

Interactive comment on “Carbonyl Sulfide: Comparing a Mechanistic Representation of the Vegetation Uptake in a Land Surface Model and the Leaf Relative Uptake Approach” by Fabienne Maignan et al.

Anonymous Referee #2

Received and published: 17 December 2020

[1] Maignan et al., develop a parameterization for including OCS uptake within a well-known land surface model (ORCHIDEE). Since OCS has been proposed as a proxy for GPP (currently not observable at large scales), this work is an important step towards estimating global GPP. The authors do a very thorough job of combing the OCS literature for published values of leaf relative uptake (LRU) and validating modeled uptake at two temperate sites where OCS flux is measured. I think this work should eventually be published in Biogeosciences. However I am currently a little confused with regard to how the paper is framed. The authors pose their formulation of OCS flux using a conductance based approach against the well-known LRU based relationship between FOCS and GPP. The LRU approach is also based on conductance, with simple assumptions regarding the relative role of mesophyll and stomatal conductances (Seibt et al., 2010). Thus the differences in flux resulting between the two approaches are not surprising, and don't really manifest themselves once transported and compared against OCS flask measurements from NOAA. This is an interesting finding, but I think the authors must focus on a more scientific question.

Answer: We thank the Referee for this analysis. We must precise here that what we call the “LRU approach” in this study, is based on a former work with ORCHIDEE (Launois et al., 2015b), which does not consider a varying LRU as modelled in Seibt et al. (2010, their equation (8)), but uses constant values for each Plant Functional Type (PFT). We have thus clarified the context of this study and formulated our goal in a more scientific way: “In a former study, Launois et al. (2015b) simply defined the COS uptake by vegetation as the CO₂ gross uptake simulated by LSMs, scaled with a constant LRU value for each large vegetation class. The goal of this study is to now simulate the uptake of atmospheric COS by continental vegetation in a more complex and realistic way using a mechanistic approach within an LSM, and apply this model to evidence the shortcomings or pertinence of the LRU concept, depending on the studied scales”. In the revised manuscript we have thus emphasized the study of the LRU variability both at hourly and site scales, and at monthly and global scales. This can be seen in more details through our answers to both Referees' comments.

[2] The other main comment I have is about modeled $g_{s,ocs}$ and $g_{i,ocs}$. I agree with the authors that the role of g_i is important and often ignored, but I am somewhat skeptical of the large diurnal variation in modeled g_i , which is related to temperature. As I understand, g_i is estimated from V_{max} of Rubisco, but Rubisco response to temperature is not thought to be that large, particularly at temperatures observed at the temperate NH sites (see Sage and Kubien 2007). Moreover, while estimates of diurnal variability of mesophyll conductance hasn't been reported much, a recent study showed that the diurnal variability in g_m (which is similar to g_i in this study) is much smaller than g_s (Strangl et al., 2019; which is also a high latitude coniferous forest, so somewhat similar to Hyytiälä). This could serve as a mechanism for plants to modify (increase) water use efficiency and therefore continue assimilating carbon even as g_s declines due to high VPD commonly observed after mid-day (see also Buckley and Warren, 2014). See also recent work by Gimeno et al., (2020). Thus, I am quite surprised that at Hyytiälä it seems like gas exchange is most often limited by g_i and not g_s . I believe some sensitivity analyses could be done with regard to the formulation of g_i . The temperature dependance of g_i is also not uniform across plant species (von Caemmerer and Evans, 2015), with obvious implications for the global formulations presented here. I think a more in-depth discussion of the implications of gas exchange most limited by g_i should be added, replacing the current and mostly qualitative discussion currently.

Answer: We first need to say that we have corrected a problem in the computation of the total internal conductance (which was wrongly systematically scaled with the maximum LAI of 12 instead of the actual LAI, but did not affect the computation of fluxes) and have redrawn Figure 1.

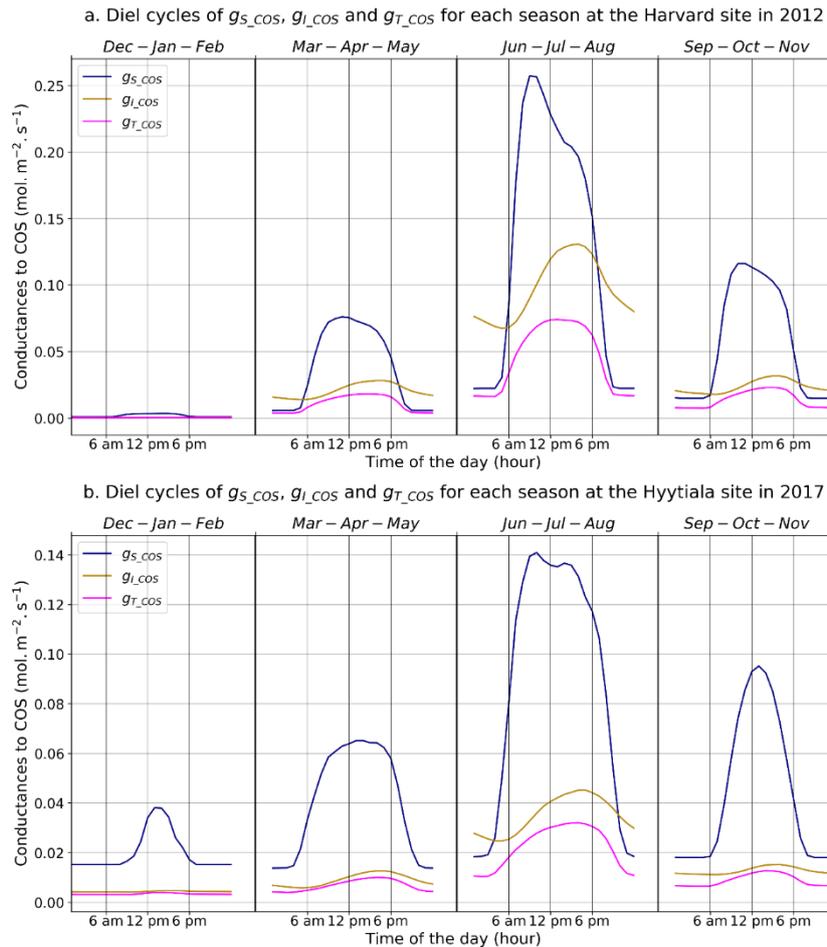


Figure R2.1: Updated Figure 1

The amplitude of the diel variation of the internal conductance is then much reduced (Figure R2.1). The diel variation of the V_{max} variable is based on a modified Arrhenius function calibrated on experimental data (Medlyn et al., 2002).

We thank the Referee for drawing our attention to the Stangl et al. (2019) study, and recall that the internal conductance includes both the mesophyll conductance and the biochemical CA activity. Stangl et al. (2019) studied the diurnal variations of stomatal and mesophyll conductances on mature *Pinus Sylvestris* trees in northern Sweden. Interestingly they found that the stomatal and mesophyll conductances estimated from measurements in June and July 2017 have significant diurnal variations (their Figure 2), with the stomatal conductance peaking between 9 and 10 am, earlier in the day than the mesophyll conductance. Albeit we don't know about the biochemical conductance, this is similar to what we see at Hyytiälä in the June-August period, and Kooijmans et al. (2019) found indications from branch measurements that the internal conductance may be limiting COS uptake during daytime.

The cited references encourage us to have a closer look at the mesophyll conductance, and we have updated our discussion on limiting conductances: “We have to acknowledge the large uncertainty regarding the modelling of the internal conductance. In parallel to optimizing the parameters of the internal conductance, an improvement could thus also be to replace it by the two factors it represents, i.e. the mesophyll conductance and CA activity. A model for the mesophyll conductance is already implemented in ORCHIDEE, with a simple parameter depending on temperature through a multiplication by a modified Arrhenius function following Medlyn and al. (2002) and Yin & Struik (2009). The impact of mesophyll conductance on photosynthesis and water use efficiency is now more studied (e.g. Buckley and Warren, 2014), even if its modelling remains challenging too: the temperature response has notably been reported as highly variable between plant species (von Caemmerer and Evans, 2015), which would imply having PFT-dependent parameters. Regarding measurements, ^{13}C discrimination of the isotopic composition of CO_2 exchanges allows for an estimation of the mesophyll conductance (Stangl et al., 2019).”

We agree a sensitivity analysis is required too, but this is a further large effort to be led in a separate study (see answer to comments [6] and (7)).

Medlyn, B. E., Dreyer, E., Ellsworth, D., Forstreuter, M., Harley, P. C., Kirschbaum, M. U. F., Le Roux, X., Montpied, P., Strassmeyer, J., Walcroft, A., Wang, K. and Loustau, D.: Temperature response of parameters of a biochemically based model of photosynthesis. II. A review of experimental data, *Plant, Cell Environ.*, 25(9), 1167–1179, doi:10.1046/j.1365-3040.2002.00891.x, 2002.

[3] One way to understand the relative roles of g_s , g_i , and the differences between the mechanistic model and the LRU approach is to look at values of intercellular and chloro-plast CO_2 concentrations (c_i and c_c respectively). These should be standard outputs of the model. ORCHIDEE c_i and c_c could be compared with “inferred” c_i and c_c using modeled A (GPP) and $g_{i,cos}$ and $g_{s,cos}$ following Seibt et al., 2010. It would be interesting to see if those differences can explain the difference in FOCS flux obtained from those two approaches (for e.g., in Figure 9).

Answer: We again clarify that what we called the “LRU approach” in this study was using a constant LRU value per large vegetation class. The equation (8) derived in Seibt et al. (2010) describing the variation of LRU as a function of the $g_{s,cos}$ to $g_{i,cos}$ ratio, and the C_i to $[CO_2]_a$ (C_a) ratio, is also valid in our mechanistic framework. So yes, the LRU variability can be mostly explained by these two ratios (the leaf boundary conductance has been neglected). The partial correlations analysis between the LRU and these two ratios yields similar values larger than 0.6 in absolute values at both sites, the Random Forest analysis also shows that both are equally important (not shown). We now indeed look at the C_i to C_a ratio (but not at C_c , as our modelled internal conductance includes both the mesophyll conductance and the biochemical CA activity). Considering a constant LRU thus may be seen as neglecting the variability of the C_i to C_a ratio.

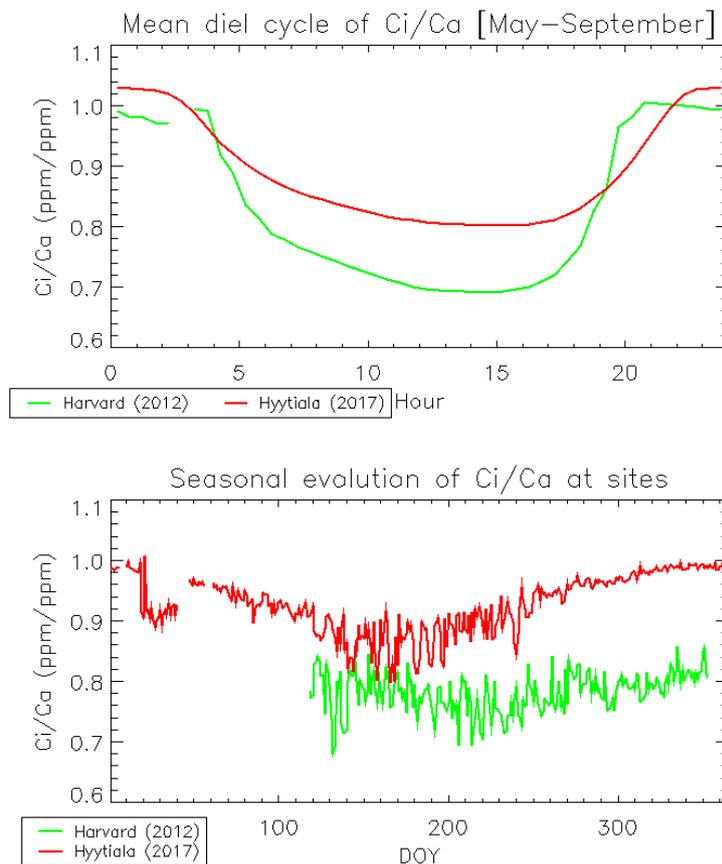


Figure R2.2: Top: Seasonal evolution of the simulated C_i to $[CO_2]_a$ (C_a) ratio at the Harvard Forest site in 2012 (green curve) and the Hyttialä site in 2017 (red curve). Bottom: Mean diel cycle over May–September

The modelled daily mean values for the C_i to C_a ratio computed at the two sites vary between 0.68 and 1.00 (Figure R2.2 top), on the upper part of the values reported by Seibt et al. (2010). The variations are also in agreement with Prentice et al. (2014) who state that the C_i to C_a ratio is pretty stable with only $\pm 30\%$ variations. The mean diel cycle between May and September has a U-shape with values decreasing during daytime (Figure R2.6 bottom), in coherence with former findings (Tan et al., 2017).

Given the non-linearity of the problem, the link between the C_i to C_a ratio and the LRU is less strong when dealing with monthly means at global scale, the partial correlation for all grid-cell-PFT drops to 0.3.

Prentice, I. C., Dong, N., Gleason, S. M., Maire, V. and Wright, I. J.: Balancing the costs of carbon gain and water transport: Testing a new theoretical framework for plant functional ecology, *Ecol. Lett.*, 17(1), 82–91, doi:10.1111/ele.12211, 2014.

Tan, Z. H., Wu, Z. X., Hughes, A. C., Schaefer, D., Zeng, J., Lan, G. Y., Yang, C., Tao, Z. L., Chen, B. Q., Tian, Y. H., Song, L., Jatoi, M. T., Zhao, J. F. and Yang, L. Y.: On the ratio of intercellular to ambient CO₂ (ci/ca) derived from ecosystem flux, *Int. J. Biometeorol.*, 61(12), 2059–2071, doi:10.1007/s00484-017-1403-4, 2017.

[4] In general, I find the manuscript too long and perhaps some figures (e.g. Figure 2) can be removed. Similarly, perhaps figures 3 and 4 can be combined. I greatly appreciate all the work that has gone in to this, but perhaps the authors ought to split this in two papers. One that describes the modeling framework in ORCHIDEE and another that focuses on the transport modeling? This would allow the authors to delve more deeply in the important findings such as Fig 8. Similarly, I can imagine that Fig 11 discussion can be greatly expanded upon.

Answer: As suggested by the Referee, we removed Figure 2 and combined Figures 3 and 4. We also moved Figure 10 in the Appendices.

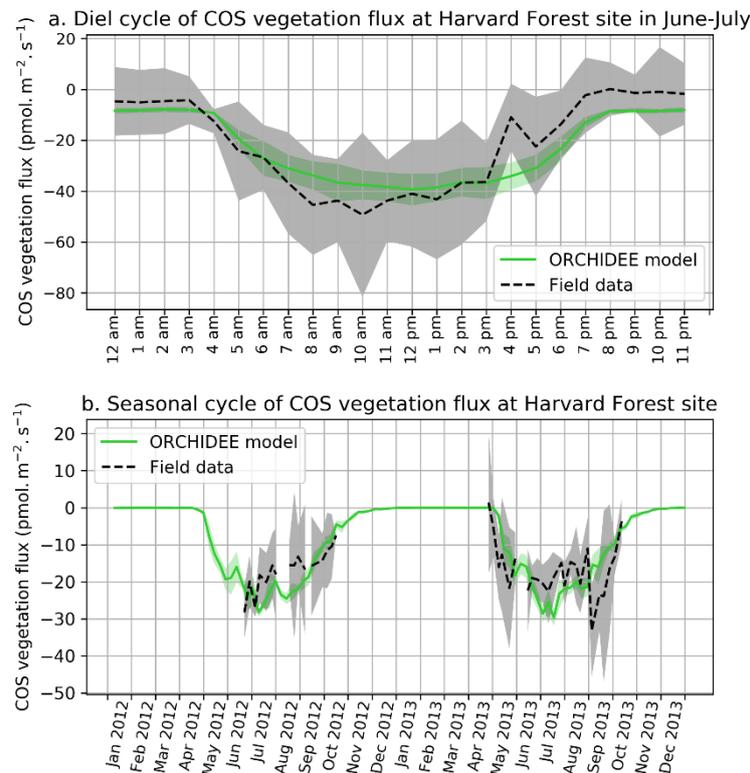


Figure R2.3 Combination of former Figures 3 and 4 for the Harvard Forest site

We acknowledge the fact that the manuscript is rich, but we would prefer to not split it. The transport part is not the focal part in this study, and Figure 11 is here to show that, at large scales, the mechanistic and LRU approaches provide similar results as evaluated by atmospheric COS concentrations. The result is important when it comes to use the LRU approach to estimate the GPP through inverse modelling of atmospheric measurements. However, we agree that there is much to say regarding transport and concentrations, and the transport model errors for COS will be assessed at NOAA sites by performing an intercomparison experiment with several transport models in a separate study; the other components of the COS budget (ocean, soil...) will be transported and a more complete comparison between observed and simulated concentrations will be made.

[5] I find it a little odd that LMDZ doesn't match the seasonal cycle of CO₂ at MLO. Is this a known issue of the transport model? In summary, I am not entirely convinced of the transport analyses since inferences could be flawed due to erroneous transport. Some quantification of transport error/uncertainty should be presented (perhaps using withheld/independent observations?).

Answer: A preliminary analysis between the TM5 and LMDz transport models suggests that transport errors are of second importance on the seasonal cycle at the NOAA stations compared to the impact of COS fluxes (Figure R2.4). The LMDz transport model is nudged toward ERA5 wind reanalysis, which prevents the large-scale advection of tracers from diverging. Note that the NOAA surface observations are independent as they have not

been used to calibrate the ORCHIDEE land surface parameters. Note also that the time lag in the CO₂ seasonal cycle at MLO appears when considering the land use change, harvest terms of the respiration. These terms can be poorly represented in land surface models and explain the seasonal lag.

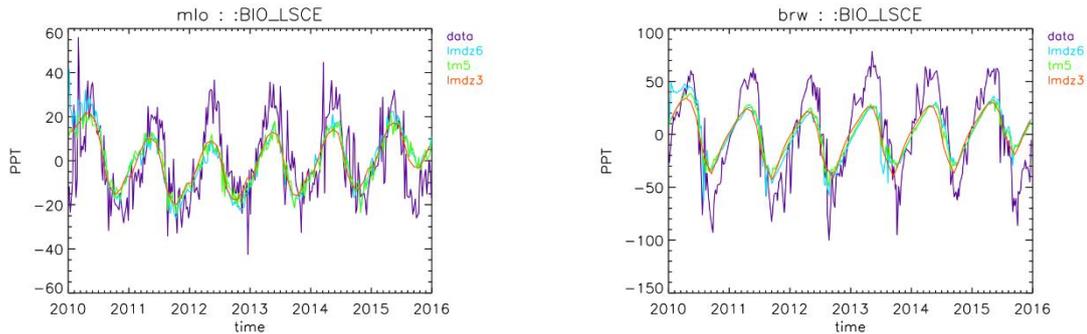


Figure R2.4: Detrended temporal evolutions of simulated and observed (purple curve) COS concentrations at two selected sites, for the mechanistic model ORCHIDEE, simulated with the LMDz3 (orange curve), LMDz6 (cyan curve) and TM5 (green curve) transport between 2010 and 2016. Left: Mauna Loa station (MLO, Hawaii), right: Barrow station (BRW, Alaska). The curves have been detrended beforehand and filtered to remove the synoptic variability (see Sect. 2.2.4 of the paper).

[6] *The writing in the results section is often very qualitative and not informative (e.g., line 329: “The conductances drop in the afternoon to reach minimum values at night”). I find most of the discussion unnecessary. It seems like a re-hash of the methodology and results section (e.g., Sec. 4.1.1. and 4.1.2), with inferences that I don’t think are always supported by the results (for instance lines 653-656. This is an example where the writing could be improved to make a very compelling argument about the use of OCS flux). Some of the work suggested (e.g in Sec. 4.1.3.) is well within the purview of this study and can be performed (ie., some sensitivity analyses of min gs and gi, and impact on simulated flux.).*

Answer: We usually describe what is seen on the Figures, as readers may not all be COS experts. Also, we provide numerous statistics (including RMSD, correlation coefficient, bias) to quantify model-data discrepancies. However, we tried to tighten the manuscript, where we thought it was possible without jeopardising the understanding.

We disagree with the Referee regarding the Discussion. The discussion is oriented on how to best exploit the results of this study in the optic of a further data assimilation to optimize parameters, and really use COS information to improve the simulated GPP.

We agree a sensitivity study of the fluxes to the parameters, including minimal stomatal conductance, is further needed, but as stressed by the Referee, the manuscript is already long and this sensitivity study will be led in a subsequent study, associated with the optimisation of the most significant parameters identified in the sensitivity study.

[7] *The writing overall really needs to be tightened and the conclusions seem a bit weak. This could be improved with better framing. Thus I believe that with a much clearer presentation of figures and text, this could be a much more compelling manuscript.*

Answer: The number of Figures has been reduced from 11 to 8. The manuscript is now more clearly focused on the evaluation of the mechanistic model, and its use to study the LRU variability. We have rephrased and tightened the beginning of the “Conclusions and Outlooks” section to better emphasize our main findings:

“We have implemented inside the ORCHIDEE land surface model the mechanistic model of Berry et al. (2013) for COS uptake by the continental vegetation. Modelled COS fluxes were compared at site scale against measurements at the Harvard temperate deciduous broadleaf forest (USA) and at the Hyytiälä Scots pine forest (Finland), yielding relative RMSDs of around 40% at both diel and seasonal scales. **We found that the mechanistic model yields a lower and thus more limiting internal conductance as compared to former works (Seibt et al., 2010; Wehr et al., 2017).** The next step is to perform a sensitivity analysis (Morris, 1991; Sobol, 2001) and to optimize the most sensitive parameters related to the modelled fluxes and conductances, to get a better agreement with observations.

Our global estimate of COS uptake by continental vegetation of -756 Gg S yr⁻¹ is in the lower range of former studies. **An important finding is that the LRU computed from monthly values of the COS and GPP fluxes yields values lower than monthly means of high-frequency LRU values.** This has consequences for atmospheric studies where COS concentrations integrate influences from fluxes at large spatial and temporal scales.”

Morris, M. D.: Factorial Sampling Plans for Preliminary Computational Experiments, *Technometrics*, 33(2), 161, doi:10.2307/1269043, 1991.

Sobol, I. M.: Global sensitivity indices for nonlinear mathematical models and their Monte Carlo estimates., 2001.

[8] *Comments about figures and tables: Figure1: Conductance seems to be highest at 12 am ie., midnight? Growing season midday at Harvard Forest is shown to be limited by g_i but Wehr et al (2017) measurements show that g_s is the limiting conductance to OCS transfer. How do these numbers compare with Wehr and Kooijmans measurements (of g_i)? You could also add total conductance to OCS (the quantity multiplied to $[OCS]_a$ in eq 3). In general, since you don't seem to be explaining month by month variations, maybe compress these to show 3 month means (e.g. JJA, SON etc)? Currently, it is impossible to see in detail the diurnal variations, specially when one is trying to discern at what times of day $g_i > g_s$ and vice versa.*

Answer: We indeed made a mistake and have corrected '12 am' into '12 pm'. We have now grouped the months per season, and added the total conductance, as suggested by the Referee (see new Figure in comment [2]).

We have to remember that the modelled internal conductance gathers both the mesophyll conductance and the biochemical conductance representing the Carbonic Anhydrase (CA) activity. We have now detailed more precisely the comparison with Wehr et al. (2017) in the "Modelled conductances" section: "For the Harvard Forest site, Wehr et al. (2017) computed the stomatal conductance using both a water flux method and a COS flux method, and obtained a close agreement between two different methods; the mesophyll conductance is modelled using an experimental temperature response, and the biochemical conductance, representing CA activity, is modelled using a simple parameter ($0.055 \text{ mol m}^{-2} \text{ s}^{-1}$), both scale with LAI to get canopy estimates. Wehr et al. (2017) found similar maximum values around $0.27 \text{ mol m}^{-2} \text{ s}^{-1}$ during daytime, from May to October, for the stomatal conductance and for the biochemical conductance (their Figure 4); adding the slightly larger mesophyll conductance (peaking around $1.0 \text{ mol m}^{-2} \text{ s}^{-1}$) to the biochemical conductance would thus also lead to a more limiting role of the internal conductance (peaking around $0.21 \text{ mol m}^{-2} \text{ s}^{-1}$) during daytime, albeit not as strong as for the modelled one (peaking around $0.13 \text{ mol m}^{-2} \text{ s}^{-1}$); the simulated stomatal conductance exhibits minimum and maximum values similar to the observations-based ones, but peaks more sharply in the morning". It is more difficult to compare absolute levels of conductances for Hyytiälä as the observations are made at branch level.

[9] *Figure 2. I believe this figure should be replaced by scatterplots or a table listing correlations between g_s , g_i and aux variables (which already exists as Table 4). Why would g_i be expected to scale with soil moisture? It is strange that PAR is highest in May at Hyytiälä.*

Answer: We have removed Figure 2, updated Table 4 and added a Random Forest analysis (see our answer to comment [10] below). We double-checked for PAR and found the same maximum in May. It can be observed in Kooijmans et al. (2019), their Figure S1 shows a long series of high PAR in May, followed by a series of much more varying PAR. This is not always the case, but in 2017 the other summer months were very cloudy and rainy in Southern Finland, reducing PAR (both midday value and monthly average); in May midday PAR values can indeed be very high and in recent years have been higher than June/July midday PAR in at least 2013, 2016 and 2017 (K.M. Kohonen, personal communication). We looked at soil moisture because g_{I_COS} is linearly related to V_{max} , which is modulated by a water stress factor depending on the soil moisture (de Rosnay and Polcher, 1998). However, both sites did not experience a strong water stress during the examined years (Table R1.1).

[10] *Table 4: g_s is more related to T_{air} than to VPD. Stomatal conductance has been shown to be related to VPD (See for e.g., Oren et al., 1999). This would be worth examining.*

Answer: Indeed equations (15) and (15a) in Yin and Struik (2009) give the explicit dependency of the stomatal conductance on VPD, but g_{S_COS} also depends on CO_2 assimilation (A), which depends primarily on PAR, and on air temperature. Table 4 was updated following the clarification that we were discussing canopy-scale conductances, which should then also depend on LAI (Table R2.1).

Table R2.1: Partial correlations linking stomatal and internal conductances to photosynthetically active radiation (PAR), air temperature (T_{air}), vapour pressure deficit (VPD), soil moisture (SM) and leaf area index (LAI), computed at a half-hourly time step over year 2012 at the Harvard Forest site and 2017 at the Hyytiälä site

Conductance	Site	PAR	T_{air}	VPD	SM	LAI
g_{S_COS}	Harvard	0.66	0.46	-0.61	-0.04	0.33
	Hyytiälä	0.59	0.49	-0.47	-0.03	0.25
g_{I_COS}	Harvard	-0.06	0.68	0.30	-0.27	0.15
	Hyytiälä	-0.13	0.74	0.65	0.32	0.49

For stomatal conductance, this updated Table gives the second role to VPD at Harvard and similar second roles to T_{air} and VPD at Hyytiälä. The Random Forest analysis, which we added given the non-linearity of the problem stressed by Referee 1, confirms the main role of PAR , and ranks VPD as the third most important variable at both sites (Figure R2.5).

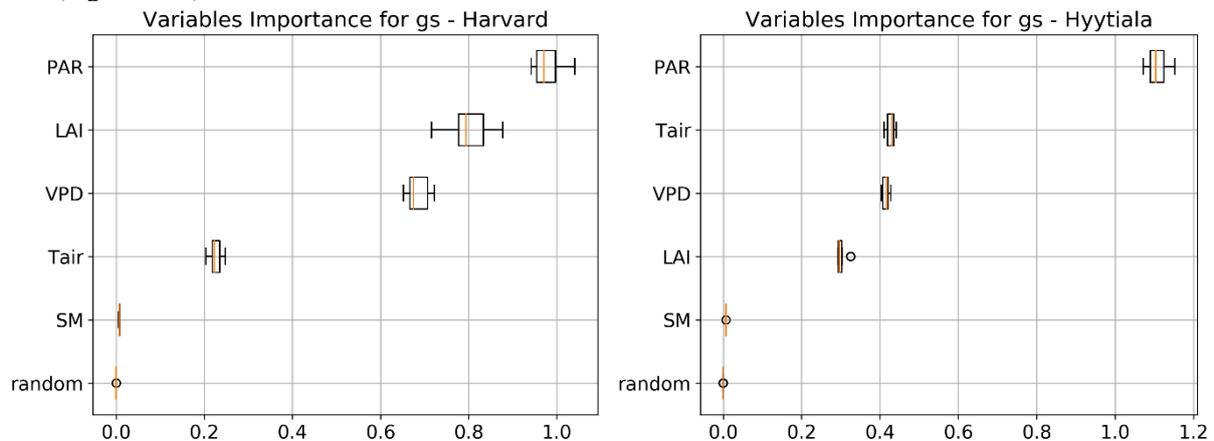


Figure R2.5: Variables importance computed using Random Forests for the stomatal conductance (g_s) at the Harvard Forest site in 2012 (left) and at the Hyytiälä site in 2017 (right). The considered predictors are air temperature (T_{air}), leaf area index (LAI), soil moisture (SM), vapour pressure deficit (VPD) and photosynthetically active radiation (PAR). A random predictor is added to prevent over-fitting.

[11] Figure 4. Doesn't seem that Harvard Forest 2013 observed fluxes match simulations all that well. For instance, fluxes seem to peak in May-June and Aug-Sep in 2013, but peak in July in the model. There is a mention of "noise" in EC based measurements in the text but these should be quantified or at least described.

Answer: We have detailed a bit the description following the Referee's comment: "The simulated weekly seasonal vegetation COS uptake roughly follows the same trend as the observed one ($r=0.53$). COS uptake increases in spring when the vegetation growing season starts and decreases in autumn at the end of the forest activity period. Simulated and observed fluxes also take similar values over the two years. There are however differences: in 2013 the start of the season is simulated about two weeks too late in May instead of late April, and measured fluxes peak in May-June and August-September, while the modelled fluxes peak in July". We have also clarified our meaning regarding the uncertainty of ecosystem flux measurements: "Kohonen et al. (2020) have quantified the relative uncertainty of weekly-averaged ecosystem COS fluxes at 40%, which is coherent with the large standard-deviation computed for field data".

[12] Figure 5. It would be easier to view this figure as two separate panels instead of one plot with two y axes.

Answer: We have split the Figure in two separate panels.

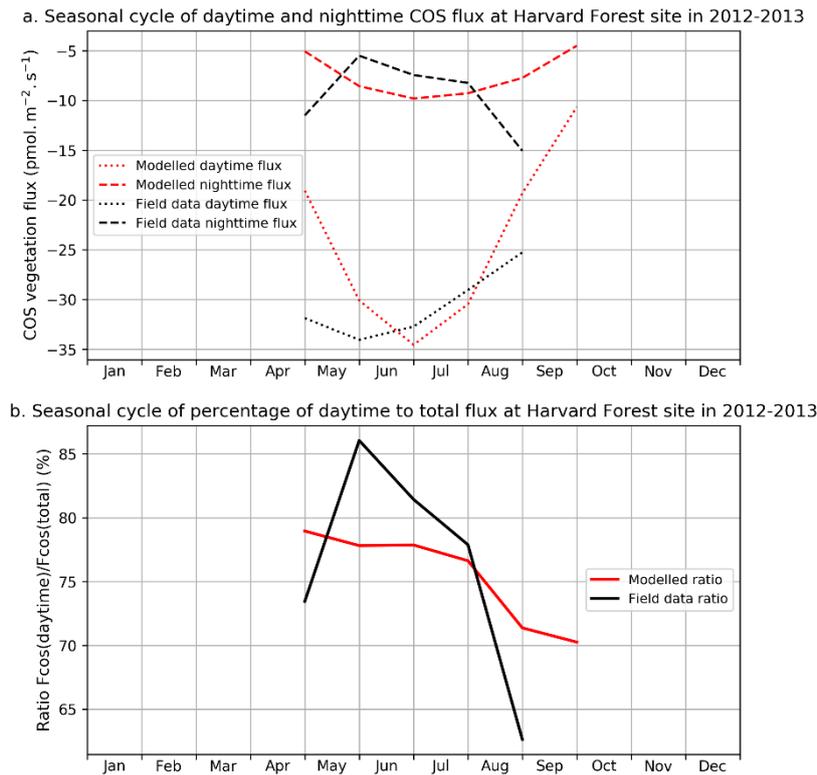


Figure R2.6 Updated Figure 5

[14] *Minor Comments: Note, I stopped providing minor comments because of the length of the manuscript.*
 Answer: Thanks anyway for providing them.

[15] *Line 70: remove “then” in COS is then hydrolyses*
 Answer: We have removed it.

[16] *Line 77: mention that soils can be a sink or a source, and cite appropriate studies (cite appropriate studies e.g., Maseyk et al, 2014; Berkelhammer et al, 2014; Kitz et al., 2020). Also mention the role of epiphytes in the OCS budget (and cite Kuhn et al., 2000; Gimeno et al., 2017 and Rastogi et al, 2018) .*

Answer: Regarding soils and epiphytes, we have expanded the presentation on LRU estimation: “LRU can be estimated experimentally, and then used as a scaling factor for estimating GPP, if F_{COS} , $[COS]_a$ and $[CO_2]_a$ are available. Measurements can be made at leaf level using branch chambers (Seibt et al., 2010; Kooijmans et al., 2019); LRU can also be estimated at ecosystem level: eddy-covariance flux towers measure the ecosystem total COS flux (Kohonen et al., 2020), removing the soil contribution gives access to the vegetation part (Wohlfahrt et al., 2012; Wehr et al., 2017). Soil can absorb and emit COS (Whelan et al., 2016; Kitz et al., 2020), the magnitude of their flux being generally much lower than that of vegetation fluxes (Berkelhammer et al., 2014; Maseyk et al., 2014; Wehr et al., 2017; Whelan et al., 2018). Epiphytes (lichen, mosses) could also have a significant contribution to the ecosystem COS budget (Kuhn and Kesselmeier, 2000; Rastogi et al., 2018)”. Gimeno et al. (2017) was cited earlier in the introduction, following a comment by the other Referee: “It is however to be noted that Gimeno et al. (2017) reported COS emissions by bryophytes during daytime”.

[17] *Lines 78-85: Add some discussion about possible scaling issues for LRU from leaf to canopy and ecoregion scales. I think some of this framework can be found in Wohlfahrt et al., 2012*

Answer: Regarding the scaling issue in measurements, see in the above comment [16] the modifications that were made in the manuscript. We have also clarified how we integrate fluxes and conductances at canopy and ecoregion scales in ORCHIDEE. In the presentation of ORCHIDEE, we have added: “The canopy is discretized in several layers of growing thickness, the number depending on the actual Leaf Area Index (LAI). The CO_2 assimilation, the stomatal conductance and the intercellular CO_2 concentration C_i are computed per LAI layer, provided LAI is higher than 0.01 and the mean monthly temperature is higher than $-4^{\circ}C$. The CO_2 assimilation, the stomatal conductance are further summed-up over all layers to compute GPP and the total conductance at canopy level. The scaling to the grid cell is made using means weighted by the Plant Functional Types fractions”. Similarly, in the presentation of the Berry model, we have added: “The vegetation COS flux and related conductances are computed

for each LAI layer, and then summed-up to get total values at canopy level. Unless specified otherwise, fluxes, conductances and LRU are further presented and discussed at canopy level”.

[18] Line 90: *summer is likely only valid for ecosystems that exhibit seasonality. Explain diurnal and seasonal variability in LRU.*

Answer: Yes, we have clarified the sentences: “Because of these different responses of COS and CO₂ uptake in leaves, LRU varies with light conditions, and decreases sharply with PAR increase (Stimler et al., 2010, 2011; Maseyk et al., 2014; Commane et al., 2015; Wehr et al., 2017; Yang et al., 2018). Consequently, LRU values are smaller at midday or in seasons with high incoming light (Kooijmans et al., 2019)”.

[19] Line 95: *What do you mean by ‘time’?*

Answer: We refer here to variability of LRU over the day and season inferred by changes in light-conditions. We have changed the sentence “The variability of LRU with plant type, light, and time should therefore...” in “The variability of LRU with plant type and over a day and season (inferred by changes in light-conditions) should therefore...”.

[20] Line 112-13: *remove these lines.*

Answer: Albeit this is the classical way to present an article structure in many journals, we removed these lines in our effort to tighten the narration.

[21] Line 121: *Briefly describe improvements in the Farquhar model.*

Answer: We added: “A main novelty is the introduction of a mesophyll conductance linking the CO₂ concentration at the carboxylation sites, C_c , to the intracellular concentration, C_i .”. Later is also specified: “The temperature-dependence of the maximum photosynthetic capacity follows Medlyn et al. (2002) and Kattge and Knorr (2007).”

[22] Lines 208-210: *Provide scientific names for these species at Harvard Forest.*

Answer: Yes, we have added them: “red oak (*Quercus rubra*), red maple (*Acer rubrum*) and hemlock (*Tsuga canadensis*)”.

[23] Lines 218-220: *Awkward phrasing. Also, please elaborate what you mean by “when possible”.*

Answer: We were referring to the uncertainties that are later inferred from using different LRU datasets (70% line 472) or different GPP datasets (40% line 464). We removed the part “evidencing some uncertainties when possible” for the sake of simplicity.

[24] Line 339: *change ‘air surface temperature’ to ‘air temperature’.*

Answer: We made the change.

[25] Line 339: *Very minor comment: ‘modelling’ and ‘vapor’. The first is a “British” spelling and the second is American”. Please pick one and be consistent throughout.*

Answer: Thanks, the British spelling has been selected, we corrected “vapor” into “vapour”.

[26] Line 348: *This isn’t true based on Fig 2 as PAR peaks in May but gs in June at Hyttiälä.*

Answer: The Referee is correct. Regarding the PAR seasonal cycle, some additional information was provided in our answer to comment [9]. Following comments by both Referees, this section was largely modified: Figure 2 was removed and a Random Forest analysis was added to the initial partial correlations. The text for $g_{S_{COS}}$ and PAR now simply reads: “PAR is the most important variable for the stomatal conductance at the two sites. Due to the way of how $g_{S_{COS}}$ is simulated according to Yin and Struik (2009), there is a linear relationship with the CO₂ assimilation, which depends mainly on PAR.”

[27] Lines 356-358: *Needs citations.*

Answer: We added Berry et al. (2013) regarding the modelled internal conductance, and Yin and Struik (2009) for the dependency of V_{max} on air temperature.

References:

Berkelhammer, M., Asaf, D., Still, C., Montzka, S., Noone, D., Gupta, M., Provencal, R., Chen, H. and Yakir, D., 2014. Constraining surface carbon fluxes using in situ measurements of carbonyl sulfide and carbon dioxide. *Global Biogeochemical Cycles*, 28(2), pp.161-179.

Buckley, T.N. and Warren, C.R., 2014. *The role of mesophyll conductance in the economics*

of nitrogen and water use in photosynthesis. *Photosynthesis research*, 119(1-2), pp.77-88.

Gimeno, T.E., Company, C.E., Drake, J.E., Barton, C.V., Tjoelker, M.G., Ubierna, N. and Marshall, J.D., 2020. Whole-leaf mesophyll conductance reconciles isotopic and gas-exchange estimates of water-use efficiency. *New Phytologist*.

Gimeno, T.E., Ogée, J., Royles, J., Gibon, Y., West, J.B., Burlett, R., Jones, S.P., Sauze, J., Wohl, S., Benard, C. and Genty, B., 2017. Bryophyte gas-exchange dynamics along varying hydration status reveal a significant carbonyl sulphide (COS) sink in the dark and COS source in the light. *New Phytologist*, 215(3), pp.965-976.

Kitz, F., Spielmann, F.M., Hammerle, A., Kolle, O., Migliavacca, M., Moreno, G., Ibrom, A., Krasnov, D., Noe, S.M. and Wohlfahrt, G., 2020. Soil COS exchange: a comparison of three European ecosystems. *Global Biogeochemical Cycles*, 34(4), p.e2019GB006202.

Maseyk, K., Berry, J.A., Billesbach, D., Campbell, J.E., Torn, M.S., Zahniser, M. and Seibt, U., 2014. Sources and sinks of carbonyl sulfide in an agricultural field in the Southern Great Plains. *Proceedings of the National Academy of Sciences*, 111(25), pp.9064-9069.

Oren, R., Sperry, J.S., Katul, G.G., Pataki, D.E., Ewers, B.E., Phillips, N. and Schäfer, K.V.R., 1999. Survey and synthesis of intra- and interspecific variation in stomatal sensitivity to vapour pressure deficit. *Plant, Cell & Environment*, 22(12), pp.1515-1526.

Rastogi, B., Berkelhammer, M., Wharton, S., Whelan, M.E., Itter, M.S., Leen, J.B., Gupta, M.X., Noone, D. and Still, C.J., 2018. Large uptake of atmospheric COS observed at a moist old growth forest: Controls and implications for carbon cycle applications. *Journal of Geophysical Research: Biogeosciences*, 123(11), pp.3424-3438.

Sage, R.F. and Kubien, D.S., 2007. The temperature response of C3 and C4 photosynthesis. *Plant, cell & environment*, 30(9), pp.1086-1106.

Seibt, U., Kesselmeier, J., Sandoval-Soto, L., Kuhn, U. and Berry, J.A., 2010. A kinetic analysis of leaf uptake of COS and its relation to transpiration, photosynthesis and carbon isotope fractionation. *Biogeosciences*, 7(1).

Stangl, Z.R., Tarvainen, L., Wallin, G., Ubierna, N., Röntfors, M. and Marshall, J.D., 2019. Diurnal variation in mesophyll conductance and its influence on modelled water-use efficiency in a mature boreal *Pinus sylvestris* stand. *Photosynthesis research*, 141(1), pp.53-63.

von Caemmerer Susanne and Evans, J.R., 2015. Temperature responses of mesophyll conductance differ greatly between species. *Plant, Cell & Environment*, 38(4), pp.629-637.

Wohlfahrt, G., Brilli, F., Hörtnagl, L., Xu, X., Bingemer, H., Hansel, A. and Loreto, F., 2012. Carbonyl sulfide (COS) as a tracer for canopy photosynthesis, transpiration and stomatal conductance: potential and limitations. *Plant, cell & environment*, 35(4), pp.657-667.

Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2020-381>, 2020.