Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2022-318-RC1, 2022

Comment on acp-2022-318

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Referee comment on "Residence times of air in a mature forest: observational evidence from a free-air CO2 enrichment experiment" by Edward J. Bannister et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-318-RC1, 2022

The authors present an observational study of air residence times generated from CO2 data during a FACE experiment and use these to investigate impacts of environmental conditions on air residence times. Inspired by a previous paper (Gerken et al. 2017, GCF17) that sought to describe the probability density function (PDF) of air parcel residence times, the authors note a generally similar behavior to GCF17 of their data and use the data to fit an inverse gamma distribution to their calculated air residence times. In general, I find the paper to be interesting especially given that air residence time is a fundamentally unsolved problem, which may be of some importance for modeling air chemistry of BVOCs. At the same time, I have several comments that should be addressed before publication.

As a side note, I am the first author of GCF17 and it is nice to see that this paper is being used as a basis for discussion.

General comments:

- Eulerian vs. Lagrangian approach: The authors' approach is in essence a Eulerian approach to calculating a mean air residence time, while Gerken et al. 2017 (GCF17) applies a Lagragian model within an LES to generate a PDF of air parcel residence times, which is used to then estimate the parameters of a first passage process (Schrödinger 1915). It appears that the main difference between the PDF of CO2 residence times generated in this work and in GCF17 is that GCF17 predicts a heaver tail of long residence times, which is not found in the data for this manuscript and the authors attribute this to limitations of the eddy diffusivity approach and the homogeneity assumption embedded in GCF17 (I hope that this is a fair reading). First of all, I don't want to dismiss these explanations, given that GFC17 was developed using LES data rather than direct observations of air parcel residence times, which cannot be observed in the field. At the same time, I am wondering about whether there is a mismatch between the Eulerien approach in this manuscript and the Langrangian approach in GFC17. It is my understanding that the approach employed in this manuscript and represented by eq. 5 of this manuscript provides a singled mean air residence time for the entire air volume within the control volume (sorry for the wordy description of this). I believe that this is not directly comparable to the air parcel residence time as defined in GCF17, which arises from tracking of individual Lagrangian particles that are thought to represent a hypothetical air parcel. So in my view the air residence time (tau) calculated in this manuscript might be more akin to the mean of the air parcel residence time distribution from GCF17. In this view each calculated \tau would be another sample of the mean air parcel residence time rather for a given instance of turbulent conditions rather than the residence time of an air parcel that together make up the PDF that are used to fit equation 1 in GCF17. In other words, the tau values in this work might represent a multitude of air parcels over a given averaging length (5 minutes in this case). This interpretation would account for the fact that tau values found in this work lack the heavy tail found in GCF17. I would appreciate additional discussion of this.

At this point I would like to reiterate that, it might very well be true that GCF17 overestimates the heavy tail of the tau PDF, but at least for the LES applied in GCF17 we did not find this to be true (see Figure 5b in GCF17).

This is also not to say that the analysis done in this work is not useful on the path to develop air residence time parameterizations and I certainly find the discovered gamma distribution fit (eq. 7 and Fig 9) a valuable contribution.

- Advection: If I understand correctly, the authors assume that advection and therefore losses to the side of the control volume are negligible. Given the setup of the FACE (and admitting that I am not very familiar with it), I am wondering to what extent this would be true given the open sides and the fact that the FACE experiment enriches CO_2 inside the control volume, thus producing an artificial horizontal CO_2 gradient. I would appreciate additional discussion on this and potentially some evidence that advection or horizontal loss of CO_2 is indeed neglegible.
- Role of LAI: Generally speaking, it might make sense in the introduction and discussion to reference LAI and leaf area density profiles more frequently since the interaction between leaf drag and turbulence penetration into the canopy are likely one of the main reasons for widely varying estimated residence times.

Specific comments:

Title: In the light of my general comment 1, it might be better to remove the 'parcel' from the title.

L 67: "Gerken et al. (2017) (hereafter GCF17) offer ..." > I suggest to also reference Katul et al. 2005 in this context, since their model was used as a starting point and includes similar assumptions. One should also note that this formulation was only proposed for neutral conditions.

L 87: "In an LES investigation of flow over forested hills, residence times of Lagrangian air parcels emitted in the lower part of the canopy were shorter than those moving over flat terrain (Chen et al., 2019)." > I suggest to expand on this since the impacts of terrain are very important for real world applications and there is ample evidence (albeit andecdotal) for preferential venting due to terrain

L90: "Researchers have also used Eulerian frameworks to investigate residence times in forests." > It might be good in this context to discuss some of the limitations on Eulerian vs. Lagrangian methods regarding their implications for air chemistry. This also goes along with my general comment on the comparability of Lagrangian and Eulerian approaches. Given that aur parcel residence time is a fundamentally Lagrangian process, the Eulerian description has some limitations such as that it is in my mind a mean air residence time, which can underestimate the tail ends, which might be important for air chemistry.

L200: "because broadleaf forests uptake little carbon below this threshold" > this sounds off. 'because carbon uptake is negligible' (?)

Section 2.3: What is the time scale of the \tau calculation? (P.S. I see that this is answered in the data processing section. I suggest to move this forward). Additionally and given the importance of release height pointed out in Gerken et al. 2017, it would also be good to hear more about at what heights CO2 is being releases. I

L 237-240" "Therefore, rather than trying to assign a numerical value to ð□□¹ð□□□ð□□¢ð□□i(â□□ð□□□ð□□□) , we identify meteorological conditions under which ð□□¹ð□□□ð□□¢ð□□i(ð□□ið□□□ð□□□) â□« ð□□¹ð□□□ð□□¢ð□□i(â□□ð□□□ð□□□) , and therefore ð□□□ = ð□□□ð□□¶ð□□□2 /ð□□¹ð□□□ð□□□ ≈ ð□□□ð□□¶ð□□□2 /ð□□¹ð□□□ð□□¢ð□□i(ð□□ið□□□ð□□□) . Figure 3 presents probability density functions of ð□□□ during the lowest 50% of wind speeds of the leaf-on period (solid black), during the highest 25% of wind speeds of the leaf-on period (dashed),... " > This seems like an abrupt transition. It might be good to give provide a sentence or two on who these are related to advection. On a broader note, I appreciate the advection problem in the sense that this is something that has been challenging in high vegetation with CO2 accumulation within the canopy airspace.

L 241" zrel > I am wondering whether it would make sense to adjust z_rel, given that it is not clear to me what the real release height of the CO2 is would be to minimize the difference between the observational results and the theoretical result by adjusting z_rel.

Figure 3a: It seems to me that all the curves in Figure 3 should have the same integral. Could you confirm this and check whether all curves are properly normalized, since it seems (by eyeballing) that the GCF17 might not have the same area under the curve.

Figure 6 should have a colorbar and possibly a trendline to better gauge the underlying density distribution

The stability classes in Section 3.3.2 should probably be moved to methods section.

L 384: "The distributions of ð□□□ remain positively skewed for each stability class (e.g., the right whiskers are longer than the left in Figure 7a)." > It might be a good idea, here and in general to report the skewness.

Section 3.4.: I am not sure how informative this section and the associated figure is. I think that it is important to discuss the edge effect and impacts of heterogeneity, but I am not sure whether this section currently does this in the optimal way. Especially since I think that Figure 8 is pretty hard to read. What dominates the differences in the different wind sectors. Is it heterogeneity or some other influence such as time of day coupled with stability?

L445: "However, although GCF17's model generates modal values similar to those we observed, it appears to overpredict the likelihood of long residence times in the upper canopy." > this might be true, but also there might be the issue of comparing an essentially Eulerian and Lagrangian method (see general comments).

L463: "These eddies create significant turbulent transport, meaning that the eddy-diffusivity model underestimates turbulent forest-atmosphere exchange in the upper canopy and therefore overestimates residence times." > Turbulent diffusivity approaches do have issues within forest canopies. One thing to note about GCF17 is the fact eddy diffusivities are estimated from the LES and adjusted for the release height by taken the mean modeled diffusivity (either arithmetic or geometric mean) between release height and canopy top. It is not clear to me that this would lead to an effective underestimation of turbulent transport. Some additional thoughts by the authors would be appreciated.

Section 3.7: This section seems a bit tacked on in the sense that it is not clear how it relates to the previous section of the model, especially given that the main conclusion from the previous section seemed to point to an overestimation of residence times in GCF17. While the information presented here is an interesting case study, it might make sense to either tie this directly to the analysis before or to remove/ move to an appendix. On a side not and also with respect to long residence times. The original GFC17 study was motivated by air exchange within Amazon rainforest canopies with large LAI and limited penetration of turbulent eddies into the lower half of the canopy. Evidence for the limited coupling of canopy airspace to the above canopy air can be for example found in Freire et al. (2017).

References:

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