

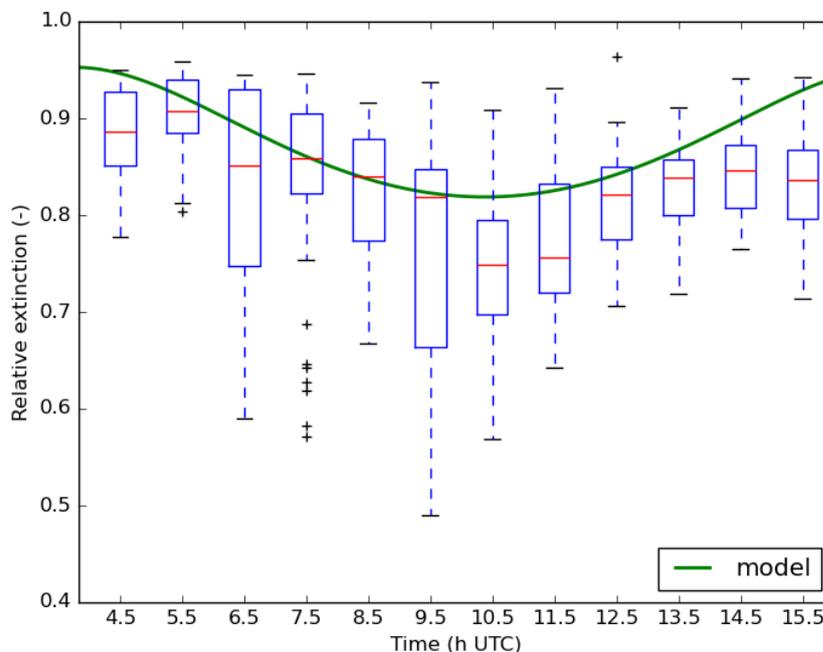
## 1 Reply to reviewer #2

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

### 6 Major Comments

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

15 To provide additional evaluation of the SilCan model, we used PAR observations from Hyytiälä in July  
16 2015, to calculate the relative extinction in the canopy (influences key drivers of COS and CO<sub>2</sub>  
17 uptake). We have only measurements at one height in the canopy (0.6 m), but from 4 different  
18 locations. For the measurements we define relative extinction as  $1 - \text{PAR}_{0.6\text{m}} / \text{PAR}_{\text{above\_can}}$ . In  
19 this,  $\text{PAR}_{0.6\text{m}}$  is PAR at 0.6m height.  $\text{PAR}_{\text{above\_can}}$  is measured PAR above the canopy. For the  
20 model we take the node located closest to the observation height. We show the plot here, using  
21 measurements for the 8 days that we included for averaging in Sect. 3.1 of the manuscript (binned  
22 per hour):



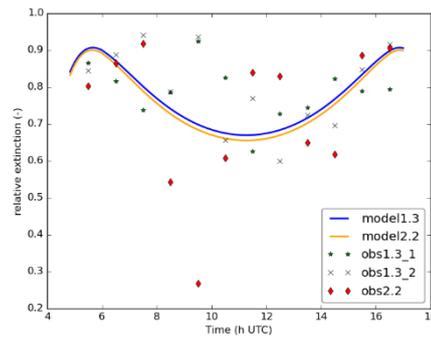
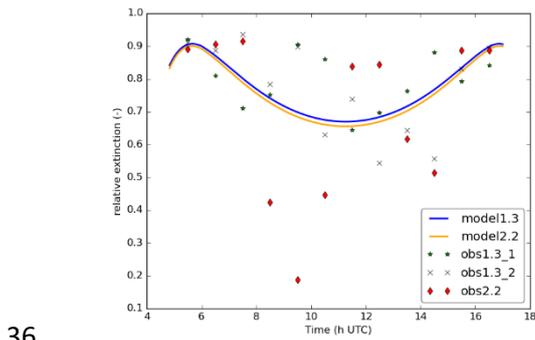
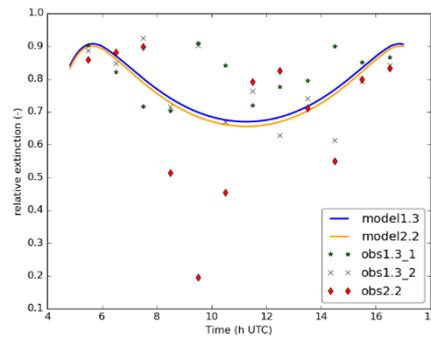
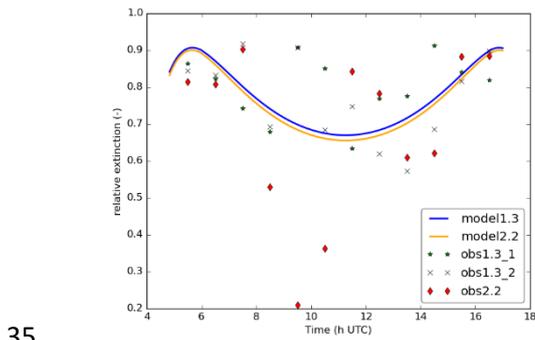
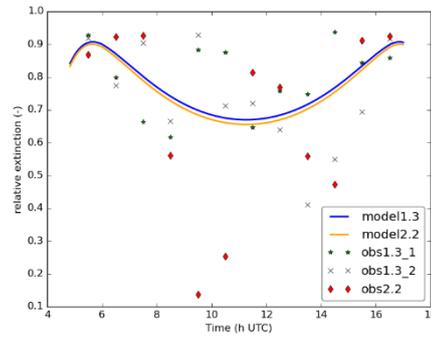
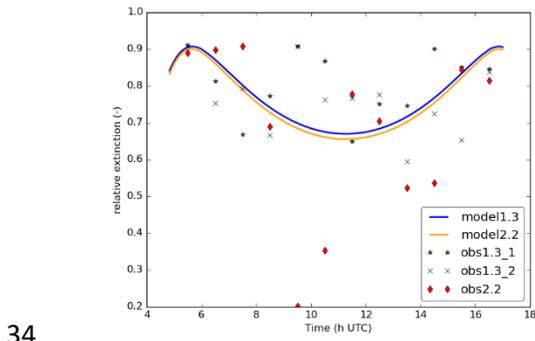
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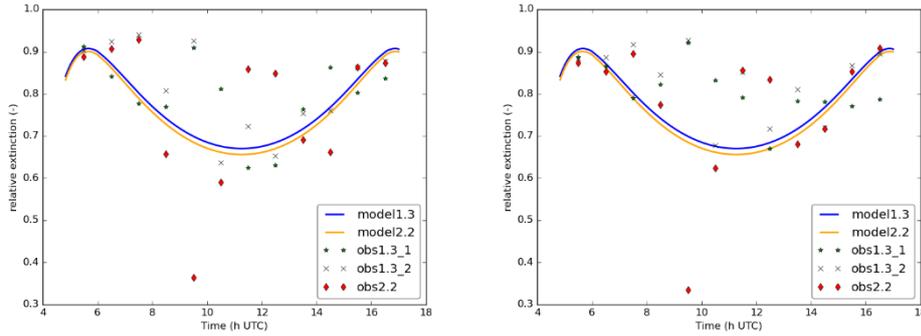
24 *Figure 1: relative extinction at 0.6 m height above the forest floor for Hyytiälä, for the 8 days in July 2015 that were included*  
25 *for averaging observations in Sect. 3.1 of the manuscript (binned per hour). The green line is the model, the boxplots show*  
26 *the observations. A box extends from the first quartile (Q1) to the third quartile (Q3) of the data, with a red line at the*

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

29 The measurements indicate that at some locations, quite some PAR remains available at 0.6 m above  
30 the forest floor (openings in canopy, sunflecks). These plots illustrate that overall the extinction of  
31 PAR is fitted relatively well, although extinction is sometimes somewhat overestimated. We have  
32 added this information to the manuscript now (not the plots themselves).

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





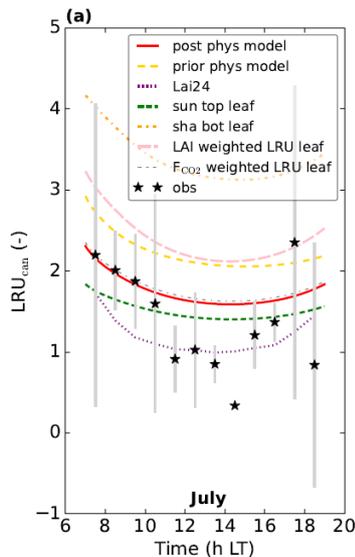
37

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

41 **Note that in Table A1 we provide evaluation of the (coupled) model, by quantifying the fit with the**  
 42 **26 assimilated observation streams for July 2015 in Hyytiälä (using partial reduced chi-square**  
 43 **statistic). A subset of these observation streams is shown in Figure 2 of the manuscript.**

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

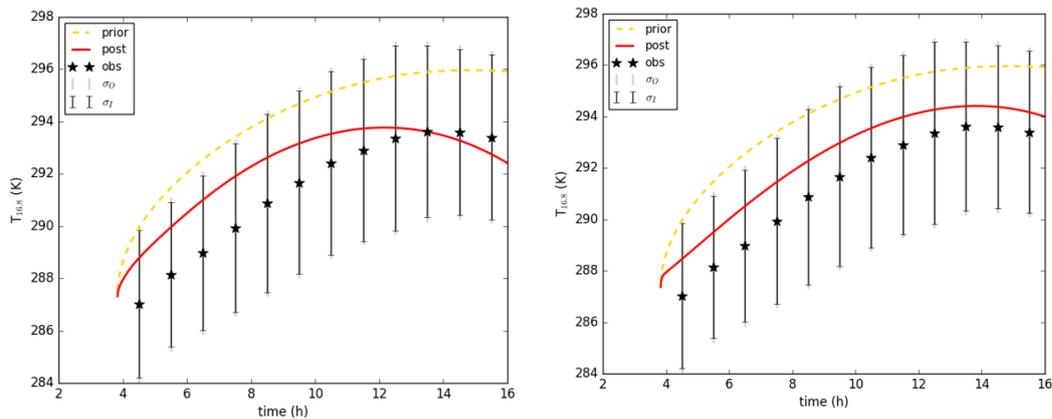
49 **Advection is set to zero indeed. A motivation for setting advection to zero is that we do not have**  
 50 **information on it, and we tried to keep the number of parameters that we optimise limited, to**  
 51 **reduce the complexity of the optimisation problem. Furthermore, the averaging over multiple days is**  
 52 **likely to reduce the influence of advection of different air masses to some extent, as this is likely to**  
 53 **differ between days. We have now performed an additional optimisation in which we include**  
 54 **advection of COS, CO<sub>2</sub>, H<sub>2</sub>O and heat in the state that we optimise. In this new optimisation, the cost**  
 55 **function is slightly lower (75 vs 77), as the model has more freedom to fit the observations, given the**  
 56 **extra advection parameters that are optimised. We show below a picture of the LRU for the new**  
 57 **optimisation:**



58

59 *Figure 3: As Figure 7a in the manuscript, but for the optimisation including advection*

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



67

68 *Figure 4: Temperature at 16.8 m height for the optimisation with advection (left) and without advection (right). Time is in*  
69 *hour UTC*

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

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

76 Chemistry modelling is indeed absent as well. At the remote locations that we model we do not  
77 expect a large influence of e.g. chemical conversions of anthropogenic CS<sub>2</sub> emissions into COS.  
78 Furthermore, our model simulations have a relatively short timescale (less than one day) compared  
79 to the relevant chemical timescales involved in the CO<sub>2</sub> and COS budgets.

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

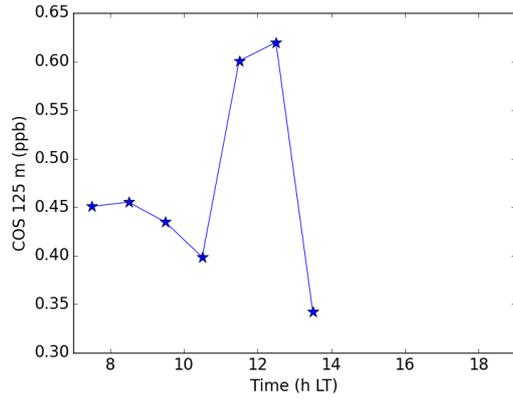
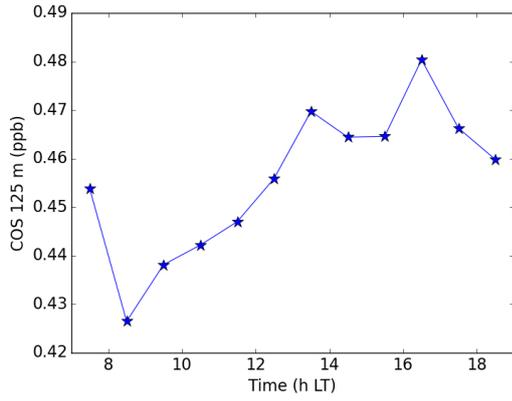
85 The COS variables are indeed very noisy, most likely caused by measurement uncertainty given the  
86 very small mole fractions compared to e.g. CO<sub>2</sub> (i.e. ppt vs ppm). Additionally, the averaging over  
87 multiple days might introduce a bit of noise due to a few missing data points. As the error bars for  
88 the COS variables are large, those measurements indeed contribute less to the cost function  
89 compared to observations with a small observational error. Thus, the fitted parameters are indeed to  
90 a large extent determined by other variables. This is not necessarily a problem, the COS observations  
91 are mostly fitted well within their uncertainty bounds. This can even be considered a strength of the  
92 framework, i.e. multiple information sources are taken into account. We do not optimize LRU itself,  
93 but, amongst others, COS and CO<sub>2</sub> fluxes and mole fractions that (to a large extent) determine LRU.

94 The difficulty with fitting LRU well also originates from the form of the equation, e.g. a small positive  
95 bias in COS plant flux and a small negative bias in the CO<sub>2</sub> plant flux can lead to a relative large  
96 deviation in LRU due to the division exacerbating small differences.

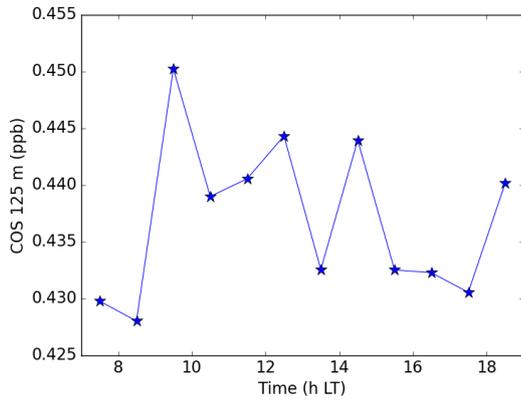
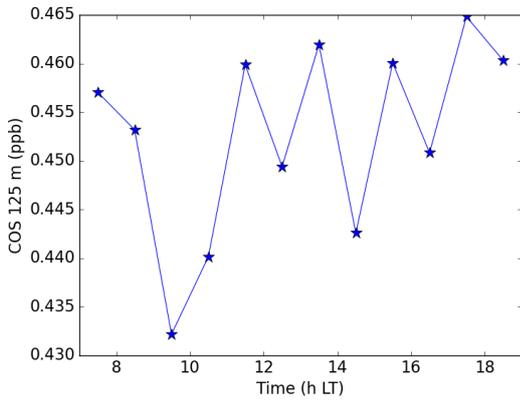
97 An increase in COS mole fractions in the morning continuing around midday could be explained by  
98 vegetation COS uptake at night, followed by entrainment of free tropospheric COS-rich air, see also  
99 Fig. 2a of Rastogi et al. (2018). However, the sudden increase in COS mole fraction in the 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

104

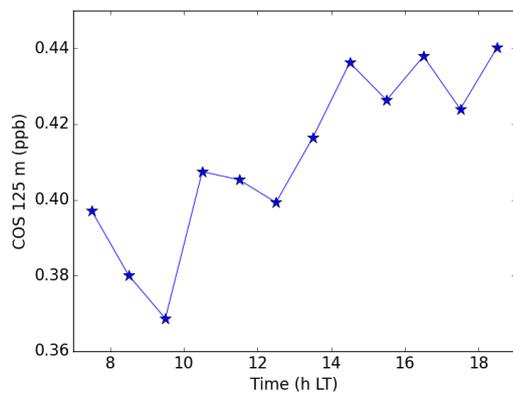
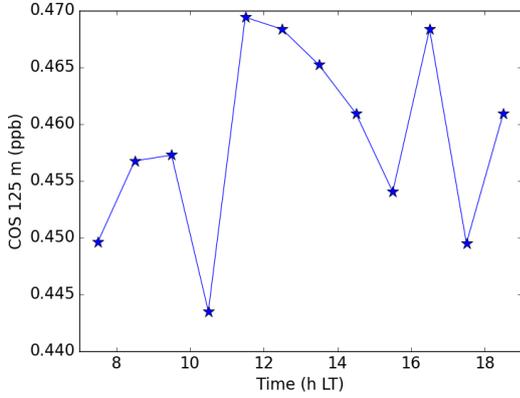
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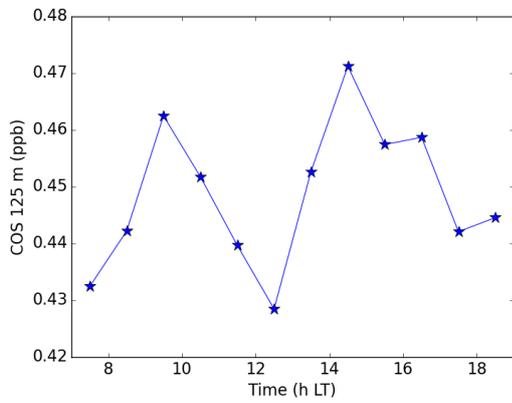
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109  
110

Figure 5: Observed COS mole fraction at 125 m height for the individual days in July 2015 (Hyytiälä) included for averaging in the manuscript

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

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

119

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

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

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

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

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

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

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

146 Figure 7 shows that the Lai24 parameterisation has a stronger variation between midday and  
147 evening/morning, especially in September. The observed large LRU values in early morning/late  
148 evening are (probably at least to a large extent) caused by open stomata while PAR is low. This leads  
149 to a CO<sub>2</sub> flux becoming close to zero (as the CO<sub>2</sub> uptake is strongly light dependent, given that PAR  
150 supplies the energy and reduction equivalents for the carboxylation process), while COS uptake  
151 continues as a diffusion process as long as stomata are open (destruction by carbonic anhydrase  
152 maintains a leaf-air gradient). The parameterisation from Lai24 seems to better capture this effect.

153 As mentioned around line 475 (line number referring to non-revised manuscript), in the A-gs  
154 photosynthesis model that is part of our modelling framework, the internal CO<sub>2</sub> concentration does  
155 not directly respond to photosynthesis and thus PAR. This ‘weakness’ of A-gs might be a reason why  
156 our model (and thus also our LRU parameterisation) does not capture the above-mentioned low-PAR  
157 effect well. However, one should realise that very low PAR conditions only occur shortly during our  
158 simulation period, and CO<sub>2</sub> uptake is very low in the early morning/evening. Therefore, this can be  
159 expected to be of limited importance when estimating CO<sub>2</sub> uptake using LRU.

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

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

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

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

179 6. A few points on discussion and conclusions: a. More emphasis should be put on the points raised  
180 at the end of Section 4.2, such that the inverse modelling framework is suitable to estimate COS and  
181 CO<sub>2</sub> uptake across different biomes. However, abundant measurements are required at such sites  
182 and that the results likely show that a separate exercise of establishing parameterisations at each site  
183 is necessary. Until the model achieves improved transferability.

184 The reviewer, with this comment, is mostly supporting the statement we make at the end of Section  
185 4.2 about representativeness of the results for the (needleleaf) Hyytiälä and Mieming sites in terms  
186 of COS/CO<sub>2</sub> uptake for other biomes. With our inverse modelling system we can at least better  
187 identify the role of processes involved in this COS/CO<sub>2</sub> uptake. But we also make a clear statement  
188 that for optimal application of our framework, we require more measurements than generally  
189 available for those other biomes to impose sufficient constraints on the inverse modelling system.  
190 Complete transferability of LRU parameterisations/ A-gs parameters between very different biomes  
191 might not be achievable, since vegetation characteristics might be too different among very different  
192 biomes. Our inverse modelling framework might also be useful for developing an optimal  
193 measurement strategy for other sites.

194 We added the following sentences: ‘Given that the Lai24 parameterisation seems to perform better  
195 at Hyytiälä, while our parameterisation seems to perform better at Mieming, it is likely that different  
196 parameterisations should be used for different locations to reach the best accuracy. Furthermore,

197 complete transferability of  $LRU_{can}$  parameterisations between vastly different biomes might not be  
198 achievable, as the response to PAR and VPD might differ, and vegetation-specific  
199 physiological/biogeochemical drivers of COS and CO<sub>2</sub> uptake might exist. However, the transferability  
200 between the Hyytiälä and Mieming locations suggests that at least within similar biomes, reasonable  
201 results can be obtained with a single parameterisation.'

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

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

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

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

218 c. In conclusions: "*For Hyytiälä, both the physical and regression model generally somewhat*  
219 *overestimated  $LRU_{can}$  with respect to the (noisy) observations. We found that the LRU of sunlit top*  
220 *leaves provides a relatively good estimate of  $LRU_{can}$ , which is encouraging for the use of canopy COS*  
221 *fluxes to estimate canopy CO<sub>2</sub> uptake. At the same time, we find that the simple leaf-scale*  
222 *parameterisation obtained in Hyytiälä by Kooijmans et al. (2019), rolled out globally by Lai et al.*  
223 *(2024), does not perform well in a more southerly needleleaf forest (Mieming, Austria).". This feels  
224 insincere to mention Lai24's underperformance at Mieming, while omitting its good performance at  
225 Hyytiälä. Particularly given the third research question raised in Section 1.*

226 Good point, we changed the sentence as follows: "we find that the simple leaf-scale  
227 parameterisation obtained in Hyytiälä by Kooijmans et al. (2019), rolled out globally by Lai et al.  
228 (2024), performs well in Hyytiälä, but does not perform well in a more southerly needleleaf forest ..."

229

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

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

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

#### 242 Minor Comments

243 1. Title: "Relative uptake of carbonyl sulphide to CO<sub>2</sub>: insights from a coupled boundary layer -  
244 canopy inverse modelling framework". Consider aligning the use of chemical abbreviation and  
245 written form.

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

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

251 Indeed the description is rather short. We now added the following information: 'In an optimisation,  
252 the framework aims to find the values of the parameters to optimise (the state), such that the cost  
253 function approaches its minimum. This is done starting from an initial guess of parameter values,  
254 after which the state is improved iteratively, thereby calculating the cost function and its gradient.  
255 For calculating the gradient of the cost function, an analytical gradient is available using the model  
256 adjoint.'

257 And for the forward model we added: 'Above the mixed layer, a discontinuity occurs in the scalar  
258 quantities, representing an infinitely small inversion layer. Above the inversion, the scalars are  
259 normally assumed to follow a linear profile with height in the free troposphere. The information on  
260 the free troposphere is used for calculating exchange between the mixed layer and the free  
261 troposphere.' We also added 'Above-canopy shortwave radiation is calculated using the date and  
262 time, cloud cover, and albedo.' We also added 'Upper soil temperature and moisture are simulated  
263 based on a force-restore model'

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

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

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

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

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

283 Technical Comments

284 Line 7: include - includes\*

285 Adapted, thanks for spotting this typo

286 Line 18: optimisations – optimisations\*

287 Adapted, thanks for spotting this typo

288 Line 24: Define VPD

289 Adapted

290 Line 38: subscript 2 in CO<sub>2</sub> – CO<sub>2</sub>\*

291 Adapted

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

295 Added

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

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

301 Line 121: moisture -> H<sub>2</sub>O. As you are explicitly referring to the tracers, I think it would be best to be  
302 clear exactly which molecule you are referring to.

303 Adapted

304 Line 149: Figure -> Fig.

305 Adapted

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

307 Adapted, we now write “The formula for LRU<sub>can</sub> is given in Eq. 1, after rearranging.”

308 Line 237: Define VPD

309 Adapted

310 Line 315: small T for the

311 Adapted

312 Line 409: full stop after Fig - Fig.

313 Adapted

- 314 Check instances of capitalised LAI24, for example in the legend of Figure 10. Important to  
315 differentiate between Leaf Area Index (LAI) and Lai24 the parameterisation
- 316 **Adapted, thanks for spotting this!**

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