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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal variability and long-term evolution of tropospheric composition in the tropics and Southern Hemisphere" *by* K. M. Wai and S. Wu

Anonymous Referee #1

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Review for Seasonal variability and long-term evolution of tropospheric composition in the tropics and Southern Hemisphere.

===== General comments:

This manuscript is split into two sections. Firstly, back trajectories and a chemical transport model are used to discuss seasonal variability in CO observed at surface stations located in remote oceanic regions, with particular attention being paid to the impact of biomass burning emissions. The model is then used to investigate future tropospheric chemistry over the Southern Atlantic Ocean with respect to changing emissions and

climate.

The subject of this paper appears to be appropriate to ACP as the paper aims to provide a better understanding of the current impacts of biomass burning in the Southern Hemisphere and future changes in ozone, which is important in terms of both air quality and climate. They also consider the impact of changing biomass burning emissions in addition to anthropogenic emissions in the future, which will be particularly important in the SH. However, I am unclear as to what emission estimates the study has used for future biomass burning.

My main concern is that whilst the contents of the paper are interesting, grammar, particularly poor sentence structure, resulted in a paper that was hard to read. Therefore a lot of effort is required by the authors to make this paper of publishable quality. I have pointed out some examples in my comments below, however, the list is not exhaustive.

Whilst the outline of the sections in the paper is clear and well ordered and they have used a variety of tools to properly investigate the seasonality of CO in the SH, there are some areas where the author's conclusions/arguments are not sufficiently backed up by the Figures. This can most likely be rectified by the inclusion of additional figures and some changes to the representation of results (it seems the analysis/simulations may already exist but the authors have chosen not to include it in some cases). In particular, I think Section 3.1 would be much clearer if the authors changed Figures 2 and 3. It would be useful in this section to see a seasonal climatology of back trajectories started at the station locations so the reader can get a feel of where the air comes from throughout the year. It would also be useful to have an additional present day simulation which excludes biomass burning emissions so you can separate the impacts of OH and biomass burning on CO at the stations. The study also mentions the use of MOPITT data , however, no results are shown. It would be useful if the authors add some figures showing some model/satellite comparisons which would be useful in terms of evaluating the model export pathways of the biomass burning.

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I would like the authors to consider my questions and revise the manuscript before I recommend the publication of this paper. Details of my comments will be found in the following.

===== Major comments:

Figures - Text is too small.

###Block 20015### L5-9: Description of MOPITT isn't required as you don't use MOPITT data in any of your analysis (unless you add some figures). Some description of the satellite data used for lightning flashes would be useful.

###Block 20016###

L13-15: Why have you used a CTM to investigate future climate changes in tropospheric chemistry? Maybe add a sentence here to say the benefits of using a CTM for this study.

L17-22: "we apply the IPCC A1B scenario for the 2000–2050 changes in anthropogenic emissions of ozone and aerosol precursors. Natural emissions of ozone precursors including NOx from lightning and soil, and NMVOCs from vegetation, are computed locally within the model on the basis of meteorological variables and hence allowed to change in response to climate change. The potential effects of climate change on biomass burning (e.g., Westerling et al., 2006; Spracklen et al., 2009) are not considered in this study." - What biomass burning (BB) emissions did the authors use for the future scenario? From Table 1, BB emissions differ between the present day and future run. In the above paragraph you only mention anthropogenic emissions.

###Block 20017###

L3: Check lifetime of CO.

Section 3.1: It would be useful if there was a table or some description of the seasonal max/min CO concentrations at the three surface observations and in which season

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they occur. This would help the reader compare the stations and would be beneficial for the discussion of when BB emissions are important at each station.

Section 3.1: It would be useful if there was a map of tropical BB emissions at different times of the year to aid the discussion of where and when emissions are important.

Section 3.1.1: Why not show the MOPITT comparisons in Figure 2 along with the model output? It would be useful to see some observed CO to compliment the model results. This would further consolidate your arguments if it can be shown that the model captures the export patterns.

###Block 20018###

L1-2: "Based on the above discussion, the impacts of burnings in Northern Africa on the CO variations of Mahe Island (Fig. 1) are expected to be minimal".

L8-10: "The spring peaks in measured CO at Mahe Island are thus attributed to the burning activities from India." I do not find the argument convincing from what you have shown in Figures 2. Specifically, why have you chosen January 2005 to investigate BB impact on Mahe Island when according to Figure 1, CO at Mahe in January 2005 is actually quite low in comparison to other years and months (e.g. February 2005 or January 2006). This therefore suggests that the time period you have chosen for your trajectory analysis and model CO maps may not be representative. I would therefore suggest choosing a different time period when observed CO is higher. Your arguments would also be more convincing if you could somehow show a climatology of trajectories for different years and months to give a better overall picture, demonstrating that the trajectories originate from India for the majority of time when CO is high.

###Block 20019###

L24: "The satellite retrievals indicate that deep convection occurred more frequent in September (Fig. 4) compared to August within the 5 yr period." – Figure only shows September. You need to show some sort of climatology of lightning flashes to show

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that this is the month where lightning peaks.

###Block 20020###

L7: “it is expected inter-seasonal variations of CO at Easter Island are minimal.” – you can see this from figure 1 as the range in CO concentrations are small in comparison to the other stations. Having a simulation that didn’t include biomass burning emissions would allow you to remove any influence of BB on this station.

L10: “It is attributed to the persistent westerlies about 30S or further southward pick up the CO-laden air masses from Africa and Latin America burnings” – what about OH seasonality?

L10-18: “It is attributed to the persistent westerlies about 30S or further southward pick up the CO-laden air masses from Africa and Latin America burnings. The air masses are firstly transported over the Indian Ocean and then reached Australia aloft, advent over the Pacific Ocean and eventually reached the Easter Island. Previous study (Edwards et al., 2006) found that a band of high CO concentrations developed which circumscribed the globe around during Austral spring and elevated CO levels from background were observed in Australia and New Zealand. The elevated CO background is attributed to be originated from Southern Africa/Latin America (Rinsland et al., 2001).” – Badly written.

L18: “Our backward trajectories launched from Easter Island reach Australia/New Zealand aloft during these periods (figure not shown), which support the CO transport pathway by the westerlies.” – Why not show these?

###Block 20021###

L13: “The increasing trend of CO at Ascension Island appears to be driven by the increases in biomass burnings from Latin America/Southern Africa and increase of ambient CH4 in recent years.” – How do you know this? I do not see any evidence in your work that this is the case. Again, a comparison between a simulation with long-

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term changing biomass burning emissions and one with either fixed or removed BB emission would identify if this is the case for fires. What about OH changes? Does your model capture this trend?

###Block 20023###

L7: "troposphere show little contribution from biomass burning" – how do you know this?

L22: The anti-correlation between change of O3 and water vapor (Fig. 6 middle and bottom right panels) within the area is due to O3 photolysis with water vapor to produce hydroxyl radicals with the presence of UV radiation. For example, the increase of water vapor at 10S facilities O3 photolysis and reduces the O3 concentrations there." – This doesn't make sense. Rewrite. Also mention Brasseur et al., (2006) in J. Clim. as they showed something similar over the tropics. Can you show OH difference plots in Fig 6 alongside ozone changes?

###Block 20039###

Figure 1: It would be nice to see a model run without BB emissions to prove seasonality is driven by BB emissions.

###Block 20040###

Figure 2: - What are the colours of the different trajectories? - It would be nice to add MOPITT at 700 hPa over the same region as shown for the model and MODIS hotspots to see if the satellite captures the same export patterns as the model. - Why not use February 2005 or January 2006 – as mentioned above, the observations show low CO in January 2005. - Extend the region shown for the model CO map so you can see Mahe Island. It seems that some CO is transported from North Africa in the direction of this station. ###Block 20040-41###

Add station locations to maps of model CO in Figures 2 and 3. This would make it easier to see if the CO plumes reach the stations.

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###Block 20041###

Which month is the model CO shown over South America? Add to Figure description.

###Block 20043###

It would be interesting to add the same analysis for the model output to the figure (both with and without BB emissions).

###Block 20044###

- Text is too small on the Figures. - Difference plot of what from what (i.e. present day minus future run?). - You say UT and MT, need to know specific heights.

===== Minor comments:

###Block 20012###

L2-3: “are studied by global chemical transport model (GCTM), satellites retrievals and surface measurements” - Insert “a” -> “studied by a chemical. . ..” - satellite not satellites

L25: Emission -> Emissions

###Block 20013###

L6: “There is much less emission from fossil fuel combustion in the Southern Hemisphere (SH)” -> Emissions from the combustion of fossil fuels are much lower in the Southern . . .

L9: “On the other hand, there are more biomass burnings in” -> Most of these emissions occur in

L15: Define SAO

L23: “transport was reported” -> transport have been reported”

L24: “Better understanding. . .” -> However, better understanding . . .

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L27: “interest to the potential” -> interest in the . . .

###Block 20014###

L2-3: “The 2050 climate change alone was estimated to increase the global lightning NOx emission by 18%” – I don’t know what you mean.

###Block 20016###

L16: “driven” -> drive

###Block 20017###

L14-18: “The CO plumes from Northern African burnings are transported westward or south-westward by the Harmattan flow and are lifted above the planetary boundary layer (PBL) when the plumes encounter the cool monsoon air from the Gulf of Guinea, as well as the Inter-tropical Convergence Zone (ITCZ).” -> Shorten/split into two sentences.

###Block 20018###

L26-28: “The trajectories further suggest two main exit pathways of the CO plumes that one is from southern tip of the continent to Indian Ocean and another is from western part of the continent” - > bad sentence.

###Block 20019###

L7: concentration -> concentrations

L10: remove ‘historical’

L12-12: “Since CO plumes followed the exit pathway from the southern tip of the continent to Indian Ocean are transverse at higher southern latitudes (e.g. 10_ S–20_ S), no elevated CO were measured at Mahe Island.” – rewrite.

L16-18: “The fire maps shown most of the burning activities in Latin America were undertaken at Brazil, Bolivia, Paraguay and Argentina and intensified in August to Oc-

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tober in 2002–2006 and were resulted in the so-called “smoke corridor” over these countries.” – this doesn’t make sense, needs rewriting. Plus I don’t know what fire maps you are referring to.

###Block 20021###

L1: “with a polynomial equation in the form as shown below” -> with the following polynomial equation:

L11: “A statistically significant ($p < 0.01$) increasing trend is identified with the increase rate of 0.33 ± 0.24 ppbyr-1 and the r2 of 0.61.” – rewrite.

L15: “An inverse modeling study with observational constraint on CO emissions from MOPITT retrievals during 2000–2009 suggested a significant increase of CO emissions since 2000 from 137 Tgyr-1 (in 2000) to 198 Tgyr-1 (in 2007) in Latin America, although the emissions was lower at about 130 Tgyr-1 in 2008 and 2009.” – rewrite.

L24: “when the burning smoke transported from the Amazon regions was also found over Latin America” –> rewrite.

###Block 20022###

L6: “The CH4 increase of $\sim$7.5 ppbyr-1 was observed from the” ->An increase of $\sim$7.5 ppbyr-1 in CH4 was observed by the. . ..

L21: “In future January, more intense CO plumes from Northern Africa just north of the equator follow the easterlies and is transported towards Latin America over the Atlantic Ocean (Fig. 6), due mainly to the burnings (Table 1).” – how do you know that this is due to biomass burning emissions? What do you mean by ‘intense’?

###Block 20023###

L3: “The future trends” – this is not a trend, maybe say ‘future composition’.

L7: “troposphere show little contribution from biomass burning” – how do you know

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this?

###Block 20024###

L3: “In Southern Africa, the reduced emissions (Table 1) might not well reflect in the ambient CO concentrations because of the large growth of CO emissions due to fossil fuel in South Africa, Republic of the Congo and Gabon, such that only 15% reduction within 10–20_ S in Southern Africa is resulted.” – Bad sentence, rewrite.

L5: “such that only 15% reduction within 10–20S in Southern Africa is resulted”

L7: “results in more than 450 ppb in lower troposphere”

L10: “increased more than 80% to 120 ppb” - Where are these figures from? Do you show it? If so refer to Figures.

###Block 20025###

L17: “stabilize the PAN” -> stabilizes PAN.

###Block 20026###

L2: “It is predicted a general increase of O3 concentrations throughout the tropospheric column over the SAO in future.” – rewrite. e.g. An increase in future O3 concentrations is predicted throughout the tropospheric column over the SAO.

L7: “reduced up” -> reduced by up. . ..

L23: “is the highest OH concentration near the tropics due to the highest UV and high water vapour found there.” - Bad sentence, rewrite.

L26: “The concentrations are then decreased with latitudes in both hemispheres.” -> The concentrations decrease with increasing latitude in both hemispheres.

L26: “Northern Africa just north of the equator..” -> Northern Africa. . ..

L28: “high NOx and O3 and is offset” -> remove the ‘and’ after O3

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###Block 20027###

L2-3: “leads to a relatively small changes of OH concentrations” -> leads to relatively small changes in OH”

L8: “is attributed by the increase of” -> is attributed to the increase in

L18-21: “While in the continent of Southern Africa, the combined effects of change of emissions of CO, NOx and NMVOCs and geographical shift of land use, as well as the change in ambient CH4 concentration, result in a larger reduction (∼30%) of lower tropospheric OH concentrations there in future.” – rewrite.

###Block 20028###

L21: “Combining the increase of different sources emissions, increase of O3 by 20–35% across the Atlantic Ocean just north of the equator is predicted.” – rewrite.

L25: “a general increase of O3 levels of the entire SAO” -> a general increase in O3 levels over the entire SAO.

###Block 20028###

L 3-4: “complicated spatial change of lightning flash rates found over the SAO in future” – this wasn’t shown.

L8-9: “contribute to less than 45% of the total tropospheric ozone. . .” -> contribute less than 45% to the total trop. . ...

L18: “In future January, increase of CO, NOx and O3 concentrations in lower troposphere due to the increase of the burning emissions leads to a relatively small changes (< 10 %) of OH concentrations over. . .” – rewrite.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 20011, 2013.