RC2: 'Comment on egusphere-2023-2833', Anonymous Referee #2, 10 Mar 2024

The O_2/CO_2 exchange ratio above a forest canopy is a valuable tracer for understanding carbon exchange at the air-sea and air-land interfaces. We conventionally used a constant for global application, but this ratio can change significantly on a regional scale, and the mechanisms are still unclear. This manuscript presents an insightful analysis of the dominant mechanisms that determine the O_2/CO_2 exchange ratio by either using observations from a single height or from multiple heights. This study highlights the complexity of only using CO_2 and O_2 measurements at a single height to quantify ecosystem carbon flux, pointing out the advantage of using measurements from multiple heights for a precise ratio estimate.

The manuscript provides a comprehensive study, and the line of thought is mostly clear. I believe this paper is of interest to the general audience of Biogeosciences. I only have several concerns regarding the model experiment designs and the readability of the paper. I recommend a minor revision before this paper can be considered for publication.

We thank the reviewer for their detailed assessment of our manuscript. We will address the remaining issues below.

Main concerns:

1. It appears to me that the CLASS model was run for only one day with a prescribed initial condition. These runs clearly do not reach a steady state. In this case, the result strongly stands on the initial condition (e.g., the initial jump).

The reviewer is indeed right that the diurnal cycle modelled by CLASS depends strongly on the used initial conditions. The study by Pino et al. (2012) also confirms this. Understanding the initial conditions is therefore crucial to understand the results of the model during the day. The high dependency on the initial conditions also means that a diurnal cycle does not reach a steady state.

I am curious about the rationale for the initial jumps used in this study.

The initial jumps are determined based on fitting the diurnal cycle of the model to the observations of the mole fractions of O_2 and CO_2, due to lack of direct observations of the jumps themselves for which one would need an aircraft or very tall tower. To make this clearer we added some extra information to line 249.

To make sure we do not fully depend our results on the initial jumps that are fitted to our data, we did the sensitivities analyses in which we changed the initial jumps (see section 4.3.1) and we discuss this in section 5.3.

I am also curious if the authors have tried to run the model for multiple days to reach a semi-steady-state and check if there are different results.

Our sensitivity analyses (section 4.3) are an illustration of how different initial conditions would influence the O_2 and CO_2 concentration, based on variations in the soil moisture and the jumps. These results therefore correspond to runs for multiple "days". The results of the sensitivity analyses are shown in the form of the budget analyses in figures B1 and B2 and in the form of the final ERatmos/ERforest ratios (figure 7). These sensitivity analyses show indeed a wide range of results, but the message remains the same: ERatmos only rarely directly represents ERforest.

To strengthen this argument, for a manuscript in preparation we are currently investigating O_2 and CO_2 measurements from a campaign that was done in the Netherlands in 2022. Measurements for both O_2 and CO_2 where made from flask samples taken below and above the boundary layer height (using aircraft). In our response to Reviewer #1, we show in figure 3 some first results from that new analysis. The jumps in the morning of this measurement campaign are 47.2 ppmEq and -38.0 ppm for O_2 and CO_2 respectively. Based on those measurements we have designed a numerical experiment with a new CLASS case (Figure 4 of our reply to Reviewer #1). The analysis shows similar results as our "lower jump ratio case" from Figure B1. This new campaign confirms that the sensitivity analyses from our study are a physically-sounded representation of the variability of ERatmos during days with different initial jumps of O_2 and CO_2.

To make it more clear that the sensitivity analyses can be seen as representative for other days we included some extra information in line 346.

2. I appreciate the detailed study on factors modulating the ER_{atmos} and ER_{forest}. The result part, however, contains too many details and reads more like a technical report. I suggest improving the readability, by either revising the leading sentence of each section to sharply focus on the main result or adding a leading paragraph summarizing the main findings. The abstract section also contains too many technical details. I suggest shortening it significantly.

We agree with the reviewer that the manuscript is relatively technical, making it challenging to read. Since this type of study has not been done before for O_2, we have chosen to include quite some explanations and conceptual figures, which we hoped would facilitate the understanding of the material we present (e.g. section 2 on the fundamental concepts). We acknowledge that this might make it technical to read the manuscript. We have therefore initially aimed to not include too many detailed figures in the main text and have rather shown the elaborated details only in the supplement. Based on this comment, we have tried to improve the readability further by removing technical details from the abstract and by adding some explanatory notes in the beginning of certain sections.

Minor comments:

1. Figure 1 is a little bit unclear. Could you label thick arrows with F(O_2)s and F(CO_2)s. It is also not clear in the figure that ER_{forest} is actually calculated from the gradient. The two-sided arrow across BL seems to suggest that O2 and CO2 entrainment are of similar magnitude.

We have adjusted Figure 1 following the comments from the reviewer. It is important to note that the size of the arrows of the fluxes in Figure 1 are not a representation of the real ratio between the O_2 and CO_2 fluxes. The sensitivity analyses clearly show that the ratio between the entrainment fluxes can change per day. We did not want to make Figure 1 only a representation of the 2019 case that was modelled with CLASS, but rather represent a typical diurnal cycle.

2. L61: Expand on 'small scale process' upon its first mention for clarity.

Small scale processes are first mentioned in line 45 and are specified as forest exchange that occurs in and below the canopy. With large scale processes we mean mainly the influence of entrainment and this was already clarified in line 58.

3. L74: Please elaborate on extreme conditions (i.e., low SMI, etc.)

We have added some elaboration on what we mean with extreme conditions to line 74.

4. L132: Modify Eq. 5 to reflect that ER_{forest} is derived from a gradient.

We have added gradients to Equation 5, to make it clear that ERforest is derived from fluxes calculated from vertical gradients.

5. L139: It is not clear how you calculate DtO2 and DtCO2. Based on fit to high-resolution data? What's the time window?

The values for DtO_2 and $DtCO_2$ in Equation 6 are normally not directly determined, rather, ERatmos is the result from a linear regression of O_2 and CO_2 data over a certain time window (Ishidoya, 2013; Keeling and Manning, 2014). The slope of this linear regression represents ERatmos and therefore represents the change of O_2 over time (DtO2) compared to CO_2 (DtCO2). The fit of the linear regression can be applied to concurrent O_2 and CO_2 values when values for more than 1 time step are present. This does not necessarily need to be high resolution data.

The resolution of the data and the time window are important to understand which processes and how much detail is included in the resulting ERatmos value.

The high time resolution of the CLASS model (1 data point per 10 seconds) shows a more detailed diurnal cycle of ERatmos with higher values compared to the observational data (1 value per 30 minutes). The time window over which the linear regression is applied also changes the resulting ERatmos value (see the different periods in Table 1), because different processes become important during the day.

We added some clarification in Line 139 about the time resolution of our observations and our model output, as well as on the time window of the linear regression.

6. L336- 339: According to Fig. 4b, 2018 features a very low ER_{forest} during the night. Could you comment on whether it is related to elevated soil temperatures that only matter at night?

As described in Lines 306-310, the diurnal cycle of ERforest quantifies the ratio between TER and GPP. In Line 336 we explain that the low ERforest in 2018 is caused by a higher air and soil temperature. In the early morning in the 2018 case, the high soil temperature increased the TER while the GPP stayed relatively low just after sun rise. An increased TER flux compared to the GPP flux results in a lower ERforest value (Figure 3).

To make this clearer, we have added some extra information to Line 337.

7. Figure 8: This figure needs extra details. Arrows indicate sunrise to sunset. Better labeled on the figure to make this point clear.

We agree with the reviewer that this figure could be improved. We have adjusted the legend, and added clarifications in the figure, including arrows that indicate sunset to sunrise. The darker colors represent the vertical profile just after sunset and the lighter color indicates the vertical profile at the end of the night, just before sunrise.

8. L527-528: The finding that ER_{atmos} can be so large compared to ER_{forest} stands on the assumption that there is no vertical gradient in CO_2 and O_2 within the BL. If the resolved ER_{atmos} is based on using data that is very close to the top of the BL, this ER_{atmos} can be more sensitive to entrainment, compared to other studies.

We agree with the reviewer that the vertical gradient of CO_2 and O_2 within the BL, specifically close to the surface, indeed affects ERatmos. Under convective conditions 70 to 80% of the boundary layer will be well-mixed and only a gradient will be present near the canopy and in the entrainment zone. Closer to the canopy the ERatmos value would be less influenced by entrainment compared to further away. In our previous study (Faassen et al. 2023) we discuss this and show that the measurements at 125 m height gives an ERatmos value of 3.40, whereas the measurements at 23 m result in an ERatmos value of 2.28 for the same day. The ERatmos value closer to the canopy is lower and closer to ERforest. However, the ERatmos value of the 23 m level still is influenced by entrainment.

. We have added some information in Line 511 to make it clear that the height at which ERatmos is determined also influences the difference between ERatmos and ERforest.

9. Figure A2: Why the model results are not extended toward the beginning and the end of the day

The CLASS model is only valid when the sensible heat flux (SH) is larger than zero, because it uses the mixed-layer theory. The mixed layer theory assumes that the boundary layer is fully mixed, which happens during the day when the atmosphere is unstable. The SH needs to be larger than zero to produce buoyancy and therefore mixing. During the night, the SH is negative and therefore the mixed-layer theory does not hold.

To make clear that the model needs a SH larger than zero and therefore only runs during the day, we added some extra information to Line 436.

10. Figure A3: It seems like the model overestimates the BL height in the afternoon and around the sunset. The simulated O_2 concentration also seems to have a clear phase lag compared to observation. How would these affect the simulated ER?

We agree with the reviewer that the CLASS model indeed shows an overestimation of the boundary layer height in the afternoon and shows a delayed onset of the increase in the O_2 values compared to the observations (Figure A3). The overestimation of the boundary layer growth between 13:00 and 14:30 (Figure A3a) results in an overestimation of the entrainment velocity within this time frame (Equation 4). This will however hardly influence the O_2 or the CO_2 budget because the surface is already the most dominant process in this time frame (P3, see Figure 4d and 4e). The overestimation of the BL height is therefore not significantly influencing the ER signals.

The later onset of the increase in the O_2 mole fraction could indeed influence the ERatmos values, and the delayed increase in O_2 means that it does not completely match the timing of the change in CO_2 for the model, and therefore the ERatmos signal deviates compared to the observations. Table 1 also shows that there are differences between ERatmos values of the observations and the model, with the largest difference in P2 where the later onset has the most influence. However, in both cases we find that the ERatmos values can become very large (above 2), even without a perfect match between the model and the observations.

11. The title reads like the paper is trying to falsify the idea of using single-height CO_2 and O_2 observations to constrain surface carbon exchange. However, this approach is still valid if it only uses nighttime data. Given the detailed model experiments conducted in this study, I suggest modifying the title to better represent the comprehensive details of this work.

We agree with the reviewer that the current title does not reflect all comprehensive results from the study. However, we did not want to use a title that is too detailed or technical, and we therefore avoided words like entrainment or ERatmos versus ERforest in the title. Considering the suggestions of the reviewer to reflect our work more comprehensively, we have changed the title to:

'Separating above canopy CO_2 and O_2 measurements into their atmospheric and biospheric signatures.'

We would also like to add that we are not sure if ERatmos measurements during the night could represent the surface processes and that a one measurement height approach is still valid during nighttime. Measurements above the canopy could decouple from the surface during nighttime or advection could influence the measurements.

References:

Faassen, K. A., Nguyen, L. N., Broekema, E. R., Kers, B. A., Mammarella, I., Vesala, T., Pickers, P. A., Manning, A. C., Vilà-Guerau de Arellano, J., Meijer, H. A., et al.: Diurnal variability of atmospheric O2, CO2, and their exchange ratio above a boreal forest in southern Finland, Atmospheric Chemistry and Physics, 23, 851–876, 2023.

Ishidoya, S., Murayama, S., Takamura, C., Kondo, H., Saigusa, N., Goto, D., Morimoto, S., Aoki, N., Aoki, S., and Nakazawa, T.: O2:CO2 exchange ratios observed in a cool temperate deciduous forest ecosystem of central Japan , Tellus B: Chemical and Physical Meteorology,750 65, 21 120, https://doi.org/10.3402/tellusb.v65i0.21120, 2013.

Keeling, R. F. and Manning, A. C.: Studies of Recent Changes in Atmospheric O2 Content, vol. 5, Elsevier Ltd., 2 edn., https://doi.org/10.1016/B978-0-08-095975-7.00420-4, 201

Pino, D., Vilà-Guerau de Arellano, J., Peters, W., Schröter, J., van Heerwaarden, C. C., and Krol, M. C.: A conceptual framework to quantify the influence of convective boundary layer development on carbon dioxide mixing ratios, Atmos. Chem. Phys., 12, 2969–2985, https://doi.org/10.5194/acp-12-2969-2012, 2012.