Dear reviewer,

We sincerely thank you for the reviewing work and appreciate your effort to read and evaluate our study, especially during the epidemic time. We have copied the comments of reviewers below, and respond to the comments point by point. The text in blue color is from reviewer and the black text is our response.

J. Elliott Campbell (Referee)

We would like to thank Professor Campbell for his time and effort to read our paper and evaluate our research.

Major Comments

To what extent can you discuss and analyze the importance of implementing CS2 as a separate tracer? 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.

The inclusion of CS2 in our model gives the possibility to validate the geophysical distribution and vertical profiles in the future. It is also worth to note that CS2 has a different source distribution compared to COS. E.g., CS2 direct emissions are mainly from the industrial production of rayon. Another issue is the atmospheric lifetime of CS2. As referee #2 also discussed, the lifetime of CS2 is more likely shorter than what we used in the current implementation (Kahn et al., 2017). It is more likely less than 4 days, and we will test the sensitivity of the CS2 lifetime in TM5. In the revised manuscript, we will include CS2 sensitivity tests by varying the lifetime from ~6 days (current setting), to 3 days (Kahn et al., 2017). Another sensitivity test that we will report on is the instantaneous transfer of CS2 to COS, and compare it with the current model settings.

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.

Thanks for the suggestion. In order to better investigate the inter-annual variations of the missing source, we have done further time-series analysis for regions. Globally, the COS unknown posterior flux is shown, together with the trend, the seasonal signal and the residual (Figure 1). It can be seen that, globally, the seasonal signal is highest during NH spring and lowest during NH fall. The global flux was then split into 8 regions (Figure 2), and the regional COS unknown flux analyzed for these regions is shown in Figure 3. The region NH1 (North America plus part of Pacific and Atlantic Oceans, orange) shows a negative “unknown” flux, indicating that more sinks are needed. This likely points to an underestimation of the biosphere

uptake in the prior, since this region (that is well constrained by observations) depicts a clear seasonal cycle in the optimized "unknown" flux. NH2 (Europe, green) and NH3 (Asia, red) have almost the same trend and seasonality, perhaps because they are difficult to separate using the available observations. Tropical regions TR0-3 have similar trend and seasonality, and generally shows a positive flux signal, with little seasonal cycle. This could represent and oceanic signal (underestimated emissions of COS or COS precursors in the prior), a signal from biomass burning, or an overestimated biosphere sink. The ocean-dominated region SH (blue) has a near neutral flux, with a seasonal cycle that shows higher emissions in local fall and early winter.

We will add some of these analyses in the supplement or in the main text of the revised manuscript.

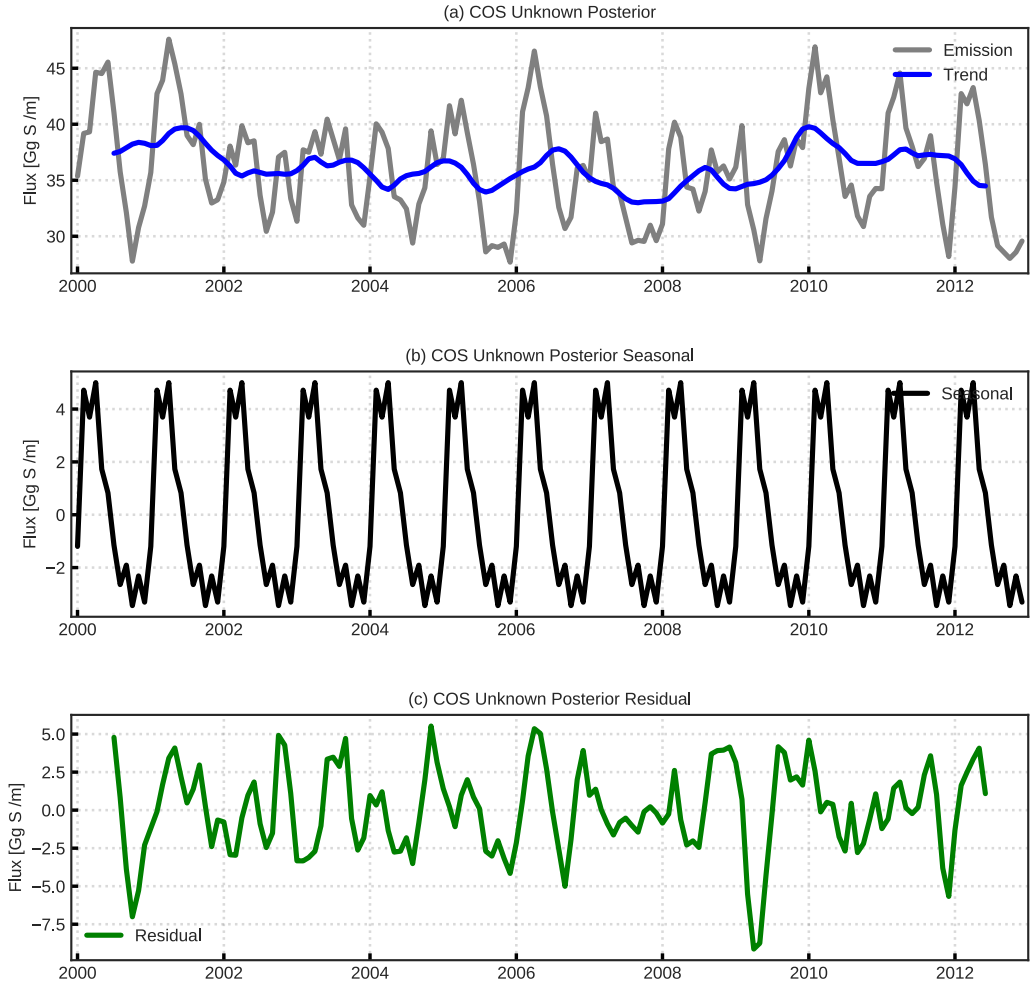

Figure 1. COS posterior flux of the optimized category "unknown" in inversion Su, with its seasonal signal and residual for the years 2000—2012.

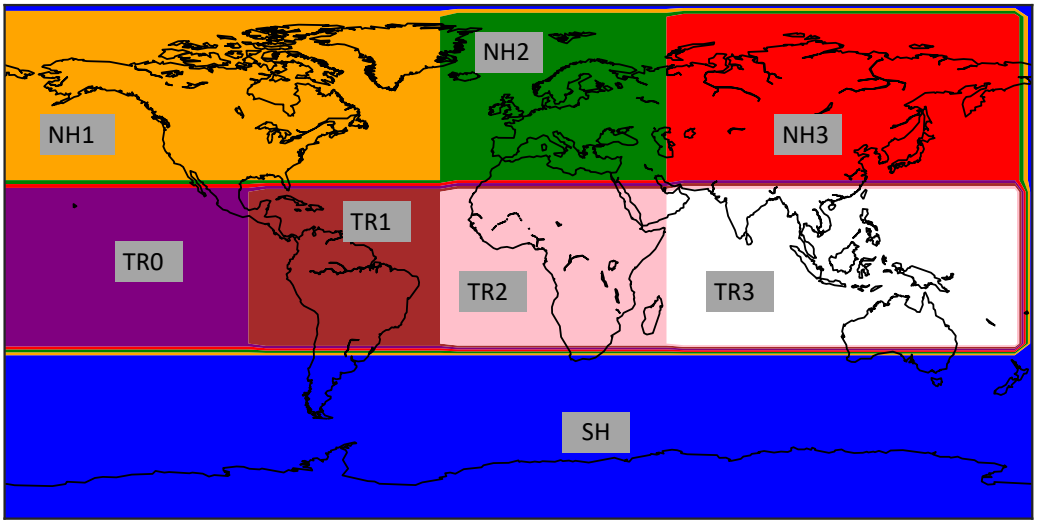

Figure 2. The regional map for analysis of COS fluxes. NH1-3 are areas in Northern Hemisphere. TR0-3 are regions in Tropics. SH refers to the Southern Hemisphere.

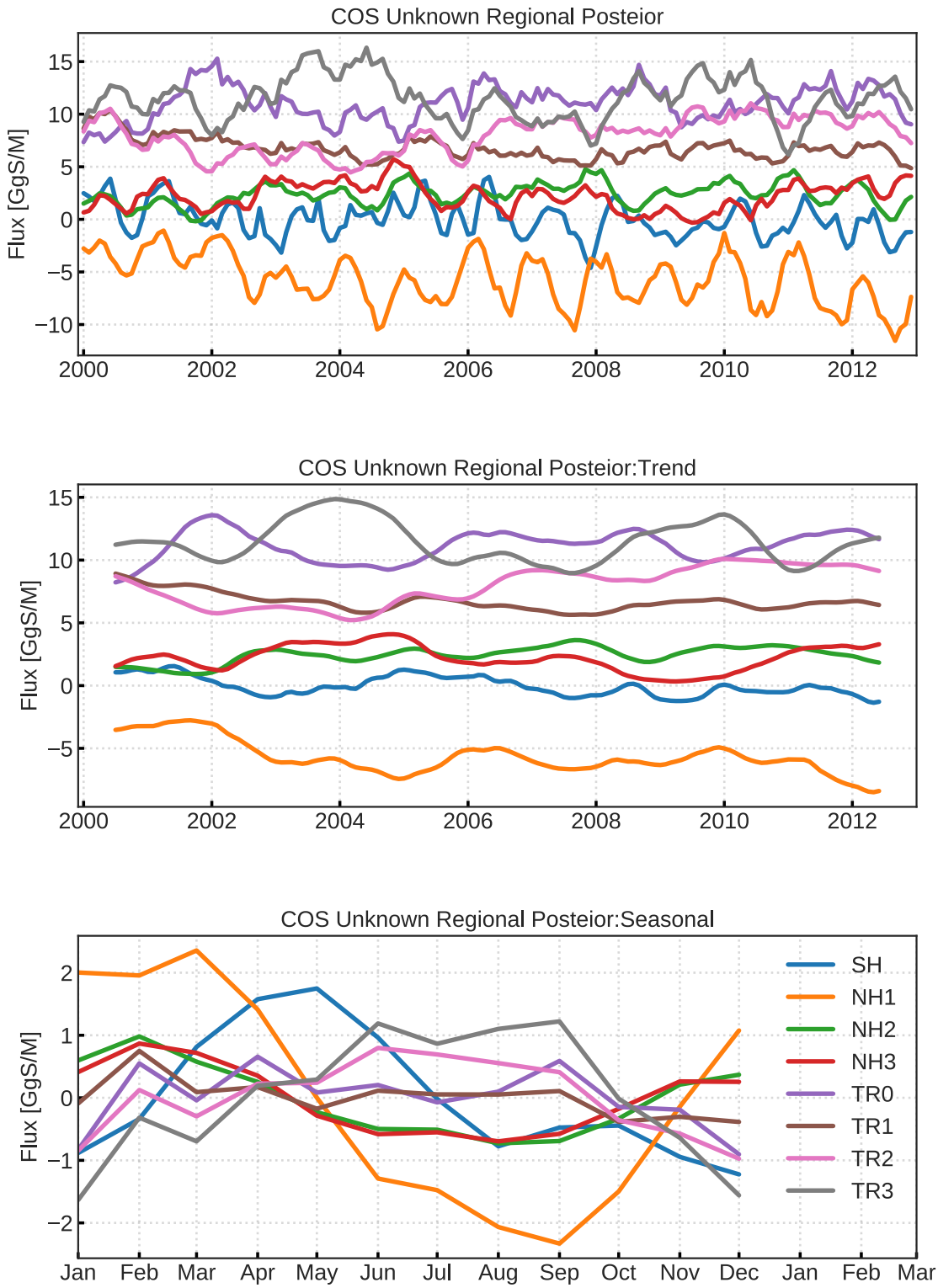

Figure 3. The regional signals of the optimized COS posterior flux (inversion Su). The upper panel shows flux contribution in each region, and the middle panel shows the trend by filtering out the seasonal and residual signals. The lower panel is the mean seasonal cycle of the COS posterior flux in each region. The colors of the lines are the same as in Figure 2.

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.

We will add this message to the abstract.

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.

This is a good suggestion. We indeed have the feeling that inversion S3 projects the total missing source on the biosphere, and this should impact the longitudinal MIPAS comparison. We will consider some extra figures in the Supplement.

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.

We realize that the SIB4 model uses 500 ppt merely as a placeholder, leading to a large uptake. We agree that our current manuscript presents this too much as a deficiency of the SIB4 model, and will adjust this in the discussion, section 2.2.3, and the abstract. The preferred (but more difficult) implementation of a first-order removal in the 4DVAR framework will be addressed in a future study.

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. . ."

This will be fixed in the manuscript.

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.

We thank the reviewer to point out the publication. This will be included in the manuscript.

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.

We thank the reviewer to point out the publication, which will be included in the revised manuscript.

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.

We implemented anthropogenic emissions largely based on Zumhehr et al. (2018) and Campbell et al 2015. We separated the COS emission from the Zumkehr et al. (2018) work according to Table 1 in Lee and Brimblecombe (2016). In Table 1 the authors reported a detailed emission budget for COS and CS2 in anthropogenic categories. Then we used the ratio of this budget to roughly estimate the direct and indirect COS anthropogenic emissions. In this way we were able to separate the COS and CS2 direct anthropogenic emissions, which should be correct within the uncertainties. We will outline the followed procedure more clearly in the revised manuscript.

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.

We will clarify this better in the revised manuscript. With more measurements available, we could try to reduce these uncertainties using our inverse modelling framework.

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).

We will mention a few differences in the revised manuscript. The major difference is that we considered new biofuel emission factors in South Asia.

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.

We thank the reviewer to point out the CO2 paper concerning THD. It is true that THD as discussed in the paper of Riley et al. (2005) is more affected by the local coastal effect and the diurnal cycle of CO2 fluxes. The current TM5 inverse modelling effort applies a coarse resolution of $6^{\circ} \times 4^{\circ}$globally, and thus the effect of costal meteorology at THD is not well captured. Another point is that biosphere flux of COS is applied on a monthly-average basis, and does not account for a diurnal cycle. The NWR station is probably also affected by local land effects that are not well resolved in the coarse simulations.

To compare with CO2 inversions, we have investigated CarbonTracker North America data and found similarities between COS and CO2 inversions. For example, at THD, CO2 has relatively large model-data mismatches of ~11 ppm in Summer (Figure 4). For NWR, the CO2 model-data mispatch is about 3.37 ppm in Summer (Figure 5). In comparison, at MHD the CO2 model-data mismatch is only 2.04 ppm in Summer inferred from Flask observations. Note that CarbonTracker North America employs a resolution of $1^{\circ} \times 1^{\circ}$ degree, compared to $6^{\circ} \times 4^{\circ}$degree in our COS study.

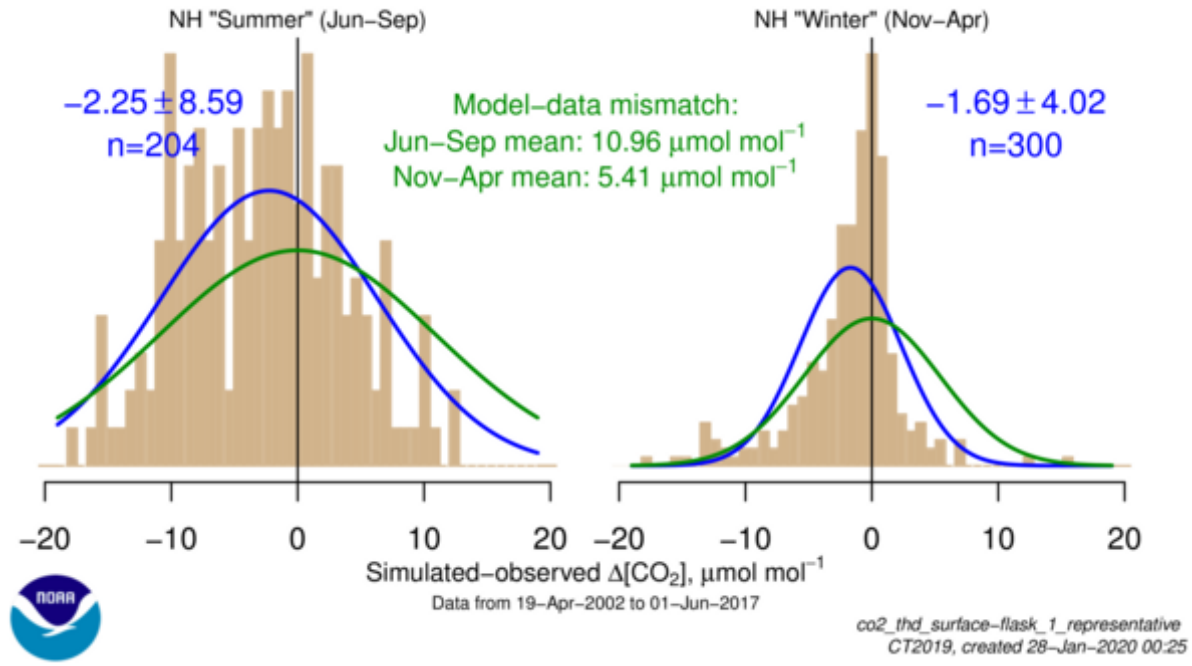

Figure 4. CO2 observation and simulation by CarbonTracker at THD (source: https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/co2tser.php?ds=co2_thd_surface-flask_1_representative&ed=assim&lastds=co2_nwr_surface-flask_1_representative).

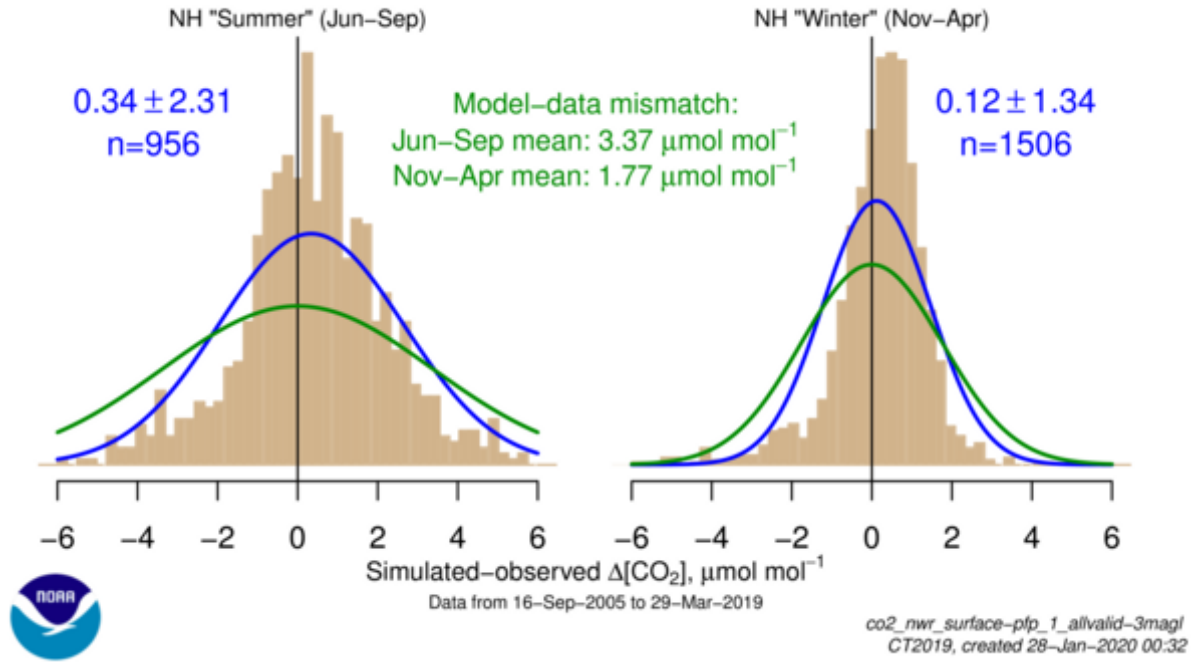

Figure 5. CO2 observation and simulation by CarbonTracker at NWR (source: https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/co2tser.php?ds=co2_nwr_surface-pfp_1_allvalid-3magl&ed=assim&lastds=co2_nwr_surface-insitu_3_nonlocal).

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)

We will include the reference.

References:

1. Khan, A., Razis, B., Gillespie, S., Percival, C., & Shallcross, D. (2017). Global analysis of carbon disulfide (CS2) using the 3-D chemistry transport model STOCHEM. Aims Environ. Sci, 4, 484-501.
2. Zumkehr, A., Hilton, T. W., Whelan, M., Smith, S., Kuai, L., Worden, J., & Campbell, J. E. (2018). Global gridded anthropogenic emissions inventory of carbonyl sulfide. Atmospheric Environment, 183, 11-19.
3. Lee, C. L., & Brimblecombe, P. (2016). Anthropogenic contributions to global carbonyl sulfide, carbon disulfide and organosulfides fluxes. Earth-science reviews, 160, 1-18.