

Response to reviewer 2

“Rastogi et al. present observed patterns of OCS uptake in an old-growth forest during 2015. Their observations are consistent with previous studies in similar ecosystems, and are valuable in corroborating those studies and in confirming that the community’s general understanding of OCS uptake holds in old-growth forests. The methods seem valid, subject to some concerns noted below. The manuscript is easy to read and clearly organized. This is not a manuscript that presents new insights or methods.”

We thank the reviewer for a very thoughtful and detailed response to our submitted manuscript. However, we would like to disagree that this method presents no new insights or methods. In this work, we propose a simple model to estimate ecosystem- scale leaf OCS fluxes from concentration measurements, using other novel measurements, such as direct measurements of canopy skin temperature, using a thermal camera. This model is based on a theoretical framework laid out by Seibt et al., (2010) and Wohlfahrt et al., 2012), and supported by other seminal ecosystem-scale studies relating OCS uptake to plant productivity or GPP (Commane et al., 2015) and stomatal conductance (Wehr et al., 2017). In addition, we show the response of inferred OCS fluxes to the diffuse fraction of downwelling radiation, as well as the response of OCS fluxes to sequential heatwaves. These responses have not been reported for any ecosystems as yet, and we hope they provide important constraints on the use of OCS as a tracer for stomatal conductance and ultimately GPP.

“Unfortunately, the inferences drawn from the observations in the manuscript are not quantitatively supported. In particular, the inferences about stomatal responses to soil moisture and heat waves seem to be not only unsupported but also incorrect (see below). The core weakness of the manuscript, which contributes to the inference problem just mentioned, is that it is overly descriptive in terms of both the analysis and the writing. In terms of the analysis, 5 of the 6 figures (and all but subsection 3.4 of the text) present time series of data, and the associated analysis is restricted entirely to ‘eyeballing’ correlations between those time series. The authors do not calculate correlation coefficients, or use multiple regression or a simple model to support their causal inferences. In terms of the writing, many patterns in the data are described in the text even though they aren’t clearly connected to any conclusions. The manuscript would be more effective if it were to focus on what was learned from the data, referring to the data as necessary to support those findings. Other patterns could be gleaned from figures or tables by any reader with a particular interest.”

We appreciate these suggestions and have reworked specific parts of the manuscript to provide more quantitative comparisons, as well as changed the language of our study that relates to soil moisture. We have also tidied up the manuscript so that it reads more cleanly.

Specific Comments:

“- lines 193-195: This justification doesn't make sense to me. The resistance to turbulent eddy transport through open air from 70m to 60m should be much less than the resistance to eddy transport through the dense canopy from 60m to the leaf surfaces. If the aim is to establish the gradient across only the stomata, then using 60m instead of 70m hardly helps. The full transport resistance from the tower top to the substomatal cavity of some particular leaf is $r_{ac} + r_{wc} + r_{lbl} + r_s$, where r_{ac} and r_{wc} are the above-canopy and within-canopy turbulent eddy resistances, r_{lbl} is the leaf boundary layer resistance, and r_s is the stomatal resistance. Of these, r_{ac} is probably negligible, r_s is probably most limiting, and r_{lbl} is probably second most limiting. The authors appear to have neglected r_{lbl} and r_{wc} , so that their G_c is not exactly the canopy-scale stomatal conductance but rather a canopy-scale combination of the stomatal, leaf boundary layer, and within-canopy turbulent eddy conductances, i.e. $G_c = 1/(r_{wc} + r_{lbl} + r_s)$. It is possible to measure r_{ac} and some portion of r_{wc} by comparing concentration measurements within the canopy to those above the canopy and using flux = conductance x gradient; this approach ought to be superior to using the theoretical u^2/u . In any case, the authors should clarify what they mean by “the” boundary layer, as the boundary layer that is usually discussed in the context of stomatal uptake is the leaf boundary layer, i.e. the thin layer of stagnant air against the surface of individual leaves, through which gas transport is substantially diffusive rather than advective or convective. Transport through the canopy airspace, or the 10m above the canopy, on the other hand, will not be diffusive at all. “

We would again like to thank the reviewer for such a carefully detailed and clear comment regarding conductance. We have rephrased this text in the original manuscript, which we acknowledge was incorrect. We agree with the reviewer that the transport between 70-60m is in fact turbulent (and therefore more related to r_{ac} than to r_{lbl}). The choice to use the canopy top mixing ratios is following previously published literature (Fares et al., 2012; and references therein). We are, however, not ignoring the leaf boundary layer in our formulation (i.e. eqs, 1-3). Here we argue (following previous studies, cited above) that the ratio of fluxes of two scalars (in this case, OCS and H₂O) across the leaf surface is proportional to the gradient between the ambient air and the leaf intercellular spaces, i.e.

$$\frac{F_{OCS}}{F_{H_2O}} = \frac{OCS_a - OCS_i}{(e_i - e_a) \cdot P^{-1}} \cdot 1.94^{-1} \quad (1)$$

where F_{OCS} and F_{H_2O} are fluxes of OCS and H₂O (in units of $\text{pmolm}^{-2}\text{s}^{-1}$ and $\text{mmolm}^{-2}\text{s}^{-1}$ respectively), OCS_a and OCS_i represent ambient and intercellular mixing ratios of OCS respectively (ppt), where $VPD_1 = e_i - e_a$ (e_i and e_a are intercellular and actual vapor pressure; kPa), and P is atmospheric pressure (kPa). We had incorrectly labelled saturated leaf pressure

obtained from leaf temperatures as e_s , and have now correctly labelled this as e_i (since it is the leaf intercellular spaces that are assumed to be saturated with water vapor, not the leaf surface). Finally, the factor 1.94 reflects the diffusivity ratio of OCS and H₂O.

To investigate the reviewer's concern regarding various resistances, we additionally estimated OCS fluxes according to the following equation

$$F_{OCS} = \left(\frac{1.56}{G_{bw}} + \frac{1.94}{G_{sw}} \right)^{-1} \cdot OCS_a \quad (2)$$

where G_{bw} and G_{sw} are the canopy- scale boundary layer and stomatal conductances for water vapour transport. The constants 1.56 and 1.94 are the ratios of diffusivities of OCS and H₂O under turbulent and diffusive flow (Seibt et al., 2010).

We derived G_{bw} by first estimating roughness parameters following Monin-Obukhiv similarity theory (Foken, 2006). These were then used to obtain stability parameters for momentum transport, which was finally used to estimate G_{bw} following Su et al., (2001). Code and further information can be found within the R package "bigleaf" and accompanying manual (<https://bitbucket.org/juergenknauer/bigleaf>). G_{sw} was estimated as

$$G_{sw} = (G_{cw}^{-1} - G_{bw}^{-1})^{-1} \quad (3)$$

where G_{cw} is the canopy (surface) conductance to water vapor transport. To address the reviewer's comments regarding the use of the Penman- Monteith method to estimate G_{cw} , we used a simple flux-gradient method to infer this conductance as follows:

$$G_{CW} = F_{H2O} \cdot \left(\frac{VPD}{P} \right)^{-1} \quad (4)$$

Where, VPD and P are the vapor pressure deficit and atmospheric pressure (both in units of kPa). Estimated conductances and F_{OCS} calculated using both approaches is shown in figure 1. Similar to Wehr et al., (2017), we find that the boundary layer conductance is not limiting at our site, and therefore $G_{sw} \sim G_{cw}$. Consequently, the resulting flux of OCS from the two methods of estimates of G_s are in fact not dissimilar (especially considering the variability around the means shown in Fig. 1b). Therefore, we decided to trust our simple method since it does not depend on theoretical formulations of stability.

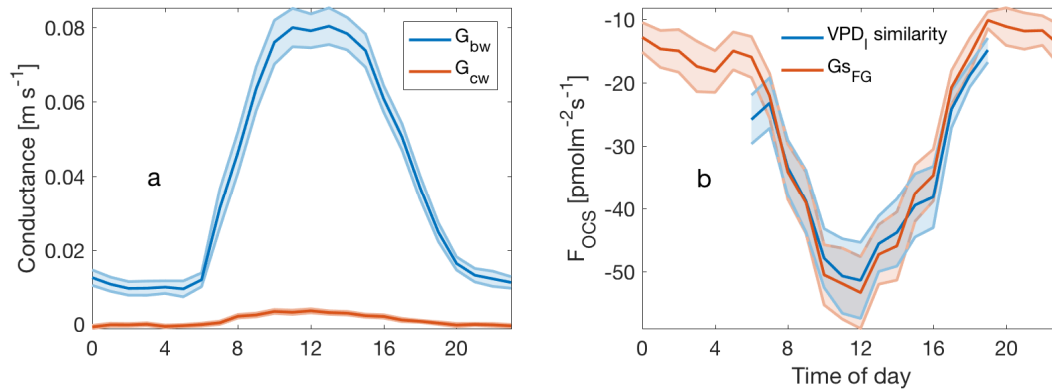


Figure 1. Mean diurnal cycles of boundary layer and canopy conductance to water vapor transport (a), and resulting OCS flux (b).

“line 223: You are talking about computing the change in canopy storage, which is a good idea, but why not do that at all times? In general, the flux through the stomata should be equal to the flux past the eddy flux sensor plus the flux into the canopy storage airspace, i.e. Eq. 1 should have a storage term appended to the right hand side (perhaps you used to have one there, as suggested by your reference to the “first term in right hand side of equation (1)” on line 221?). Here you are saying that when the eddy flux term was near zero, you considered the storage flux term. But the storage flux term might be substantial even when the eddy flux term is not near zero.”

We have revised this to exclude nighttime data, and periods when the eddy flux is near zero. In tall canopies such as our site, the portion of canopy that is coupled to the overlying atmosphere changes considerably during the day, and parts of the lower canopy are likely to be always decoupled from the upper canopy as well as above canopy air (Pyles et al., 2004). This has obvious consequences on canopy storage and venting of gases such as CO_2 and OCS. We have therefore excluded the storage flux entirely from our estimates of F_{OCS} . Moreover, change in storage flux leads to a change in mixing ratios (increase during the day), and is implicit in our formulation of F_{OCS} . We acknowledge that storage fluxes provide an important portion of the ecosystem exchange of gases such as CO_2 (and OCS) that is missed by the eddy flux measurement, but our approach doesn’t aim to infer a turbulent flux. Instead, the goal behind this study is to estimate a ‘leaf- flux’, assuming that the canopy acts like a big-leaf.

“line 236 (Eq. 6): Given the considerations about energy imbalance and the PM equation raised by Wohlfahrt et al., 2009 (Agricultural and Forest Meteorology 149, 1556– 1559) and by Wehr et al., 2017 (Biogeosciences 14, 389–401), it should be stated why this particular form of the PM equation was used (or why the PM equation was used at all instead of just using sensible heat flux measured from the tower). Those papers indicate that retrieved values of stomatal

conductance can be substantially affected by the choice of equation.”

We acknowledge the reviewer’s concern regarding the use of the Penman-Montieth method to estimate canopy conductance and have now changed the analyses to use the equation presented in eq. 4.

“- line 329: Regarding “declined precipitously with soil moisture”, it is a bit hard to tell from the color scale in Fig. 3a, but it looks like the decline in OCS (which matches the decline in G_c), is better correlated with the rise in VPD than with the drop in soil moisture. People often assume that soil moisture drives seasonal patterns in stomatal conductance (and it surely does at some sites), but it is also possible that the seasonal pattern in G_c and F_{OCS} is explained entirely by VPD (that was the finding for the mesic Harvard Forest site used in the Wehr et al., 2017 paper you cite). It would be interesting to try to disentangle those two water-related drivers here, at least with a simple regression approach.”

Yes, we agree with the reviewer on this. However, since we explicitly use VPD to estimate F_{OCS} , it would be circular for us to explain variability in F_{OCS} as a function of VPD. The idea behind showing the relationship with soils moisture was a way to link overstory canopy processes, with changes in soil moisture.

“- line 380-1: Did the estimation of ER from the tower include measurements of canopy CO_2 storage? If turbulence is low at night, most of the respired CO_2 is probably accumulating in airspaces below the eddy flux sensor.”

Flux tower estimates of CO_2 flux at the site do not incorporate storage (as computed by a profile). This is in part due to large horizontal advective losses that we are unable to estimate (Sonia Wharton, *pers. comm*) within the tall old-growth canopy. This is another reason why we chose to ignore storage estimates of OCS from this analysis.

“- line 427-9: I don’t see how this inference is connected to the preceding observations, and I don’t see any evidence in the manuscript that soil moisture (as opposed to VPD) is limiting gas exchange.”

We have changed the language in the manuscript. We also provide a simple linear regression that quantifies the relationship of F_{OCS} with soil moisture (Fig. 3b).

“- line 452-5 and 483-4: These inferences are flawed. It is not true that “canopy scale stomatal conductance during these events is dramatically reduced”. Figure 6 shows that G_c was not reduced at all during the first heat wave, and was not reduced until the end of the second heat wave, at which point the water flux also dropped. During the third heat wave, G_c was reduced,

but the water flux did not increase. Even more importantly, Gc was estimated based on the assumption that the water flux was exclusively transpiration, so it makes no sense to say that the behavior of Gc implies the increased water flux was not transpiration. If the approach used to calculate Gc is valid, then the increased water flux was indeed due to increased transpiration, on account of an increased VPDL.”

We agree with the reviewer that higher water flux is likely due to increased transpiration under high VPD, and have changed the language to reflect this (lines 425-432).

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