Review : Long-term fluxes of carbonyl sulfide and their seasonality and interannual variability in a boreal forest

1 GENERAL COMMENTS

This paper analyses the seasonal and interannual variabilities of 5-years biospheric COS fluxes at a site located in a boreal pine forest in Finland. To explain these variability modes, the relationships between the COS biospheric sink and environmental drivers (vapor pressure deficit, light, air temperature) are described. A linear regression model is used to select the main environmental drivers of COS biospheric sink variability. They further develop an empirical model of the COS biospheric sink that is function of PAR, LAI and VPD. The model calibrated with the observations successfully reproduces the observed seasonal and interannual variabilities. Then, they optimize the parameters of the model using the LAI, VPD, PAR simulated by the SIB4 Land Surface Model along with the observed COS fluxes at the site of interest. The calibrated model is then applied to the evergreen boreal needleleaf forests over the whole northern hemisphere. The total COS biospheric sink is greater than the simulated one by the SIB4 LSM, in agreement with a missing sink inferred by most top-down studies.

Overall, the paper provides a valuable advance in reconciling the bottom-up and top-down COS budget at high latitude. However, the manuscript seems to be written in a rush. The many references in the Supplementary makes it difficult to follow sometimes and some clarifications are needed. In particular, I have a few questions that need to be addressed.

1. In agreement with reviewer 2, the empirical relations 2-5 call for more explanations of their provenance and their physical

- meanings in the main text. In the equation 5, it is unclear how the parameter e is fixed. For the observations, how and why was the parameter e fixed before optimizing the other parameters (Page 4, line 107)? For the SIB4 LSM, why is e multiplied by 2.1, the average ratio of Hyytiala and SIB4 LAI data (Page 5 line 2)?
 - 2. This concerns the text around Table 1, where the authors attempt to disentangle the drivers of the COS biospheric sink variability. Given the high non-linearity of the underlying equations 2-5, it is not clear how well the simple multiple linear regression analysis presented in Table 1 is able to capture such highly non-linear interactions. To my opinion, the statistical analysis would require a different approach, such as a decision tree or a neural network in order to deal with the highly non-linear interactions between variables.
 - 3. To upscale the flux observations to evergreen needleleaf forests in the whole boreal region, it would have been more straightforward to use the MODIS products of PAR, LAI along with the surface air temperature, VPD from the MERRA reanalysis. The MODIS-derived LAI and PAR should be more realistic spatial variability than the ones simulated by the LSM. A processed-based alternative would have been to (i) calibrate the parameters of the SIB4 LSM with the

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observed LAI, PAR, VPD, FCOS at the closest grid point of the eddy covariance site and (ii) to run the newly calibrated LSM to simulate the COS biospheric fluxes over evergreen needleleaf forests in the whole boreal region. Such analysis would have shed light on the specific LSM parameters that need to be calibrated. The use of the SIB4 LSM would be better justified if the authors evaluate the performance of the LSM to reproduce the COS biospheric sink at Hyytiala (for instance Fig.4). Also, the statistical analysis in Table 1 could be also done for the SIB4 LSM and compared with Maignan et al., 2021 who did a similar analysis.

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2 SPECIFIC COMMENTS

Page 2, line 41-42: "The terrestrial ... 1360 GgS/y" I would add Remaud et al. (2021) who used a more recent anthropogenic inventory to infer the net biospheric sink over the globe through inverse modelling. The missing sink in the high northern latitudes was also shown in Remaud et al. (2021); Hu et al. (2021).

Page 3, line 7: "of which is gap-filled ..." The method for gap-filling needs to be explained for the sake of clarity.

Page 3, line 91: I would add "at 23 m height, where the EC measurements were made".

Page 3, line 95: Where are located the 5 locations?

Page 4, line 106: Here, I would present the parameterization of Carbonyl sulfide fluxes by showing and explaining the equations

15 1-5.

Page 6, line 146, "However, the snow ... region." What is the link with the former sentence?

Page 6, line 153: "We analyzed ... FCOS" What is the dependence of the CO2 flux to the VPD?

Page 7, line 165: Epiphytes can also significantly take up the atmospheric COS during the night when the soil is wet as shown by Rastogi et al. (2018).

- 20 Page 7, line 182: What does "unfiltered" data mean? Was the Fig. S6 drawn with the data that have not been gap-filled? Page 7, line 201: The set-up of the wavelet analysis should be explained in the method. Figure S7 should be in the main text. Page 7, lines 194-209: See Maignan et al. (2021) (Fig. 3). Based on the ORCHIDEE LSM, they showed that the internal conductance was mainly driven by Ta. In the afternoon, the internal conductance limits the total conductance and reaches a maximum 3 hours after the peak in stomatal conductance.
- Page 9, Table 1: I wonder how the result would look like if the measurements were not gap-filled.Page 10, equations 1-5 : A plot associated with each equation (e.g FPAR as function of PAR) would give a better idea of their contribution to FCOS.

Page 10, line 247: It should be mentioned earlier in Part 2.4 that the parameterization is not applied to the gap-filled data. Page 10, line 252: It is worth discussing here the reasons why the empirical model and the observations for July 2014 are in

30 disagreement for July 2014 (Figure 1). Does the empirical model underestimate the sensitivity of the surface fluxes to vapor pressure deficit? Does the net CO2 fluxes exhibit such decrease during the same period? Page 11, line 263: "The cum.. respectively." What are the numbers within parenthesis? IS FCOScum computed using the gap-

filled measurements? Page 11, lines 265-267: I don't understand the link between the COS sulfur deposition and the Carbonyl

sulfide balances and their interannual variation. Please explain.

Page 12, Figure 4: I find Figure 4 particularly interesting, showing that the COS biospheric sink increases between 2013-2017 while the average atmospheric COS concentration decreases over the whole northern hemisphere during the same period. Has the length of the growing season been increasing over the years? It would be interesting to investigate the reasons underlying

5 this increase, for instance by looking at the environmental drivers of FCOS. It would also be interesting to reproduce the Figure 4 for the SIB4 LSM.

Page 12, line 273: "..improve the .. sink estimate." and line 285 "... could help to improve the representation of gross primary production in biospheric models as well.". The paper falls a bit short of really providing a parameterization that could help to improve the LSMs. This is more true that this study highlights a LSM bias which is an underestimation of the simulated

10 biospheric sink in the high latitudes, in agreement with recent inverse modelling studies (Ma et al., 2021; Remaud et al., 2021). Page 12, line 273: I would rather say: "with the recent top-down studies of the COS atmospheric budget (Ma et al., 2021; Remaud et al., 2021; Hu et al., 2021)"

References

- Hu, L., Montzka, S. A., Kaushik, A., Andrews, A. E., Sweeney, C., Miller, J., Baker, I. T., Denning, S., Campbell, E., Shiga, Y. P., Tans,
 P., Siso, M. C., Crotwell, M., McKain, K., Thoning, K., Hall, B., Vimont, I., Elkins, J. W., Whelan, M. E., and Suntharalingam, P.:
 COS-derived GPP relationships with temperature and light help explain high-latitude atmospheric CO2 seasonal cycle amplification,
- 5 Proceedings of the National Academy of Sciences, 118, https://doi.org/10.1073/pnas.2103423118, https://www.pnas.org/content/118/33/ e2103423118, publisher: National Academy of Sciences Section: Physical Sciences, 2021.
 - Ma, J., Kooijmans, L. M. J., Cho, A., Montzka, S. A., Glatthor, N., Worden, J. R., Kuai, L., Atlas, E. L., and Krol, M. C.: Inverse modelling of carbonyl sulfide: implementation, evaluation and implications for the global budget, Atmospheric Chemistry and Physics, 21, 3507–3529, https://doi.org/10.5194/acp-21-3507-2021, https://acp.copernicus.org/articles/21/3507/2021/, publisher: Copernicus GmbH, 2021.
- 10 Maignan, F., Abadie, C., Remaud, M., Kooijmans, L. M. J., Kohonen, K.-M., Commane, R., Wehr, R., Campbell, J. E., Belviso, S., Montzka, S. A., Raoult, N., Seibt, U., Shiga, Y. P., Vuichard, N., Whelan, M. E., and Peylin, P.: Carbonyl sulfide: comparing a mechanistic representation of the vegetation uptake in a land surface model and the leaf relative uptake approach, Biogeosciences, 18, 2917–2955, https://doi.org/10.5194/bg-18-2917-2021, https://bg.copernicus.org/articles/18/2917/2021/, publisher: Copernicus GmbH, 2021.
- Rastogi, B., Berkelhammer, M., Wharton, S., Whelan, M. E., Itter, M. S., Leen, J. B., Gupta, M. X., Noone, D., and Still, C. J.:
 Large Uptake of Atmospheric OCS Observed at a Moist Old Growth Forest: Controls and Implications for Carbon Cycle Applications, Journal of Geophysical Research: Biogeosciences, 123, 3424–3438, https://doi.org/https://doi.org/10.1029/2018JG004430, https:
- //agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2018JG004430, 2018.
- Remaud, M., Chevallier, F., Maignan, F., Belviso, S., Berchet, A., Parouffe, A., Abadie, C., Bacour, C., Lennartz, S., and Peylin, P.: Plant gross primary production, plant respiration and carbonyl sulfide emissions over the globe inferred by atmospheric inverse modelling,
- 20 Atmospheric Chemistry and Physics Discussions, pp. 1–43, https://doi.org/10.5194/acp-2021-326, https://acp.copernicus.org/preprints/ acp-2021-326/, publisher: Copernicus GmbH, 2021.