RC1: 'Comment on egusphere-2023-2833', Anonymous Referee #1, 28 Jan 2024

Thank you for the opportunity to review this manuscript. The authors present a thorough comparison between ERatmos and ERforest methodologies in quantifying the exchange of O2 and CO2 above a forest canopy. They demonstrate that ERatmos could be significantly influenced by entrainment, which results in unrealistic values. Consequently, the authors recommend against using ERatmos for constraining O2 and CO2 exchanges at a local scale, advocating instead for measurements at multiple heights to more accurately derive ERforest.

Entrainment significantly influences atmospheric composition within the boundary layer and is a well-researched phenomenon. However, this study stands out as the first, to my knowledge, that specifically addresses the impact of entrainment on O2 and CO2 exchanges. This represents a notable contribution to the field. This study suggests careful selection of O2 and CO2 measurements at single heights is required to correctly represent the biological exchange between O2 and CO2 in forest setting. This consideration is equally important in urban and other backgrounds, particularly for studies focusing on exchange ratios over smaller spatio-temporal scales. Given its importance and novelty, I recommend the acceptance of this study after the following issues are addressed.

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

Major comments:

1. Is it possible for the effects of advection to be counterbalanced by those of entrainment? The observed discrepancies between ERatmos and ERforest might stem from both entrainment and advection processes (Equation 8). In Appendix A1, the authors analyze the influence of the entrainment coefficient (β) on ERatmos signals and discuss instances where ERatmos aligns with ERforest. However, the role of advection remains unclear. Can we rely on measurements taken at a single height when advection's impact is potentially neutralized by entrainment? This interaction might explain why ERatmos and ERforest yield similar results.

We agree with the reviewer that advection of CO_2 and O_2 can influence ERatmos significantly, as we already briefly discuss in section 5.2. To show in more detail how advection can impact the ERatmos signal we did two extra analyses where we added advection to the 2019 CLASS case, in which the advected air either originates from (1) another forest or (2) from a fossil fuel source.

The amount that is advected of a certain scalar (φ) into the CLASS model can be determined with the following equation:

$$adv(\varphi) = \frac{d\varphi}{dx} \cdot u \qquad (1)$$

Where dφ is the net horizontal gradient of either CO_2 or O_2 between the control volume solved by CLASS and a location outside the control volume (either the other forest or fossil fuel location), dx is the distance between the location of the CLASS run and the other location and u is the wind speed. Note that we take a net advection term, representative for the transport driven by the wind vectors u and v in the x and y direction respectively. For these two extra analyses we made in total 6 theoretical cases, where we assumed a distance of 50 km for dx and 6 m s^{-1} for u. We used different values for dφ for the 6 cases, based on the difference between the 2019 CLASS run and another CLASS run (Table 1).

Table 1 The dφ of Equation (1) that we have used to detemine the size of advection of O_2 and CO_2 for two extra analyses (either advecting air from another forest or from a fossil fuel source) and their three theoretical cases. The advection for the other forest cases changes over time and the dO_2 values therefore indicate the start value and the end value. The advection for the fossil fuel source stays from 12:30 with the value indicated in the table.

Advection case	*dO_2 [ppmEq] (t=0, t=11:30)*	*dCO_2 [ppm]*
	(1) Other forest:	
Adv less entr O_2	(0.0,-16.2)	0
Adv lower init O_2	(-15, -15)	0
Adv both O_2	(-15.0, -31.2)	0
	(2) Fossil fuel source:	
Same advection	-19.01	17.1
Lower advection	-10.2	10.7
Higher advection	-30.8	25.6

(1) Advection of air from a forest location

We did 3 simulations with CLASS including advection from another forest source. In these cases, the resulting O_2 mole fractions are lower than in the base case. We have implemented this in three ways: 1a) using a lower initial O_2 jump (adv less entr O_2), 1b) using a lower initial O_2 mole fraction (adv lower init O_2) and 1c) using both a lower jump and a lower initial O_2 mole fraction (adv both O_2).

Figure 1 shows that advection of less O_2 from a location with a forest, decreases the O_2 mole fraction compared to the 2019 base case (figure 1b) and as a result decreases the ERatmos value (figure 1c). We implemented the advection to start at the beginning of the run and to stop after 11:30.

In case advection from another forest would counterbalance the effect of entrainment as the reviewer suggests, the advected air needs to have less O_2 (negative dO_2) compared to the 2019 base case (Table 1). Advection of air with less

O_2 would reduce the steep increase of O_2 in P2 and therefore decrease ERatmos and bring it closer to ERforest.

We find that from the 3 cases, the “adv both O_2“ case results in the lowest ERatmos values (1.68) and relatively comes the closest to ERforest (0.94), but still does not reach the same value. For reaching similar values of ERatmos compared to ERforest, the advection has to be of almost similar size as the fossil fuel cases (see below). We can therefore say that advection can only bring ERatmos closer to ERforest under very specific conditions. It is therefore highly unlikely that advection can bring ERatmos to a value that is representative for the ERforest signal.

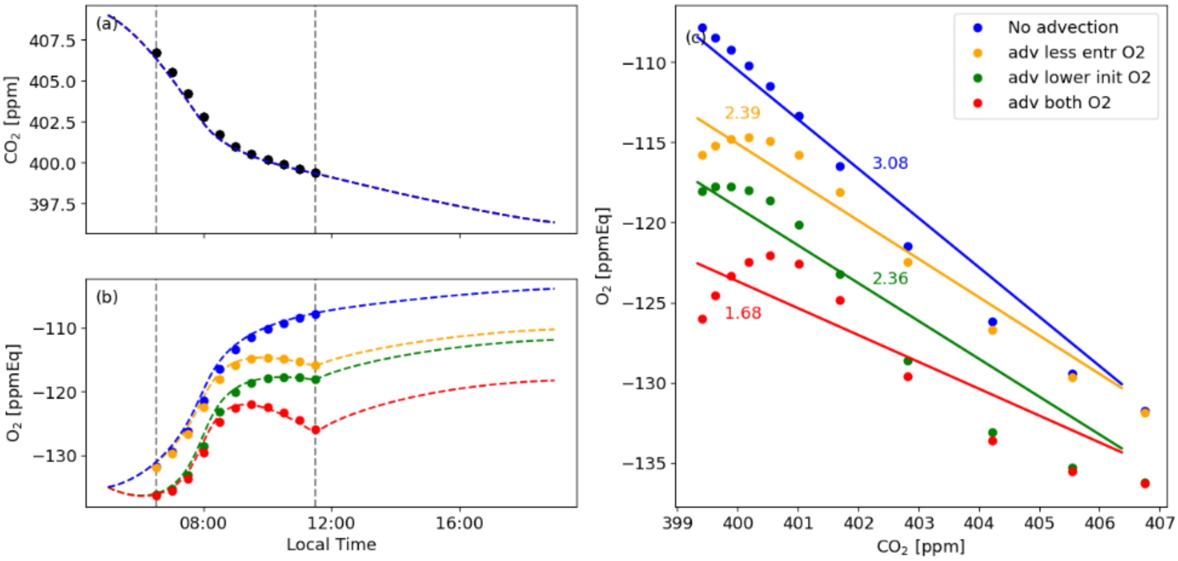

Figure 1 Results from advection from another forest for three theoretical cases (Table 1) and its impact on the CO_2 mole fraction (a), O_2 mole fraction (b) and the ERatmos (c). The advection starts when the run starts and ends at 11:30 Local Time. The numbers in figure (c) are the slopes of the linear regression lines for the period between 6:30 LT and 11:30 LT (P2 in manuscript).

It is important to note that the advected air does not only have a forest surface exchange signature, but it also includes the effects of entrainment that occurred on the location where the air is advected from, and entrainment can therefore never be fully excluded.

(2) Advection of air from a fossil fuel source

For the case with advection from a fossil fuel source, we have advected air from a fossil fuel source that has an exchange ratio of -1.38, which is similar to the global average fossil fuel mix. We have simulated 3 cases, with either low, middle or high source strengths. We now advect both O_2 (dO_2) and CO_2 (dCO_2) (Table 1).

Figure 2 shows that advection can create an ERatmos signal that is not directly representative of the ER signal of the fossil fuel source. It is rather a mixture of the advected air ($O2_{advected}$), surface forest exchange ($O2_{forest}$) and atmospheric mixing ($O2_{entrainment}$)and it therefore requires cautious interpretation, as it cannot be straightforwardly associated with the advected air due to non-linearity. This can be seen in Figure 2c, in that the linear regression lines do not reach the value of -1.38. The ERatmos value even becomes lower than 1 with our “low advection case”.

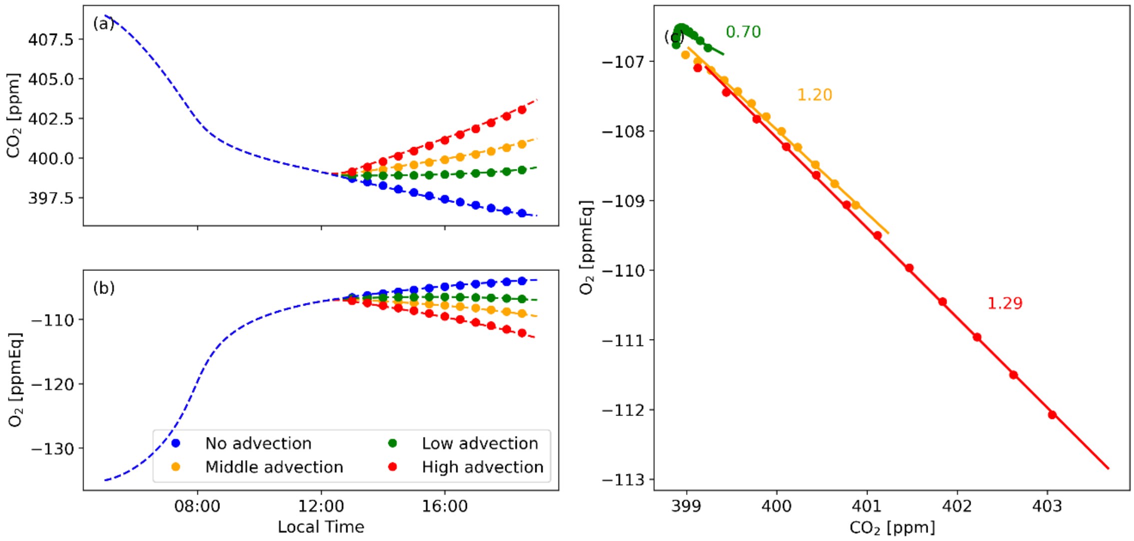

Figure 2 Results from advection from a fossil fuel source for three theoretical cases (Table 1) and its impact on the CO_2 mole fraction (a), O_2 mole fraction (b) and the ERatmos (c). The advection starts at 12:30 LT. The numbers in figure (c) are the slopes of the linear regression lines for the 3 cases.

We have decided not to include these sensitivity analyses in the manuscripts, since the manuscript was already dense in content.

2. Does entrainment exert a more pronounced impact during typical days? This modelling study is generally based on the mixed layer theory. In studies by Ishidoya et al. (2013, 2015), their analysis did not specifically distinguish between measurements on ‘typical days’ and ‘non-typical days’, and derive similar ERatmos and ERforest values.

In the studies by Ishidoya et al. (2013, 2015), these authors do not analyze individual days but average several days into a composite day to analyze ERatmos and ERforest. By creating a composite (or representative) day, different scenarios of entrainment are included and as a result the extreme values of ERatmos could be averaged out. For example, the initial jump changes per day which results in different ERatmos values per day (Figure B1). By including different scenarios with different initial jumps, the high ERatmos values will be averaged out. We discuss this in section 5.4.

In our previous study (Faassen et al. 2023) we also made a composite day that was the average of seven days. Within these seven days typical and non-typical days were included. For example, two days that had a high amount of clouds during the day are part of our composite day. Even by including non-typical days, we showed that the ERatmos (-2.28) could still significantly deviate from ERforest.

After reading this work, I am fully convinced the impact of entrainment should be considered on 'typical day'. However, it remains uncertain how this applies to specific instances, such as heavily polluted urban days or during extraordinary events like COVID-19 lockdowns, where mixed layer theory may not always hold. It would be beneficial for readers to understand the frequency and significance of entrainment during these atypical periods.

Entrainment also occurs on non-typical days. Entrainment is mainly the result of the combination of the growth of the boundary layer and the difference (jump) of a scalar between the boundary layer and the free troposphere (Equation 2). This holds for both typical and non-typical days and for scenarios where mixed-layer theory may not fully apply. Entrainment occurs for each of these options, it is only easier to be study under weak synoptic and mesoscale conditions (typical days). However, when a frontal system passes, entrainment occurs, but its contribution is less.

The strength and the significance of entrainment depends on the day. A clear example of the importance of entrainment of CO_2 during a polluted day is given by the study of Casso-Torralba et al., (2008). They analyzed diurnal cycles of CO_2 measured above a grassland at the Cabauw station in the Netherlands. The study shows that during a (typical) day without advection, entrainment is dominating the CO_2 signal in the early morning. However, during a polluted day when advection of air with higher CO_2 levels is present (non-typical day), the advected air masses with high CO_2-concentrations balance the combined effect of entrained air or low CO_2 and the uptake of CO_2 by the surface. As a result, the CO_2 mole fractions hardly change during the day. During the polluted day, entrainment is still important, but it is not the dominant process during the morning anymore.

In Figure 2a above in this reply, we show a first analysis of how ERatmos could respond during 'non-typical' days with polluted advected air, and show that ERatmos should also be handled with care on polluted days.

When assessing single-height measurements on non-typical days, can we still depend on ERatmos for accurate representation? While modeling these atypical days using the CLASS model might be challenging, I suggest the authors discuss these considerations, possibly in Section 5.2, to provide a more comprehensive perspective.

For a new study focusing on measurements in a forest in the Netherlands, we are currently studying an example of a 'non-typical' day that includes subsidence. During this day we measured O_2 and CO_2 from flask samples (Figure 3a), as well as the jumps based on aircraft sampling (Figure 3b). During this non-typical day, the ERatmos comes closer to ERforest and this gives the illusion that ERatmos starts to represent the surface processes.

However, when analysing the different components of Equation 8, we find that the large jumps of O_2 and CO_2 create large entrainment fluxes (Figure 4). Together with low surface fluxes due to a heat wave during the campaign day, the beta values for both O_2 and CO_2 are high. Therefore, Eratmos is not representing ERforest, but ERatmos rather represents the ratio between the entrainment fluxes. ERatmos can therefore also not give a correct representation of ERforest during non-typical days.

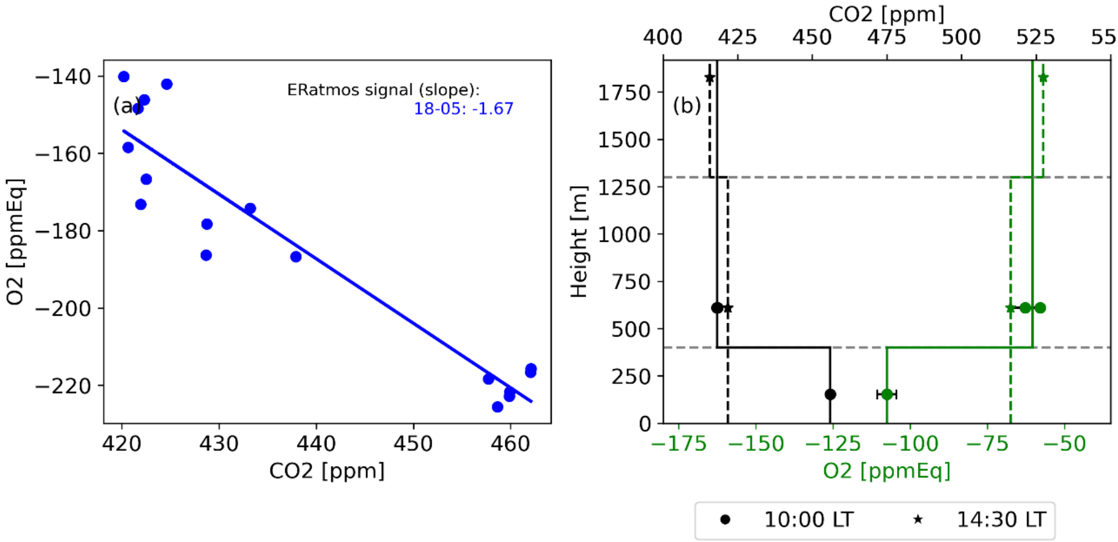

Figure 3 Measurements during our campaign in a forest the Netherlands on 18 May 2022 for ERatmos (a) and vertical profile measurements from aircraft samples, including their jumps based on the measured boundary layer height (b). The jumps in panel b are -38.0 ppm and -6.1 ppm for CO_2 for 10:00 LT and 14:30 LT respectively and the jumps for O_2 are 47.2 ppmEq and 10.6 ppmEq for 10:00 LT and 14:30 LT. The figure is based on a manuscript that is currently in preparation.

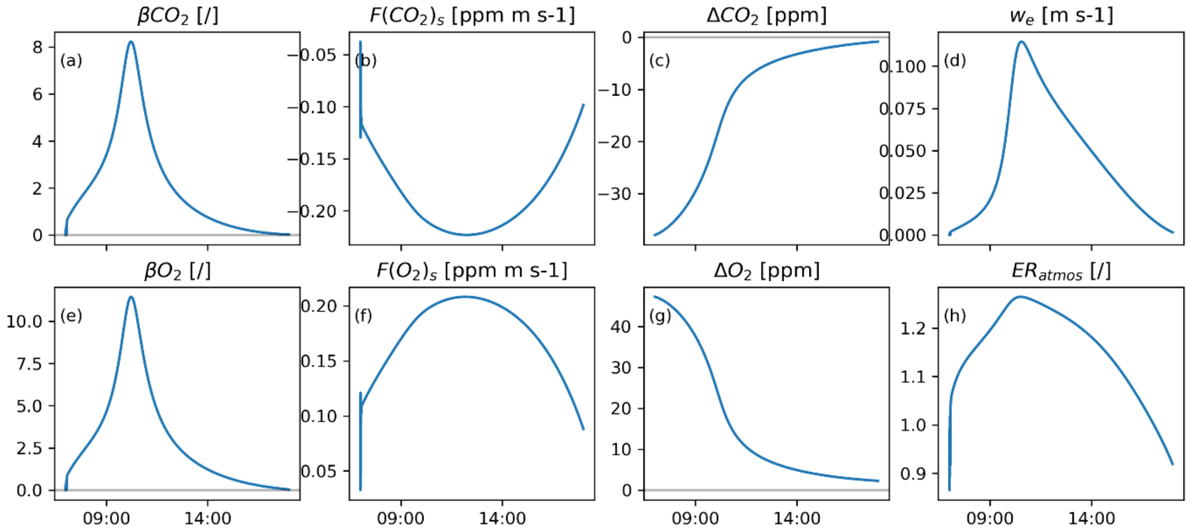

Figure 4 The different components of equation 8 (similar to Figure 5 in the manuscript) for the campaign day in the Netherlands that was modelled with CLASS. The figure is based on a manuscript that is currently in preparation.

To clarify this point we have added a few sentences on the effect for non-typical days to section 5.2.

References

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