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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inverse modelling of carbonyl sulfide: implementation, evaluation and implications for the global budget" *by* Jin Ma et al.

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In this study, the authors present a global source/sink inversion for atmospheric OCS. The study is particularly notable for using a 4DVar approach with OCS, implementing the indirect sources of OCS as separate tracers, and applying a broad suite of observations as constraints, and considering alternative state variables for the inversion. This study advances the understanding of OCS sources and sinks using large-scale data and provides a foundation for critical next steps to implement a first-order sink inversion with satellite data.

Major Comments

To what extent can you discuss and analyze the importance of implementing CS2 as a

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separate tracer? Previous previous studies assume CS2 emissions convert on emission and only have a single OCS tracer. I wonder if one region this may be important is the Asian anthropogenic outflow. In previous studies without the separate tracers, the high mixing ratios should be more immediately over Asia while in your TM5 simulations the highest mixing ratios should be somewhat downstream of the Asian source. Perhaps a feature like this can be seen in TES data. It may be relevant to draw comparisons to related studies with CO2 such as: Suntharalingam, P., Randerson, J. T., Krakauer, N., Logan, J. A. and Jacob, D. J.: Influence of reduced carbon emissions and oxidation on the distribution of atmospheric CO2: Implications for inversion analyses, Global Biogeochem. Cycles, 19, GB4003, 2005.

In the abstract, the authors find that the missing source shows little inter-annual variation but large seasonal variation. While figure 6e provides some information on this seasonality, further plots and discussion would be helpful to explore this seasonality. Consider adding maps to the supplement of the optimized fluxes from 6e for 4 seasons and time series of regional averages.

The abstract notes that an overestimated sink cannot be ruled out but the manuscript notes that tropical land constraints are not available. This note about missing tropical continental boundary layer data should be added to the abstract for clarity.

Can the authors reconsider this statement about the overestimate sink by further use of the MIPAS data? I don't think you need another inversion run but just a few plots to compare the S1 and S3 runs to the geographic variability in MIPAS. Note that the MIPAS data clearly show global minimums in the convective outflow of the Amazon? I don't think Fig10 is sufficient to explore the MIPAS constraint because the critical dimension is longitude. In the MIPAS tropics there is high mixing ratios in the western tropical Pacific and low mixing ratios over Amazon/Congo. Maps of TM5 at high altitude along with maps of MIPAS are needed to test the validity of the large changes in the biosphere flux from inversions S1 and S3.

Section 2.2.3 could use some additional explanation with respect to: “The SiB4 model was constrained by a prescribed COS mole fraction of 500 pmol mol−1 outside of canopy.” This 500 pmol mol-1 is a placeholder. The actual boundary layer mixing ratio is lower and this SiB flux is best implemented in a model using a first-order dependency on ambient levels. Thus the 1053 GgS/y is likely an overestimate which is consistent with the lower flux reported in Berry et al of 738 GgS/y in which the first-order relationship is used. I think its fine that this study uses a zero-order approach but I think this should be carefully distinguished in the methods from future work that will need to implement the first-order approach. Furthermore it could be noted later in the abstract when the correction is made to obtain 851 GgS/y that this result is closer to the Berry et al result of 738 GgS/y.

Minor Comments

Maybe adjust the abstract wording to slightly improve clarity that plant sink in the TM5 runs are zero order (not first order). For example, “We finally find that the biosphere flux dependency on surface COS mole fraction (which was not modeled in this study) may substantially. . .”

The introduction notes Suntharalingam et al. (2008) study which attempted to fit background data by increasing the plant sink but you may also want to reference the Campbell et al. (2008) finding that this upward revision could be validated using direct observations from the continental boundary layer from the intensive INTEX-NA airborne campaign.

Campbell, J. Elliott, et al. "Photosynthetic control of atmospheric carbonyl sulfide during the growing season." Science 322.5904 (2008): 1085-1088.

The authors site Lennartz et al. (2017) for the bottom-up ocean emissions but please also note the upward revision in Lennartz et al. (2019).

Lennartz, S. T., von Hobe, M., Booge, D., Bittig, H. C., Fischer, T., Gonçalves-Araujo,

R., Ksionzek, K. B., Koch, B. P., Bracher, A., Röttgers, R., Quack, B., and Marandino, C. A.: The influence of dissolved organic matter on the marine production of carbonyl sulfide (OCS) and carbon disulfide (CS2) in the Peruvian upwelling, Ocean Sci., 15, 1071–1090, https://doi.org/10.5194/os-15-1071-2019, 2019.

Section 2.1.1 should note the lack of observatories in the tropical continental boundary layer.

Regarding section 2.2.1, how did you divide the anthropogenic emissions into direct COS and indirect CS2? Did the Zumkehr data file present emissions separately for direct and indirect? If they didn't then how did you back this out? You may want to look at the emission inventory in Campbell et al (2015) which does present separate emission estimates for direct COS and indirect CS2. The Zumkehr approach was an extension of Campbell et al 2015.

Campbell, JE, Whelan, ME, Seibt, U, Smith, SJ, Berry, JA, and Hilton, TW (2015), Atmospheric carbonyl sulfide sources from anthropogenic activity: Implications for carbon cycle constraints. Geophys. Res. Lett., 42, 3004– 3010. doi: 10.1002/2015GL063445.

Section 2.2.1 discussed uncertainties in molar yield. It might be worth noting that the uncertainty in the anthropogenic inventory is much larger than the uncertainty in molar yield.

Section 2.2.2 might want to draw comparison of this studies results to previous estimates from biomass burning in Campbell et al (2015) and open burning in Stinecipher et al. (2019).

Regarding the poor posterior fit at NWR and THD, are there references in the CO2 inversion literature that had the same difficulty? These sites are designed to capture background mixing ratio but sometimes they suffer from local influence which would be one reason for the poor posterior fit. One helpful paper you may want to reference is Riley et al.

Riley, W. J., Randerson, J. T., Foster, P. N., and Lueker, T. J. (2005), Influence of terrestrial ecosystems and topography on coastal CO2 measurements: A case study at Trinidad Head, California, J. Geophys. Res., 110, G01005, doi:10.1029/2004JG000007.

Line 421: “However, observations clearly show a large drawdown of COS near the surface (Hilton et al., 2017; Spielmann et al., 2020).” You may want to reference the INTEX-NA data which is an intensive sampling of vertical profiles in the continental boundary layer (Campbell et al.,2008)

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