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 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 light and 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:

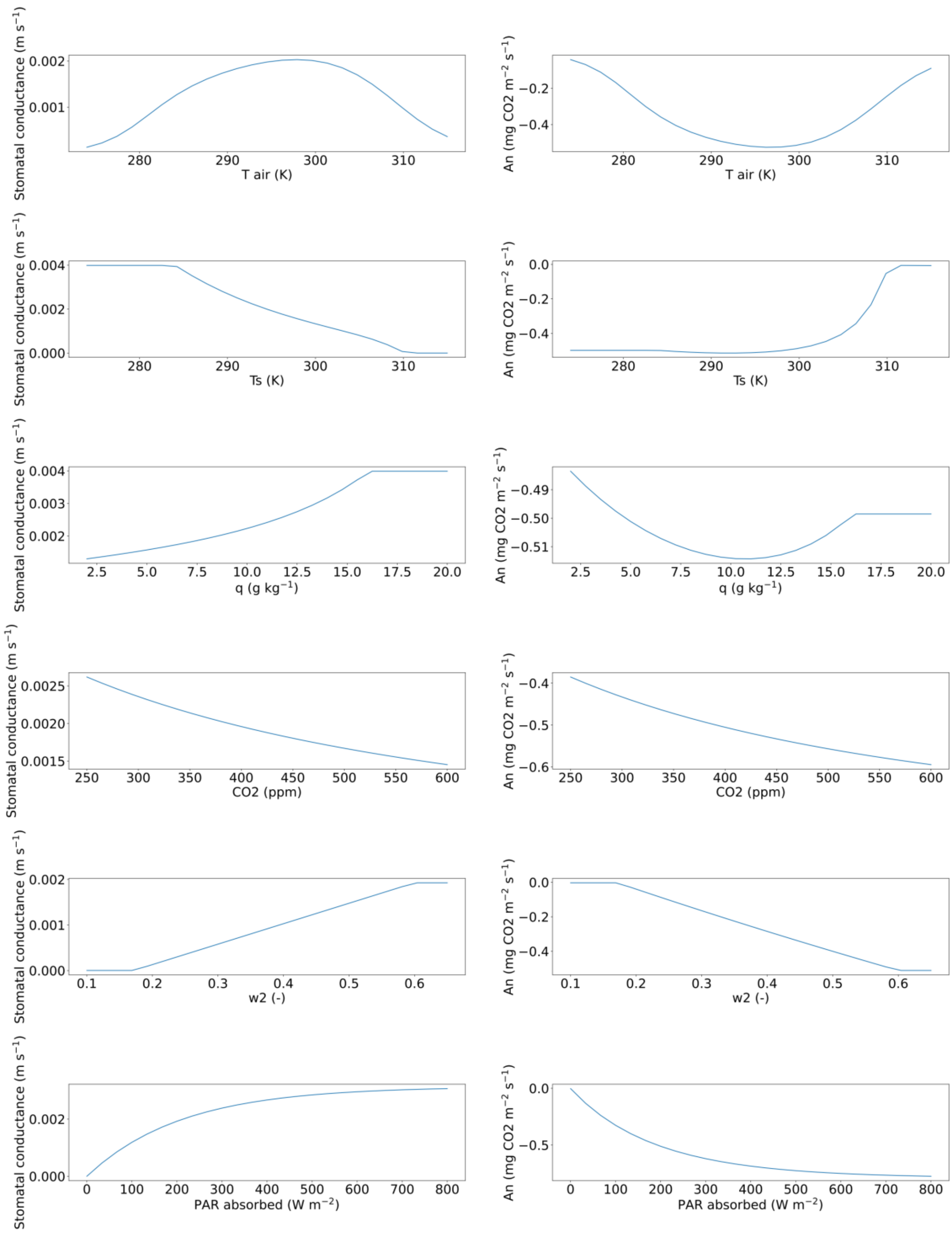

Figure 1: Response characteristics of our A-gs model using the uptake parameters obtained for Hyytiälä. w2 is the volumetric moisture content of the deep soil layer, q is specific humidity, and Ts is leaf temperature.

The Kooijmans et al. (2019) manuscript does not explicitly simulate leaf-scale CO_2 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 CO_2-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 CO_2 and OCS fluxes and concentrations (e.g., Stimler et al. 2011; Kooijmans et al. 2019). In this study, the [OCS] and [CO_2] 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, CO_2, 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 CO_2 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 CO_2 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 CO_2 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.

References

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