Dear reviewers,

Thank you both very much for the time you have spent on reading our manuscript, and in particular for your constructive comments, which helped to improve our manuscript. Please find a point-to-point reply to your comments below, sorted per reviewer.

Note that, besides the reviewer suggestions, we also made some additional changes to the manuscript to improve general readability and quality.

Reply to reviewer 1, Dr. Joseph Berry:

This paper analyzes OCS exchange in pine canopies using inversion of an intermediate-scale model that simulates vertical gradients of temperature, humidity, and wind speed, along with trace gas concentrations in the canopy, coupled to a convective boundary layer and the overlying troposphere. The main focus is on OCS exchange and on the parameter LRU, which relates the deposition velocities of CO2 and OCS to their local concentrations. The inversions indicate a vertical gradient of LRU within the canopy, which is qualitatively consistent with previous studies showing that LRU responds to PAR and VPD. This modeling framework is unique and potentially valuable for interpreting eddy covariance observations of OCS exchange.

We would like to thank the reviewer for the time invested in reading our manuscript and for the constructive comments, which helped to improve our manuscript. The point-to- point response to the comments follows below, the reviewer's text is given in black, and our response in red.

However, the emphasis on the absolute values of LRU produced by the model appears overstated. Robust evaluation of these values requires careful consideration of (i) the observational constraints used in the inversion and (ii) the realism of the model parameterizations. While the study uses a widely applied parameterization for OCS exchange, the descriptions of CO2 uptake and stomatal conductance—both critical for determining LRU—are unfamiliar, non-standard, and not well explained in the main text. Beyond the list of equations in the supplementary material, there is no clear description of the response characteristics of this parameterization or how it compares with more established approaches. A direct comparison with the parameterization used by Kooijmans et al. at this site would be particularly informative.

For the photosynthesis and stomatal calculations we follow the A-gs approach. Although it is indeed not the most widely used model for leaf photosynthesis, it is still a well-established model, e.g. it has been used in land surface models of ECMWF and in the Earth system model operated by the Centre National de Recherches Météorologiques (CNRM-ESM1). Van Diepen et al. (2022) recently made a comparison between the photosynthetic responses to light and intercellular CO2 predicted by the leaf photosynthesis models of Farquhar et al. (1980) and Goudriaan et al. (1985). The latter model is used in the A-gs approach. They found both models calculate near-similar responses of photosynthesis to changes in intercellular CO2. They also found near-similar responses to light (at different fixed levels of intercellular CO2). We added this information to the manuscript, as this is indeed important information that was lacking.

We have also added response characteristics of our A-gs model using the uptake parameters obtained for Hyytiälä as an appendix figure to the paper. We also show the figure below:

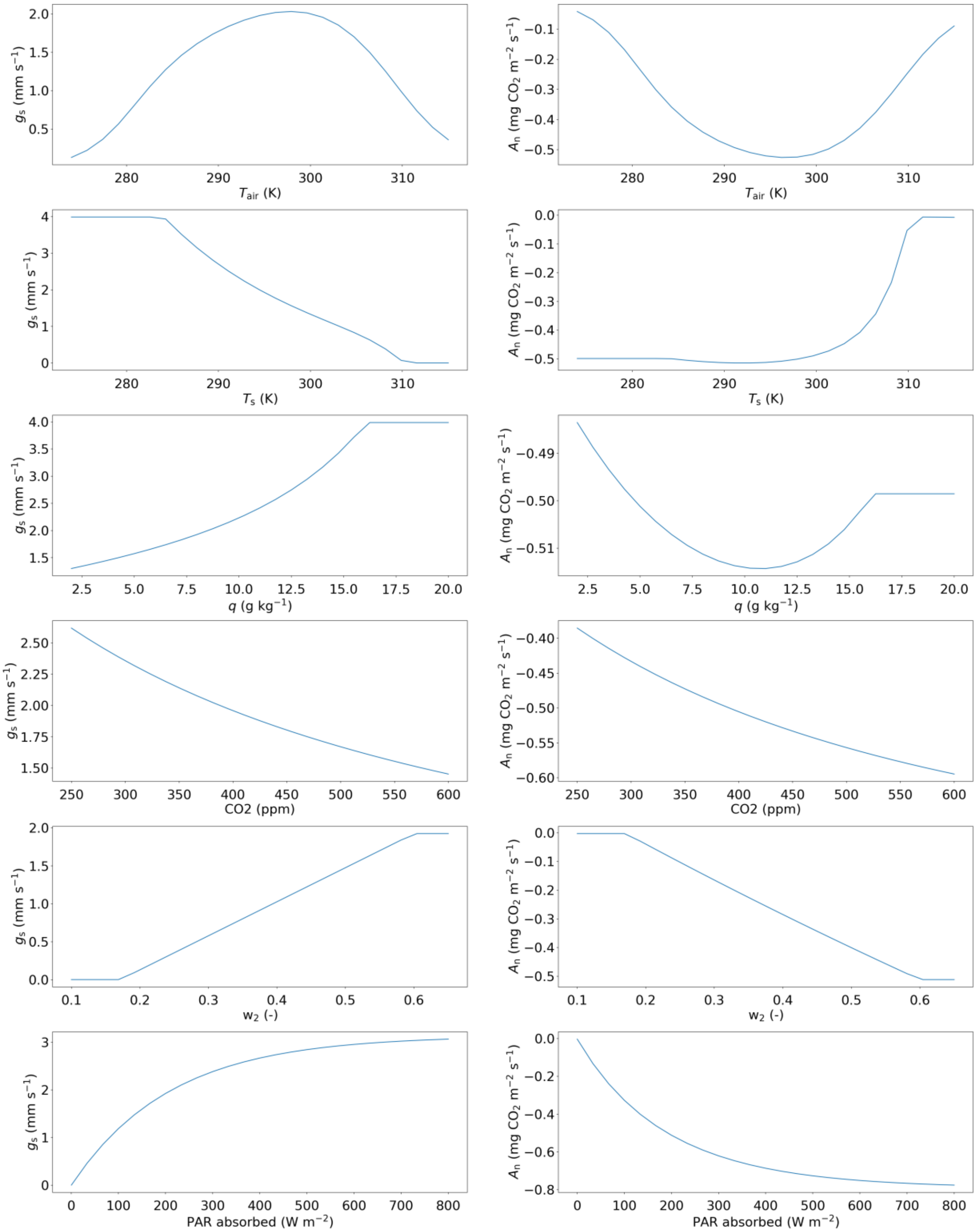

Figure 1: Response characteristics of our A-gs model using the uptake parameters obtained for Hyytiälä. w2 is the volumetric moisture content of the deep soil layer, q is specific humidity, and Ts is leaf temperature.

The Kooijmans et al. (2019) manuscript does not explicitly simulate leaf-scale CO2 uptake, but uses measurements. For stomatal conductance of COS, Kooijmans et al. (2019) used the Ball-Berry parameterisation applied to chamber measurements of FCO2. These measurements are rather uncertain, and might not be fully representative for the conductance outside of the chambers. Note also that we evaluate our model against CO2-flux measurements in Fig. 2e and Fig. 9e.

We agree that both the model and the observations are a source of uncertainty in the modelled LRU values. We have explicitly mentioned this in the text now.

The reported LRU values fall within a reasonable range, but it is not clear that they represent independent estimates directly comparable to those in the literature. The most reliable way to determine LRU remains direct gas-exchange measurements of CO2 and OCS fluxes and concentrations (e.g., Stimler et al. 2011; Kooijmans et al. 2019). In this study, the [OCS] and [CO2] measurements appear sparse and, at times, show gradients with the opposite sign to what would be expected. This raises doubts about whether there is sufficient information to constrain LRU directly from the observed concentrations and fluxes. Instead, it seems likely that the inversion primarily fits stomatal conductance and photosynthesis to the vertical profiles of temperature, VPD, CO2, and PAR, with LRU then emerging as an implicit consequence of applying the chosen OCS parameterization. In that sense, the regression that they propose linking LRU to VPD and PAR may mainly reflect the built-in response behavior of the parameterization rather than the physiological behavior of the leaves themselves.

We agree that accurately measuring the four components of LRU directly is the most reliable way to obtain LRU for a specific location (and specific time of day/year etc.). However, datasets providing enough information to derive LRU at the canopy or leaf scale are very sparse, and our LRU parameterisation aims to offer an alternative that allows for COS-based GPP estimates at more locations and larger scales than just the few locations with measurements. Lai et al. (2024) used a parameterisation for LRUcan, derived in Kooijmans et al. (2019), that is based on measurements of the leaf-scale relative uptake of COS and CO2 at a boreal forest location (Hyytiälä), to estimate global terrestrial GPP. We evaluate their parameterisation for a different location (Mieming), and we tried to offer an improvement to the existing parameterisation.

We are aware that there is considerable uncertainty in the modelled LRU, but given the sparse knowledge on LRU currently available, we believe our LRU parameterisation has added value in this respect.

Regarding the constraints on LRU using observations, the general aim of the framework is to allow the assimilation of various streams of observations simultaneously (fluxes, mole fractions at multiple heights, temperatures, humidity etc.) to estimate model parameters, thereby obtaining a physical model that is consistent with a diverse set of observations. We thus aim for a holistic approach for modelling LRU, by aiming to obtain parameters consistent with all these diverse observation streams.

Note that we deliberately made the choice to not (directly) use LRU observations for optimising our model parameters, but instead use (amongst others) observations of COS and CO2 fluxes and mole fractions. In this way we try to fit the physical essentials (fluxes and mole fractions) well, instead of directly optimising a derived quantity such as LRU. A quantity such as LRU could in theory be fitted well while e.g. both the COS and CO2 flux are strongly overestimated. With our approach we try to avoid this.

Indeed the regression equation is derived based on model output, which is not necessarily fully aligned with true physiological behaviour, as also mentioned in the discussion around line 475 (line number referring to non-revised manuscript). Ideally, we would have measured LRU data available at many locations to check the validity of our parameterisation. However, as mentioned before, these data are sparse. For the two locations modelled in the manuscript we do compare with LRU derived from observations, as shown in figures 7 and 10. These figures show an acceptable fit with observations, given the large uncertainty present in these observations. The difficulty with fitting LRU also originates from the form of the equation, e.g. a small positive bias in COS flux and a small negative bias in the CO2 flux can lead to a relative large deviation in LRU due to the division exacerbating small differences.

The study nevertheless provides a useful illustration of how vertical gradients in light, CO2, and humidity can generate vertical structure in LRU, and it demonstrates an interesting modeling capability to quantify the gradients in [OCS] betwee the bulk atmosphere and the leaf surface that confound estimation of GPP from the OCS flux and LRU. From this perspective, the work is valuable. However, it should not be presented as an alternative to direct gas-exchange measurements for determining LRU, and the manuscript should be revised to clarify this distinction. The Kooijmans et al. paper cited above provides code and data that could be used to calibrate, test, or possibly replace the current parameterization, and the manuscript would benefit from more extensive explanation of the parameterizations and inversion framework in the main text. Finally, the comments regarding the failure of the Lai et al. model to reproduce the study's results are not currently supported by data and should either be removed or substantiated with appropriate analysis.

We now made clear in the text that we do not aim to provide LRU estimates that are equally accurate as directly measuring the components of LRU for a specific location. Instead the goal of the parameterisation is to allow use of LRU on a larger scale and on more locations. As mentioned earlier, we included additional information on the CO2 uptake model in the manuscript.

We believe some of the comments might be related to a misunderstanding: the LRU parameterisation derived in Kooijmans et al. (2019) is the 'Lai24' parameterisation used for comparison in our manuscript. Therefore, we already compare our parameterisation with the one from Kooijmans et al (2019), for both locations. Lai et al (2024) apply the parameterisation that was derived for one site by Kooijmans et al. (2019) on a global scale (including vastly different ecosystems compared to Hyytiälä) to estimate global GPP. Our study results suggest that the Lai24 parameterisation underestimates LRU_{can} at Mieming, and similar biases might be present in other ecosystems. In the discussion (Sect 4.2) we discuss potential reasons for the observable underestimation of LRU_{can} by this parameterisation.

As mentioned earlier, we included additional information on A-gs. We also added some information on the inverse modelling framework and coupled forward model to section 2.2.

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Reply to reviewer #2

We would like to thank the reviewer for the time dedicated to reading our manuscript and for the very constructive and detailed comments, which helped to improve our manuscript. The point-to-point response to the comments follows below, the reviewer's text is given in black, and our response in red.

Major Comments

1. The complexity of the canopy model, SilCan, used in the modelling of PAR at the top and throughout the canopy, is somewhat downplayed. An extensive summary is provided in the supplement, but little is discussed of its evaluation. While the results of this work do somewhat suggest it is performing adequately, i.e. LRU values are sensible in the canopy. A more elaborate evaluation of its performance is recommended or should be presented if done so. How does it compare with other canopy models? Has there been any comparison with estimates of PAR from remote sensing or in-situ observation? Has a separate publication specifically detailing this model been considered?

To provide additional evaluation of the SiLCan model, we used PAR observations from Hyytiälä in July 2015, to calculate the relative extinction in the canopy (influences key drivers of COS and CO2 uptake). We have only measurements at one height in the canopy (0.6 m), but from 4 different locations. For the measurements we define relative extinction as 1 - PAR_0.6m / PAR_above_can. In this, PAR_0.6m is PAR at 0.6m height. PAR_above_can is measured PAR above the canopy. For the model we take the node located closest to the observation height. We show the plot here, using measurements for the 8 days that we included for averaging in Sect. 3.1 of the manuscript (binned per hour):

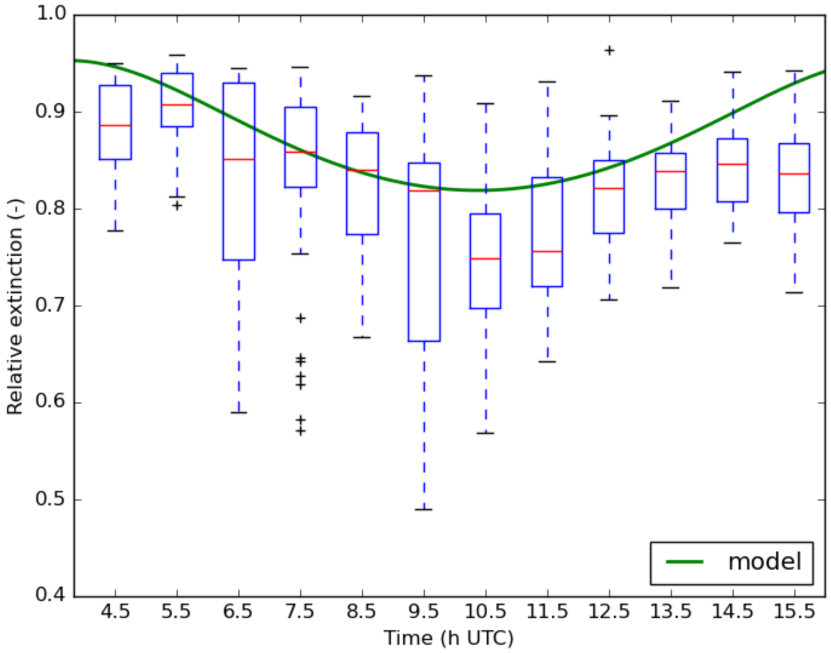
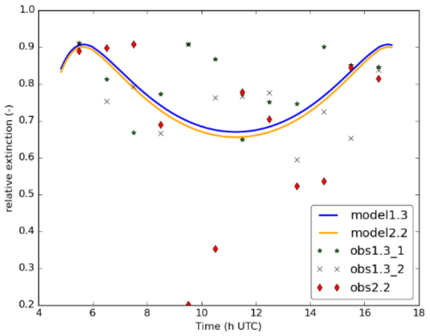


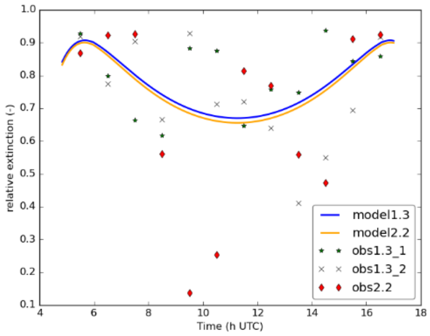
Figure 2: relative extinction at 0.6 m height above the forest floor for Hyytiälä, for the 8 days in July 2015 that were included for averaging observations in Sect. 3.1 of the manuscript (binned per hour). The green line is the model, the boxplots show the observations. A box extends from the first quartile (Q1) to the third quartile (Q3) of the data, with a red line at the

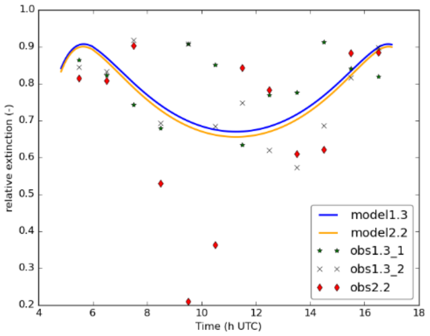
median. The whiskers extend from the box to the farthest data point lying within 1.5x the inter-quartile range (IQR) from the box. Flier points are those past the end of the whiskers.

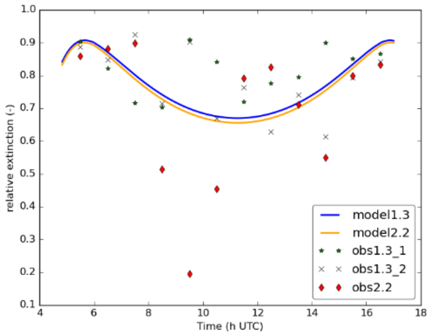
The PAR observations indicate that at some locations, a substantial amount of PAR remains available at 0.6 m above the forest floor (due to openings in the canopy or sections with low plant area density). The comparison indicated that overall the relative extinction of PAR is fitted relatively well, although extinction is sometimes somewhat overestimated. We have added this information to the manuscript now (not the plots themselves).

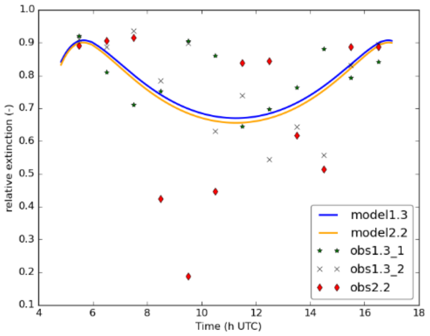
We did a similar check for August in Mieming, with a similar result (now we plot the individual days):

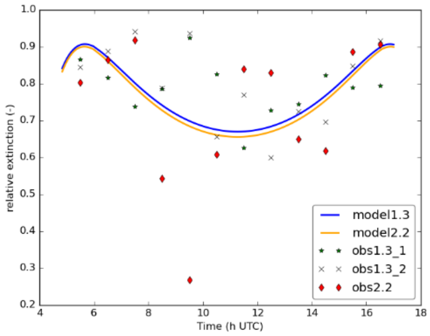












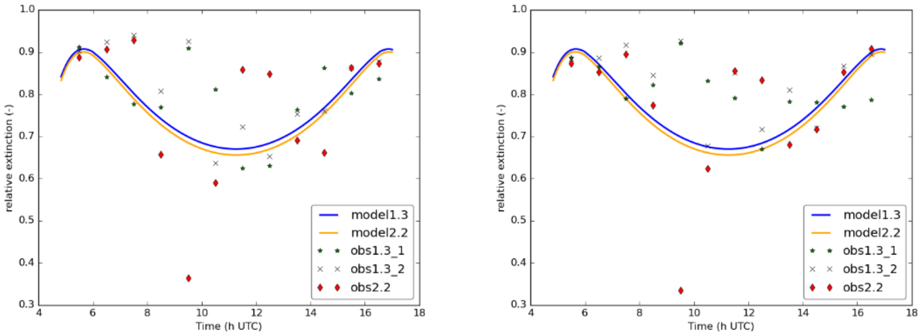


Figure 3: relative extinction at 1.3 and 2.2 m height above the forest floor for Mieming, for the 8 days in August 2023 that were included for averaging observations (see Sect. 2.4 of the manuscript). Measurements at 1.3 m height are shown for two locations (obs1.3_1 and obs1.3_2), and measurements at 2.2 m height are shown for one location (obs2.2).

Note that in Table A1 we provide evaluation of the (coupled) model, by quantifying the fit with the 26 assimilated observation streams for July 2015 in Hyytiälä (using partial reduced chi-square statistic). A subset of these observation streams is shown in Figure 2 of the manuscript.

2. To what extent does the absence of advection and chemistry modelling impact the applicability of the resulting LRU even for the same biome elsewhere? It is mentioned that advection is set to zero for all simulations. A sentence or two is required to highlight the limitations associated with this. For example, on the day scale, changes to air temperature or precipitation would substantially affect LRU. How does this scale up to application of LRU on an annual basis?

Advection is set to zero indeed. A motivation for setting advection to zero is that we do not have information on it, and we tried to keep the number of parameters that we optimise limited, to reduce the complexity of the optimisation problem. Furthermore, the averaging over multiple days is likely to reduce the influence of advection of different air masses to some extent, as this is likely to differ between days. We have now performed an additional optimisation in which we include advection of COS, CO2, H2O and heat in the state that we optimise. In this new optimisation, the cost function is slightly lower (75 vs 77), as the model has more freedom to fit the observations, given the extra advection parameters that are optimised. We show below a picture of the LRU for the new optimisation:

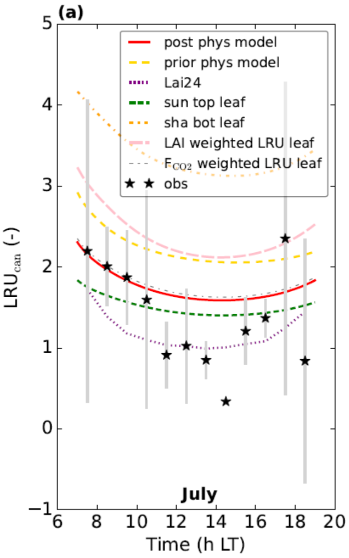


Figure 4: As Figure 7a in the manuscript, but for the optimisation including advection

When comparing with Fig. 7a of our manuscript, it is clear that differences are small. Even without advection, the model already has quite some capabilities for fitting temperature observations etc., e.g. by adjusting the free tropospheric lapse rates. To disentangle the effects of advection and free-tropospheric entrainment, more specific observations such as vertical soundings might be useful, but this was not the focus of our work (see also Sect. 9.6 of Bosman and Krol, 2023). As an example of changes to the model output, we show here the temperature at 16.8 m height with and without advection:

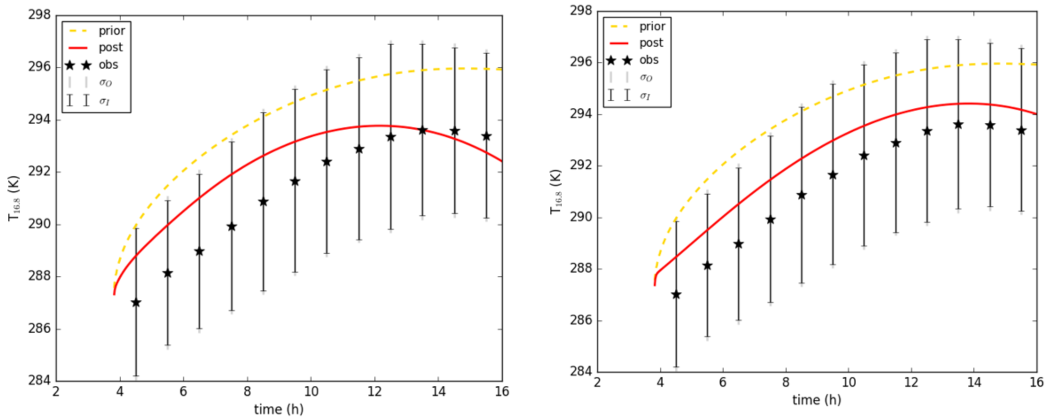


Figure 5: Temperature at 16.8 m height for the optimisation with advection (left) and without advection (right). Time is in hour UTC

The changes in 16.8 m temperature are rather limited. Note that our framework is less suitable to estimate LRU in winter/late fall, as the assumption of a well-mixed layer is expected to be violated more often (and vegetation uptake will be small anyway due to reduction/absence of photosynthesis). Therefore we do not attempt to scale up the LRU to an annual value.

We have added some information on the additional optimisation that includes advection to the discussion on model performance in the manuscript.

Chemistry modelling is indeed absent as well. At the remote locations that we model we do not expect a large influence of e.g. chemical conversions of anthropogenic CS2 emissions into COS. Furthermore, our model simulations have a relatively short timescale (less than one day) compared to the relevant chemical timescales involved in the CO2 and COS budgets.

3. The COS mole fraction and COS flux in Figure 2 (b, d and f) are exceptionally noisy compared to the other variables (additionally, I don't understand an increase in mole fraction around midday). While this is highlighted in the text, has the full extent of this knock-on effect been considered? This is likely contributing to the positive bias seen in the LRU output. But to what extent does this allow for other variables to dominate in the inversion calculations?

The COS variables are indeed very noisy, most likely caused by measurement uncertainty given the very small mole fractions compared to e.g. CO2 (i.e. ppt vs ppm). Additionally, the averaging over multiple days might introduce a bit of noise due to a few missing data points. As the error bars for the COS variables are large, those measurements indeed contribute less to the cost function compared to observations with a small observational error. Thus, the fitted parameters are indeed to a large extent determined by other variables. This is not necessarily a problem, the COS observations are mostly fitted well within their uncertainty bounds. This can even be considered a strength of the framework, i.e. multiple information sources are taken into account. We do not optimize LRU itself, but, amongst others, COS and CO2 fluxes and mole fractions that (to a large extent) determine LRU.

The difficulty with fitting LRU well also originates from the form of the equation, e.g. a small positive bias in COS plant flux and a small negative bias in the CO2 plant flux can lead to a relative large deviation in LRU due to the division exacerbating small differences.

An increase in COS mole fractions in the morning continuing around midday could be explained by vegetation COS uptake at night, followed by entrainment of free tropospheric COS-rich air, see also Fig. 2a of Rastogi et al. (2018). However, the sudden increase in COS mole fraction in the observations of Fig. 2 in our manuscript around midday is indeed remarkable. We don’t have a clear explanation for this. I attach here for the COS mole fraction at 125 m, the plots of the 7 individual days (remember that we generally average over 8 days – but one of them has no data for COS at 125 m):

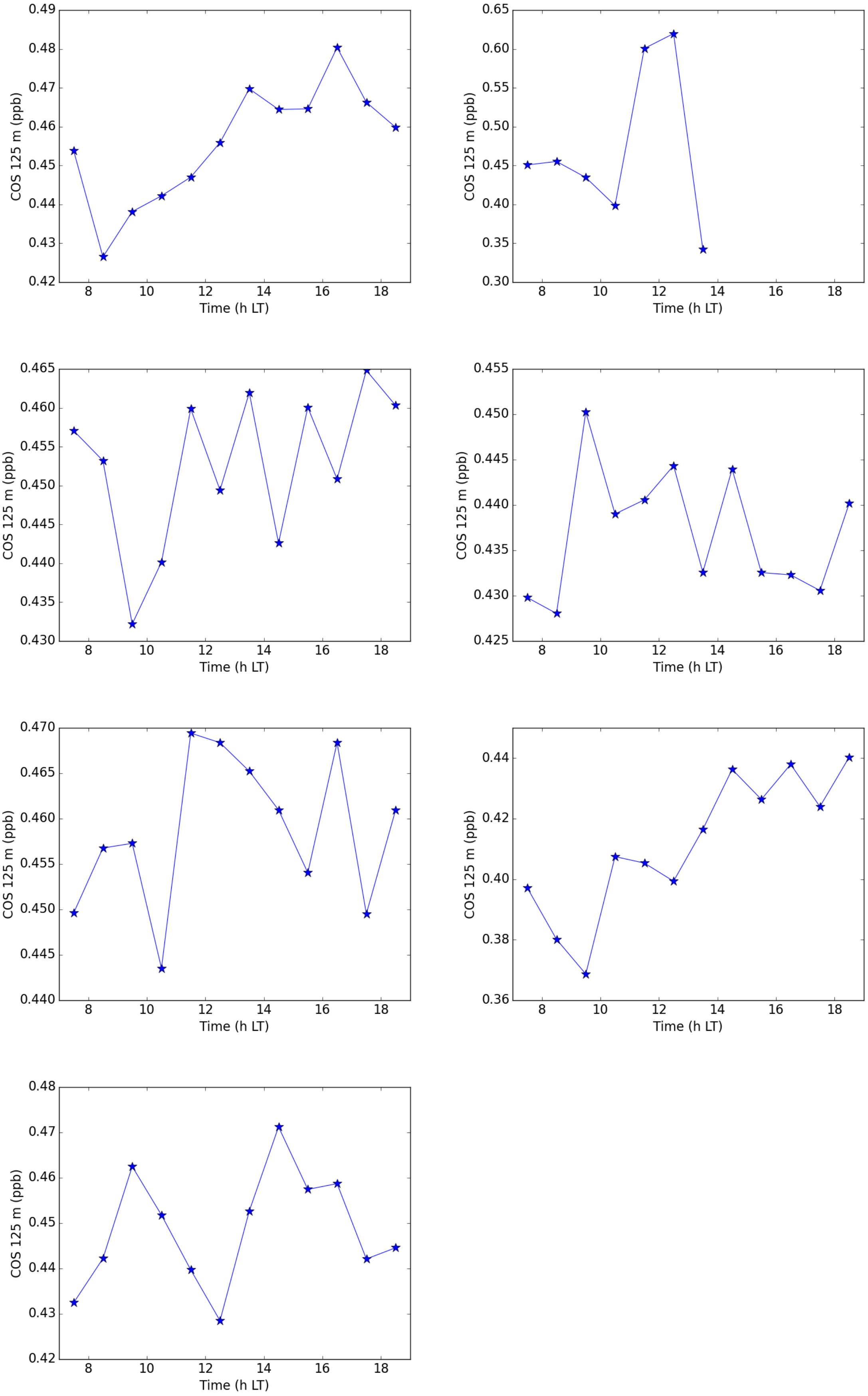


Figure 6: Observed COS mole fraction at 125 m height for the individual days in July 2015 (Hyytiälä) included for averaging in the manuscript

It seems that these high values around midday are mostly caused by two very high values at one of the measured days, i.e. the top right subplot (3 July 2015). The very high values cannot simply be discarded as measurement errors, as for the same times (11.30 and 12.30 h on 3 July 2015) we find very high values for the COS mole fraction at 4 m height as well. As this is a specific analysis for explaining the high values around midday, we will not include these plots in the manuscript itself.

4. Section 3.1.3 contains a particularly interesting discussion and explanation of how the modelled variables affect one another. However, it's very difficult to follow, particularly as the plots are not particularly easy to interpret. Some specific points on this include:

a. In Figure 3: having a solid blue line in multiple panels on the same plot would often suggest they are the same variable, but for different circumstances, say for morning, noon and evening or similar, but in this case, they are for different variables all together, this could be misleading or confusing.

The solid blue lines in the three subplots are indeed for totally different variables, we changed the colors of the blue line in subplots (b) and (c) to avoid confusion.

b. Line 305: “This can to a large extent be explained by the profiles of absorbed PAR (Fig. 3b, **red and green dashed lines**).”. Include specific references to the plot, so it is easier for the reader to identify.

Thanks for this useful suggestion, for most of the references to figures 3 and 4, we have now added information on which specific lines to look at.

I recommend a restructuring or re-write of this section. And making the colours more reader friendly and interpretable (specifically with regards to colour-blindness). Or even adjust the structure of the plots. A final note is to consider moving the legends off the plot entirely.

Indeed these plots are rather complicated, given the large amount of info in it. We have adjusted the whitespace between the subplots, removed the annotated text in the figures, and placed the legends outside the figures to make it look less busy. Regarding colour blindness, please note that every plotted line is per subplot unique in terms of dashes as well, so even without reference to colours, the lines should all be distinguishable.

5. The parameterisations for estimating LRU from Kooijmans et al. (2019) are used to estimate LRU – Lai24. As these results seem to perform better at Hyytiälä, which is addressed around Line 510. A more thorough explanation of why this is the case, perhaps with further analysis would be interesting. The shape of the Lai24 model run appears to fit the measurements better, particularly the inflections at the start and end of the day, which is not as apparent in the physical or regression model. Further, the models designed in this work might lead to better correlation values at Mieming, but does this necessarily mean it is performing ‘better’? Are there sufficient measurements at Mieming to understand the shape of diurnal LRU? Would we expect the same ‘bowl’ shaped LRU we see at Hyytiälä here?

Figure 7 shows that the Lai24 parameterisation has a stronger variation between midday and evening/morning, especially in September. The observed large LRU values in early morning/late evening are (probably at least to a large extent) caused by open stomata while PAR is low. This leads to a CO2 flux becoming close to zero (as the CO2 uptake is strongly light dependent, given that PAR supplies the energy and reduction equivalents for the carboxylation process), while COS uptake continues as a diffusion process as long as stomata are open (destruction by carbonic anhydrase maintains a leaf-air gradient). The parameterisation from Lai24 seems to better capture this effect.

As mentioned around line 475 (line number referring to non-revised manuscript), in the A-gs photosynthesis model that is part of our modelling framework, the internal CO2 concentration does not directly respond to photosynthesis and thus PAR. This ‘weakness’ of A-gs might be a reason why our model (and thus also our LRU parameterisation) does not capture the above-mentioned low-PAR effect well. However, one should realise that very low PAR conditions only occur shortly during our simulation period, and CO2 uptake is very low in the early morning/evening. Therefore, this can be expected to be of limited importance when estimating CO2 uptake using LRU.

We also have to keep in mind that the Lai24 parameterisation was obtained by directly fitting LRU measurements of sunlit leaves in Hyytiälä. We derived our LRU_{can} parameterisation by fitting components of LRU_{can} (which has a non-linear dependence on some of the components). This provides a potential reason why the Lai24 parameterisation provides better results for the specific location the parameterisation was derived for (taking into account that the LRU of sunlit leaves is not very different from LRU_{can}, as discussed in Sect. 4.2). We added some information on this to the manuscript.

In Mieming we would expect the same low-PAR effect in the early morning/evening, however, the leaf scale observations we show in Figure 10 are not available during early morning/evening.

Based on the limited observations shown in Figure 10, we conjecture that the bias in LRU in Mieming is larger for the Lai24 parameterisation, although we are aware that there is considerable uncertainty and only partial coverage. We do not intend to claim that our parameterisation is generally ‘better’ than the Lai24 parameterisation, but based on the limited information available in Mieming, our parameterisation performs better at that location.

At line 535 (line number referring to non-revised manuscript) we now write the following ‘Our developed parameterisation, although not performing equally well as Lai24 in Hyytiala, shows better transferability to Mieming.’ We changed it into ‘Our developed parameterisation, although not performing equally well as Lai24 in Hyytiala, shows better transferability to Mieming, based on our model results and limited observational data.’

6. A few points on discussion and conclusions: a. More emphasis should be put on the points raised at the end of Section 4.2, such that the inverse modelling framework is suitable to estimate COS and CO2 uptake across different biomes. However, abundant measurements are required at such sites and that the results likely show that a separate exercise of establishing parameterisations at each site is necessary. Until the model achieves improved transferability.

The reviewer, with this comment, is mostly supporting the statement we make at the end of Section 4.2 about representativeness of the results for the (needleleaf) Hyytiälä and Mieming sites in terms of COS/CO2 uptake for other biomes. With our inverse modelling system we can at least better identify the role of processes involved in this COS/CO2 uptake. But we also make a clear statement that for optimal application of our framework, we require more measurements than generally available for those other biomes to impose sufficient constraints on the inverse modelling system. Complete transferability of LRU parameterisations/ A-gs parameters between very different biomes might not be achievable, since vegetation characteristics might be too different among very different biomes. Our inverse modelling framework might also be useful for developing an optimal measurement strategy for other sites.

We added the following sentences: ‘Given that the Lai24 parameterisation seems to perform better at Hyytiälä, while our parameterisation seems to perform better at Mieming, it is likely that different parameterisations should be used for different locations to reach the best accuracy. Furthermore, complete transferability of LRU_{can} parameterisations between vastly different biomes might not be achievable, as the responses to PAR and VPD might differ, and (additional) vegetation-specific physiological/biogeochemical drivers of COS to CO2 relative uptake might exist. However, the transferability between the Hyytiälä and Mieming locations suggests that al least within similar biomes, reasonable results can be obtained with a single parameterisation.'

b. A little too much emphasis is put on the physical and regression models outperforming Lai24 at Mieming. Particularly the authors have been slightly overcomplimentary in the performance of the physical and regression models at Mieming site. Figure 10 shows both models and Lai24 still underestimating observations.

Indeed our modelling framework underestimates the Mieming observations as well. Our statement '... our parameterisation outperforms Lai24 at this location.' is indeed perhaps somewhat too confidently written. We have changed it into ''our parameterisation outperforms Lai24 at this location, based on limited observations and model output''

But note that our fit to LRU is based on fitting observational streams that form components of LRU, while the Lai24 parameterisation directly fitted LRU measurements. The bias is likely to a large extent related to the non-linear LRU definition (non-linear dependence on some of the components we optimise).

And an important point we want to make based on our study is that the approach by Lai24, i.e. using an LRU parameterisation obtained at a single site (Hyytiälä) to estimate global GPP leads to uncertain results. Applying our own parameterisation globally would lead to uncertain results as well, unless extensive validation at multiple biomes is performed.

c. In conclusions: "*For Hyytiälä, both the physical and regression model generally somewhat overestimated LRUcan with respect to the (noisy) observations. We found that the LRU of sunlit top leaves provides a relatively good estimate of LRUcan, which is encouraging for the use of canopy COS fluxes to estimate canopy CO2 uptake. At the same time, we find that the simple leaf-scale parameterisation obtained in Hyytiälä by Kooijmans et al. (2019), rolled out globally by Lai et al. (2024), does not perform well in a more southerly needleaf forest (Mieming, Austria).*". This feels insincere to mention Lai24's underperformance at Mieming, while omitting its good performance at Hyytiälä. Particularly given the third research question raised in Section 1.

Good point, we changed the sentence as follows: "we find that the simple leaf-scale parameterisation obtained in Hyytiälä by Kooijmans et al. (2019), rolled out globally by Lai et al. (2024), performs well in Hyytiälä, but does not perform well in a more southerly needleleaf forest ..."

The strength of this work is the improved understanding of LRU variations within the canopy and on the development of a simple and well represented regression model. More work is required if the goal is to make model parameterisations transferrable between biomes (in further publications, not this one). While the performance of the physical and regression models at Mieming are slightly overplayed, it is worth noting that results do not necessarily have to be sold as 'good' or 'better' to be interesting and valuable. A little more in-depth discussion on the cause of the discrepancy between the physical model and Lai24, and the performance against measurements would enhance this already well-written publication.

Thanks for these constructive comments. We have addressed these issues in the individual major comments above.

Please see the supplementary document for more in-depth and specific comments. Note they are written up chronologically, not ranked by importance or significance.

Minor Comments

1. Title: “Relative uptake of carbonyl sulphide to CO2: insights from a coupled boundary layer - canopy inverse modelling framework”. Consider aligning the use of chemical abbreviation and written form.

We changed it into ‘’ Relative uptake of carbonyl sulphide to carbon dioxide: insights from a coupled boundary layer - canopy inverse modelling framework”

2. Lines 104-105: overall I think the coupled modelling framework and inverse system could do with a more thorough explanation here. It appears the appropriate publications and documentation have been cited, but an additional sentence or two may help readers outside of the modelling community.

Indeed the description is rather short. We now added the following information: ‘In an optimisation, the framework aims to obtain values of the parameters to optimise (the state) that minimise the cost function. This is done starting from an initial guess of parameter values, after which the state is improved iteratively, thereby calculating the cost function and its gradient. For calculating the gradient of the cost function, an analytical gradient is available using the model adjoint.’

And for the forward model we added: ‘Above the mixed layer, a discontinuity occurs in the scalar quantities, representing an infinitely thin inversion layer. Above the inversion, the scalars are normally assumed to follow a linear profile with height in the free troposphere. The information on the free troposphere is used for calculating exchange between the mixed layer and the free troposphere.’ We also added ‘Above-canopy downward shortwave radiation is calculated as a function of time, location, and cloud cover.’ We also added ‘Upper soil temperature and moisture are simulated based on a force–restore model’

More details can be found in the cited publications. Note that we also added additional information about the photosynthesis model in an appendix.

3. Figure 1: The diffuse and direct radiation is a little misleading in this diagram. Assuming the direct radiation is that which is incident directly on the vegetation and diffuse is from reflection and scattering processes in the atmosphere. Having the direct arrows going diagonal through the atmosphere is counter-intuitive to the idea that a direct path would be the shortest path possible. I think resolved if the diffuse radiation arrows emphasise the randomness of scattering processes in the atmosphere. Perhaps coming off the direct beam..?

The direct radiation is indeed incident directly on the vegetation and diffuse is from reflection and scattering processes in the atmosphere (including above-canopy and, closer to the ground, in-canopy scattering). Note that we have drawn the direct arrows such that they come directly from the sun, which is also included in the sketch. Given that the sun is not drawn directly overhead, the diagonal is the shortest path here. The diffuse radiation in the figure, in contrast, does not point directly from the direction of the sun.

4. References to the supplementary material should include specific section or equation references, given the length of the supplementary document (examples include 135 and 228)

We now added specific references to parts of the supplement when possible. Note that line 135 (line number referring to non-revised manuscript) refers to the canopy model in general, without reference to a specific aspect.

Technical Comments

Line 7: include - includes*

Adapted, thanks for spotting this typo

Line 18: optimations – optimisations*

Adapted, thanks for spotting this typo

Line 24: Define VPD

Adapted

Line 38: subscript 2 in CO2 – CO2*

Adapted

Line: 102-103: add in link to the canopy model in question: “We have added a relatively simple canopy model to the ICLASS framework **(SiLCan, see Section 2.2)**, in order to simulate gases and atmospheric conditions in forest canopies in more detail.”

Added

Lines 108-111: as this is directly discussed in Bosman and Krol, 2023, I think it would help the reader to direct them to it specifically, i.e. “see Section 3.1 in Bosman and Krol, 2023”.

We have now added a reference to Section 3.1 of Bosman and Krol (2023) at line 101. Lines 108-111 do not directly relate to section 3.1 in Bosman and Krol (2023). (line numbers referring to non-revised manuscript)

Line 121: moisture -> H2O. As you are explicitly referring to the tracers, I think it would be best to be clear exactly which molecule you are referring to.

Adapted

Line 149: Figure -> Fig.

Adapted

Line 215-216: “The formula for **LRU is found by rearranging Eq. 1**”

Adapted, we now write “The formula for LRU_{can} is given in Eq. 1, after rearranging.”

Line 237: Define VPD

Adapted

Line 315: small T for the

Adapted

Line 409: full stop after Fig - Fig**.**

Adapted

Check instances of capitalised LAI24, for example in the legend of Figure 10. Important to differentiate between Leaf Area Index (LAI) and Lai24 the parameterisation

455 Adapted, thanks for spotting this!

References

Bosman, P. J. M. and Krol, M. C.: ICLASS 1.1, a variational Inverse modelling framework for the Chemistry Land-surface Atmosphere Soil Slab model: description, validation, and application, Geoscientific Model Development, 16, 47–74, https://doi.org/10.5194/gmd-16-47-2023, 2023.

Lai, J., Kooijmans, L. M., Sun, W., Lombardozzi, D., Campbell, J. E., Gu, L., Luo, Y., Kuai, L., and Sun, Y.: Terrestrial photosynthesis inferred from plant carbonyl sulfide uptake, Nature, 634, 855–861, https://doi.org/10.1038/s41586-024-08050-3, 2024.

Rastogi, B., Berkelhammer, M., Wharton, S., Whelan, M. E., Itter, M. S., Leen, J. B., et al. (2018). Large uptake of atmospheric OCS observed at a moist old growth forest: Controls and implications for carbon cycle applications. Journal of Geophysical Research: Biogeosciences, 123, 3424–3438. https://doi.org/10.1029/2018JG004430