of Sandoval-Soto et al. and then later confrimed by multiple topdown studies (Campbell et al, Sunthralingam et al, Berry et al, etc.) and other bottomup studies using chamber (Stimler et al) and canopy (Asaf et al, Maseyk et al) approaches.

The top down evidence from the global scale should be better specified. First it should be clear that there are four independent lines of five independent lines of evidence that point to a tropical source: MIPAS satellite (Glatthor et al), TES satellite (Kuai et al), FTIR (Wang et al, ACP, 2016... this ref isn't in the manuscript but might be added), NOAA and HIPPO observations (Berry et al, Kuai et al, Suntharlingam et al).

A critical point should be raised to alert the reader to alternative explanations for the top-down trends. In particular, the MIPAS remote sensing data is the upper troposphere ($\sim$10km) and transport from Asia to the upper troposphere in the deep tropics (e.g. Ashfold et al ACP 2015). Recent anthropogenic emission estimates for Asia are not yet sufficient to explain the missing source but they are based on very little bottom-up data from Asia (Campbell et al 2015). Other hypotheses could be mentioned as well such as a soil source which has been shown in a recent survey of global soils but not particularly large in the tropics (Whelan et al ACP 2016). Biomass burning is another but the most recent review of emission factors shows a relatively small source (Campbell et al 2015).

The Van Hobe study was included but more could be done to explain what other cruise

data is available. The introduction needs to explain how the cruise measurements and ocean box modeling fit within the context of previous cruise measurement and ocean box modeling. Were these cruises in seasons or locations that have others have not gone?

The introduction or discussion could also compare the modeling approach here to what has been done previously. In particular the recent paper by Launois et al.

Some comments may be helpful on alternative approaches for validating these flux estimates. Spatial gradients in atmospheric mixing ratios have been used recently (Berkelhammer et al below). Are other approaches also possible? M. Berkelhammer, H.C. Steen-Larsen, A. Cosgrove, A. Peters, R. Johnson, M. Hayden and S.A. Montzka (Minor Revisions, July 2016) Radiation and atmospheric circulation controls on carbonyl sulfide concentrations in the marine boundary layer. Journal of Geophysical Research (available upon request).

Section 2.3 should describe how the box model relates to the measurements. This is done in the results section "Following an earlier study (von Hobe et al., 2003), we use our observations ..." but belongs in the methods. A few additional sentences of explanation may be helpful. Why was the parameter p chosen for fitting the model as opposed to the numerous other parameters. Were other parameters also examined? If not then perhaps this should be stated as an important next step for future work. Why was the von Hobe et al., 2003 study used but not other studies? What is the spatial and temporal extent of the Von Hobe data?

"global radiation I was" not sure what "I" is

page 6, explain what you mean by "case study simulations"

define "CTD profiles"

The methods section should also include a summary of the time and location of the 3 cruises.

The section "2.1 Measurement set-up for trace gases" present a different method for each cruise. It would be helpful if this section also summarized the impact of having different methods on the different cruises in terms of different precision and other factors that may or may not influence the quality of these measurements.

Table 3 missing kmˆ-2 in TransPEGASO flux

"an non-negligible" to "a"

Some description is needed of the error associated with assuming a constant atmospheric mixing ratio on TransPEGASO. Seasonal and spatial variation in atmospheric mixing ratios can be on the order of 100 ppt.

"leaving the missing source still explained" should be "unexplained"?

Again this is an important contribution of new, high quality data and a well written manuscript. The authors present a compelling approach and with further data could provide a key to closing the global COS budget.

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