

In this manuscript, Faassen et al. tackle two questions:

- 1) How is it possible that the exchange ratio of atmospheric oxygen and carbon dioxide (inferred from the covariation of measurements of the abundance of these species at a single height) might be far higher than expected?
- 2) How does the covariation-based exchange ratio relate to the exchange ratio of the underlying forest, and is the latter really captured by a flux-based (two-height) set of measurements?

The answers are derived primarily from the CLASS model, characterizing the dynamics of the atmospheric mixed layer, along with measurements made at the Hyttiala research site in Finland.

Overall, this paper is a thorough and thoughtful investigation of a complicated phenomenon. It is a natural continuation of the work done by Faassen et al. (2023) and addresses many of the questions that were either left open, or were answered somewhat speculatively in that work. I feel the conclusions are well-supported and the paper is worthy of publication.

The manuscript is long and dense and includes appendices that are themselves substantive. I have the feeling that the authors might be able to improve the efficiency and clarity of their presentation, but this would require a wholesale re-write, and the current version is more than adequate (with some minor revisions suggested below). That said, I do find the abstract overly detailed – it would benefit from substantial shortening. In addition, the discussion could benefit from an effort to make the structure clearer to the reader.

We thank the reviewer for their assessment of our manuscript. We acknowledge that it is indeed dense in content and fundamental concepts. We struggled especially to keep it short because we also wanted to introduce some theory that is not commonly used in the field of atmospheric mole fraction observations.

We have shortened the abstract and tried to remove technical details from it. Also, based on other reviewers' comments, we have introduced some lines in the beginning of the results sections to better structure it. Likewise, we have added a few lines with an overview of the structure in the beginning of the discussion section.

My main scientific concerns/questions

In this work, ERatm is based on a single altitude well above the canopy. What if ERatm were based instead on a single altitude within the canopy (or even just above the canopy)? i.e. How would the conclusions of this paper change if ERatm were to be replaced with an ER based on the slope of an O<sub>2</sub> vs CO<sub>2</sub> plot made with data collected within the canopy? Perhaps a covariance-based ER is not intrinsically limited, but instead the limitations arise when the covariance data are collected far above the canopy. This might explain why the Hyttiala data show ERatm values that are not observed at other locations.

This is an interesting point. Unfortunately, it is not possible anymore to verify in-canopy values retrospectively for our campaigns in Hyttiala, that were done in 2018 and 2019. However, as we also wrote in response to point 8 of reviewer #2, we did compare the ERatmos values of the 125 m level with the 23 m level measurements. The canopy height at Hyttiala is around 18 meters. We note that the influence of entrainment on the measurements is smaller closer to the canopy at 23 m, but still present, and we get high values of 2.28 for ERatmos at that level. We have included this in line 511. The influence of entrainment on canopy level is for example also shown by the study of Patton et al. (2016) where they indicate that entrainment is also important at canopy level.

We would recommend to always use a height above the canopy to determine ERatmos, to make sure that the fluxes of the complete canopy are taken into account in the calculation. When measuring inside the canopy there is a risk that the fluxes above the measurement height are missed and the ER signal does not incorporate the complete GPP and TER fluxes. More research should be done on the implications of measuring the ERforest inside the canopy compared to just above the canopy.

My other questions center on the “jumps” between the atmospheric boundary layer and the free troposphere. These jumps are central to the explanation of the diurnal cycles in ERatmos (and by extension, the difference between ERatmos and ERforest). The sensitivity analysis done is very valuable. My concern is that these jumps seem to be chosen ad hoc. I don’t doubt that jumps exist, and I also am convinced by this work that they, along with entrainment, are the explanation for the observed values of ERatmos, but the empirical evidence of jumps shown in Figure 2 is far from compelling. The data are consistent with the black-line conceptual models shown, but they are equally consistent with smaller (or non-existent) jumps and smoothly-varying lapse rates within the free troposphere. For example, the 10:25 LT trace from 2018 shows a “jump” at 1000m that’s at least as big as some of the jumps depicted in the conceptual models. Likewise, the 9:56 LT trace from 2019 has a much bigger discontinuity at 1900m than it does at 1450m.

We acknowledge that we do not have observational evidence of the O<sub>2</sub> and CO<sub>2</sub> “jumps” which would have strongly improved our analysis. Note that in Figure 2, we only show observational evidence for the potential temperature profiles, which we could measure with radiosondes, but this is not possible for O<sub>2</sub> and CO<sub>2</sub> (even though we have actually tried for CO<sub>2</sub> in this campaign with very light weight CO<sub>2</sub> sensors, but they did not give accurate results). Since we don’t have observed values for the O<sub>2</sub> and CO<sub>2</sub> jumps, and as the reviewer says, there can also be variable, we assess the influence of different values for the jumps in our sensitivity analyses in Section 4.3.1.

In a recent new campaign, we did manage to get observations for the jumps for O<sub>2</sub> and CO<sub>2</sub>, based on flask samples collected from aircraft. This was in a forest in the Netherlands. In our response to reviewer #1, we have included preliminary results from that campaign, which confirm that the values that we use here are realistic.

The jumps of the potential temperature profile are based on the difference between the mixed layer and the lapse rate value at the boundary layer height. The boundary layer height is not chosen ad hoc, but determined with the parcel method (Kaimal and Finnigan,

1994). We added the reference to Kaimal and Finnigan, (1994) in Line 199. The radiosondes also do not represent the full extent of the boundary layer at the time specified in Figure 2, because they only go through one specific trajectory and therefore do not measure the complete mixed boundary layer above the forest. The vertical profile of potential temperature is therefore not perfectly representing the mixed layer.

Furthermore, the authors show that the jump ratios depend quite strongly on the composition of the free troposphere background air. Yes, if the background/free-troposphere air has very different O<sub>2</sub> and CO<sub>2</sub> values from what's in the boundary layer, then entrainment will result in extreme ERatmos values, but are the free-troposphere values explored in section 5.3 (and the sensitivity analyses) realistic? At least the authors acknowledge the need for direct O<sub>2</sub> and CO<sub>2</sub> measurements in future campaigns (line 473) but perhaps this need should be emphasized.

As we stated above, indeed we think that our values are realistic, based on the new campaign. We have also introduced this around line 473 in the manuscript.

Beyond these concerns, here are numerous minor editorial comments and suggestions:

In section 2, the authors refer to "mixed layer theory". In my mind, this isn't a theory- it's a model. It's a particular representation of the mixed layer, choosing to include some processes and not others, based on educated judgement of what is important and what isn't. If "mixed layer theory" is a widely used term (with which I'm simply not familiar), then stick with it, but otherwise, in the section title and throughout the paper, please replace "theory" with "model".

Mixedlayer theory is the theory on which the mixed layer model is based. It is a commonly used term in boundary layer meteorology and we therefore keep it as it is in the manuscript.

The first time "surface" is used, it should be explicitly defined. Is it the surface of the soil? The top of the canopy? Likewise, on line 134, how is "above the canopy" defined?

We have added the following line to the caption of Figure 1:

Note that the term "surface fluxes" refers to the fluxes from the surface layer, which includes the vegetation layer including the top of the canopy. The surface layer is the lowest 10% of the boundary layer where the surface directly influences the atmospheric boundary layer.

With "above the canopy" we mean that it is not inside that canopy. We explain this in line 134.

L8: should read "measured at a single height"

Changed accordingly.

L9: Should read “with the goal of relating the ERatmos signal to the ERforest signal and understanding the”

This sentence was changed while rewriting the abstract.

L20: should read “rarely represents ERforest directly and”

Changed accordingly (in a rewritten sentence).

L23: should read “we recommend always measuring”

Changed accordingly.

L26: should read “land use change emissions, moderated by uptake”

Changed accordingly.

L27: comma after “oceans”

Changed accordingly.

L28: should read “a valuable tracer, enhancing”

Changed accordingly.

L29: remove the comma after “exchange”

Changed accordingly.

L32: comma between “CO2” and “represents”

Changed accordingly.

L33: should read “allow us to”

Changed accordingly.

L44: should read "available instruments do not allow eddy covariance (EC)"

Changed accordingly.

Fig1 caption: should read “over time can lead to”

Changed accordingly.

L74: should read “that have not yet been measured”

We removed this part of the sentence, while adding some more details based on a comment from another reviewer.

L76: should read “In this study, we aim to”

Changed accordingly.

L77: should read “and we propose a new”

Changed accordingly.

L78: should read “measurements can be employed”

Changed accordingly.

L79: should read “aforementioned limitations.” And “ whether the ERatmos signal constrains boundary layer dynamics, and we identify”

Changed accordingly.

L89: should read “with the model CLASS (Sect. 3.) We then show the model”

Changed accordingly.

L91: should read “represents forest exchange (Sect. 4.) Next, we place”

Changed accordingly.

L92: should read “(not) be used (Sect.5.) Finally”

Changed accordingly.

Eq. 1: The “primed” terms should be explained/defined.

Changed accordingly.

L109: should read “represent large scale”

Changed accordingly.

L115: should read “associated normally with high pressure systems. We assume wsub is negligible.”

Changed accordingly.

Eq.3: Shouldn't there should be a subscript on the phi that is in time derivative? We are left wondering if this is phi<sub>bl</sub> or phi<sub>ft</sub>?

This is the boundary layer value (well-mixed tracer), as defined with equation 1. We repeated it now here as well.

Line 123: should read “layer height ( $dh/dt$ ) effectively determines the entrainment velocity, and by extension the entrainment”

Changed.

Eq. 4: the last  $w$  should have a subscript consistent with eq. 2 (i.e. both should be either  $w_{sub}$  or  $w_s$ ). Also, in the denominator, it's not clear what the delta refers to. The difference between where and where?

Changed accordingly.

Line 127:  $w_s$  or  $w_{sub}$  (like equations 2 & 4)?

Changed accordingly.

Line 130: As mentioned above, “Theoretical” versus “Modeled”

This is theory, not modelled.

Line 143: Should read “According to the mixed-layer model described above”

Changed, but get theory, as this is not the model.

Line 147: should read “term in Eq. 1 here, but we will add it later (Eq. 9).

Changed accordingly.

Line 149/150: should read "definition of ERforest (Eq. 5) with Eq. 2 allows us to rewrite Eq. 7 as:"

Changed accordingly.

Line 154: should read “and ERforest within the mixed layer model.”

Changed but kept theory.

Line 156: should read “effect of other large scale processes such as advection of O<sub>2</sub>”

Changed accordingly.

Eq. 9: Somehow, the authors should indicate that they're just introducing advection as an example of what one could do. They can refer to section 5.2 in which they explicitly neglect

advection. Whatever they decide, it should resolve the peculiarity of introducing terms which never get used.

Changed accordingly.

Line 160: should read “values are of particular importance here: When the”

Changed accordingly.

Line 163: The section that follows doesn’t describe the CLASS model; it describes measurements.

Changed accordingly.

Line 188: should read “et al., 1988). We use conserved”

Changed accordingly.

Line 188-190: I simply don’t understand this sentence. How does the use of mole fractions imply that tracers are well-mixed?

Agreed, we have removed this sentence since it was unclear and not necessary.

Line 197: should read “time of 6 seconds, effectively averaging over 10m of altitude.

Changed accordingly.

Line 213: should read “law of diffusion, based on the difference”

Changed accordingly.

Line 217: should read “TER fluxes. The differences between”

Changed accordingly.

Line 237/238: should read “sizes and signs, each with their own ER”

Changed accordingly.

Line 247: what is meant by “The final initial and boundary conditions”? Is “final initial” something in particular, or is it a list of three things (final conditions, initial conditions and boundary conditions)?

We have removed the word “final”.

Line 259: should read “Combe et al., 2015), given that”

Changed accordingly.

Line 265 and following: should read “and ERforest. Specifically, we looked at changes in ERatmos resulting from changing the different components of Eq. 8. The first sensitivity analysis uses the 2019 base case and investigates the effect of background air with a different composition by altering the initial jumps of O<sub>2</sub> and CO<sub>2</sub>. By only changing”

Changed accordingly.

Line 283: should read “While there is limited”

Changed accordingly.

Line 295: I’m puzzled by “an increasing net CO<sub>2</sub> flux out of the forest” since in panel d of Fig. 4, CO<sub>2</sub> is dropping during P1.

We have changed “out of” to “into” since indeed the net flux of CO<sub>2</sub> is negative (uptake). See also Figure A3.

Figure 4: In panel B there are two lines across the bottom. Presumably these are the values of ERforest, but they are unlabeled and the axis is ERatm (rather than a generic ER), so it’s confusing. Please clarify.

Good point, these lines are indeed ERforest. We improved Figure 4 by making the legends and axes more clear.

Figure 5: The caption doesn’t correctly describe what is in panel E. Please correct.

Changed accordingly.

Line 229: should read “exhibit higher values than the model predictions of Sect. A1 because”

Changed accordingly.

Line 345-346: should read “cases where ERatmos could equal ERforest if large-scale conditions were to change.”

This was rewritten.

Line 347: should read “(Sect. 4.3.1), and changes in climate (soil moisture”

This was rewritten.

Line 356: should read “dominant and closer to”

Changed accordingly.

Line 383: I'm pretty sure Figure 6 does not show energy balance closure.

Changed this.

Line 384: add a comma after "result"

Changed accordingly.

Line 385: should read "the respiration, up to a threshold"

Changed accordingly.

Line 386: should read "surface fluxes and an enhanced sensible heat flux. This will increase the boundary"

Changed accordingly.

Line 392-393: should read "focus on two particular locations in the parameter space shown in Figure 7:"

Changed accordingly.

Line 394: Begin a new paragraph with "A lower soil moisture..."

Changed accordingly.

Line 395: should read "decreases ERatmos during P2 and increases ERatmos during P3"

Changed accordingly.

Line 398: should read "and CO<sub>2</sub> change more slowly and remain"

Changed accordingly.

Line 401: add a comma after "result"

Changed accordingly.

Line 402: should read "similar to the"

Changed accordingly.

Line 426-427: I am not sure if you are saying ERa is set by nitrogen content in the leaf and light striking the leaf (i.e. "leaf level" applies to both nitrogen and light), or whether you are saying ERa is set by nitrogen content of the whole plant and light that strikes the leaf. Please clarify.

Changed accordingly.

Line 430: should read "how ERatmos can change during"

Changed accordingly.

Line 437: should read "ERforest. This also has the potential to improve estimates of the global biospheric ER, currently taken to be 1.1 (Severinghaus, 1995)"

Changed accordingly.

Line 450: should read "2022a). However, caution should"

Changed accordingly.

Line 452-453: should read "adverted air. In addition to the surface and entrainment influences, ERatmos also depends on the magnitude of the advected flux. This is because mixing two ER"

Changed accordingly.

Line 454: should read "of two sources with"

Changed accordingly.

Line 455: should read "ERatmos values. A solution could be to include other tracers in the"

Changed accordingly.

Line 459: remove the comma after "processes"

Changed accordingly.

Line 460: should read "signal. During the day,"

Changed accordingly.

Line 465: should read "ERforest may be due to either"

Changed accordingly.

Line 466-467: should read "ratio. If the cause is the former (low BetaCO<sub>2</sub>), the ERatmos signal during P3 should be closer to ERforest. If the latter (a high jump ratio), ERatmos should remain well above ERforest in P3."

Changed accordingly.

Line 473: should read “recommend that future measurement campaigns include”

Changed accordingly.

Line 479: should read "In the absence of observational"

Changed accordingly.

Line 485: What exactly is caused by mesoscale and synoptic processes? Subsidence, or the existence of the jump? Is it correct to say that they can cause subsidence that in turn creates a jump? Please clarify.

We meant the subsidence. Changed accordingly.

Figure 8: This is a valuable and information-rich figure, but the legend in Panel A is a bit confusing since there's no dark green in that panel. I suggest changing the green bar in the legend to light green (to match the light gray), and add something to the caption describing the difference between dark and light colors. Also, the arrows to/from the trees are a bit confusing since they only apply to a period of net respiration. I suggest taking out the green arrow and adding a few words to the caption description of Panel A saying it depicts only CO<sub>2</sub> fluxes/abundances during a period of net respiration.

We have modified the figure according also to comments from reviewer #2. The flux arrows are indeed for net respiration, since it is during the night time. We have also updated the caption to clarify.

Line 492: By eye, ERforest seems to be less than 1.0 in all of the panels (the green arrows are always longer than the gray arrows.) Isn't this the case?

The reviewer is indeed correct that the green arrows (indicating O<sub>2</sub>) are always longer than the grey arrows (indicating CO<sub>2</sub>). However, this means that ERforest is larger than 1.0 because ERforest is O<sub>2</sub>/CO<sub>2</sub>. This decision was made because several studies have shown that ER<sub>r</sub> (ER of respiration, the ERforest during the night) is larger than 1.0 (Ishidoya et al., 2013; Angert et al., 2015; Hilman et al., 2022). This was briefly mentioned in line 504. We added the above-mentioned studies for extra clarity.

Line 495: should read “This can occur for example, when the”

Changed accordingly.

Line 504-505: should read "found a non-linear relationship between O<sub>2</sub> and other tracers that was difficult to explain. While"

Changed accordingly.

Line 519: should read “constant ERatmos value.”

Changed accordingly.

Line 522: remove the comma after “that”

Changed accordingly.

Line 526: should read “ERforest are likely rare.”

Changed accordingly.

Line 546: should read “Additional tracers can strengthen this approach. Del13C, “

Changed accordingly.

Line 554: should read “ERatmos determined from the time dependence of O<sub>2</sub> and CO<sub>2</sub>”

Changed accordingly.

## References:

Angert, A., Yakir, D., Rodeghiero, M., Preisler, Y., Davidson, E. A., and Weiner, T.: Using O<sub>2</sub> to study the relationships between soil CO<sub>2</sub> efflux and soil respiration, *Biogeosciences*, 12, 2089–2099, <https://doi.org/10.5194/bg-12-2089-2015>, 2015.

Hilman, B., Weiner, T., Haran, T., Masiello, C. A., Gao, X., and Angert, A.: The apparent respiratory quotient of soils and tree stems and the processes that control it, *Journal of Geophysical Research: Biogeosciences*, 127, e2021JG006 676, 2022.

Ishidoya, S., Murayama, S., Takamura, C., Kondo, H., Saigusa, N., Goto, D., Morimoto, S., Aoki, N., Aoki, S., and Nakazawa, T.: O<sub>2</sub>:CO<sub>2</sub> exchange ratios observed in a cool temperate deciduous forest ecosystem of central Japan , *Tellus B: Chemical and Physical Meteorology*, 65, 21 120, <https://doi.org/10.3402/tellusb.v65i0.21120>, 2013.

Kaimal, J. C., & Finnigan, J. J. (1994). *Atmospheric boundary layer flows: their structure and measurement*. Oxford university press.

Patton, E. G., Sullivan, P. P., Shaw, R. H., Finnigan, J. J., & Weil, J. C. (2016). Atmospheric stability influences on coupled boundary layer and canopy turbulence. *Journal of the Atmospheric Sciences*, 73(4), 1621-1647.