

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 Hyytiälä 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.

My main scientific concerns/questions

In this work,  $ER_{atm}$  is based on a single altitude well above the canopy. What if  $ER_{atm}$  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  $ER_{atm}$  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 Hyytiälä data show  $ER_{atm}$  values that are not observed at other locations.

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  $ER_{atmos}$  (and by extension, the difference between  $ER_{atmos}$  and  $ER_{forest}$ ). 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  $ER_{atmos}$ , 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.

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 ER<sub>atmos</sub> 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.

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”.

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?

L8: should read “measured at a single height”

L9: Should read “with the goal of relating the ER<sub>atmos</sub> signal to the ER<sub>forest</sub> signal and understanding the”

L20: should read “rarely represents ER<sub>forest</sub> directly and”

L23: should read “we recommend always measuring”

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

L27: comma after “oceans”

L28: should read “a valuable tracer, enhancing”

L29: remove the comma after “exchange”

L32: comma between “CO<sub>2</sub>” and “represents”

L33: should read “allow us to”

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

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

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

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

L77: should read “and we propose a new”

L78: should read “measurements can be employed”

L79: should read “aforementioned limitations.” And “whether the ER<sub>atmos</sub> signal constrains boundary layer dynamics, and we identify”

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

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

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

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

L109: should read “represent large scale”

L115: should read “associated normally with high pressure systems. We assume  $w_{sub}$  is negligible.”

Eq.3: Shouldn't there should be a subscript on the  $\phi$  that is in time derivative? We are left wondering if this is  $\phi_{bl}$  or  $\phi_{ft}$ ?

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

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?

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

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

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

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

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

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

Line 156: should read “effect of other large scale processes such as advection of  $O_2$ ”

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.

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

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

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

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

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

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

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

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

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)?

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

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”

Line 283: should read “While there is limited”

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.

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.

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

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

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

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

Line 356: should read “dominant and closer to”

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

Line 384: add a comma after “result”

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

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

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

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

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

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

Line 401: add a comma after “result”

Line 402: should read “similar to the”

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.

Line 430: should read “how ERatmos can change during”

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)”

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

Line 452-453: should read “advected 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”

Line 454: should read “of two sources with”

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

Line 459: remove the comma after “processes”

Line 460: should read “signal. During the day,”

Line 465: should read “ERforest may be due to either”

Line 466-467: should read "ratio. If the cause is the former (low BetaCO2), 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.”

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

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

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.

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 CO2 fluxes/abundances during a period of net respiration.

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?

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

Line 504-505: should read "found a non-linear relationship between O2 and other tracers that was difficult to explain. While”

Line 519: should read “constant ERatmos value.”

Line 522: remove the comma after “that”

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

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

Line 554: should read “ERatmos determined from the time dependence of O2 and CO2”