We thank both referees for their valuable and constructive comments. We have carefully followed the reviewers' comments and suggestions to revise our manuscript and have provided point-to-point responses below.

Anonymous Referee #1

===== 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.

Response

We have added clarification on the emissions in the MS –
""Following Wu et al. (2008a), we apply the IPCC A1B scenario for the 2000–2050 changes in anthropogenic emissions of ozone and aerosol precursors based on data from the IMAGE socioeconomic model [IMAGE Team, 2001; Streets et al., 2004]. Anthropogenic emissions include those associated with fossil fuel, biofuel, and human-induced biomass burning."

Comment
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.

Response
We really appreciate the referee's help on this. We have followed the referee's comments and suggestions for revision throughout the manuscript.

Comment
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. 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.

Response
We have carried out some additional model simulations and analysis following the reviewer's suggestion. We have also updated the figures in the MS. A figure showing the seasonal climatology of back trajectories has been added. More detailed responses are provided below for specific comments.

===== Major comments:

Comment
Figures - Text is too small.

Response
We have enlarged the text size in the figures.

Comment
###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.

Response
Point well taken. We have removed the description on MOPITT data. We have some figures on CO from MOPITT, but are not sure about the copy-right issue on MOPITT data so decided to not include them in the MS (The relevant MOPITT products are easily accessible at http://www2.acd.ucar.edu/mopitt). We have also removed the LIS lighting plot since that is not necessary for the discussion in the MS.

Comment
###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.

Response
We have clarified this sentence as: "To examine in details the potential impacts of global change on atmospheric chemistry and composition in the SH and the equatorial region, we carry out GEOS-Chem simulations driven by meteorological fields archived from the GISS GCM simulations. "

Comment
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.

Response
We accounted for the changes in "anthropogenic biomass burning" such as agriculture burning based on data from the IMAGE socioeconomic model [IMAGE Team, 2001]. We have revised/clarified this part as –
"Following Wu et al. (2008a), we apply the IPCC A1B scenario for the 2000–2050 changes in anthropogenic emissions of ozone and aerosol precursors based on data from the IMAGE socioeconomic model [IMAGE Team, 2001; Streets et al., 2004]. Anthropogenic emissions include those associated with fossil fuel, biofuel, and human-induced biomass burning."
We have moved the sentence "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." to the discussion section.

Comment
###Block 20017###
L3: Check lifetime of CO.

Response

We have revised/clarified this part as: "The atmospheric lifetime of CO in the troposphere ranges from weeks to months (Duncan et al., 2007). The relative long lifetime of CO makes it a tracer suitable for studying the long-range transport in the troposphere"

Comment

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 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.

Response

Point well taken. We have added descriptions of the CO seasonal variations at the three sites in the text:
"The CO concentrations usually peak in September-October, January-February and September-November for the Ascension Island, Mahe Island and Easter Island, respectively. The minimum CO concentrations are generally found in January-March, May-July, and February-March respectively at these three sites. "

Comment

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.

Response

The location of the BB emissions has been added to Fig. 1 in the MS.

Comment

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.

Response

We have decided to remove the description on MOPITT data in the MS. We have some figures on CO from MOPITT, but are not sure about the copy-right issue on MOPITT data so decided to not include them in the MS. However, we have enclosed a plot here for the reviewer's reference - see Figure A1 in the Appendix at the end of the document.

Comment

###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.

Response

We have revised/clarified this part as –
"Since the biomass burning plumes from Northern Africa are generally transported westward or south-westward, they are not expected to significantly affect the atmospheric CO over the Mahe Island located in the Pacific Ocean (Fig. 1). To examine the spring peaks of CO at Mahe Island, we carry out back-trajectory analysis which indicates that air masses at Mahe Island are dominated by those originate from India in January and February with additional contribution from Europe via Middle East, Sahara Desert and Indian Ocean (Fig. 2). The burning emissions in India are intensive in January to March, as shown by the GFEDv2 emissions and findings of Galanter et al. (2000). The spring peaks in measured CO at the Mahe Island are thus attributed to the biomass burning emissions from India."

Comment

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.

Response

Yes, we have examined the climatology of trajectories. We have updated Fig. 2 to include the trajectory frequency plot.

.

Comment

###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 that this is the month where lightning peaks.

Response

Point well taken. Indeed, deep convection/lightning should be common for the season, not just for September; so we have removed our earlier discussion specifically on September.

Comment

###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.

Response

We have followed the reviewer's suggestion and carried out a sensitivity model simulation without biomass burning emissions. Now the contribution of BB is shown in Fig. 1.

Comment

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.

Response

We have revised this part to –

"Our modeling results show that, with the prevailing north-westward atmospheric transport, biomass burning plumes from Latin America have little impacts on atmospheric composition over the Pacific Ocean at higher southern latitudes (> 10° S)". Therefore, the Easter Island which is located sub-tropically (~ 27° S) to the west of Latin America is not directly impacted by the plumes. However, both measurement and model simulations show significant seasonal variations in CO concentrations at the Easter Island, with peaks in September-November (Fig. 1). A major contribution to the seasonal variation of CO appears to be the seasonal variation of OH, the major sink of CO. OH peaks in summer (January/February in the Southern Hemisphere) and reaches the lowest level in winter (July/August in the Southern Hemisphere) (Seiler et al., 1984). To further examine the potential impacts of biomass burning on the seasonal variation of CO at Easter Island, we perform a sensitive model simulation by excluding biomass burning sources in the model ("no_bb" in Fig. 1). Figure 1 also shows the CO levels contributed by biomass burning (i.e. CO_{bb}: total CO minus CO_{no_bb}) at the Easter Island which indicates that both the OH seasonality and biomass burning emissions contribute to CO variability at the Easter Island. The "bb" component of CO at the Easter Island is attributed to biomass burning emissions from Africa and Latin America being picked up by the persistent westerlies about 30°S or further southward. These biomass burning plumes are first transported across the

Indian Ocean towards Australia. Then they are advected over the Pacific Ocean eventually reaching the Easter Island. This transport pathway is consistent with previous studies. Rinsland et al. (2001) attributed the elevated CO background over Australia during October to December in 1997 to emissions from Southern Africa/Latin America."

Comment

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?

Response

We have added the backward trajectory plot to Figure 4 in the MS.

Comment

###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-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?

Response

We have added discussion and clarification in this part -

"The increasing trend of CO at Ascension Island appears to be driven by the increase in biomass burning from Latin America/Southern Africa and increase in ambient CH_4 in recent years. Zhao et al. (2008) derived a positive trend up to 0.03 per decade for aerosol optical depth (AOD) over Southern Africa (lat: 15.0 to 5.0° S; lon: 5.0 to 15.0° E) in the fall seasons for the priod of 1980–2005, which was mostly attributed to biomass burning. A slightly positive trend during austral spring was also found over Latin America (lat: 20.0 to 10.0° S; lon: 40.0 to 30.0° W) with biomass burning plumes transported from the Amazon regions (Zhao et al., 2008). The AOD measurements from MODIS in 2001-2007 reported an increasing trend of 0.0012 yr^{-1} within 30° S–equator latitudes (Yu et al., 2009), with biomass burning in Latin America/Southern Africa as the major source of aerosols in these regions. Bevan et al. (2009) also reported an increasing trend of AOD due to biomass burning on Amazon derived by the ATSR during 2000 to 2005.

Long-term variations of CO can also be affected by the variations of atmospheric CH_4, which provides an important source for CO. Significant increases in atmospheric CH_4 concentrations have been reported since 2007 after staying relatively stable in earlier years of the past decade. An increase of ~ 7.5 ppb yr^{-1} in CH_4 was observed by the satellite instrument SCIAMACHY over the tropics (Schneising et al., 2011). The measurements from the AGAGE and CSIRO networks show renewed growth both in the SH and NH (Rigby et al., 2008). Observation with the NOAA's global sampling network also showed increases in global CH_4 during 2007 (by 8.3 ppb yr^{-1}) and 2008 (by 4.4 ppb yr^{-1}), especaially for the SH and the tropics (Dlugokencky et al. 2009)."

For OH changes, our model simulation does not indicate any trend for the study period of 2001-2010. Earlier studies based on methyl chloroform observations also derived relatively stable global OH for this period (two references listed below).

I. S. A. Isaksen, S. B. Dalsøren (2011). Getting a Better Estimate of an Atmospheric Radical. Science Vol. 331 no. 6013 pp. 38-39.

S. A. Montzka, M. Krol, E. Dlugokencky, B. Hall, P. Jöckel, J. Lelieveld (2011) Small Interannual Variability of Global Atmospheric Hydroxyl. Science Vol. 331 no. 6013 pp. 67-69.

Comment

###Block 20023###
L7: "troposphere show little contribution from biomass burning" – how do you know this?

Response
We have removed this sentence.

Comment
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?

Response

We have added the difference plot for OH concentrations at mid-troposphere over the SAO to Fig.6. We have cited/compared with the results from Brasseur et al., (2006) in J. Clim. .
We have revised this part to –
"Brasseur et al., (2006) reported similar findings over this area in their study although their results were for July. The changes in O3 appear to be primarily driven by the changes in cloudiness and therefore solar radiation. The decrease in clouds around 25-30° S increases the solar radiation and hence O3 production there. The changes in humidity can also contribute to the changes in O3 by affecting the photochemical O3 destruction. We find significant decreases in specific humidity around the 25-30° S region over the SAO."

Comment
###Block 20039###
Figure 1: It would be nice to see a model run without BB emissions to prove seasonality is driven by BB emissions.

Response

We have carried out additional sensitivity model simulations by turning off biomass burning emissions and updated the MS.

Comment
###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.

Response
The colors of different trajectories indicate that they are launched from different locations. As explained above, we have decided to exclude MOPITT plots from the MS for the copy-right issue. However, we have enclosed a plot here for the reviewer's reference - see Figure A1 in the Appendix at the end of the document.
The BB plumes from North Africa are mainly transported westward (which is evident by the trajectory plot in Fig.2), thus the CO emitted from Northern Africa should have a minimal effects to Mahe Island.

Comment
###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.

Response
Point well taken. The station locations have been added to maps of modeled CO in Figs. 2 and 3.

Comment
###Block 20041###
Which month is the model CO shown over South America? Add to Figure description.

Response
It is for September 2006; we have added this info. to the Figure description.

Comment
###Block 20043###
It would be interesting to add the same analysis for the model output to the figure (both with and without BB emissions).

Response
Unfortunately for the modeling results on CO, we have only archived monthly averages for the 10-year period but not daily data, so we are not able to make plot on daily CO.

Comment
###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.

Response
The Figures and the associated text have been enlarged in the revised manuscript. The difference is defined as 2050s – 2000s, as shown in the figure caption. Specific height have been clarified for "UT" (7.6 – 12.8 km) and "MT" (2.1–7.6 km).

Comment
===== 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 . . .
L27: "interest to the potential" -> interest in the . . .

Response
We appreciate the reviewer's help and have rectified all of these issues.

Comment
###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.

Response

We have revised this sentence to:

"On the other hand, natural emissions of ozone precursors would also be affected by climate change. For example, NOx emission from lightning, which is a significant source for NOx in the tropics, is generally expected to increase in a warmer climate (Price and Rind, 1994; Grenfell et al., 2003; Shindell et al., 2006; Wu et al., 2008a)"

Comment

###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.

Response

We agree with all of these comments and have revised them in the MS.

Comment

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 October 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.

Response

We have revised this sentence to "Most of the burning activities in Latin America occur in Brazil, Bolivia, Paraguay and Argentina (Fig. 1, Area 3 in top panel) and generally peak in August to October."

Comment

###Block 20021###

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

Response:

Done.

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.

Response

We have revised this sentence to: "The regression analysis yields an increasing trend ($\boldsymbol{a_1}$=0.33 ± 0.24 ppb yr^{-1}) for CO at Ascension Island which is statistically significant ($p < 0.01$)".

Comment

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.

Response

We have removed this sentence.

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

Response
We have removed this sentence.

###Block 20022###
L6: “The CH4 increase of _7.5 ppbyr−1 was observed from the” ->An increase of _7.5 ppbyr-1 in CH4 was observed by the. . ..

Response
Done.

Comment
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’?

Response
We have clarified this part as “Higher CO emissions from biomass burning over Northern Africa are projected for 2050s following the IPCC A1B scenario (Table 1). This results in higher continental outflow of CO from Northern Africa towards Latin America across the Atlantic Ocean (Fig. 6).”

Comment
###Block 20023###
L3: “The future trends” – this is not a trend, maybe say ‘future composition’.

Response
“The future trends” has been revised to “The future changes”.

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

Response
We have removed this sentence.

Comment
###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”

Response
This sentence has been revised to “Over Southern Africa, reductions in CO concentrations by up to 15% are calculated within 10–20°S (figure not shown) for September in the 2050s. This results from the combination of decreasing biomass burning emissions and large growth in fossil fuel combustion (Table1) in South Africa, Republic of the Congo and Gabon.”

Comment
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.

Response
We have added the figures in the MS (Figure 7).

Comment
###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
###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.

Response
Point well taken. We have done the revisions/clarifications in the MS.

Comment
###Block 20028###
L 3-4: "complicated spatial change of lightning flash rates found over the SAO in future" – this wasn't shown.

Response
This sentence has been removed in the revised MS.

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

Response
Done.

CommentL18: "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.

Response
This sentence has been revised to -"With the compensating effects associated with increasing tropospheric CO, NOx and O3 from biomass burnings, we find relatively small changes (< 10 %) in OH concentrations over the burning areas in Northern Africa and the adjacent Atlantic Ocean to the west of the continent."

Anonymous Referee #2

The authors have examined how current and future anthropogenic and biomass burning emissions influence tropospheric constituents, such as CO, O3, and OH in the tropics. Using the GEOS-Chem model, driven by meteorological fields from the GISS GCM, they have quantified how changes in emissions associated with the A1B emission scenario might impact tropospheric composition by 2050. There is a clear need for studies of this nature. However, I cannot recommend the manuscript for publication in this present form. As it is currently written, it is difficult to tell what experiments were conducted and, as a result, it is difficult to assess the interpretation of the results presented in the manuscript. There are also several places where statements are made without evidence to support the claims. For example, at the top of page 20025, they state that "the contribution of fossil fuel emissions is _10% of total O3 to the east of the continent (< 60E) over the Indian Ocean in the future," but it is not clear how they obtained this estimate. Similarly, in the penultimate sentence of the manuscript they state that the reduction in OH in the boundary layer "is due to the lack of OH sources

to offset OH loss from increased of assumed CH4 and calculated CO in future." I am suspicious that CH4 and CO could be the cause of the decreased OH and no evidence was given to support the claim. The manuscript also needs significant editing to improve the grammar. I editing some of it, but stopped after page 20017. I think this is an interesting study that would be of interest to the community, but the manuscript needs to be better written to properly describe the work that was done. I encourage the authors to consider revising the manuscript to address my comments below.

Response

We appreciate the reviewer's constructive and very helpful comments. We have extensively revised the manuscript. We have also carried out some additional model simulations and analysis. For the 1st example (regarding the contribution of fossil fuel emissions), we have clarified that part to: "Our sensitivity model simulations by excluding fossil fuel emissions from South Africa indicate that in the future scenario, the fossil fuel emissions would enhance the total O3 over South Africa by up to 30%".

For the second one (on the changes in OH), our earlier discussion was on the wide-spread OH reduction. Now we have added further analysis on the localized OH decreases which indeed appears to be driven by increases in biogenic isoprene emissions. We have revised this part to –

"The decreases in OH over the remote oceans reflect the future increases in methane and CO which are primary sinks for OH. The multi-model study by Voulgarakis et al. (2013) also reported large reduction (by 24% to 36%) in their simulated OH over these SH regions in 2100 following the RCP8.5 scenario. In addition, we find even higher OH reduction (by up to 60%) over South Africa, which appears driven by the additional effects associated with increasing biogenic VOC (in particular isoprene) emissions in the future climate. Biogenic VOC emissions in our model follow the MEGAN scheme (Guenther et al., 2006) and increase with temperature."

General Comments

Comment

1. Page 20015, line 6: How are the MOPITT data used in the analysis? Care should be taken when using MOPITT version 4 (V4) for trend analyses. As noted in Worden et al. (Atmos. Chem. Phys., 13, 837–850, 2013), MOPITT V4 data have a positive bias as a result of the assumption in the retrieval algorithm that the instrument characteristics were constant. I would encourage the authors to use V5 MOPITT data instead.

Response

We have removed the use and description of MOPITT data and plots in the MS since we are still not sure about the copy-right issue on MOPITT products – in earlier analysis, we made some plots on MOPITT CO using the interactive tool at http://www2.acd.ucar.edu/mopitt.

Comment

2. Page 20021, line 12: How do you reconcile the positive trend in CO estimated at Ascension with the negative trend reported by Worden et al. (2013) at all latitudes in the northern and southern hemispheres? They found a

weaker trend in the southern hemisphere than in the north, but it was negative. A concern with using the results of Fortems-Cheiney et al. (2011) to support the positive trend estimated at Ascension is that they used the biased V4 MOPITT data in their analysis.

Response

We have added discussion in the MS –

"We note that Worden et al. (2013) identified a slight decrease in the total CO column over the Southern Hemisphere (0^o-60^oS) for the period of 2000-2011. The discrepancy compared to the positive trend at the Ascension Island identified in this study could reflect the different metrics used for atmospheric CO (column vs. surface values) as well as the different spatial coverage in these two studies. Further study would be desirable to explore in detail the causes for this discrepancy."

We have removed reference to Fortems-Cheiney et al. (2011) and added a new reference (Bevan et al.2009; listed below) that supports our finding. They reported an increasing trend of AOD due to biomass burning on Amazon based on the ATSR data for 2000 to 2005.

Bevan, S. L., P. R. J. North, W. M. F. Grey, S. O. Los, and S. E. Plummer (2009), Impact of atmospheric aerosol from biomass burning on Amazon dry-season drought, J. Geophys. Res., 114, D09204, doi:10.1029/2008JD011112.

Comment

3. Page 20024, lines 1-6. I don't understand this first sentence. It suggests that the reduction in biomass burning will be significantly offset by an increase in fossil fuel emissions, but according to Table 1 biomass burning will decrease by 8.3 Tg CO (38%) in Southern Africa, whereas the fossil fuel source will increase by 1.31 Tg CO. How did the authors conclude that the net reduction in CO emissions will be only 15%?

Response

Indeed, we didn't express it clearly – we meant the mixing ratios of CO would decrease by up to 15%. We have revised this sentence to: "Over Southern Africa, reductions in CO concentrations by up to 15% are calculated within 10– 20°S for September in the 2050s. This results from the combination of decreasing biomass burning emissions and large growth in fossil fuel combustion (Table1) in South Africa, Republic of the Congo and Gabon."

Comment

4. Page 20026, line 9: How did the authors estimate that "> 55%" of the ozone at each model level in both the present and future time slices is not affected by emissions from Southern Africa and Latin America? Did they run the model separately without biomass burning and lightning NOx emission to isolate their impact on ozone abundances? Based on the discussion at the end of page 20025 I believe that was done, but it is not clear. The authors need to better explain what sensitivity analyses were conducted.

Response

We have revised this section to –

"We carry out a series of sensitivity model simulations to examine the contributions to total ozone over the remote SAO from various sources. Results from a specific sensitivity simulation by excluding certain emission source (e.g., biomass burning, lightning, fossil fuel combustion, soil emissions) are compared with the control run to derive the contribution from that source. Figure 9 shows the vertical profiles of total O3 over the remote SAO as well as the contributions from biomass burning and lightning sources. The most effects on tropospheric O3 from biomass burning and lightning are found in the lower and upper troposphere, respectively. Lightning NOx enhances O3 in the upper troposphere by up to 24 ppb. The contribution of soil NOx emissions is found to be negligibly small. We also find that the total emissions from Southern Africa and Latin America can only account for less than 45% of the total ozone over the SAO at any level in the troposphere; i.e. more than 55 % of the total O3 throughout the troposphere over the SAO is attributed to emissions outside of Southern Africa and Latin America as well as the stratosphere-troposphere exchange."

Comment

5. In Figure 9 the ozone profile shows a sharp decrease across the tropopause, which is not physical. I assume this is due to the use of Synoz for stratospheric ozone? What is the impact of this on their analysis of the ozone budget? Because of the upward vertical motion across the tropical tropopause, such a strong decrease in ozone could make

vertical transport across the tropopause a significant sink of upper tropospheric ozone, which would make it difficult to meaningfully interpret the results of the study. The authors should try to quantify this sink.

Response

Thank you for catching this. It was simply due to some artificial effects with the ploting. We have double checked the data and updated the figure – now the profile looks normal (continuous and smooth as expected).

Comment

6. Page 20027, lines 11-16: I don't understand how increases in background CH4 and CO could produce the large localized changes in OH in the boundary layer as shown in Figure 10. Is this due to changes in biogenic emissions of shorter-lived gases, such as isoprene, as a result of changes in surface temperatures? I would like to see a more detailed analysis that better explains the OH response shown here.

Response

Thank you for pointing this out. Our earlier discussion was on the wide-spread OH reduction. Now we have added further analysis on the localized OH decreases which indeed appears to be driven by increases in biogenic isoprene emissions. We have revised this part to –

"The decreases in OH over the remote oceans reflect the future increases in methane and CO which are primary sinks for OH. The multi-model study by Voulgarakis et al. (2013) also reported large reduction (by 24% to 36%) in their simulated OH over these SH regions in 2100 following the RCP8.5 scenario. In addition, we find even higher OH reduction (by up to 60%) over South Africa, which appears driven by the additional effects associated with increasing biogenic VOC (in particular isoprene) emissions in the future climate. Biogenic VOC emissions in our model follow the MEGAN scheme (Guenther et al., 2006) and increase with temperature."

Voulgarakis, A., Naik, V., Lamarque, J.-F., Shindell, D. T., Young, P. J., Prather, M. J., Wild, O., Field, R. D., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Folberth, G. A., Horowitz, L. W., Josse, B., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Stevenson, D. S., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: Analysis of present day and future OH and methane lifetime in the ACCMIP simulations, Atmos. Chem. Phys., 13, 2563-2587, doi:10.5194/acp-13-2563-2013, 2013.

Comment

Technical Comments

1. Page 20013, line 2: "total CO sources" should be "total CO source".
2. Page 20013, line 7: "regions than the northern" should be "regions than in the northern"
3. Page 20013, line 9: Change "more biomass burnings" to "more biomass burning".
4. Page 20013, line 14: Remove "the" before "tropospheric composition".
5. Page 20013, line 15: Add "the" before "ozone anomaly".
6. Page 20014, line 1: Please restructure the sentence. Maybe along the lines of "global increase in emissions of ozone precursors, including emissions from biomass burning."
7. Page 20014, line 3: Remove "alone" after climate change.
8. Page 20014, line 6: Remove "the" before "tropical composition".
9. Page 20014, line 7: Change "on the impact" to "of the impacts".
10. Page 20017, lines 13-14: Please rewrite the sentence starting with "It is evident by. . ." as "This is evident by the numerous fire events present in the MODIS fire map and by the elevated CO observed by MOPITT."
11. Page 20017, line 20: Change "transports" to "transport".

Response

We agree with all of the above technical comments and have revised them accordingly.

Comment

12. Page 20019, lines 25-26: It is not obvious to me from the information presented that there is more frequent deep convection in September than in August. Why not show the map for August as well?

Response

Point well taken. Indeed, deep convection/lightning should be common for the season, not just for September; so we have removed our earlier discussion specifically for September.

Comment

13. Page 20020, line 15: Replace “circumscribed” with “circumnavigated” or “circled”.

Response

Done.

Comment

14. Figures 6, 7, and 8 are too small. Why not plot the whole tropical region rather than just selected regions? It would be helpful for the reviewer to see the modeled response across the whole tropics and subtropics. Maybe show -180W to 180W and 50S to 30N.

15. Figure 9 is too small. Please make it larger so that the reader can actually see what is plotted.

Response

Point well taken. We have enlarged all the Figures in the revised manuscript. We have also expanded the spatial coverage of Figures 6, 7, and 8.

Appendix

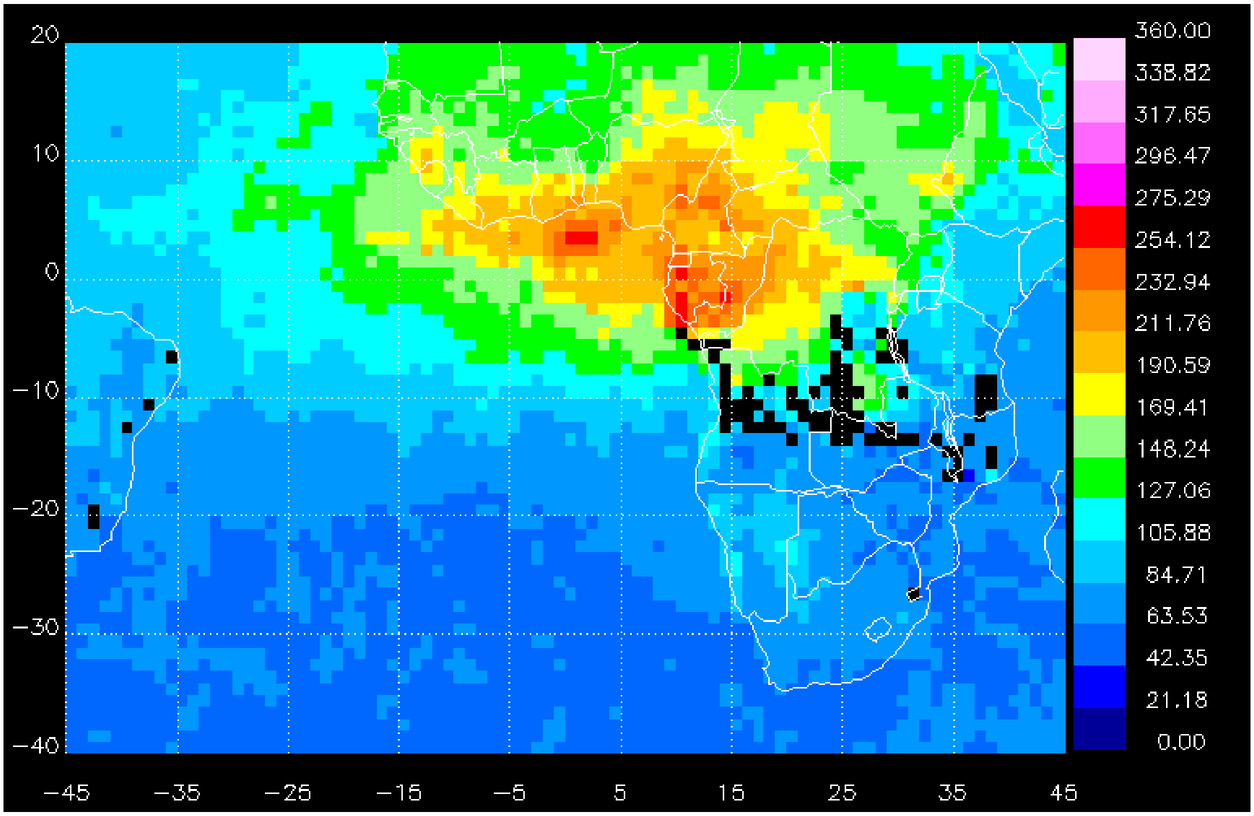

Figure A1. The MOPITT monthly averaged CO concentrations (ppb) at 700 hPa in January 2005. (produced by the Interactive Tool available at http://www2.acd.ucar.edu/mopitt)