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Atmospheric Chemistry and Physics
Discussions
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Interactive comment on "Multi-model simulation of CO and HCHO in the Southern Hemisphere: biogenic emissions and model uncertainties" *by* G. Zeng et al.

Anonymous Referee #2

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General comments

This paper presents the results of a modelling study simulating carbon monoxide and formaldehyde concentrations in the Southern Hemisphere, using two different biogenic emissions estimates and four atmospheric chemistry and transport models, over a period of 5 years (2004-8). Model performance was evaluated through inter-model comparisons as well as observational data from surface stations.

This is a timely and well-structured study. The use of atmospheric columns of a variety of marker compounds to constrain biogenic emissions estimates and elucidate the skill of atmospheric chemistry models in simulating the oxidation and transport of these species and their products is an important method in regions of sparse observations (ironically those with the highest emissions). The inclusion of simulations with artificial tracers to disentangle the contributions of transport, primary emissions and secondary production of CO is particularly nice. Given recent work indicating the influence of biogenic precursors and their climatically and air quality active products, such an evaluation is urgently needed. This study marks and important first step in this process.

Overall, the study is well presented, described and discussed. I do have concerns over the level of detail provided regarding certain aspects of the work, and these are outlined below. In particular, I feel that the analysis is not of sufficient depth, especially with regard to an attribution of the observed differences to the chemistry mechanisms included in the four atmospheric chemistry models used. There are also sections where the English is poor, particularly earlier in the paper, and the manuscript is in need of a thorough editing.

Specific comments

2. Model simulations and observations

One of my chief concerns with the paper is the level of detail provided regarding the model set-up and simulations. The information provided here for the processes, boundary conditions and driving data for each model is not consistent, making comparison difficult. The authors refer to previous model descriptions too often with regard to key processes. While it is extremely difficult to ensure full consistency between models, the paper would benefit from greater elucidation of some of the inconsistencies that might affect the conclusions drawn from their results.

2.1 Simulations

Biogenic emissions estimates are strongly dependent on land cover (i.e. vegetation distribution), and driving meteorology. Presumably, although the two inventories are generated off-line, it is done with the "default" input fields for MEGAN-CLM and LPJ-GUESS. The authors should state clearly here the input data used, as well as the spatial and temporal resolution of the emissions inventories generated, as this is a clear source of inconsistency with the chemistry models that are then driven with these emissions. For some of the model set-ups, the emissions will be more compatible with the land surface and meteorology driving data for the chemistry and transport than others. Differences in land cover, for example, will also affect deposition of the primary emissions and their oxidation products; differences in meteorology will affect chemical production and loss rates. It is possible that some of the observed differences between simulated concentrations of oxidation products are a result of these inconsistencies. The use of model specific natural emissions of ocean CO is inconsistent with the use of specified biogenic emissions. Given that one aim of the study is to assess the impact of different biogenic emissions on chemistry transport model output and that anthropogenic emissions are the same for each model, surely this would have been a simple difference to eliminate.

Furthermore, in the descriptions of the GEOS-Chem and CAM-chem model simulations, the authors state that diurnal variability was imposed on the biogenic emissions inventories. Was this also done for NIWA-UKCA and TM5? And why? What was the temporal resolution of the biogenic emissions calculated by MEGAN and LPJ?

Likewise the use of model specific lightning NOx emissions is inconsistent with the approach of harmonizing emissions used to drive the four chemistry models. NOx levels are critically important to the atmospheric oxidation of volatile organic compounds. Again this would have been a simple source of discrepancies between models to eliminate from the study. For both lightning NOx and ocean CO emissions, I would like to see further justification of the use of model specific inventories. How substantial are the differences between the models, and how sensitive are the models to these inputs?

2.2 Models

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The details provided about the four atmospheric chemistry and transport models used in this study are not consistent. In addition to the temporal and spatial resolution of the model and the driving meteorology, other aspects off particular importance to VOC oxidation and atmospheric concentrations are imposed methane concentrations, the dry deposition scheme and the treatment of partitioning of gas-phase condensable species to the aerosol phase. The level of detail of the chemistry mechanism is also critical.

At the very least, the authors should specifically (i.e. not simply by referring the reader to previous model descriptions) provide information of all of these in either the text or Table 2 (or both). Details of the chemistry mechanisms should include the number of chemical species (transported, reactive, fixed) and reactions (photolysis and thermal) in the full mechanism, and in the isoprene and monoterpene oxidation schemes. Furthermore, details of relevant compound lumping (e.g. are methacrolein and methyl vinyl ketone treated separately or lumped as a single species) should be provided, particularly with regard to species within the biogenic chemical degradation paths, and related species such as methanol and acetone. I would also like to see the authors elucidate further on the treatment of organic nitrates, PAN, peroxy radicals and (in the case of isoprene) epoxides. Monoterpene chemistry, in particular, varies considerably between atmospheric chemistry models and a full description of the schemes used within each model would be useful in the form of an Appendix or as Supplementary Information.

2.3 Observations

Please give details of the temporal resolution of the raw measurement data so that this can also be clearly compared to that of the models.

Sections 3-6

My other main concern is the lack of detailed consideration and discussion of the differences in chemistry between the models and the impact this has on the simulated

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HCHO and CO concentrations. It seems that it should be straight-forward enough to provide details of the main chemical sources and sinks of HCHO and CO in the various chemistry schemes, and the relative production and loss rates to each. How do these differ between low and high NOx environments in the models?

This lack of detail makes these sections extremely unsatisfying. The authors speculate on numerous possible causes for the differences between modelled and measured HCHO, but do not explore any of these further. I recommend that they select the most likely causes and perform some sensitivity tests to establish whether it is indeed a contributor to the persistent under-estimation of HCHO in these models.

3. Comparison between models and observations

Again, there is a lack of consistency, this time between sites. While it is unavoidable that Darwin has only total column data, while partial columns are preferable for this application, the authors need to consider the likely effect that this has on the comparisons they are making and the conclusions that can be drawn. Ideally, I would like to see the authors show comparisons of total columns for another site to assess how model performance differs between total and partial column values. It could be expected that the use of total columns might tend to smooth differences between models as they have greater spatial coverage.

4. Model differences in chemistry and transport

Again there is an unnecessary inconsistency in approach. Previous evaluations of model performance had used partial (or total in the case of Darwin) columns. Here the authors choose to use tropospheric columns, with total heights set by the model-specific tropopause.

Technical corrections

Title - The authors do not, to my mind, currently address model uncertainties in a quantitate enough way to justify including this in the title.

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Abstract – the Abstract contains far too many undefined acronyms (e.g. NOAA GMD, NDACC, etc.). Please either spell these out in full or define them here.

1. Introduction – the English is particularly difficult to understand in this section of the paper. I suggest the authors read it through and edit it appropriately.

Here, and throughout the paper, abbreviations and acronyms (particularly for the models) are used without having been previously defined (e.g. MEGAN).

The section on p2618, L4-10, seems rather contradictory. The authors state that global models are suitable tools for investigations such as these, and follow this up with the assertion that "For instance, global atmospheric models systematically overestimate observed . . .", which seems to indicate their lack of suitability. Perhaps some rearrangement would help – the authors are right to highlight the shortcomings of the tools they are using, and to identify them as suitable.

P2618, L17 – are the authors aware of any atmospheric chemistry models that are still using a value of ∼200 Tg y-1 for isoprene emissions? I was under the impression that as model chemistry had improved, this value had not been used since TAR.

P2618, L24-29 – I'm not sure that I understand the point the authors are making here, regarding the Marais et al. study and the sparsity of direct observations. Again, rephrasing would be helpful.

P2619, L1-4 – Again, this section appears to contradict the previous section. Further details of the model set-up used would perhaps help. Were biogenic emissions included in this study? If so, were they also generated by MEGAN?

P2619, L14 – does the "They" used here refer again to the Morgenstern et al., study

2. Model simulations and observations P2621, L2 – Please state the % difference in monoterpene emissions as has been done for isoprene. P2622, L10 – Figure 2 does not show differences; it shows the absolute values from two models side-by-side.

P2623, L13 – Please provide further information here (or elsewhere in this paper) of the simple monoterpene oxidation scheme used. P2623, L13-4 – Describe further how this gradient is implemented.

P2624, L19-21 – How does this assumed yield of CO from monoterpenes compare with yields in the other chemistry schemes? What is the assumed timescale of this conversion, and again how does this compare?

P2628, L20 – The sites are not shown anywhere in the paper. However, I would very much like to see them shown on a map to better understand their locations and the differences between them.

3. Comparison between models and observations

P2628, L17-21 – Could the differences in direct CO emissions from the ocean (different in the various models) also be a factor?

P2628 – Please include a direct comparison between modelled and FTIR CO columns for LPJ simulations, either as an addition to Figure 3 or as a new figure.

P2629, L2-3 – Were the emissions inventories generated from the same meteorological driving data (in which case, the model columns might be expected to show similar seasonal and inter-annual variations)? Or, if not, please could the authors indicate how seasonal and inter-annual differences in relevant meteorology compare between the different models used here.

P2629, L5 Was the multi-annual ensemble mean data derived as daily or monthly averages? If monthly, why (given that the earlier comparisons were made with daily data)? If daily, why is daily data not presented in Figure 5?

P2629, L17 – Please make clear in the text that these are annual averages (presumably).

P2629, L18-19 – Why should the magnitude of the model biases have this order? Could

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the authors speculate here, or refer the reader to the section in which this is explored.

P2629, L20-22 – Again please speculate on the possible reasons for this seasonal difference between model variability.

P2629, L26 – Is the greater inter-model variability due to the relative scale of the biogenic emissions from each of MEGAN and LPJ? Have the authors tried normalizing the biases by the magnitudes of the emissions inventories? P2630, L8 and elsewhere – Please provide % differences as well as absolute.

P2630, L19-25 – It seems that this could be easily checked by selecting only the model data from dates that coincide with the sampling dates at Cape Grim. This would provide a much more rigorous evaluation in any case.

P2630, L26-7 – Please speculate as to why NIWA-UKCA and GEOS-Chem should perform so well when compared against surface observations in spite of the fact their deviations were not always the lowest when assessed against CO columns.

P2631, L5-7 – What are the characteristics of the sites for which LPJ performs better than MEGAN? Why might this be the case?

P2631, L9-13 – See above regarding the anomalies at Cape Grim. The strong positive bias here provides further motivation for removing polluted dates from the analysis and selecting only the sampling dates from the model output data. For the purposes of transparency, the authors could show the current time series (i.e. with all dates included) on the same plot.

P2631, L22-28 – Does Figure 8 show model simulations driven with emissions data from MEGAN or LPJ? Please make it clear here and in the caption of Figure 8.

P2632, L1 – Again what are the key differences between these sites that might drive that difference in variability.

P2632, L6-7 – 10-15% is not a small difference. What is the % underestimation?

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P2632 – Again please show the comparisons between the modelled and observed columns for the chemistry models driven by the second emissions inventory (presumably LPJ again) either as an addition to Figure 8 or as an additional figure.

P2632, L10 – the multi-annual monthly mean (?)

P2632, L23-5 – Have the authors tried to scale methanol emissions input into the chemistry models to determine whether this makes a substantial difference to HCHO concentrations?

P2632, L25 – P2633, L1 – This section is highly confusing and poorly explained. I would strongly urge the authors to include the full set of reactions to which they are referring. The authors jump from a discussion of methanol emissions to a reaction yield of HCHO from CH_3OOH (acetic acid, rather than methanol). Please explain the connection between a reaction commonly included in methane oxidation in chemistry models and the methanol budget. Are the specific reactions referred to here included explicitly in all of the models used in this study?

P2633, L11 – What % of global terrestrial isoprene emissions?

P2633, L21-29 – Please give details of the chemistry included in the IMAGES model. How does it compare to that included in the models used in this study, and how do the conclusions drawn by Vigouroux et al. apply to the results here?

P2634, L1-2 – Why should a reduced oxidative capacity result in an increase in HCHO?

P2634, L9 – “differnces” should read “differences”

P2634, L21 – Please explain here rather than later in the text why January and September have been chosen for this analysis.

P2635 – Please provide the same figure for CO_OH as for CO_25 (Fig. 11)

P2635, L25 – “towars” should read “towards”

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P2636, L5 – “reflects” should read “reflects”

P2636, L15-6 – Please explain/speculate here why NIWA-UKCA should show such a small contribution from primary emissions, and relate this to the results/discussion already presented from comparisons of the partial CO and surface CO data.

P2636, L21 – “agreement” would read better than “correspondence”

P2637, L4 – “deduce” would read better than “derive”

P2637, L25-9 – As noted previously, the authors should include full details of the isoprene and monoterpene oxidation schemes of the four chemistry models used in these simulations. Presumably, the authors do not mean that isoprene itself is lumped, but it would be instructive to see which of its products are lumped and how this differs between models.

P2637, L25-29 – It would be helpful if the authors were to refer the reader back to the bottom row of panels in Figure 13.

P2638, L1 – Please state in the text that the profiles shown in Figure 14 are again in latitudinal zones.

P2638, L1-3 – Why are there no observations included in Figure 14?

P2638 – Although I appreciate that the authors have chosen to focus on output from the MEGAN simulations, I would suggest that it would be useful to include at least a discussion of the LPJ simulations and how they compare. Are the results in keeping with those from MEGAN (even if of lower magnitude)? Are there any notable differences that might highlight the behaviour of the chemistry mechanisms under different NOx:VOC ratios?

P2638, L8 – Again, inclusion of the detailed isoprene and monoterpene oxidation schemes employed by each of the models would greatly facilitate the discussion here. What are the relative rate constants for the photolysis of HCHO in each of the models?

What are the production rates of CO and HCHO from the oxidation of the biogenic VOCs in each?

P2638 – See above comment. It would be useful to see the production rates of CO and HCHO from OH oxidation of biogenic VOCs for each model. Ditto loss rates of HCHO to OH-initiated reactions.

P2638, L17-8 – What is the correlation coefficient for each model? Or the ratio of HCHO to OH?

P2638, L25 – I suggest that the authors could easily produce vertical profiles of H2O vapour for each model, or H2O/OH in line with the HO2/OH profiles in Figure 14.

P2637, L28-9 – Again, the authors should calculate the production/loss rates for these reactions to determine which is the cause, or if it is a combination of the two.

P2639, L1-3 – Given the complexities of the chemistry involved, it is hard to see how the authors can draw this conclusion without having conducted a more detailed analysis than is presented here.

5. Sensitivity of modelled SH CO and HCHO to uncertainties in biogenic emissions

Please could the authors explain why they have (again) introduced an unnecessary inconsistency into their analysis. In the previous section they chose to focus on January and September; here January and July. Please justify fully this decision, or ideally, change one of he sections so that the same months are discussed in both.

P2639, L17-19 – The right-hand panels in Figure 15 appear to show increases in the CO columns in LPJ relative to MEGAN in some parts of northern and central Africa. However, the choice of colour scale is poor. Furthermore, as this is a discussion of the columns over the Southern Hemisphere, there is no reason for the plots to extend north of the equator.

P2640, L2 – “reginos” should read “regions”

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P2640, L4-6 – What is the relative change in biogenic emissions between the models for each season? Does this correlate with the changes in columns? What are the seasonal differences of NOx emissions in the region?

P2640, L15 – Please could the authors explain what they mean by spatial variations of the biogenic emissions. Is that variations between the two inventories? Are these the only two regions with marked spatial variations? At what resolution are these variations apparent?

P2640, L23-29 – As these are both oxidation products of biogenic VOCs perhaps the authors could explain more clearly why one should be correlated and the other anti-correlated with OH concentrations. Again, this would be best done via an analysis of production and loss rates. What are the correlation coefficients?

P2641, L1-4 – Again, please quantify the relative speeds of the isoprene oxidation processes in each model, and present the mechanisms used in each.

P2641, L5-14 – Given the limited nature of the discussion of the zonal vertical profiles presented in Figure 18, I would suggest that showing the panels for January and July only would suffice. There is little difference between those for October and January in particular.

6. Summary and conclusions

P2641, L23 – I would not describe -19.2% as comparing well, certainly not relative to the other locations.

P2642 – The word “significant” has a specific statistical meaning. As the authors have not performed any tests of significance they should not use it in this context.

P2642, L9-12 – While the complexities of the system preclude a simple solution to the discrepancies between modeled and observed HCHO, a more detailed analysis of the differences between the chemical mechanisms and the HCHO production and loss rates to the various reactions included in each would at least indicate where future research should be directed.

P2642, L17-20 – Please state here in the text the % difference in biogenic emissions between the MEGAN and LPJ simulations.

P2642, L21-2 – “neither” . . . “nor” should read “either” . . . “or” as the sentence is already negative.

P2642, L25-26 – “reduced” would read better as “reductions in”

L2642, L25-28 – What is the % difference between biogenic emissions in MEGAN and LPJ for these source regions?

P2642, L1-2 – As before, the inclusion of details of the oxidation processes and a more thorough analysis and discussion of how the differences between the models affect the production/loss of HCHO and CO would strengthen this assertion.

P2642, L6 – Without the in-depth analysis of the chemical production and loss rates of HCHO in the various models, the authors are over-stating the case, by saying that they “show”. Their results may suggest this, but they have not conclusively demonstrated causality.

P2643, L8-12 – To what extent do the models include the latest findings on isoprene/OH in low NOx environments?

P2644, L1-2 – It would be instructive for the authors to perform sensitivity tests with the models using the same methane fields for all. This would allow them to assess the effect of NMVOC chemistry differences alone.

P2644, L3 – “distribuitons” should read “distributions”

P2644, L3-18 – This entire section would be strengthened if the authors included the analyses of the biogenic VOC oxidation mechanisms, together with production and loss rates, as suggested previously.

P2644, L19-20 – a more detailed analysis of the sources of the differences in modelled CO and HCHO concentrations between the mechanisms, might result in an improvement in these mechanisms, allowing a more robust use of HCHO and CO columns to constrain biogenic emissions and reduce this uncertainty.

Table 1 – The text in section 2 states that the models use internally generated emissions of lightning NOx and ocean CO emissions. These emissions should be clearly stated somewhere either in the text or in this table, which currently implies identical CO emissions across all models.

Table 1 – While C5H8 and C10H16 have been identified within the Introduction as isoprene and monoterpenes respectively, these compounds are referred to almost exclusively by name rather than formula in the text. Please use the names here.

Figure 3 – The title of 3rd panel should read "Wollongong"

Figure 3 – The fourth panel would benefit from being (vertically) larger so that differences between the models can be more clearly seen.

Figure 5 – Why are the observations plotted as symbols and the model output by lines? If the observations and model output is equivalent then they should all be depicted in a consistent way (i.e. all as symbols or lines).

Figure 15 – The choice of colour scale is poor. It is particularly hard to distinguish between the different reds used for positive changes, and for the greens used for changes of -5-10% and -10-15%. Why do the panels extend north of the equator when this paper is focused entirely on the Southern Hemisphere? Ending the plots at 0S would help with the issue of the scale, as well as removing the area in northern Africa where the changes are strongly positive.

Figure 15, Caption – What do the authors mean by "multi-annual and ensemble mean"? Surely the plot only shows the multi-annual mean for each of the four models?

Figure 16 – See the comments regarding the colour scale and geographical extent of

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Figure 15.

Figure 17 – As above.

Figure 18 – While I realise the contours are clearly labelled this figure would again benefit from a better choice of colour scale.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 2615, 2015.

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