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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Isoprene chemistry in pristine and polluted Amazon environments: Eulerian and Lagrangian model frameworks and the strong bearing they have on our understanding of surface ozone and predictions of rainforest exposure to this priority pollutant" *by* J. G. Levine et al.

Anonymous Referee #2

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General comments

This paper presents the results of a modelling study simulating atmospheric composition and in particular the concentration of ozone, above the Amazon during the SAMBBA field campaign. The study, employing two very different atmospheric chem-

istry models (one an Eulerian 3-D chemistry and transport model; the other a Lagrangian box model applied in trajectory mode), explores how differences in model input data, chemistry mechanisms and dynamics affect concentrations of key species (ozone and its precursors).

The study is generally well-designed and thorough, providing useful insights into both the potential drivers of ozone production and loss over this region, and the implications of model choice. However, I do feel that the results and conclusions presented here would be far more robust were the authors to provide a quantitative assessment of the performance of the models in terms of capturing the magnitude and variability of the observed concentrations. Without this, it is hard to determine the true extent to which introducing "Eulerian-style" mixing into a Lagrangian model improves model skill.

While the background to this study is well covered, I am concerned at the level of self-citation; many other research groups have made substantial contributions to modelling isoprene chemistry above the Amazon but have received relatively scant attention here. Many of the references to others' work are rather dated (e.g. why cite Guenther et al. 2006 rather than Guenther et al., 2012?) but see below for specific instances. The section (2.3) detailing the changes in chemistry mechanism from CheT to CheT2 appears to have been copied almost verbatim from an earlier paper from co-authors and requires changing prior to publication.

The key findings of the research are at times rather lost in the painstaking and overly verbose presentation style. The manuscript would benefit from substantial reduction before publication. The figures are in general far too small (requiring viewing at 400% in order to distinguish features) and have poor choice of colour; see below for suggested improvements. I would recommend the manuscript be published once my comments (above and following) have been suitably addressed.

Specific comments

The approach taken in the study was clearly extremely thorough and painstaking in

terms of covering all possible permutations of model and input data. However, I feel that it is unnecessary for all of these to be presented here (see specific points below). In addition to finding the manuscript hard to follow in place due to the repetitious and verbose style, I did find the use of the word "recall" was rather insulting and would recommend its removal.

1 Introduction

P24254, L5 – Why not cite Guenther et al., 2012?

P24254, L7 – Anthropogenic emissions have changed markedly in many world regions since 1995; why not refer to e.g. Lamarque et al., 2010?

P24254, L7-13 – Please provide references for lifetimes and the importance of OH chemistry.

P24254, L15 – Why not WHO 2005 guidelines? Or even the 2014 update on health impacts?

P24254, L19 – I'm not sure that Avnery et al is the most appropriate reference here: (1) Avnery et al. was a purely modeling study, experimental evidence of the effect would be far better; (2) the study here focuses on ozone and its impact on a forest, not crops. How about output from CLRTAP (e.g. ICP Vegetation or ICP Forests groups)?

P24255, L2-4 – This has also been shown for other forest ecosystems, see e.g. Rohrer et al., 2014

P24256, L14-19 – However, Squire et al. did not have a sonde profile for the Amazon basin, and the closest one they analysed showed a substantial MBE . . .

P24256, L21 – Please add "thought to be" before "responsible", and use a more recent reference, e.g. Arneth et al., 2011.

P24257, L5 (and again P24268, L14 and P24269, L5) – The cited report (Jenkin, 2012) is not available for the reader to refer to.

P24257, L19 – Please remove "Recall"

p24259, L3 – I do not feel that "caricature" is an appropriate word to use in a scientific paper; what's wrong with characterise or describe?

2 Method

P24259, L25 – Please re-iterate the SAMBBA reference at this point.

P24260, L11-15 – I feel the authors do protest too much! There are no OH measurements, full-stop. Comparison of modelled O3 concentrations is useful, full-stop. While it is true that reproducing OH but not O3 would highlight a problem within the model, it is irrelevant here as the authors are unable to compare both; furthermore the converse is not true, matching O3 well does not indicate OH is well captured as numerous studies have shown.

P24260, L23-24 – While the specific point in space may be "arbitrary" the authors clearly had reason(s) for selecting it; please explain the reasoning here. It would also be useful to state the lat-lon of Manaus for reference here and in the caption of Fig. 1.

P24262, L11 – "In-flight"

P24264, L24 – Please elucidate this artificial "flight track"; is this a flight track consisting of a single point? If not, it should be presented on Fig. 1.

P24266, L24 – Please present these equations on separate lines.

P24267, L1 – Why use the limits suggested by Pugh alone? Why not combine the two to span the full range, i.e. 0.3-1.5 m2/s?

P24267, L8-9 – Some recent observations suggest that even short-lived species can have non-zero concentrations in the free troposphere.

P24267, L10-25 – It is not entirely clear how the authors distinguish a simulation as a distinct model setup (either here or in Table 2). To claim 8 implies that different mixing

does not constitute a different setup. However, in that case setup 5 is the same as setup 1 (which could be considered as Mix0); likewise 6 and 2, etc.

P24268, L6 – P24269, L11 – This entire section is almost identical to the text in the description of the CheT and CheT2 presented in Squire et al., 2014. This is not acceptable; please re-phrase.

P24270, L17 – Messina et al., 2015 is not included in the listed references. Is this paper still in preparation?

P24270, L17-19 – Please comment on the possible impact of using ECMWF for nudging and back-trajectory calculations and NCEP reanalysis for biogenic emissions estimates.

P24270, L19-22 – Please justify the use of daily average biogenic emissions for a study focusing on the temporal and spatial heterogeneity of atmospheric composition above the Amazon.

P24270, L23-24 – Please give further details of the algorithm used to apply a diurnal cycle to these daily average biogenic emissions, and justify its use by demonstrating its appropriateness.

P24270, L28 – P24271, L1 – Why show both January and July given that only biogenic emissions are seasonal? Why show January or July at all, given that the study was performed for September? It seems to me that Fig. 2 is entirely unnecessary. If the authors decide to retain Fig. 2, some improvement in presentation is required (see comments below).

P24270, L29 – Please remove "Recall".

P24271, L1-5 – While Fig. 3 is more interesting (and relevant as it shows September) than Fig. 2, not all of the panels seem necessary, particularly given that many of them do not show substantial differences between UKCA and "high" resolution emissions. Perhaps the authors could present a selection of the species rather than an exhaustive

list (see comments below).

3 Results

P24271, L8 – Please could the authors explain why they have selected this particular flight. It seems that for the most part there is little difference in the results between flights, e.g. changing chemistry mechanism shows little effect in model-measurement fit for either UKCA or CiTTyCAT, apart from the paired simulations in which the resolution of the emissions is altered from "UKCA res" to "High res". In this case, the effect on the selected flight (B735) is anomalous. It seems that it would have been preferable to have focused on one of the other four, so I would like to understand the reasoning behind the selection of this flight.

P24271, L10-11 – I feel that this would be a much stronger study if the authors were to quantify the performance of each model. Making a qualitative assessment of the effect of various sensitivity tests and using that to justify the use of a "hybrid" model in future modelling studies is less compelling. The temporal variability in the observations strongly suggests that an estimate of goodness-of-fit would be extremely useful, and seemingly relatively easy to do – I wonder why the authors chose not to.

P24271, L18-22 – The line colours should all be in the legend rather than in the text.

P24271, L19 – Please remind the reader of the "initialisation" process.

P24272, L1 – Please do not mix decimal hours and hh:mm; the latter is preferable. LT is also more meaningful than UT in a situation involving biogenic emissions and photochemistry, and I would suggest the authors employ this throughout.

P24272, L7 – Please explain what is meant by "structure" in this context: variability? heterogeneity?

P24272, L11-13 – Without a quantitative assessment, it is not apparent which is preferable.

P24273, L5 – Please remove “Recall”.

P24273, L6-9 – Is it not possible for the authors to determine which of these two processes is the most influential?

P24273, L19-28 – The authors return to discuss the O3 concentrations; why not present the NOx results first and keep all of the analysis of O3 together?

P24275, L3 – I would urge the authors not to use the term “recycling” even in quotation marks. The updates from CheT to CheT2 is NOT a coarse OH recycling “fix” in the sense in which the term “recycling” is generally understood in the atmospheric chemistry community; rather it is an inclusion of a sound theory-based set of reactions by which OH is re-generated from higher-generation isoprene oxidation products. Perhaps using “regeneration” would be a more apposite shorthand for this.

P24275, L5-7 – Please remove the high/low values. They are an unnecessary distraction.

P24275, L12-17 – Surely the authors could determine whether CiTTyCAT is producing CO at a much higher rate than UKCA? It seems more likely that it is a mixing problem given that the two models share the same primary emissions, deposition and chemistry mechanisms.

P24275, L22 – This is not particularly surprising; most chemistry mechanism intercomparisons find that O3 concentrations are impervious to choice of scheme and in this case the changes were relatively minor in terms of the overall mechanism.

P24276, L9-10 – I would swap the order i.e. relative to the measurements (and to UKCA) as surely the obs are more important in terms of model evaluation.

P24277, L1 – Please keep “High res” in quotation marks. As this went over a page break it gave the appearance of a new sentence which was a little confusing.

P24277, L2 – I thought this rather interesting as NOx emissions are little affected by

the change in resolution; it might be something the authors could investigate further (although see my comments earlier about choice of flight).

P24277, L11 – In fact none of the panels show UKCA data.

P24277, L23 – Please remove “Remember”. I would also suggest making this a new paragraph.

P24278, L4 onwards – Please could the authors explain why they have chosen to use CheT when exploring the sensitivity of the model to vertical mixing. The isoprene mechanism included in CheT has been superseded by that in CheT2. It seems illogical to return to the older scheme for subsequent analyses.

P24278, L12 – Please replace “caricatured”.

P24278, L13-14 – Please state in the text to which figure the coloured lines refer.

P24279, L18 – Please explain what “in reality” it means that Mix3 gives the best model-measurement fit.

P24279, L24 – Please remove “Recall”

P24279, L27 – I would very much like the authors to explain their choice of site. While arbitrary from the point of view that there are no measurements at a suitable location, there must have been a logical process by which the point in space was selected. Presumably, it was a nice round distance (i.e. 1 deg) from Manaus in the direction that would be downwind under prevailing conditions. Is it the centre of a grid cell?

P24280, L20 – I would suggest starting a new paragraph here at the end of the discussion of plant exposure metrics.

P24280, L21 onwards and P24821, L18 onwards – Given that the objective of this set of sensitivity tests is to demonstrate the effect of the inclusion of mixing on the plant exposure metrics, I don’t see the need to include all of the simulations here. As noted previously it seems odd to continue to use (and present) CheT when CheT2 is available

and preferable. Further, given that the (arbitrary) benchmark of the performance of CiTTyCAT is UKCA (which is run at 3.75 x 2.5 deg) it seems as if the baseline simulation should be CheT2, "UKCA res" with anything other than this presented as an additional sensitivity experiment.

P24281, L5 – Why have the authors used 50 and 75 ppbv as threshold values? While these are (roughly) the WHO levels for human health, this study is concerned with the impact on vegetation. In this context the threshold in terms of exposure metrics is 40 ppbv. Surely it is therefore the number of times 40 ppbv is exceeded that is of interest, as that is difference that will impact the calculated values of "AOT40".

P24282, L1 – As the authors have no way of calculating POD with the atmospheric chemistry models that they use in this study, I would be interested to know how they deduce the changes in O3 concentration would have less effect on POD than "AOT40".

P24282, L4 – Following on from above, it's not clear to me how the authors can give a broader message regarding all 3 plant exposure metrics when they were only in a position to calculate 1 ("AOT40"). It certainly seems intuitive that AOT40 will change similarly, but the relative size of the change is likely to be quite different; as noted before, this study is not able to calculate POD.

P24282, L11 – Please remove "recall".

P24282, L12 – While this insensitivity was noted in flight B735, it was also notable that the same was not true of the 4 other flights included in this study.

4 Summary and discussion

P24283, L13-14 – Please continue to demote this version of AOT40 as "AOT40".

P24283, L19-27 – I would be interested to know how realistic the authors think this is, i.e. that little or no horizontal mixing occurs between air parcels during long-range transport such as that shown in Fig. 1.

P24284, L12 – Please remove "Recall".

P24284, L13-19 – I'm not sure that this level of detail (i.e. giving the parameter names, rather than just saying timescales, and stating their values) is necessary or desirable in the summary.

P24285, L15 – I would suggest the authors change how they refer to the Mix1-3 simulations; the models were not actually combined. Perhaps something along the lines of the Lagrangian nudged toward Eulerian mixing or similar would be a better description of what was done.

P24285, L26 – As noted before 40 ppbv may be a more appropriate threshold in the context of this study.

P24286, L10 – Please could the authors explain what they mean by a "transfer function" here. Do they intend O3 concentrations or plant exposure metrics to be scaled by such a function?

Table 4 – As noted previously, I would suggest using 40 ppbv as the threshold.

Fig. 1 – This is far too small; I needed to zoom to 400% to make out the details of the flight tracks. As each panel has the same scale, I would suggest using a single colour bar for the figure. Each panel could be labelled (a), (b), etc. and described in the caption (removing the need for each to be individually titled). An indication of spatial scale (in km) would be useful. The biggest improvement by far however would be to condense the information provided. I see no benefit to be gained from presenting every 1-minute interval back-trajectory individually. Why not "bin" them according to sector of origin? Perhaps with line thickness indicating the relative size (i.e. number of trajectories) of the bin?

Fig. 2 – The panels are far too small (again a zoom of 400% was necessary). I would recommend removing this Figure entirely. If it is retained I would suggest, as for Fig. 1, a reduction in the number of colour bars included (many are the same) and would

remove the titles in exchange for panel labelling.

Fig. 3 – Again the panels are far too small. I would strongly urge the authors to remove some of the (less interesting) species and focus on those that (a) change markedly; and (b) could be expected to make the largest difference to modelled O3 concentrations.

Figs. 4-9 – I would suggest swapping the y-axes. The concentrations are the key data with the altitude as an "extra". I would also prefer the x-axis to show LT (in hh:mm format). If possible it would also be nice to see rough distances from Port Vuelho and Manaus represented as this could affect NOx-regime.

Fig. 5 (and Figs. 8-11) – The panels are far too small. Removing the x-axes labels from upper panels would help, as would removing the legend from each panel and showing it once to the side of the figure.

Figs. 8-9 – If the authors feel that all 5 panels should be presented, perhaps they could be stacked vertically.

Figs. 9-11 – Please be consistent with the use of each colour. Up until this point, blue has been used for UKCA, green for CiTTyCAT and red for altitude. Please do not now use blue or red for a CiTTyCAT simulation. In Fig. 11, please ensure that the line colours are consistent between the upper and lower panels.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 24251, 2015.