RC2: 'Comment on acp-2022-504', Anonymous Referee #2, 06 Sep 2022

Diurnal variability of atmospheric O_2, CO_2 and their exchange ratio above a boreal forest in southern Finland

Faassen et al. present a highly novel dataset of O_2 and CO_2 measurement in the surface layer over a boreal forest. Such measurements are technically very challenging making this study one of the very few so far that have succeeded to apply O_2 in micrometeorological land surface flux measurements. Typically, the signal to noise ratio in O_2 gradient above forests is very small which limits the application of the flux gradient methods. Here the authors make use of a 125 m tall tower to increase the O_2 gradient.

A major challenge in this study is that the measurement uncertainty of the O_2 system is below comparable systems. This limits the interpretation of the data. Nevertheless, in my view the authors found a suitable way forward by aggregating the data to a "representative day".

While the experimental design and analysis is well done, there are several aspects that need to be addressed before publication.

We thank the reviewer for their review and assessment of our manuscript. We will address the points raised below. Note that line numbers given refer to the line numbers in preprint version.

Major comments

- Footprints: A major question regarding the study is that the ERatmos values are much higher than in previous studies. Some potential reasons are discussed in lines 481 to 490. What I am, however, missing is a proper treatment of the concentration footprints. Firstly, they differ between heights, particularly if the height difference is 100 m. This could lead to situations where the bottom height sees the local land surface whereas the top height sees air influenced at a regional level. Secondly, right next to the towers (roughly 200 m) is a large lake. Given that lakes have different O_2:CO_2 exchange ratios, I am wondering how this would influence the observed signal. Some of the co-autors have published articles on eddy covariance flux measurements over that lake. For the manuscript it would be help to add a footprint analysis and evaluate and discuss the influence of these two aspects on ERatmos and ERforest.

  For a detailed explanation on the high values of ERatmos compared to previous studies, we would like to refer to our answer of point 1 of reviewer #1. We explain there why we think entrainment is the most likely explanation of the high ER_{atmos} values that we observed.

  We agree with the reviewer that a more extended explanation about the footprints of the measurements should be added. We have therefore calculated the footprint for the flux gradient (Figure 1) and we looked in more detail at the footprint of the concentration measurements.

  Looking in more detail at the footprint of the concentrations measured at 23 m and 125 m (Carbon Portal ICOS RI, 2022), we find that the measurements at both heights

are strongly characterized by background signals, mainly forest and also ocean. The influence of fossil fuel is limited at both heights. Despite the high influence of background signals on both measurement heights, both heights are still able to capture the diurnal cycle of the forest. Our measurements heights are not disconnected from the surface but they integrate the surface and the atmosphere, as we observe two clear diurnal cycles. This suggests that both heights are measuring roughly the same areas. We agree with the reviewer that the difference between the heights (100 m) is quite large compared to previous studies. This large difference was used to be sure we were able to measure a O_2 gradient. If we can improve the accuracy of our O_2 measurements, we would certainly choose a smaller difference between the two measurement heights for a next campaign or add a third measurement height.

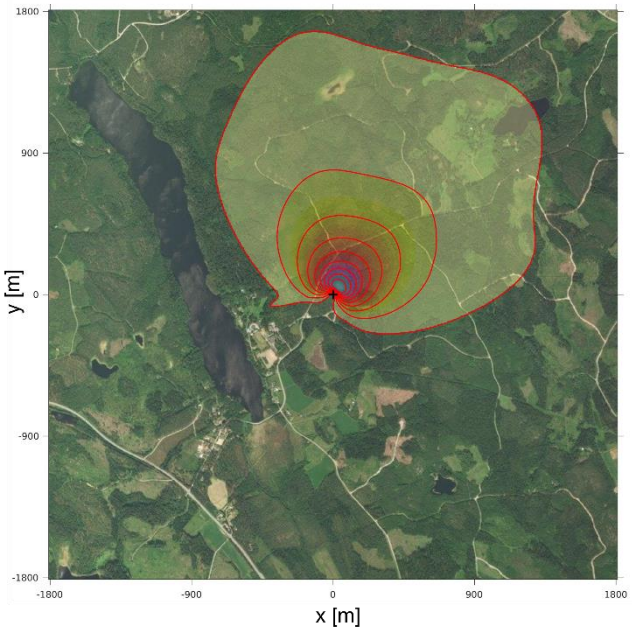

Figure 1 The footprint of the O2 and CO2 surface flux, determine with the gradient method, for the days between 7 through 12 July 2019 at Hyytiälä. The lines and contours indicate the contributions to the footprint from 10% to 90% in steps of 10%. The plus sign (+) indicates the place of the tower. The lake is located west of the measurement tower without influencing the measurements. This is the same Figure as figure B3 in the paper.

For the flux calculations based on the gradient method we calculated the footprint with the method of Kljun et al. (2015) by using the geometric height (Figure 1). Figure 1 shows that the main wind direction during the aggregate day was coming from North to Northeast and therefore the lake did not fell inside the footprint for these days.

To be clearer about the footprints of the ER_{atmos} and ER_{forest} signals we modified the text in the line 289 and added Figure 1 to the appendix.

- Flux partitioning: It I understood correctly, the exchange ratio of assimilation (ERa) is calculated based on equation 8 assuming a constant ERr and ICOS data of NEE, GPP and TER (line 276). Once ERa and ERr are retrieved for one representative day, these values are used to calculate GPP and TER on other days. For me it is not clear what we learn from this exercise as GPP is used to constrain ERa and then ERa used to constrain

GPP. Other studies such as Wehr et al. 2016 Nature have shown that NEE partitioning with an independent method using 13C in CO_2 resulted in lower TER and lower GPP compared to the temperature-based function following Reichstein et al. 2005 possibly indicating a Kok effect. If now the temperature-based GPP is used to calculate ERa, the O_2 based method does not provide additional and independent information. While I understand that the authors have no independent measurements of ERa at hand, I still miss a more careful discussion including Wehr et al. 2016 and addressing the limits of this approach.

We agree with the reviewer that our method is indeed not completely independent when estimating GPP and TER with the O_2 method. We indeed use GPP to estimate ERa and use this ERa on another set of days to estimate GPP again. However, this analysis is not completely circular, as we calculate a new ER_{forest} for a new set of days. However, due to this assumption, we could not make a completely independent comparison between the EC method and the O_2 method, but we show this to give a first estimate of the flux partitioning using O_2 to highlight the benefits of using the O_2 method. Further research will indeed focus on separately deriving ERa independently from EC.

Our goal with the analysis presented in Figure 8, was to determine if the O_2 method gave similar results compared to the EC method, by applying the ER values we determine during this study to another day. In the text we discuss that we find realistic values for ERa and ERr and therefore we are confident that they could be used in the O_2 method. Independent estimates of ERa would likely result in only minor changes compared to our estimate. Small changes in ERa would have similar effect on the uncertainty of the partitioning compared to small changes in ERforest (Figure 8). The biggest source of variability in the O_2 method is the ERforest of the new set of days. The ERforest depends on the O_2 gradient, and this gradient was difficult to determine during our measurement campaign because of the relatively low measurement precision. However, figure 8 still shows that we find a good comparison between the O2 and the EC method for the partitioning of the fluxes and we present it to show the potential of the method.

To independently determined ERa and ERr, one would use branch and soil chambers, or lab measurements to measure process-level O_2 and CO_2 exchange which were not included unfortunately in our campaigns. When chamber measurements would have been available, we could have made a more detailed comparison between the O_2 and EC method and compare this result with the study of Wehr et al. (2016). With a more detailed analysis we could potentially say of the O_2 method gives a higher or lower estimation of GPP and TER and if this differs compared to the 13C method from Wehr et al. (2016).

We added text to line 611 to make to address these points.

Minor comments

- Line 23: better “net uptake” than “uptake”:

We agree with the reviewer and changed therefore uptake into net uptake in line 23.

- Line 23 and 24: better be consistent using ether land biosphere or terrestrial biosphere:

We agree with the reviewer on this point and changed therefore land biosphere into terrestrial biosphere in the lines: 23, 44, 45.

- Line 27: Add a citation for last sentence in first paragraph.

We agree with the reviewer and have added citations the end of the first paragraph in line 27.

- Line 30-32: here I am missing a mentioning of Wehr R et al. 2016 Nature where they showed that fluxes partitioning using 13C differ from fluxes partitioning following Reichstein et al. 2005.

We agree with the reviewer and have added the citation of Wehr et al. (2016) to line 32. For a more elaborate comparison between our results and Wehr et al. (2016), please look at the discussion above of the second major point.

- Line 36/7: fluxes of O_2 and CO_2 are opposite. Here a positive ER is used. It might be helpful to indicate this by saying "indicates the amount of moles O2 consumed per mole of CO2 produced (or vice versa)".

We agree with the reviewer that this sentence should be made clearer. Therefore, we changed line 36/37.

- Figure 1: in the text of the introduction the term GPP and TER are used and in figure 1 respiration and assimilation. Please use consistent terms.

We agree with the reviewer that we have to be consistent when using these terms and have to be clear what we mean with them. Therefore, we added Gross Primary Production (GPP) and Total Ecosystem Respiration (TER) in figure 1b, to make the link with the text clearer. However, our opinion is that we cannot use GPP and assimilation interchangeably because they indicate different scales. GPP is a measurement that applies to the ecosystem level and assimilation is more related to the process/leaf level. To make this more clear, we changed the text in the caption of Figure 1.

- Line 90 to 94: personally, I prefer if the given objectives are presented with the term "objectives" for allowing speed-reading. Maybe a matter of taste

We added the word 'objective' in line 90 to make it easier for people that want to speed-read this paper.

- Line 113: what is the influence of the nearby lake on the exchange ratio. The footprints at 23 m and at 125 m are very different. How does this influence the results?

The lake has minimal influence on the measurements both at 23 m and 125 m, as they both mainly measure background signals. The difference in footprint becomes important when looking at the surface flux calculations. The footprint of the flux calculations shows that the signal we measured did not originate from the lake during our representative day (Figure 1). Air masses that are influenced by the presence of the lake are therefore absent. We elaborated more on the footprints at the first major point in this review above, and have changes the text at line 289 and added Figure 1 above to the appendix.

- Line 129/130: It seems that the sampling lines are alternatingly flushed with 120 ml/min and 2 l/min. Has it been evaluated whether these changes in flow rate lead to any effects on the O_2 signal? Or are all these effects removed by discarding the first 4 minutes after switching.

These effects are indeed removed by discarding the first 4 minutes after switching heights, as the flow rates adapt within that time. We use these different flow rates to make sure the samples lines are properly flushed before switching, preventing delays in the measurements of the new height.

- Line 210: I find it confusing that in equation 5, eddy covariance terms for the turbulent fluxes are presented, but the turbulent fluxes are obtained from flux gradient measurement. Why are not equation 5 and 6 combined?

We agree with the reviewer that it can be confusing to use the eddy covariance terms, while we calculate the surface fluxes of O_2 and CO_2 with the gradient method. Therefore, we changed the terms that indicate the surface fluxes from $w'CO_2'$ and $w'O_2'$ into F_{CO2} and F_{O2} throughout the paper to clearly show that they are inferred from gradients.

We did not want to combine equation 5 and 6, to make it as clear as possible how ER_{forest} should be calculated from the surface fluxes of O_2 and CO_2. These could (in other setups) possibly also be measured directly from EC measurements of O_2 and CO_2. However, we derived these from the vertical gradients and therefore showed in a separate equation how we did that. To make the connection between equation 5 and 6 clearer we now refer to equation 6 in line 215. (Note that an equation was added (equation 4) in the new version of the paper, and therefore the numbers of the equations in the paper changed).

- Line 218: In my view it is not the stability that characterised if in a period respiration or assimilation dominate, but it is the radiation regime. Why was here stability used and not nighttime vs. daytime?

We agree with the reviewer that indeed the radiation regime is also of importance to distinguish between the different periods. To make it more clear in the text we changed line 218.

- Line 255: unit is missing. Should be "0.4 m s-1".

We agree with the reviewer about this point and changed therefore the text in line 255.

- Line 271: here it is referred to ICOS NEE and GPP from EC measurements. It would be good to say how ICOS partitions NEE into GPP.

We agree with the reviewer and have added a reference and more information in line 275 on how the GPP is determined in Hyytiälä.

- Line 304: why was a fixed calibration time during the day selected (20:00 - 22:00). An alternative could be using a moving calibration time.

We agree with the reviewer that a moving calibration time would benefit our data, and we generally applied a moving calibration time period for both the 2018 and 2019 campaigns. The calibration tanks are measured every 23 hours (see line 169) and take 2 hours to

complete. However, between the 7th and 13th of July we decided to fix the calibration time. During this period radiosondes were launched at Hyytiälä, which we will use for a follow up study. To make sure we captured the morning transition well during this period, we decided to fix the calibration time between 20:00-22:00, to allow a smoother planning of the radiosonde launches. At the time of this decision, we did not know we wanted to make an aggregate representative day. By coincidence the period between 7th through 12th July resulted in the best O_2 gradients and therefore this fixed calibration time is present in our representative aggregate day.

To make this more clear we adjusted the text in line 304.

- Line 307: 0.70 ± 0.65: the unit is missing.

We agree with the reviewer about this point and changed therefore the text in line 307.

- Fig. 4a: for the height 23 m, the CO_2 concentration varies with a range of 15 ppm, whereas the O_2 concentration varies with a range of 35 ppmEq. Wouldn't we expect to see a similar range of variation? What is the role of the nearby lake?

If our signal was only influenced by the forest, we would indeed expect a similar range for both O_2 and CO_2. However, the concentration measurements are highly influenced by entrainment as well. How much entrainment is influencing the data depends on the boundary layer growth and the difference in concentration between the boundary layer and the free troposphere, also called the jump. If there is a difference in this jump for O_2 and CO_2, this will result in different range of the effect of entrainment. It is highly likely that the jump of O_2 and CO_2 are not the same, as the background air in the free troposphere contains different sources of e.g. fossil fuel emissions and ocean exchange. These processes affect the O_2 and CO_2 concentration differently. The air that is entrained could also be affected by the memory of the day before or advection during the night inside the residual layer. A more elaborate explanation of how we think entrainment influenced our measurements is given in the first answer to reviewer #1 (see 'Explanations for high ER_{atmos} values'), and we have update the text in line 481 to further explain this..

The lake will have had likely very little effect on our measurements, as was already discussed with the first major point, see above.

- Fig. 4b: at night we see a vertical gradient in O_2 concentration (roughly 10 ppmEq) that exceed instrument precision (roughly 4 ppmEq), but during daytime the gradient is – even averaged over multiple days – lower than instrument precision. To me it is unclear how the uncertainty of the measurements is propagated to the final fluxes and ERforest.

We agree with the reviewer that it was not clearly described how the uncertainties on our measurements were calculated. The error bars in figure 4b are not the same as the instrument precision presented in table 1. The error bars in figure 4b, the following figures, and the uncertainties in the ER signals are all based on the standard error of the 30-minute average CO_2 and O_2 concentrations. The error bars in 4b are then determined by calculating the uncertainty for the aggregate data points, based on this standard error (that we included in the revised version of the paper as equation 4) and by using error propagation. To make this more clear, we added additional information to the text, in lines 166 + 192, and the added

equation 4 after line 192. We also added this information to the caption of figure 4, together with extra text in lines 332, 380 and 401. See also point 4 of reviewer #1.

- Line 318: in P3b: O_2 and CO_2 concentration changes show the same sign, instead of the expected opposite sign. This is related to an instability of the MKS pressure regulator. It is unclear why this effect should only affect P3b and no other times of the day. How was this evaluated?

We agree with the reviewer that this was not clearly explained in the text. We give an elaborate explanation about this in the response of the first major point of reviewer #1 and added a new figure to the appendix. Please see our response to reviewer #1 for our answer and the updates to the paper.

- Fig. 5: Which regression type was used to calculate the regression?

This was a linear regression. To clarify this, we changed the text in line 208 and the caption of figure 5.

- Line 339: Given that the measurement uncertainty is so high compared to the variation during P3a, I am wondering how the uncertainty could be included via error propagation when calculated the slope and its uncertainty.

The measurement uncertainty per data point of the O_2 concentrations of the aggregate is around 2 ppmEq (see Figure 4). This is relatively low compared to the increase of O_2 during the day (30 ppmEq) or the decrease during the night (15 ppm Eq). Incorporating the uncertainty of the datapoints into the slope has therefore little effect on the uncertainty of the slope.

The largest uncertainty in the ER_{atmos} values is the definition of the time boundary of where the influence of entrainment stops, which is difficult to incorporate into the slope of the linear regression. We therefore leave the uncertainty of the ER_{atmos} values as it is, and we indicate that the ER_{atmos} value of P3a could deviate a lot when the time boundaries of this period shifts.

- Fig. 6: The units of the fluxes are given in ppm m m-1. This is very unusual for the flux community. Typically, the fluxes are reported in µmol m-2 s-1. Also, I find it confusing that the y-axis label is the covariance, but the fluxes are calculated from a flux-gradient approach and not from eddy covariance.

We agree with the reviewer about these points and changed therefore the units of the fluxes to µmol m-2 s-1 in the text and the figures, and removed the eddy covariance notations from the paper. As mentioned before, we now stated clearly that the fluxes are inferred from gradients using flux-gradient method formulations.

- Fig. 6b: Could please describe in the caption what are the error bars. Could just be moved from the main text (line 380). Also here, it is unclear to me if an error propagation incl. measurement uncertainty was carried out.

We agree with the reviewer about this point and changed the caption of figure 6. We also clarified the calculations of the error bars in the text, see our answer above and our answer to reviewer #1 for more explanation.

- Fig. 7 : It is surprising to see ERforest values at -2 to -2.5. This is much more negative that other reported data and it is unclear what this could mean physiologically. It is also surprising that the fluxes with the most negative values are also the largest fluxes, where we would expect to see large gradients and thus robust flux calculations.

We agree with the reviewer that ER_{forest} values of 2 to 2.5 are highly unlikely if they would only represent forest exchange. However, we do not think data points in figure 7 can be interpreted individually. As was explained in the manuscript, the concentration measurements have a relatively high uncertainty compared to previous studies and therefore the vertical gradient is difficult to determine. We therefore use an aggregate day to get a more robust estimate for the O_2 gradient, however with a remaining uncertainty on the individual data points. This also translate into the individual estimates for ER_{forest} per time step, where for some time steps, very high O_2 surface fluxes caused such unrealistic values for ER_{forest}. We therefore use the average O_2 and CO_2 surface fluxes to determine the final ER_{forest} values.

As the reviewer states, the large O_2 surface fluxes during the day follow from a larger vertical gradient of O_2. However, this gradient is still small compared to the gradient during the night and therefore still had a relatively large uncertainty. A large gradient during the day could therefore result from the relatively large measurement uncertainty, and as a consequence result in these high O_2 fluxes and unrealistic ER_{forest} values.

- Appendix: Personally, I prefer that the units are shown as well.

We agree with the reviewer about this point and therefore we have added units to the equations in the appendix.

Citation: https://doi.org/10.5194/acp-2022-504-RC2

References:

Carbon Portal ICOS RI. (2022). *STILT station characterization for Hyytiälä at 17m*. Carbon Portal.

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