Biogeosciences Discuss., referee comment RC1 https://doi.org/10.5194/bg-2021-281-RC1, 2021

Comment on bg-2021-281

Anonymous Referee #1

Referee comment on "Global modelling of soil carbonyl sulfide exchange" by Camille Abadie et al., Biogeosciences Discuss., https://doi.org/10.5194/bg-2021-281-RC1, 2021

Abadie et al. implemented a mechanistic and empirical soil model of COS exchange into the ORCHIDEE land surface model and compare those with observations of soil COS fluxesat several sites, representing different soil types. Through a sensitivity study they find themost important parameters for the soil COS flux and optimize those parameters with observations at two sites to improve the COS soil flux simulations. Finally, the authors provide an updated global soil COS budget, including both oxic and anoxic (wetland) soils.This is a very complete and thorough study that I find very well readable. My comments are hence minor.

Answer: The authors thank the Referee for the overall positive answer to this study and for the very helpful comments to improve the manuscript.

Abstract:

P1, L44: -576 Gg S yr-1 for vegetation+soil, or only the vegetation?

Answer: -576 GgS yr-1 corresponds to the revised budget for vegetation only. We replaced "which helped reduce the imbalance of the atmospheric COS budget by lowering COS uptake by soils and vegetation globally (-10% for soil, and -8% for vegetation with a revised mean estimate of -576 GgS y-r1 over 2009-2016)" by "which helped reduce the imbalance of the atmospheric COS budget by lowering **soil COS uptake by 10% and plant COS uptake by 8% globally (with a revised mean vegetation budget of -576 GgS yr^{-1} over 2009-2016)**".

Introduction:

P2, L62-63: This sentence reads weird.

Answer: We rephrased this sentence as shown in the answer of the next comment.

P2, L63-64: The numbers 700-1100 GgS yr-1 sound like a very large gap. If I'm correct, Berry et al. (2013) added an additional ocean flux of 600 to close the gap, and Kuai et al.(2015) added 559 Gg S yr-1. Do the values 700-1100 GgS yr-1 represent the total oceanemissions? So not only the emission gap? A reference to a more recent inversion could beadded (Ma et al. (2021) with a total gap of 432Gg S yr^{-1}).

Answer: Indeed, the values 700-1100 GgS yr-1 represent the total COS oceanic emission estimates (Berry et al., 2013; Kuai et al., 2015; Launois et al, 2015). We modified the sentence as follows "**Several atmospheric transport inversion studies have suggested that an unidentified COS source located over the tropics, of the order of 400-600 GgS yr^{-1}, was needed to close the contemporary COS budget (Berry et al., 2013; Glatthor et al., 2015; Kuai et al., 2015; Ma et al., 2021; Remaud et al., 2022)**."

P2, L78: better say something like: "they have usually not been considered in atmospheric COS budgets".

Answer: We replaced the sentence as suggested by the Referee "Although such COS emissions can be large in some conditions, **they have usually not been considered in atmospheric COS budgets**."

P3, L87: form = from

Answer: Thank you, this error was corrected.

*P3, L113: For clarification, consider adding something like: "at the different sites **that will be used for evaluation in this study**".*

Answer: We completed the sentence as suggested "To better represent the observed soil conditions at the different sites **that will be used for evaluation in this study**, we substituted the soil textures initially assigned in ORCHIDEE from the USDA texture global map with the observed soil textures corresponding to the USDA texture classes (Table S2)."

P3, L119: More important to the mechanistic model than to the empirical model?

Answer: Yes, this precision was added to the sentence "The move from the coarse Zobler classes to the finer USDA classes is found to be more important to the mechanistic model **than to the empirical model**."

P8, anoxic soil COS production: Did you consider to use the formulations of Meredith et al., 2018? These are similar to that of Whelan et al., 2016, but then with the alfa and beta parameters specific for peatland/wetland soils. That is, fca = 3700 for boreal peatland(Meredith et al., 2019) and alfa and beta for peatland from Meredith et al., (2018,). It would be interesting to compare those COS production estimates for wetland soils.

Answer: In the mechanistic approach, the f_{CA} parameter is related to COS uptake by oxic soils only, as we consider anoxic soils as COS sources (equation 18). We did not use the formulation of Meredith et al. (2018) to represent soil COS flux for boreal peatlands as we decided to use the same formulation adapted from Ogée et al. (2016) for all anoxic soils. Indeed, soil COS emissions from peatlands are difficult to characterize as they can vary by an order of magnitude depending on the observation site (see Figure 1 in Meredith et al., 2018). This large variability shows that factors other than soil temperature could be important to include to represent and parametrize soil COS emissions from peatlands. Moreover, the wetland map from Tootchi et al. (2019) used in ORCHIDEE does not distinguish between the different wetland types. Therefore, improving soil COS flux modeling for the different types of anoxic soils should be the focus of future work as adapting the parameters to each wetland type might not be sufficient to improve their representation. However, as the approach to estimate soil COS fluxes from wetlands could be refined by distinguishing boreal peatland as suggested, we added this in section 4.3. as a future improvement. We completed the sentence as follows "We could also refine our approach by distinguishing between the different types of wetlands and define a P_{ref} value for each wetland type instead of a global value of 10 pmol COS m^{-2} s^{-1}. **Then, a distinction could also be made for anoxic soil COS fluxes from boreal peatlands, as Meredith et al. (2019) give a value of f_{CA} specific to this biome**."

P10, L330: "….the same training method than the one used in Spielmann et al." should be"….the same training method ***as*** *the one used in Spielmann et al."*

Answer: Thank you, this error was corrected.

P10, L333: If I understand the description in Wehr et al. (2017) right, the soil COS fluxesat US-HA are not based on eddy-covariance fluxes. It can be better described as flux- profile measurements, connected to CO2 soil chamber measurements and profiles.

Answer: The Referee is right, we clarified the description of US-HA measurement methods for soil COS fluxes "At US-HA, soil COS fluxes in 2012 and 2013 were not directly measured but derived from **flux-profile measurements, connected to CO2 soil chamber measurements and profiles.** A sub-canopy flux gradient approach was used to partition canopy uptake from soil COS fluxes. For more information on this approach and its limitations, see Wehr et al. (2017)."

P10,L350-351:" The stations located in the northern Hemisphere sample air masses coming from the entire northern hemisphere domain above 30 degrees." The stations cover mainly North-America and actually Eurasia is hardly covered, so I would not agreewith this statement.

Answer: The stations located in the Northern hemisphere cover mainly North America, however they are sensitive to air masses coming from the entire Northern hemisphere domain above 30 degrees based on atmospheric modeling, as shown in Remaud et al. (2022; Figure 1 and part 2.2.1).

P11, L384: what does "d" stand for?

Answer: The abbreviation "80 or 667 d" was replaced by "80 or 667 days".

P12, L412: spell out "DA".

Answer: The abbreviation "DA" was replaced by "Data assimilation".

Results:

P13, section 3.1.1. I think the authors could put more emphasis on the potential role of nitrogen fertilization on soil fluxes. E.g. the results of IT-CRO, an agricultural site, could beemphasized in this context. Also the overestimated COS uptake at AT-NEU and ES-LMA could be discussed in light of nitrogen fertilization.

Answer: Thank you, we emphasized the importance of nitrogen inputs at AT-NEU, ES-LMA and IT-CRO in the description of figure 2. "**Besides, AT-NEU and ES-LMA are managed grassland sites with nitrogen inputs. Then, soil COS production could also be enhanced by a high nitrogen content as suggested by several studies (Kaisermann et al., 2018; Kitz et al., 2020; Spielmann et al., 2020), which is not represented in our models**. The mechanistic model is able to represent a net COS production at IT-CRO but overestimates it. This might highlight the importance of adapting the production parameters (α and β) in this model to adequately represent a net COS production. **In this model, the net soil COS production is related to an increase in soil temperature. However, it is to be noted that IT-CRO is an agricultural site with nitrogen fertilization. Therefore, soil COS production in the observations could also be enhanced by nitrogen inputs**."

P13, L468 (Table 3): I would consider showing Table 3 as a figure. The same also forTable 4, which could even be combined with a Fig. from Table 3.

Answer: We inserted the RMSD values in Table 3 and Table 4 in Figure 2 and Figure 3, respectively.

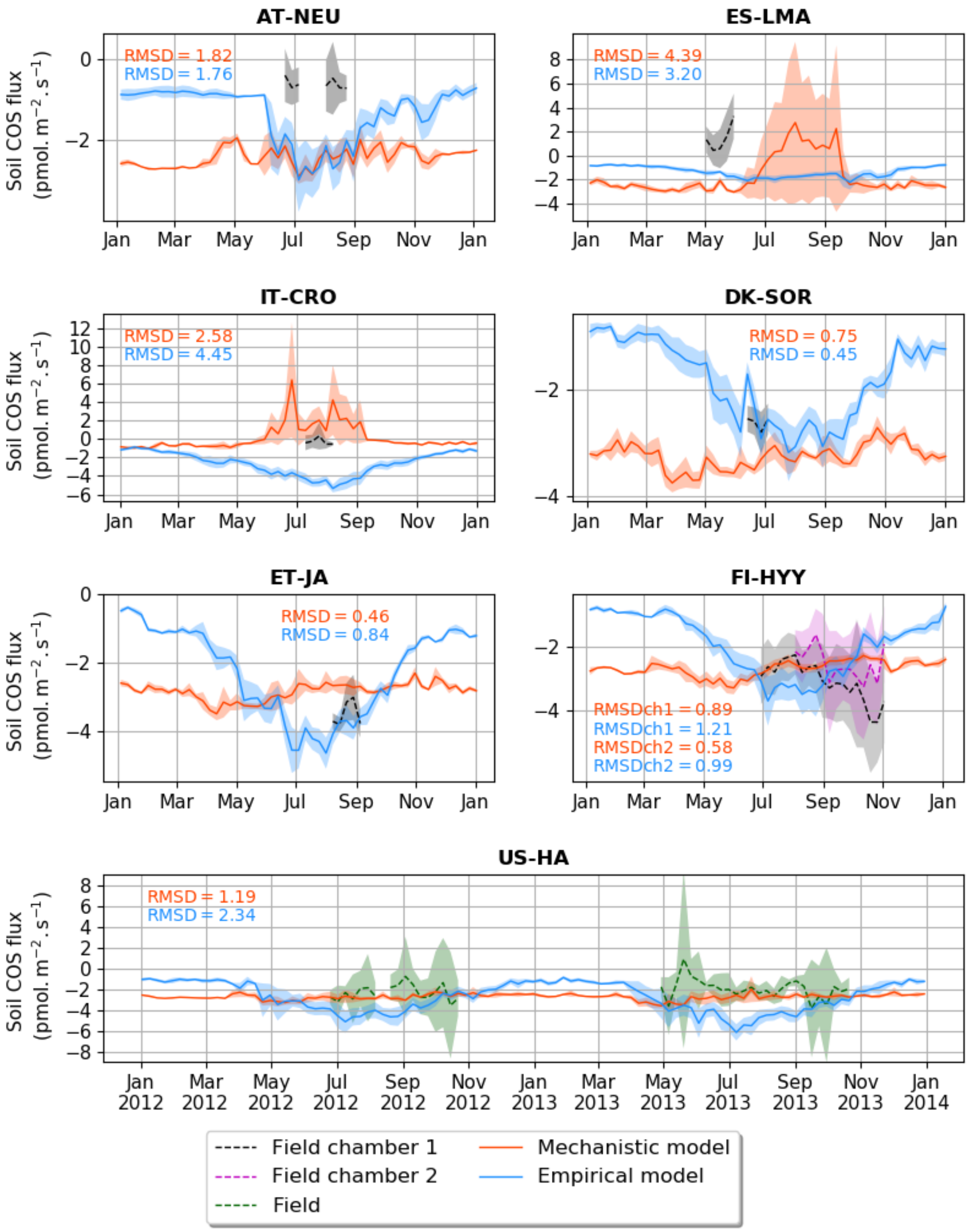

Figure 2: Seasonal cycle of weekly average net soil COS fluxes (pmol m^{-2} s^{-1}) at: AT-NEU, ES-LMA, IT-CRO, DK-SOR, ET-JA, FI-HYY and US-HA. The shaded areas around the observation and simulation curves represent the standard-deviation over a week for each site. Soil COS fluxes are computed with a variable atmospheric COS concentration. RMSD values between the simulated and observed fluxes are given with the respective model color at each site, and for both soil chambers at FI-HYY (ch1 and ch2).

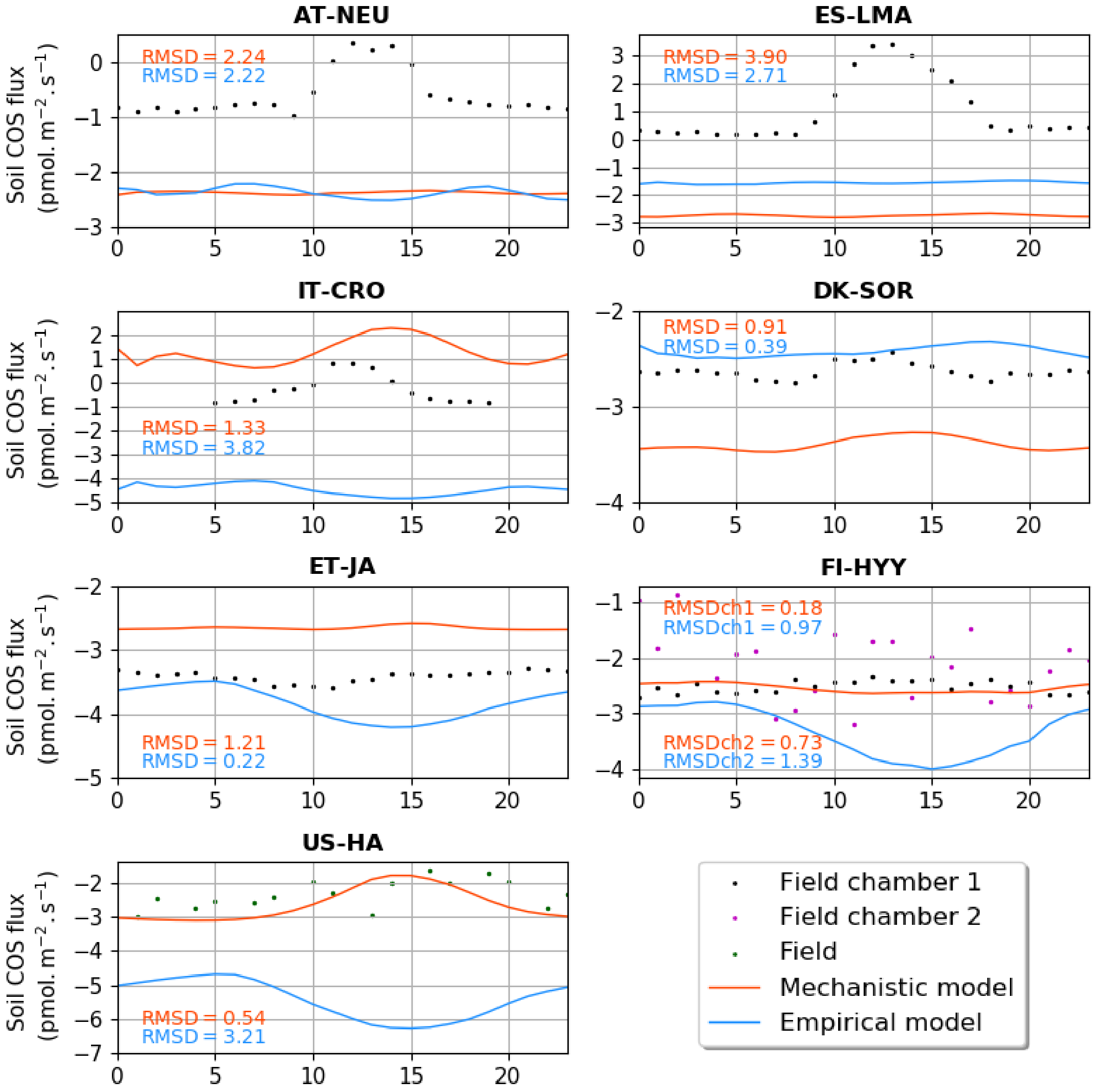

Figure 3: Mean diel cycle of net soil COS fluxes (pmol m^{-2} s^{-1}) over a month at: AT-NEU (08/2015), ES-LMA (05/2016), IT-CRO (07/2017), DK-SOR (06/2016), ET-JA (08/2016), FI-HYY (08/2015) and US-HA (07/2012). Soil COS fluxes are computed with a variable atmospheric COS concentration. The observation-based diel cycles (dots) are computed using Random Forest models at At-NEU, ES-LMA, IT-CRO, DK-SOR and ET-JA. At AT-NEU and ES-LMA. RMSD values between the simulated and observed fluxes are given with the respective model color at each site, and for both soil chambers at FI-HYY (ch1 and ch2).

P14, L486-488: Or the division by PFT is not sufficient, and more specific information on e.g. nitrogen content is needed.

Answer: Indeed, the mismatch between the observed and simulated fluxes could also be due to the PFT fractions attributed at each site, or to missing processes in the models. These assumptions were added to the one made on the need to adapt the PFT-specific parameters on site. We completed the sentence as follows "**The mismatch between the model and the observations could be due to several factors including: i) an insufficient representation of the vegetation complexity by the division in PFTs;**

ii) a poor calibration of the PFT-specific parameters (f_{CA}, α, β); or iii) missing processes in the model, such as considering the effect of nitrogen content on soil COS fluxes."

P14, L496: globally = generally?

Answer: "Globally" was corrected by "generally".

P14, L503-505: This seems to be a repetition of line 500-501.

Answer: Lines 500-501 refer to the soil moisture optimum while lines 503-505 refer to the temperature optimum, both described in Ogée et al. (2016). This was clarified as follows "Furthermore, an optimum soil water content for net soil COS uptake is found between 10% and 15%, which was also observed in Ogée et al. (2016) and in several field studies to be around 12% (Kesselmeier et al., 1999; Liu et al., 2010; van Diest and Kesselmeier, 2008). This optimum soil water content for soil COS uptake is related to a site-specific temperature optimum, which is found between 13°C and 15°C at US-HA for example. Indeed, Ogée et al. (2016) also describe a temperature optimum with a value that depends on the studied site (Kesselmeier et al., 1999; Liu et al., 2010; van Diest and Kesselmeier, 2008)."

Fig 4: Can you show the same plots for observations?

Answer: We represented below a version of Figure 4 for the observed soil COS flux. The comparison between the observed and modelled soil temperature and water content response of soil COS flux is limited by the observation periods. However, the observations do not show a net increase in soil COS uptake with soil temperature as seen at all sites for the empirical model in Figure 4. The increasing trend in net soil COS emissions with soil temperature at ES-LMA and IT-CRO represented for the mechanistic model is also found in the observations. At ET-JA, the net soil COS uptake increases with lower soil water content, which is also shown in Figure 4 for the mechanistic model. However, at AT-NEU the response of soil COS fluxes to soil temperature differs between the simulation and the observations.

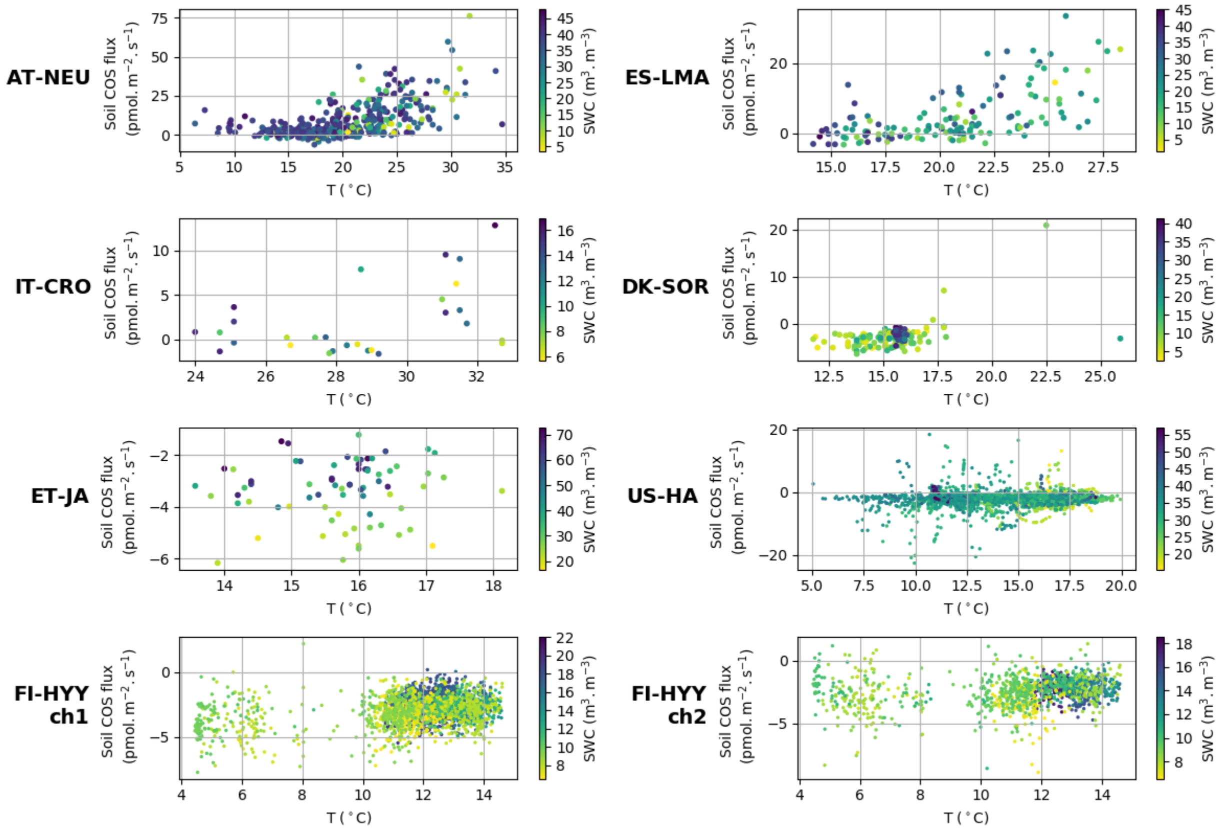

Figure RC1_1: Observed net soil COS flux (pmol m^2 s^{-1}) versus soil temperature (°C) and soil water content (SWC) (m^3.m^{-3}) at AT-NEU, ES-LMA, IT-CRO, DK-SOR, ET-JA, US-HA and FI-HYY.

This figure was added to the supplement as Figure S1 and part 3.1.3. was completed as follows "At IT-CRO and ES-LMA where a strong net soil COS production is simulated by the mechanistic model, the main driver of soil COS fluxes becomes soil temperature. At these sites, the net soil COS production increases with soil temperature, due to the exponential response of soil COS production term to soil temperature. **The increase in soil COS production with soil temperature at IT-CRO and ES-LMA is supported by the observations (Figure S1).**
Contrary to the mechanistic model, soil COS uptake computed with the empirical model is mainly driven by soil temperature, with a soil COS uptake that increases with increasing soil temperature. This response of the empirical model to soil temperature is due to its relation to soil respiration, which is enhanced by strong soil temperature. **However, this net increase in soil COS uptake with soil temperature at all sites is not found in the observations (Figure S1).**"

Fig 5: Can the numbers at the end of the parameter names be replaced with an abbreviation? It is not entirely clear to me what the numbers represent, are they a PFTánd soil texture number?

Answer: Yes, we changed the parameter names to avoid confusion. The numbers at the end of the parameter names were kept only for PFTs and removed for soil texture as the same soil texture was imposed at the two sites (texture 3 "sandy loam"). We also reminded the PFT numbers in the legend of the figure.

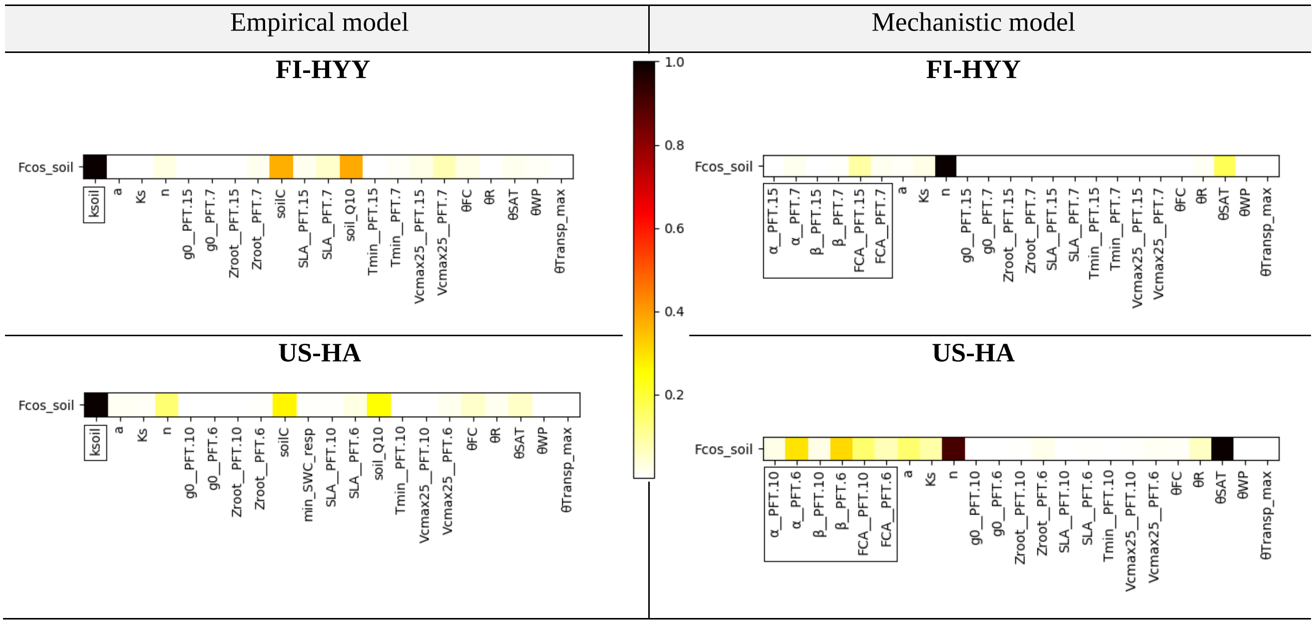

Figure 5: Morris sensitivity scores of the key parameters to which soil COS fluxes are sensitive, for the empirical (left) and the mechanistic (right) models. The two studied sites are FI-HYY (top) and US-HA (bottom). Full descriptions of each tested parameter can be found in Tables S3 and S4 in the supporting information. The PFT is indicated at the end of the parameter names for the PFT-dependent parameters (at FI-HYY: PFT7 = boreal needleleaf evergreen and PFT 15 = boreal natural C3 grassland, at US-HA: PFT6 = temperate broadleaf summergreen and PFT10 = temperate natural C3 grassland). The first-order parameters are shown in the frames.

P16, L555: Can you remind the reader what PFT 15 is?

Answer: Yes, we reminded the name of PFT 15 (boreal C_3 grass) in the sentence L555 "However, at FI-HYY the most influential uptake parameter is for PFT 15 **(boreal C_3 grass)** that only represents 20% of the PFTs at this site while PFT 7 (boreal needleleaf evergreen forest) is the dominant PFT."

Figure E1: Can you explain the green and blue points, which are prior and which areposterior?

Answer: We added the description of the prior and post optimization parameter values in the figure legend:

Figure E1: Comparison between prior and posterior optimization parameter values at FI-HYY and US-HA. The y-axis represents the normalization between the edges of the range of variation for each parameter. Prior values of the parameters are represented in blue and post optimization values are in green.

P17, L600-605: It is very interesting to read that the optimized parameters not only improve the simulated soil COS flux, but also the soil hydrology! Can you give some moredetails on the improvement of the soil moisture, e.g. with a figure or numbers?

Answer: Yes, we added information on the RMSD value change for soil water content prior and post optimization for both models to this sentence "However, while it improves the simulated water content compared to the observations for the mechanistic model at the two sites **(RMSD decreases by 28% at FI-HYY and 22% at US-HA)**, it leads to a degradation at FI-HYY for the empirical model **(RMSD increases by more than 3 times)**."

P17, L612-613: It may be worth discussing the resemblance of the global distribution ofCOS soil fluxes of oxic soils with that presented by Kooijmans et al. (2021) (see their supplementary material). It is nice to see that the implementations of both the empiricaland mechanical models show very similar global distributions in ORCHIDEE and SiB4.

Answer: Indeed, we added a comparison of the empirical and mechanistic models between SiB4 and ORCHIDEE in this section. The empirical approach shows a similar distribution in SiB4 and ORCHIDEE. However, as explained in this section the authors do not completely agree on the resemblance of SiB4 and ORCHIDEE distributions for the mechanistic model concerning the regions with a net COS uptake by oxic soils. We added this comparison to section 3.2.1: "**The distribution and magnitude of soil COS flux from the empirical approach is similar to the one presented in Kooijmans et al. (2021) (see Figure S15 in the supplementary material of Kooijmans et al., 2021), when implemented in SiB4. For the mechanistic model, the comparison of oxic soil COS flux distribution with the one in SiB4 shows a net soil COS emission in India in both SiB4 and ORCHIDEE. However, the maximum oxic soil COS flux is about 60 pmol m^{-2} s^{-1} higher in ORCHIDEE than in SiB4. The regions with the strongest net oxic soil COS uptake also differ between SiB4 and ORCHIDEE as it is concentrated in the tropics in SiB4 and in Western North and South America, and in China for ORCHIDEE."**

P19, L683-684: So the soils do not seem to explain the biases at high latitudes, so can weconclude that the vegetation sink is underestimated at the higher latitudes?

Answer: Yes, this point was mentioned in section 4.1 ("This positive net global budget could be due to an underestimation of vegetation COS uptake in the northern hemisphere, participating in the underestimation of the COS concentration drawdown (Figure 9), but the absence of anthropogenic emission seasonality could also play a role.").
We completed the reference to inversion studies in section 3.3 to support the argument that the mismatch in the high latitudes was related to an underestimation of vegetation uptake "These model-observation mismatches have led top-down studies to identify **vegetation as an underestimated sink in the high latitudes (Ma et al., 2021; Remaud et al., 2022)**, and the tropical oceanic emissions as being the missing source (Berry et al., 2013; Launois et al., 2015; Le Kuai et al. 2015; Ma et al. 2021; Remaud et al., 2022; Davidson et al., 2021)."

P19, L699-702: But at the same time it is inconsistent with comparisons at AT-NEU, ES- LMA, IT-CRO and US-HA, and the marginal model-observation biases can not explain thetoo high atmospheric concentrations, so I find this sentence out of place and would remove it.

Answer: Thank you, this sentence was removed.

P19, L709-710: Instead of showing the Launois et al. results in Fig. 10 you could *considerto include that of Maignan et al. (2021), which to me seems to be a more fair comparison.*

Answer: We clarified the difference between the simulated atmospheric COS concentrations in Figure 10. Maignan et al. (2021) used the soil fluxes from Launois et al. (2015) when transporting the surface fluxes. In Figure 10, the contribution from all components (described in Table 2) other than soils is the same for the simulated concentrations. This clarification was added in the legend figure:

Figure 10. Detrended temporal evolution of simulated and observed COS concentrations at two selected sites, simulated with LMDZ6 transport between 2011 and 2015. The simulated concentrations are obtained by transporting the surface fluxes described in Table 2, and changing only the contribution from soils, with mechanistic (Oxic soils alone, and Oxic + Anoxic soils) and empirical approaches (Berry et al., 2013; Launois et al., 2015). Top: Alert station (ALT, Canada), bottom: Harvard Forest station (HFM, USA). The curves have been detrended beforehand and filtered to remove the synoptic variability (see Sect. 2.3.3).

Discussion:

P20, L717: It would be relevant to compare also with recent global soil COS sink estimatesof Kooijmans et al. (2021) and to include those in Table 5.

Answer: Indeed, we added the results from Kooijmans et al. (2021) to the comparison in Table 5 (which became Table 3 after the removal of Table 3 and Table 4). We completed section 4.1. of the discussion "According to the mechanistic approach of this study, the COS budget for oxic soil is a net sink of -126 GgS yr-1 over 2009-2016, which is close to the value of -130 GgS yr-1 found by Kettle et al. (2002) (Table 3). **This net COS uptake by oxic soils is higher than the one found in SiB4 by Kooijmans et al. (2021) with -89 GgS yr^{-1}, also based on the mechanistic model described in Ogée et al. (2016). In SiB4 and in ORCHIDEE**, the mechanistic model gives the lowest oxic soil COS net uptake compared to all previous studies, which were using empirical approaches."

Table 3: Comparison of soil COS budget per year (GgS yr^{-1}). The net total COS budget is computed by adding all sources and sinks of COS (anthropogenic, ocean, biomass burning, soils, vegetation, atmospheric OH oxidation, photolysis in the atmosphere) used to transport COS fluxes (Table 2).

	Kettle et al. (2002)	Berry et al. (2013)	Launois et al. (2015)			**Kooijmans et al. (2021)**	This study	
			ORCHIDEE	LPJ	CLM4	**SiB4 (modified)**	Empirical soil model	Mechanistic soil model
Period	2002	2002–2005	2006-2009			**2000-2020**	2009-2016	
Plants	-238	-738	-1335	-1069	-930	**-664**	-576	
Soil oxic	-130	-355	-510			**-89**	-214	-126
Soil anoxic	+26	Neglected	+101			**Neglected**	Neglected	+96
Soil total	-104	-355	-409			**-89**	-214	-30
Net total	+64	+1	-566	-300	-161	**(-)**	**-165**	**+19**

A correction of the net total COS budget was made for the estimates of this study as the contribution from atmospheric sinks was previously omitted (OH oxidation that accounts for -100 GgS y^{-1} and COS photolysis in the troposphere representing -30 GgS y^{-1} as explained in section 2.1.3). Therefore, the net total COS budgets using the empirical or the mechanistic approach for soil contribution were corrected to -165 GgS y^{-1} and +19 GgS y^{-1}, respectively. The net total COS budget computed using the mechanistic model for soils is of the same order of magnitude as the uncertainty on the COS budget components (Table 2). We added in section 4.1 "**When computing the net total COS budget considering all sources and sinks of COS (Table 2) with the empirical soil model, we found that neglecting the potential COS production of oxic soils and COS emissions from anoxic soils leads to an overestimation of COS sink or an underestimation of COS source to close the budget (-165 GgS yr^{-1}). On the contrary, the total COS budget computed with the mechanistic soil model is closed given the uncertainties on each component (Table 2). However, despite a closed budget, the mismatch between the observed and simulated latitudinal gradients of atmospheric COS concentration highlights errors in COS flux component distributions (Figure 9).**"

P20, L738: Please, specify that this is about the lack of seasonality in the COS soil flux.

Answer: We added this clarification as suggested by the Referee "It is also to be noted that the mechanistic model better simulates the lack of seasonality **in the soil COS flux** at US-HA compared to the empirical model (Figure 2)."

P21, L759-772: The authors here talk about under- or overestimations, but it does notread as if this is compared to actual observations. So I do not think under- or overestimations are the right term here, they are simply higher or lower than other estimates.

Answer: The Referee is right, this is not a comparison with observations but between simulations. We replaced all the comparison terms as suggested in this section.

P22, The authors briefly touch upon the role of nitrogen fertilization in the discussion of section 4.3, but I think the authors could (and should) put more emphasis on the potentialrole of nitrogen fertilization in this manuscript.

Answer: We agree and we developed the discussion on the role of nitrogen and the importance of including its effect in future developments of the model in this section. "Several studies also found that soil COS production could be related to nitrogen content, which increases with nitrogen fertilizer application (Kaisermann et al., 2018; Meredith et al. 2018, 2019). **At the sites where soil is enriched with nitrogen inputs, such as agricultural fields, managed and fertilized grasslands and forests, the fertilization practices would also need to be included when representing the dynamics of soil COS fluxes. However, the soil nitrogen content and soil microbial nitrogen biomass vary not only with fertilization, but also with location. Then, in addition to indications on land use, information on the total soil nitrogen content should be included in the model to consider nitrogen impact on soil COS flux**."

P22, L788-789: More recent references such as Kaisermann et al. (2018) would beappropriate here.

Kaisermann, A., Jones, S. P., Wohl, S., Ogée, J. and Wingate, L.: Nitrogen Fertilization Reduces the Capacity of Soils to Take up Atmospheric Carbonyl Sulphide, Soil Syst., 2(4),doi:10.3390/soilsystems2040062, 2018.

Kooijmans, L. M. J., Cho, A., Ma, J., Kaushik, A., Haynes, K. D., Baker, I., Luijkx, I. T., Groenink, M., Peters, W., Miller, J. B., Berry, J. A., Ogée, J., Meredith, L. K., Sun, W., Kohonen, K.-M., Vesala, T., Mammarella, I., Chen, H., Spielmann, F. M., Wohlfahrt, G., Berkelhammer, M., Whelan, M. E., Maseyk, K., Seibt, U., Commane, R., Wehr, R., and Krol, M.: Evaluation of carbonyl sulfide biosphere exchange in the Simple Biosphere Model(SiB4), Biogeosciences Discuss. [accepted], https://doi.org/10.5194/bg-2021-192, 2021.

Meredith, L. K., Boye, K., Youngerman, C., Whelan, M., Ogée, J., Sauze, J. and Wingate, L.: Coupled Biological and Abiotic Mechanisms Driving Carbonyl Sulfide Production in Soils,Soil Syst., 2(3), doi:10.3390/soilsystems2030037, 2018.

Meredith, L. K., Ogée, J., Boye, K., Singer, E., Wingate, L., von Sperber, C., Sengupta, A.,Whelan, M., Pang, E., Keiluweit, M., Brüggemann, N., Berry, J. A. and Welander, P. V: Soilexchange rates of COS and CO18O differ with the diversity of microbial communities and their carbonic anhydrase enzymes, ISME J., 13(2), 290–300, doi:10.1038/s41396-018-0270-2, 2019.

Answer: These references were added as suggested.

Biogeosciences Discuss., referee comment
RC2 https://doi.org/10.5194/bg-2021-281-
RC2, 2022

Comment on bg-2021-281

Anonymous Referee #2

Referee comment on "Global modelling of soil carbonyl sulfide exchange" by Camille Abadie et al., Biogeosciences Discuss., https://doi.org/10.5194/bg-2021-281-RC2, 2022

Abadie et al. simulated soil COS fluxes on a global scale using a mechanistic soil model in a land surface model ORCHIDEE, and evaluated the simulation results, from both the mechanical model and an empirical model based on scaling soil respiration, against 7 sitesin the Northern Hemisphere. Furthermore, an atmospheric transport model LMDZ was used to investigate the contribution of different soil flux products to the latitudinal gradient of atmospheric COS concentrations. Moreover, sensitivity analyses were performed to reveal the importance of various parameters, which is useful to understand the control mechanisms of the soil COS fluxes. Note that the mechanistic model has been previously developed and published. Nevertheless, implementing the existing model in ORCHIDEE to study the global soil COS fluxes is desired. This is a very nice model study. The paper is well structured and very well written and is certainly suitable for the journal of Biogeosciences.

As has been noticed by the authors, the available field observations of soil COS fluxes are very limited, which is especially true when global COS fluxes are the focus of the study. Infact, all 7 sites are located in a narrow latitude range of 42 – 62 °N, and do not cover a full seasonal cycle, which makes it difficult to evaluate the simulation results on a global scale, and raises many questions around whether the presented simulation results are justified, e.g., whether smaller global COS soil flux than previous estimates is trustworthy,whether very large emissions in part of the tropics exist, not to mention the validation of the seasonal cycle and the diel cycle of the simulated results. On the other hand, these are also nice topics to be followed on. For this manuscript, I strongly feel that these pointsneed to be better clarified in the revised version before publication.

Answer: The authors thank the Referee for the generally positive comment and for the very insightful questions and suggestions to improve this manuscript.
We agree with the Referee, the scarcity of soil COS flux observations and the limited latitudinal range are important limitations to the validation of this study. However, we selected observation sites that cover the largest diversity given the available observations, which represent 7 different plant functional types in ORCHIDEE. Moreover, previous

studies had access to an even smaller number of observations to validate their estimates as many field observations of soil COS fluxes were carried in the recent years. The difficulty of performing long-term and continuous measurements of soil COS fluxes also has to be acknowledge as flux towers do not allow to measure soil COS flux only. The efforts made to collect soil COS flux observations enabled the recent study of Kooijmans et al. (2021), and this study to benefit from the increasing number of measurements. As indicated by the Referee, more observations in the tropics would help to validate the net soil COS production simulated in some regions, such as the large one found in Northern India in both SiB4 and ORCHIDEE. Soil chamber measurements of soil COS flux were performed at La Selva Biological Station, at a tropical rainforest in Costa Rica (Sun et al., 2014). When available, these observations could allow a first comparison with the simulated soil COS flux in the tropics.

We added in the abstract "Evaluation of the model against flux measurements at 7 sites yields a mean root mean square deviation of 1.6 pmol m^{-2} s^{-1}, instead of 2 pmol m^{-2} s^{-1} when using a previous empirical approach that links soil COS uptake to soil heterotrophic respiration. **However, soil COS model evaluation is still limited by the scarcity of observation sites and long-term measurement periods, with all sites located in a latitudinal band between 39°N and 62°N and no observations during wintertime in this study**." We also completed section 4.3. as follows "Moreover, one difficulty with the study of soil COS fluxes arises from the scarcity of field measurements that could be used for model validation and calibration. **Besides, the observation sites considered here are all located in a small latitudinal range between 39°N and 62°N. Measurements in the tropics and in the Southern hemisphere are needed. Especially, soil COS flux observations in Northern India could help to validate the net soil COS production simulated in both SiB4 and ORCHIDEE. In the tropical rainforest, soil COS flux measurements were performed at La Selva Biological Station in Costa Rica (Sun et al., 2014). When available, these measurements could allow a first comparison between the observed and simulated soil COS flux in a tropical region.**"

Then, a comparison to another global model can also give some strength to our results. The study of the mechanistic soil COS model presented in Kooijmans et al. (2021) enables us to compare our results to those obtained in SiB4. We emphasized this comparison in the Results part as the distribution between net soil COS sources and net soil COS sinks at the global scale shows some similarities between SiB4 and ORCHIDEE. This was added to section 3.2.1. "**The distribution and magnitude of soil COS flux from the empirical approach is similar to the one presented in Kooijmans et al. (2021) (see Figure S15 in the supplementary material of Kooijmans et al., 2021), when implemented in SiB4. For the mechanistic model, the comparison of oxic soil COS flux distribution with the one in SiB4 shows a net soil COS emission in India in both SiB4 and ORCHIDEE. However, the maximum oxic soil COS flux is about 60 pmol m^{-2} s^{-1} higher in ORCHIDEE than in SiB4. The regions with the strongest net oxic soil COS uptake also differ between SiB4 and ORCHIDEE as it is concentrated in the tropics in SiB4 and in Western North and South America, and in China for ORCHIDEE.**"

We then added the net global budget for oxic soils computed in SiB4 in the Discussion part in section 4.1., which also concludes to a smaller contribution from oxic soils than estimated in previous studies. The discussion was completed as follows "According to the mechanistic approach of this study, the COS budget for oxic soil is a net sink of -126 GgS yr^{-1} over 2009-2016, which is close to the value of -130 GgS yr^{-1} found by Kettle et al. (2002) (Table 3). **This net COS uptake by oxic soils is higher than the one found in SiB4 by Kooijmans et al. (2021) with -89 GgS yr^{-1}, also based on the mechanistic model described in Ogée et al. (2016). In SiB4 and in ORCHIDEE**, the mechanistic model gives the lowest oxic soil COS net uptake compared to all previous studies, which were using empirical approaches."

Sun, W., Maseyk, K. S., Juarez, S., Lett, C., and Seibt, U. H.: Soil-atmosphere carbonyl sulfide (COS) exchange in a tropical rainforest at La Selva, Costa Rica. AGU Fall Meeting Abstracts, 2014, B41C-0075, 2014.

Regarding the selection of the field sites, why were the soil flux measurements in anagricultural field in the Southern Great Plains by Maseyk et al., 2014 not used?

Answer: The data from Maseyk et al. (2014) are not publicly available and we were unable to obtain the original data. The agricultural field in the Southern Great Plains would be represented by C3 crops in ORCHIDEE. In the dataset we used, IT-CRO site is also represented by C3 crops.

L25: remove "budgets" after "atmospheric COS"

Answer: "Budgets" was removed as suggested.

L37 specify the region of the tropics, otherwise, it sounds like high emissions in all tropicalregions

Answer: The Referee is right, we have now replaced the sentence "The predicted spatial distribution of soil COS fluxes, with large emissions in the tropics from oxic (up to 68.2 pmol COS m-2 s-1) and anoxic (up to 36.8 pmol COS m-2 s-1) soils in the tropics mainly, marginally improves the latitudinal gradient of atmospheric COS concentrations, after transport by the LMDZ atmospheric transport model" by "The predicted spatial distribution of soil COS fluxes, with large emissions from oxic (up to 68.2 pmol COS m-2 s-1) and anoxic (up to 36.8 pmol COS m-2 s-1) soils **in the tropics, especially in India and in the Sahel region**, marginally improves the latitudinal gradient of atmospheric COS concentrations, after transport by the LMDZ atmospheric transport model."

L170: please briefly discuss why the steady-state condition is valid? what assumption hasto be made to make the steady-state valid?

Answer: We assume that, over the 30-minute time step used here to run the ORCHIDEE model, the environmental conditions in the soil (temperature, moisture, etc.) are constant. We also assume chemical equilibrium between the gaseous and dissolved COS and neglect COS adsorption on soil particles, as suggested by Ogée et al. (2016). In these conditions, gas exchange of COS in the soil column is evolving rapidly to a steady state, attained before the end of the 30-minute time step. For example, if we consider COS diffusion, the diffusion time scale is L^2/D with L the diffusion length and D the diffusivity of COS. In ORCHIDEE, we consider that COS uptake happens in the top 9 cm. With the diffusivity of COS in the air that is around $1.4*10^{-5}$ m^2/s, the diffusion time scale is around 10 minutes. Moreover, the first order reaction from the carbonic anhydrase enzyme COS consumption reduces the time needed to reach the steady-state condition. The evolution of the soil COS flux can therefore be approximated as a succession of steady states from one time step to the other. The model description was completed as follows "Under steady-state conditions and uniform soil temperature, moisture and porosity profiles, an analytical solution of Eq. 2 can be found (Ogée et al., 2016). **We assume that the environmental conditions, such as soil temperature and moisture, are constant in ORCHIDEE over the 30-minute model time step. We also assume chemical equilibrium between the gaseous and the dissolved COS, neglecting advection as suggested by Ogée et al. (2016). In these conditions, the typical time scale for COS diffusion in the upper active soil layer**

is much shorter than the 30-minute model time step.”

L457-458: Clearly, the mechanistic model predicts nearly no seasonal cycle except for large production signals in the summertime, as is shown in Figure 2. However, this impliesthat relatively large net soil uptake exists in northern high latitude in winter times, when the temperature can be rather low and the land is covered by snow. This makes me wonder what the applicable range for the parameters shown in the method section, e.g., the valid temperature range of f_{CA} in eq. 16, the valid temperature range for α and *β in eq. 17.*

Answer: We acknowledge that the impact of snow cover on soil COS flux is not represented in the soil COS models and it could be important in specific cases. However, the scarcity of COS flux observations from soils covered by snow and in winter does not allow to implement its effect. Note that Helmig et al. (2009) found that COS uptake was not zero when soil is covered by snow at Niwot Ridge, Colorado. We added in section 4.3. “**In the soil COS models, the impact of snow cover is also not represented. Indeed, due to the scarcity of soil COS flux observations in winter and with snow cover, its effect on soil COS flux could not be implemented in soil COS models yet. However, Helmig et al. (2009) found that COS uptake was not zero when soil is covered by snow at Niwot Ridge, Colorado.”**

As mentioned by the Referee, at the sites where there is no strong soil COS production for the mechanistic model, the absence of seasonality leads to a winter soil COS flux that is of the same order of magnitude as the summertime flux. However, for the simulated flux it is to be noted that despite a similar order of magnitude, the soil COS flux in summer does not result only from soil COS uptake but from the combination of soil COS uptake and production, which compensates for the increase in soil COS uptake in the summertime (Figure C1). Observations of soil COS fluxes during wintertime would be of much help to evaluate this net soil COS uptake simulated in winter.

Concerning the f_{CA} parameter for soil COS uptake, in Ogée et al. (2016) soil COS fluxes were simulated with several f_{CA} values for temperatures ranging from 0°C to 25°C. Then, for the production term with the α and β parameters, the temperature range tested in Whelan et al. (2016) was set between 10°C and 40°C. At the studied sites, the temperature can reach lower values than 10°C. However, in the absence of more information on soil COS production in winter, we kept this expression of soil COS production for lower temperatures which implies that the production term would tend to zero.

Helmig, D., Apel, E., Blake, D. *et al.* Release and uptake of volatile inorganic and organic gases through the snowpack at Niwot Ridge, Colorado. *Biogeochemistry* **95,** 167–183 (2009). https://doi.org/10.1007/s10533-009-9326-8

L471-473: Note that vegetation was also removed for the FI-HYY site, as is in Sun et al.,2018 “The moss layer or any other vegetation was removed to expose the humus layer inside the chambers.” This contradicts the statement. If the assumption would be true, what would be the mechanism for artificially enhanced COS production?

Answer: The statement was “However, the mechanistic model struggles to reproduce soil COS fluxes at AT-NEU and ES-LMA, with an overestimation of soil COS uptake or an underestimation of soil COS production at AT-NEU and a delay in the simulated net COS production at ES-LMA. We might suspect that the removal of vegetation at these sites prior to the measurements could have artificially enhanced COS production in the observations”. This assumption was based on the fact that removing vegetation can affect soil structure and increase the accessibility of plant matter residues and more generally soil organic matter to degradation and abiotic COS production. The enhancement of soil COS production by a degradation of accessible soil organic matter was discussed in

Whelan et al. (2016).
Indeed, vegetation was also removed at FI-HYY. We added this sentence in section 2.2.2. concerning the soil COS flux measurements at FI-HYY "**Any vegetation was removed from the chambers before the measurements**."
However, we think that the removal of vegetation at FI-HYY might not have the same impact on soil COS production as at AT-NEU and ES-LMA as these sites have different environmental conditions. At AT-NEU and ES-LMA, the average soil temperature (respectively 19.8°C and 20.5°C with maximums of 34.7°C and 28.3°C) during the observation periods is higher than at FI-HYY (10.9°C with a maximum of 14.9°C). At these two sites, soil is also expected to receive more direct radiations than Hyytiälä forest, as they are grasslands. Soil COS production was related to thermal or photo-degradation of organic matter (Kitz et al., 2017, 2020; Whelan et Rhew, 2015; Whelan et al., 2016, 2018). Therefore, the authors would disagree with the fact that the absence of net soil COS production despite the removal of vegetation at FI-HYY contradicts a possible enhancement of soil COS production with the removal of vegetation at AT-NEU or ES-LMA. We added this justification "We might suspect that the removal of vegetation at these sites prior to the measurements could have artificially enhanced COS production in the observations. **Indeed, the removal of vegetation could change soil structure and increase the availability of vegetation residues and soil organic matter to degradation (Whelan et al., 2016). AT-NEU and ES-LMA are grassland sites for which soils are expected to receive higher light intensity than forest soils. These sites also show a high mean soil temperature of about 20°C during the measurement periods. Therefore, high soil temperature and light intensity on soil surface could enhanced soil COS production as it was related to thermal or photo degradation of soil organic matter (Kitz et al., 2017, 2020; Whelan et Rhew, 2015; Whelan et al., 2016, 2018). This is not the case at FI-HYY, ET-JA or DK-SOR, where soil temperature is much lower (mean value about 10°C at FI-HYY and 15°C at ET-JA and DK-SOR during the measurement periods) and the forested cover decreases the radiation level reaching the soil. Note that herbaceous biomass is also likely to be higher in grasslands than in forests.**"

L478: The diel cycles of simulated COS soil fluxes by the mechanistic model shown in Figure 3 are totally not supported by the observations. Note that when relatively large uncertainties in the observations are considered, a minimum net soil COS uptake in the observations is not significant at all. Actually, the large discrepancy calls for a better understanding of the mechanistic model: what causes the diel cycles in the model but notshown in the observations.

Answer: We agree and revised our analysis for the diel cycle at US-HA that shows large variability. We removed the sentence "A minimum net soil COS uptake is also observed at US-HA but in the afternoon". As suggested by the Referee, we studied the response of soil COS flux diel cycle to soil temperature and soil water content to better characterize the difference between the simulated and observed diel cycle.

<u>Simulated soil COS flux</u>:
Figure 3 shows a diel cycle of soil COS flux simulated with the mechanistic model at IT-CRO and US-HA. We represented below the modelled mechanistic soil COS flux versus soil temperature and soil water content in a similar way as in Figure 4 but with the hourly average fluxes of the mean diel cycle represented in Figure 3. As shown in the figure below, the main driver of the diel cycle at US-HA and IT-CRO for the mechanistic model is soil temperature, leading to a decrease of the net soil COS uptake at US-HA and to an increase of the net soil COS production at IT-CRO. The simulated ranges of soil temperature at US-HA and IT-CRO correspond to the highest values compared to the other sites. It is also to be noted that the simulated soil water content at all sites shows almost no diel variations (about 1%).

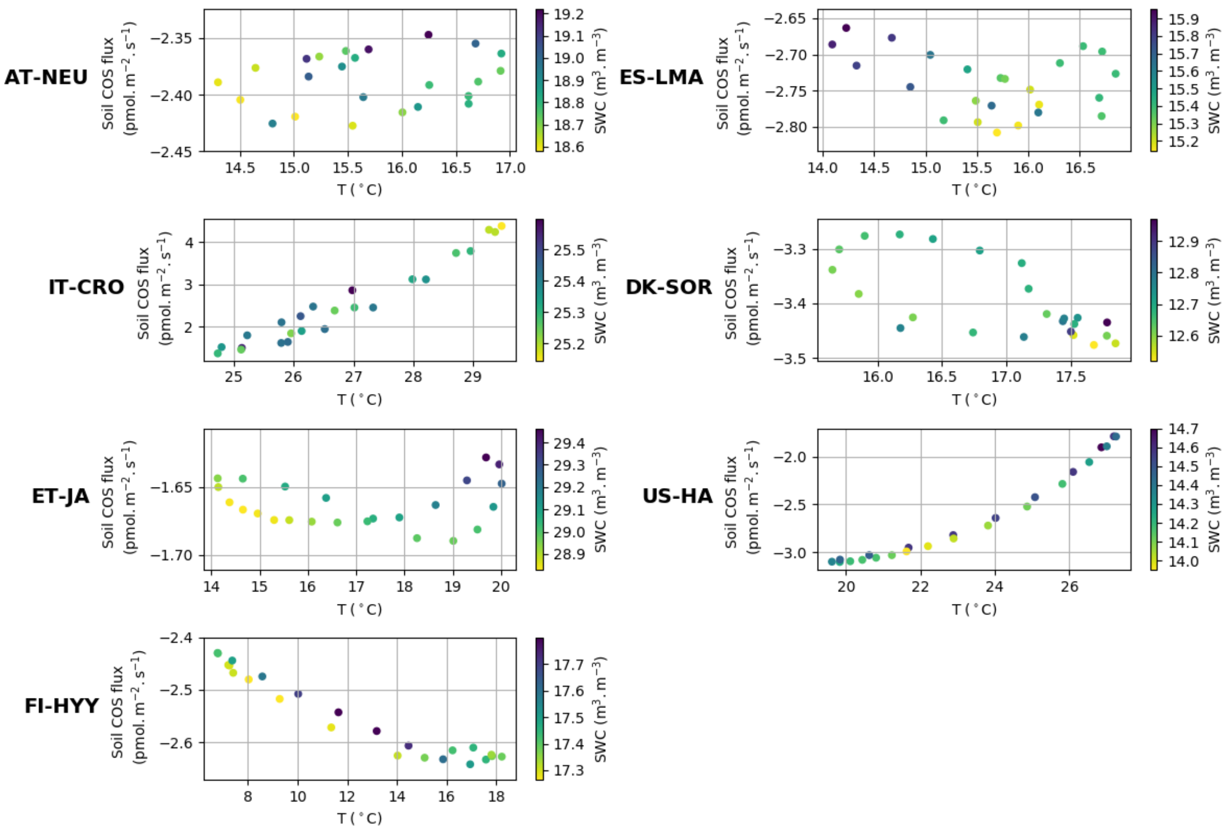

Figure RC2_1: Simulated hourly average net soil COS flux (pmol m^2 s^{-1}) versus soil temperature (°C) and soil water content (SWC) (m^3.m^{-3}) at AT-NEU, ES-LMA, IT-CRO, DK-SOR, ET-JA, US-HA and FI-HYY, for the mechanistic model. Soil COS fluxes represent the mean diel cycle of net soil COS fluxes (pmol m^{-2} s^{-1}) over a month at: AT-NEU (08/2015), ES-LMA (05/2016), IT-CRO (07/2017), DK-SOR (06/2016), ET-JA (08/2016), FI-HYY (08/2015) and US-HA (07/2012).

Observed soil COS flux:
We also represented below the hourly average of the observed soil COS flux used to compute the mean diel cycle in Figure 3 versus soil temperature and soil water content. The observed soil COS fluxes show diel variations at AT-NEU, ES-LMA, IT-CRO and DK-SOR illustrated in Figure 3. At these four sites, soil COS flux exhibits an increasing trend of the net soil COS production with soil temperature at AT-NEU, ES-LMA and IT-CRO, and a decreasing trend of the net soil COS uptake with soil temperature at DK-SOR. However, Spielmann et al. (2020) found that the random forest regression of soil COS fluxes was the most sensitive to the incident shortwave radiation, which is not represented in the soil COS models. Kitz et al. (2020) also related daytime net COS production to high light intensity reaching the soil surface at ES-LMA.
At US-HA, no diel cycle is found in the observations (Figure 3) while the mechanistic model shows a decrease in the net soil COS uptake around 3pm. As seen above, the mechanistic model diel cycle at US-HA is a response of soil temperature. However at this site, Wehr et al. (2017) indicate that they did not find a dependency of soil COS flux to soil temperature or soil water content as their ranges of variation could be too low to see an effect. Indeed, the simulated range of soil temperature at US-HA in ORCHIDEE is larger than the one measured on site. The diel variations of soil COS flux simulated at US-HA could be due to this larger range of soil temperature enhancing soil COS production, while this effect is not found in the observations, or more compensated by soil COS uptake than in the simulated flux.

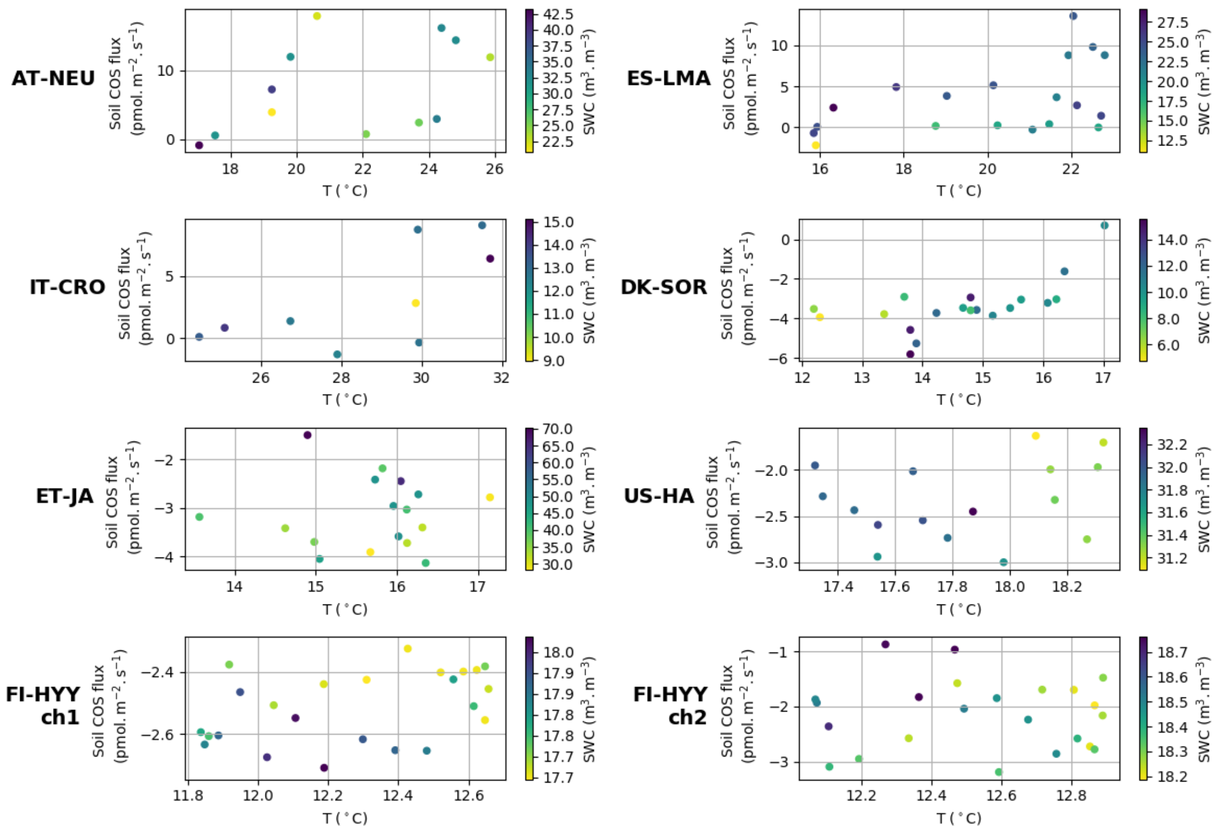

Figure RC2_2: Observed hourly average net soil COS flux (pmol m^2 s^{-1}) versus soil temperature (°C) and soil water content (SWC) (m^3.m^{-3}) at AT-NEU, ES-LMA, IT-CRO, DK-SOR, ET-JA, US-HA and FI-HYY. Soil COS fluxes represent the mean diel cycle of net soil COS fluxes (pmol m^{-2} s^{-1}) over a month at: AT-NEU (08/2015), ES-LMA (05/2016), IT-CRO (07/2017), DK-SOR (06/2016), ET-JA (08/2016), FI-HYY (08/2015) and US-HA (07/2012).

The part on soil COS flux simulated with the mechanistic model in section 3.1.2. was modified as follows "Figure 3 shows the comparison between the simulated and observed mean diel cycles over a month. The observations show a minimum net soil COS uptake or a maximum net soil COS production reached between 11 am and 1 pm at AT-NEU, ES-LMA, IT-CRO and DK-SOR. At AT-NEU and ES-LMA, neither model is able to represent the observed diel cycle. **At these grassland sites, Spielmann et al. (2020) and Kitz et al. (2020) found that the daytime net COS emissions were mainly related to high radiations reaching the soil surface, which impact is not represented in the soil COS models**. At IT-CRO and DK-SOR, the diel cycles simulated by the mechanistic model show patterns similar to the observations with a peak in the middle of the day, but with an overestimation of the net soil COS production and a delay in the peak at IT-CRO, and an overestimation of the net soil COS uptake at DK-SOR. The mechanistic model reproduces the absence of a diel cycle observed at FI-HYY and ET-JA but with an underestimation of the net soil COS uptake at ET-JA. **AT US-HA, the observed soil COS flux does not exhibit diel variations while the mechanistic model shows a peak with a decrease of the net soil COS uptake around 3 pm. Wehr et al. (2017) explain this absence of diel cycle in the observations by a range of variations for soil temperature and soil water content that is too low to influence soil COS flux. In ORCHIDEE, the simulated range of temperature at US-HA is larger than the one measured on site and temperature is the main driver of the decrease in net soil COS uptake at this site (not shown). Therefore, the enhancement of soil COS production by soil temperature could be only found in the simulated flux. Another possibility is that it could be totally compensated by soil COS uptake in the observations**."

L568: Section 3.1.5, although it is nice that the authors have made an effort to optimizesoil COS flux, it may be premature. As the results are not used in the following results, Isuggest leaving this section out or putting it into the Appendix.

Answer: Indeed, the optimized parameters are not used at the global scale as there are only few sites with enough observations to perform an optimization. However, the optimization gives useful information as it shows that for the empirical model, improving the representation of soil COS flux leads to a degradation of the simulated water content at FI-HYY, while soil water content is improved after soil COS flux assimilation for the mechanistic model. We added the RMSD change of soil water content prior and post optimization in section 3.1.5. "However, while it improves the simulated water content compared to the observations for the mechanistic model at the two sites **(RMSD decreases by 28% at FI-HYY and 22% at US-HA)**, it leads to a degradation at FI-HYY for the empirical model **(RMSD increases by more than 3 times)**". The optimization also shows how the most important parameters of the mechanistic model are affected when assimilating observed soil COS fluxes. As the optimization does not only aim at improving the simulated soil COS flux, but also at better understanding the models and their limitations, the authors would like to keep this study as part of the manuscript.

L645: Section 3.2.2 Temporal evolution of the soil COS budget. It is expected that oxic soil COS sinks would decrease when atmospheric COS concentration decreases, and one could even expect that the decrease is, to the first order, proportional to the decrease of COS concentrations. However, the sharp decrease from 2016 is far beyond this. What arethen the main reasons that can explain the sharp decrease in the mechanistic model?

Answer: Indeed, the decrease in oxic soil COS budget computed with the mechanistic model is sharper than the drop in atmospheric COS concentration. Atmospheric COS concentration is not the only driver of soil COS flux in the mechanistic model. Changes in the soil COS production term also impact COS flux from oxic soils, which depends on soil temperature. Between 2010 and 2019, the soil COS production term has increased by 7%. In this model, the production term can have a strong impact on the net soil COS flux as it is expressed as an exponential response to soil temperature. In addition to a decrease of soil COS diffusion into the soil with lower atmospheric COS concentrations, an increase in the production term contributes to reducing the net soil COS uptake by oxic soils.
It is also to be noted that the drop in atmospheric COS concentration is not homogenous around the globe as illustrated in the figure below which shows the difference in simulated atmospheric COS concentrations between 2010 and 2019. The largest simulated decreases are located in Europe and South of China. Then, in these regions, the soil COS uptake is expected to be particularly affected by the drop in atmospheric COS concentration.

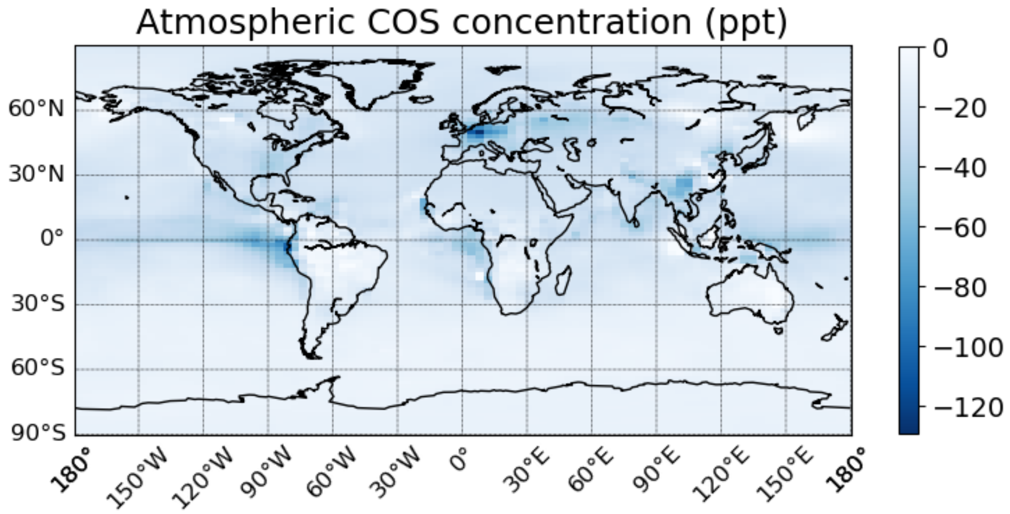

Figure RC2_3: Difference of simulated atmospheric COS concentrations (ppt) between 2010 and 2019. Negative values show a loss in atmospheric COS concentration in 2019 compared to 2010.

The simulated changes in soil temperature and moisture are also heterogenous as illustrated by the difference of soil temperatures and moisture between 2010 and 2019 (positive values represent an increase in 2019 compared to 2010). Changes in oxic soil COS budget result from the combined effect of decreasing atmospheric COS concentration and changes in the drivers of soil COS fluxes.

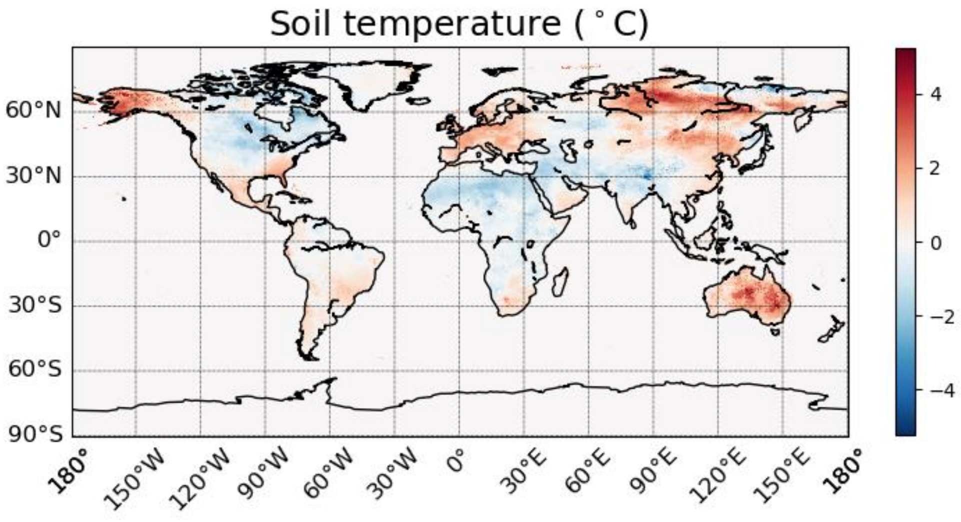

Figure RC2_4: Difference of simulated soil temperaturas (°C) between 2010 and 2019. Positive values show an increase in soil temperature in 2019 compared to 2010.

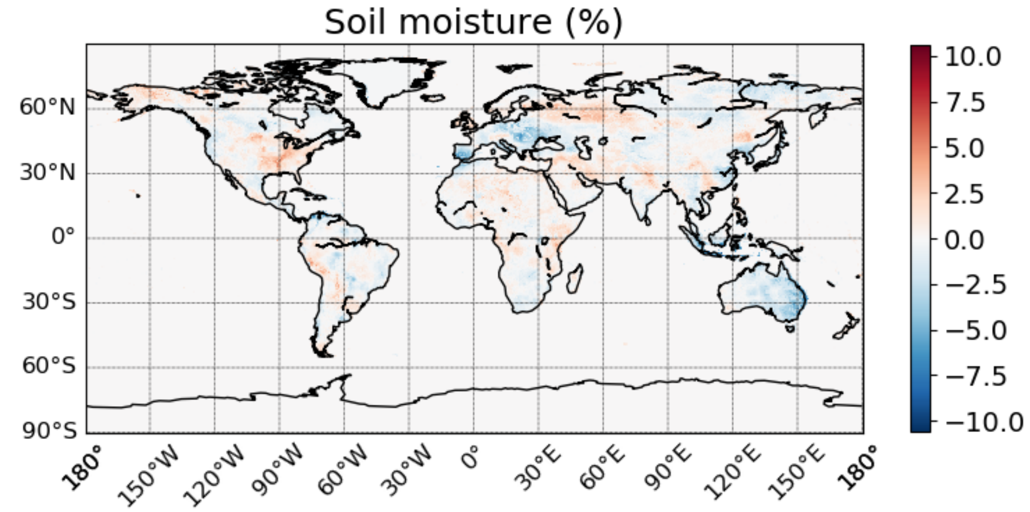

Figure RC2_5: Difference of simulated soil water contents (%) between 2010 and 2019. Negative values show a decrease in soil water content in 2019 compared to 2010.

Section 3.2.2 was completed as follows "**Note that the decrease in oxic soil COS budget computed with the mechanistic model is sharper than the drop in atmospheric COS concentration because changes in oxic soil COS budget result from the combined effect of decreasing atmospheric COS concentration and changes in the drivers of soil COS fluxes (i.e., changes in soil temperature and water content during the 10 year period which are not homogenously distributed around the globe (not shown))**."