

Review of revised paper acp-2026-161: Linking In-Canopy Chemistry to Above-Canopy O₃, BVOCs, and NO_x Gas Fluxes in the Amazon Rainforest by Brown et al.

We thank both reviewers for the attention they have given to our revised submission. We appreciate the feedback and have responded to the additional comments in red.

Reviewer 1:

Thanks to the authors for their in-depth response to my questions and comments, and the addition of the expanded Section 2, which has greatly improved the readability of the manuscript. I'm rating the manuscript as minor revisions at this point, though the latter are things that should be addressed (for example, one of their formulae has an error I missed the first time around – hopefully a typo). I also have a few further comments on the revised / added sections of the manuscript, which should be addressed prior to publication. My page and line numbers are with respect to the track-changes version of the revised manuscript:

We are pleased to have been able to answer your comments and agree they helped improve the manuscript.

Page 6, line 168. Please mention towards the end of this paragraph that the lateral tendencies were based on finding a best fit of a parameter to observations.

We have now added a sentence where suggested:

“The advection parameter is adjusted to best reproduce observations of trace gas concentrations.”

Page 7, line 181. Are there any observations to indicate an annual variation in LAI over time, or is it pretty constant year to year? i.e. could a variation in LAI explain some of the 2015/2013 differences in the model results?

Thanks for bringing this up. LAI is considered to be fairly consistent and other literature uses the same fixed value. We refer the reader to a longer-term study where LAI differences were evaluated.

“Variability in LAI at this site is not considered statistically significant (Botía et al., 2021).”

Page 10, line 270, also page 11, line 309: Add a sentence or two at the end of the deposition description to explain how the numerical values of deposition and emissions were included into the model. I think from the revised paper that the they were included into the vertical diffusion solver as individual layer rate of change terms, please confirm in the text.

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We have added a sentence to the deposition description but the combination of these terms into the vertical diffusion solver is treated in the transport and fluxes section, also addressed in the following comment.

“The total deposition velocity (v_d) at each layer is transformed into a sink tendency ($S_{dep} = -\frac{v_d}{z} c$ (s^{-1})) using the layer trace gas concentration c and combined with the emissions tendency before being passed to the vertical transport solver (described below).”

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The authors should also clarify exactly what they mean by “mass injection”, since it could be interpreted in different ways in terms of the model numerics. For example, mass injection could be interpreted as simply adding or removing mass to/from the model layer, as a separate operator. That approach has the disadvantage of introducing discontinuities in the concentration profile in the vertical, which can then lead to other numerical issues. However, adding the mass as a ->rate of change<- term to the concentration rates of change will provide a much more stable solution without discontinuities. I think (?) that this is what the authors have done – could they please confirm it in the text? It would be best if this could be stated in an equation in the main manuscript or the SI where the emissions and deposition terms are included and their units stated.

We have changed Eq 13 so that it explicitly provides the equation passed to the vertical transport solver. This equation includes units and combined with the text, we feel it is unambiguous. We have also removed the term ‘mass injection’.

“Transport and fluxes

The mass flux for gas-phase species is described by Eq. 13:

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial z} \left(K \frac{\partial c}{\partial z} \right) + S_c \text{ (Eq. 13)}$$

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where c is the mixing ratio of a chemical species, S_c represents the sum of emissions, deposition, and advection tendencies at each layer (s^{-1}), and K is the eddy diffusivity coefficient ($m^2 s^{-1}$).

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Emissions, deposition and advection contributions are summed for each layer to calculate the rate of change (S_c) at each layer and are passed to the vertical transport solver. Numerically, these tendencies are incorporated through operator splitting: the term $S_c \Delta t$ is added to the right-hand side before the implicit vertical transport solve. The chemical solver is applied subsequently. This operator splitting is described and evaluated in Wei et al. (2021).

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In this version, we move soil NO emissions from being a lower boundary condition to an emission contributing to S_c in the lowest atmosphere layer. Surface emissions and

deposition are therefore treated explicitly within the chemical source terms prior to the transport step.”

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A further line has been added to the supplementary, but this level of detail is likely not accessible to many readers. As this is not a model development paper, there is a limit to how much can reasonable be discussed beyond what is already provided in the original model documentation:

“Vertical transport solver

Equation S7 represents the continuous species continuity equation:

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial z} \left(K \frac{\partial c}{\partial z} \right) + S_c + C \text{ [Eq. S7]}$$

where c is the mixing ratio of a chemical species, S_c represents the sum of emissions, deposition, and advection tendencies at each layer (s^{-1}), C is net chemical production (s^{-1}), and K is the eddy diffusivity coefficient ($m^2 s^{-1}$).

In the numerical implementation, the terms are operator split. Emissions, deposition, and advection tendencies are incorporated into the transport step (Eq. 13 in the main text), while gas-phase chemistry is solved subsequently in a separate chemistry step.

Source/sink tendencies (S_c) are represented in FORCAsT at each layer within the species continuity equation. Numerically, these tendencies are incorporated through operator splitting: S_c is multiplied by the model timestep Δt and added to the concentration field prior to solution of the implicit vertical diffusion equation, yielding $(I - \Delta t L)c^{n+1} = c^n + \Delta t S_c$, for timestep n where L is the vertical transport operator. This means FORCAsT treats S_c as a concentration tendency, but integrates it separately from transport.”

Page 10, line 285: “We select a value of k that best improves representation of observed O3 concentrations in 2015”: was the same k value used in both years, then? I’m wondering if that might explain some of the differences in model performance between the two years. This along with the 2015-specific sigma-w value should be mentioned in the discussion of possible reasons for differences between the two years.

We do not consider advection in 2013 as there was very little fire emission so, regardless of wind strength and direction, we would not expect considerable advection. This is written in the text and included in Table 2 but we add further clarification in the description of the simulations (Sect. 2.3):

“Biomass burning advection is not considered in 2013 as there was limited fire activity during November.”

Page 11, line 297 “extrapolated”. It might be better to state this as “extrapolated and bounded by a maximum concentration of 50 ppbv above 2000m”.

We agree this is better phrasing and have changed the text.

Page 13, line 360: to what extent might the use of 2013 sigma-w values potentially influence the differences in the model results between 2013 and 2015? For example, Figure S13 was calculated from equations S3 to S5 (which do not depend on sigma-w) above the canopy – and in that above canopy region, the K values look rather different between the two years, with 2013 being more stable during the daytime compared to 2015. Equation 18 in the main manuscript is using 2015’s sigma-w for 2013, in that case. Is there any observational evidence (perhaps from other years) that might indicate that the sigma-w (55) values don’t vary much between the years? The authors should add a sentence or two acknowledging that the use of 2015 sigma-w values might have a substantial impact on the 2013 results. For example, this could be mentioned as a possible reason for model differences noted at line 535-538, page 14, and could be mentioned as a source of uncertainty prior to the new Table in section 2.3, and on somewhere between lines 634 and 665 of the revised manuscript.

Thanks, we agree this is an uncertainty and have included this throughout the text and in the discussion. We have added additional text at the locations you suggest:

Prior to the new Table in sect. 2.3:

“ATTO observations of turbulence regimes show variability across seasons and years (Botía et al., 2020; Cava et al., 2022; Mortarini et al., 2022), but there are no direct comparisons of how σ_w changes from one year to the next.”

New Line 430:

“In observations, Pfannerstill et al. (2018) also found increased turbulent features during 2015, however we also note that missing observations of 2013 σ_w may affect our results.”

**Page 13, line 372: The author’s equation (19) contains an error, hopefully just a typo in the manuscript rather than something that appears in the analysis: the denominator derivative in the storage term should be “dt” not “dx”. I went back to the Rummel et al (2007) reference to check this; their equation (1) uses “dt”. Could the authors please confirm that they did in fact use d/dt in their calculation of the storage term of equation 19, in their analysis? The equation makes much more sense dimensionally in that case, with

the final term being the rate of change of the vertical integration of the species concentration, rather than the derivative with respect to species concentration, as it currently appears in both the original and revised manuscript. This caused me some confusion on the first round of review.

Thank you for spotting this mistake! It was a typo and this explains confusion from reviewers.

Page 14, line 537: “suppressed” misspelled. Also, I agree with your reasoning here – Raupach and others noted that canopy turbulence can be intermittent under stable conditions – which make it challenging to represent using K theory without time-dependent sigma-w values.

This has been corrected.

Page 34, line 1135: Note that in the case of regional models, simplified multi-layer models (Makar et al., 2017, Nature Comms) or parameterizations to try to capture the effects while preserving the original model layer structure (Wang et al., 2025, ACP) are being adopted. That is, the community has started to recognize that this is an issue for 3D modelling – the Wang et al (2025) approach avoids the computational issues while incorporating some of the layering impacts.

This is a great point. We have added the following sentence:

“Already, simple canopy layer structures (Makar et al., 2017) and canopy layer parameterisations (Wang et al., 2025) are being implemented into large-scale models, highlighting a growing interest in representing in-canopy processes in computationally efficient ways.”

Reviewer 2:

I have checked the response to the raised comments as well as the revised paper. Many revisions have been made also tackling the comments raised by the other reviewer. The model system itself has been described in much more detail (also partly in response to the other reviewer’s comments) and most of the issues that I raised seem to properly tackled. Therefore, I deem the paper to be acceptable for publication in ACP upon addressing mostly some last minor issues.

We are very grateful for your feedback that has helped to improve our manuscript and are pleased to have addressed your comments.

Based on a previously shared comment you introduced the following modification. Line 98: “This acts as a first step towards identifying the important features of trace gas exchange required for improved representation of tropical forest in-canopy chemistry in global models” I don’t think it is mainly/only about improved representation of tropical forest in-canopy chemistry; it is about the combined role of all in-canopy interactions including biogenic emissions, chemistry, turbulence and deposition ultimately determining the effective atmosphere-biosphere exchange fluxes in large-scale (regional and global) models.

We have broadened the statement in the text:

“This acts as a first step towards identifying the important features of trace gas exchange required for improved representation of tropical forest in-canopy processes and interactions in global models.”

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Also triggered by the statement in the abstract (line 24/35): “Whilst canopy models have been applied to temperate forests, there are few studies in tropical forests”, I might refer you to another tropical forest study (lead by one of your co-authors, Atmos. Chem. Phys., 18, 3403–3418, 2018 <https://doi.org/10.5194/acp-18-3403-2018>, for which we also used a multi-layer canopy model) showing that there is more than chemistry involved in these BVOC/O₃ interactions of the tropical forest; wet and dry non-stomatal removal processes. I am also bringing this up in response to your modified model deposition description and the discussion about non-stomatal removal in NO_x exchange (lines 1076/1077). In this discussion you could shortly elaborate on not considering such differentiation between wet and dry non-stomatal removal processes (or did I miss something?). With a tropical rainforest canopy, the role of canopy wetness by rain/dewfall interception might have relevant implications for BVOC (especially the more soluble products), O₃ (and NO_x?) exchange.

Thanks for highlighting the wet deposition effect identified in this paper. We have chosen to add this to the discussion, as you suggest.

“Previous studies have identified non-stomatal deposition to wet leaf surfaces to be a potentially relevant removal process in the tropics, an aspect not considered in this study (Yáñez-Serrano et al., 2018). The effect of canopy wetness on deposition magnitude and escape efficiencies may therefore be important for soluble BVOCs and NO_x, and greater understanding of this process would enable improved parameterisations.”

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You mention in the paper that there were no O₃ dry deposition velocity measurements and which made me aware that for the diagnosed periods (since later on they collected/are still measuring (?) O₃ fluxes at ATTO) that there are no O₃ flux measurements. It would be

useful to explicitly mention this in the methods sections, also given a strong focus in your model analysis on the role of in-canopy chemistry on O₃ canopy-top fluxes but which then mostly relies on using only O₃ concentration measurements.

Thanks, we explicitly mention this now:

“There are no O₃ flux measurements available for the simulation period so subsequent evaluations only consider O₃ concentration measurements.”

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Line 166/167: In the modified section on constraining these 1-D canopy model experiments you now mention on the specifics of the nudging procedure that it secures that “the below-canopy environment is more likely to be well-represented and analysis can focus on below-canopy processes”. This is not properly reflecting some of the motivations to nudge canopy exchange model experiments. It is also very useful to compare model simulated and observed atmosphere-biosphere fluxes, e.g., seeing changes in bi-directional exchange of NO_x/BVOCs due to changing advection conditions.

Thanks for adding this, we have changed the text:

“This has the advantage of holding the above-canopy environment as close to the true values as possible, which enables evaluation of atmosphere-biosphere fluxes in response to above canopy changes, such as advection. Additionally, ~~the below-canopy environment is more likely to be well-represented and analysis can focus on below-canopy processes.~~”

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