I am pleased to resubmit for publication the revised version of ACP paper acp-2014-228. I have incorporated new figures, tables and text into the paper in response to the reviewers' comments. The introduction and background sections have been combined and condensed to better motivate the current study. The results section has been condensed and restructured to frame the discussion of meteorological and other physical and chemical processes through their effects on O_3 distributions. New analyses include an evaluation of the simulated PBL depth and mean O_3 profiles calculated over broad regions to facilitate comparison with other models. The revised paper includes discussion of the potential implications of model biases in biogenic emissions and clarifications of the implications of the aircraft-satellite-model O_3 intercomparison, and numerous other clarifications and corrections to the text. Please find below my responses to specific comments.

Reviewer #1

1. The major criticism I have of this paper is the lack of detailed information about the process controlling chemical production of O3 from the models. At present the model results are compared against the observed O3 but few details are given as to why the model results agree or disagree with the observations from a chemical point of view. There is some focus on the role of NO (from soil) and indeed comparisons are made to NO profiles, but other important O3 precursors (e.g. PAN) are neglected. Similarly there is hardly any mention of the role of VOCs in the paper. For example, isoprene acts as an important O3 precursor. How sensitive are the model results to isoprene emissions and chemistry? A cursory comparison of isoprene fluxes from observations and the MEGAN model is included. But there is little to no discussion on the impacts biases in isoprene oxidation may cause. A large amount of the model observation comparison focuses on comparison with meteorological data. Whilst this is undoubtedly a key component to the story I suggest perhaps some of this could be cut down and more analysis on the O3 budgets could be included. Or more links could be drawn between the chemistry and meteorology. What impact does biases in temperature have on O3? The wet scavenging of soluble species should impact O3 too, the effect of which can be relatively easily tested in the model simulations.

We thank the reviewer for his/her comments. We have added the following text discussing the possible model sensitivities to errors in emissions of isoprene and other BVOCs (lines 552-561 in the Revised Manuscript):

"Emissions of BVOCs can increase O_3 production by the following mechanism. Oxidation of BVOCs can lead to formation of HO_2 and RO_2●, which react with NO to form NO_2. NO_2 in turn photolyzes to form $O(^3P)$, which reacts with O_2 to form O_3. The relative sensitivities of O_3 production to NO_x or BVOC emissions depend upon the relative amounts of VOCs and NO_x present. Under clean conditions with a high VOC:NO_x ratio, O_3 production is NO_x sensitive, whereby increases in NO_x will lead to increases in O_3 while increased VOCs will have little impact. On the other hand, in polluted areas with a high NO_x:VOC ratio, the system is VOC-sensitive, that is, increased VOCs contribute to O_3 production but an increase in NO_x actually depletes O_3 (National Research Council, 1991). We expect the polluted East/South regions during BARCA A to be VOC-sensitive and the clean West, North and around Manaus regions during BARCA A and all regions in BARCA B to be NO_x-sensitive. Kuhn et al. (2010) determined via aircraft transects in the Manaus urban plume that most of the VOC reactivity was provided by isoprene emissions from the surrounding rainforest, and NO_x emissions suppressed O_3 production close to urban sources, but stimulated it downwind."

One limitation of this study is that measurements of only a few gas phase species (CO, O_3) are available from BARCA. Thus, it is not possible to evaluate other important O_3 precursors (PAN, VOCs) in the models using BARCA data. We also agree that wet scavenging of soluble O_3 precursors should impact O_3 production. However, as measurements of only relatively passive/insoluble species (CO, O_3) were taken during BARCA, a detailed evaluation of the impact of wet scavenging falls outside the scope of this study. The meteorological evaluation of the models has been shortened and rewritten to emphasize how biases in meteorological variables (e.g. temperature) impact O_3 (Section 4.2, lines 443-508).

References

National Research Council, 1991. Rethinking the Ozone Problem in Urban and Regional Air Pollution. National Academy Press, Washington, DC, 500pp.

In general the manuscript is well written, however, I think the paper could benefit from a number of changes, below, before being published in ACP.

General comments (line number, page and comment):

2. Line 1, page 14017: The authors have not included the role of VOCs (in particular BVOCs) as O3 precursors in the Amazon basin. Is this because they have no net effect on O3?

We chose to focus on NO_x as the primary O_3 precursor in the Amazon basin under clean conditions based on the study of Jacob and Wofsy (1988), who found that O_3 production in a photochemical model based on ABLE-2A was relatively insensitive to the amount of VOCs present. This was because oxidation of CO provided sufficient HO_x to generate background O_3 values of 20 ppb. Thus, the amount of additional O_3 produced in the boundary layer depended on the amount of NO_x present. However, the polluted regions during BARCA A may be VOC-sensitive, and we have added the following text to discuss the possible implications (lines 736-739):

"In polluted, VOC-sensitive conditions, approximately the correct net amount of O_3 is generated in the PBL. This suggests there is insufficient VOC reactivity in the models, since the correct amounts of O_3 deposition velocities and NO_x emissions would both decrease O_3 production."

3. Line 20-27, page 14020: Are there likely to be any issues with using land use data from c.a. 2000 when comparing to observations made in 2008/9?

Conversion of forest to pasture land cover reduces surface latent heat fluxes and increases sensible heat fluxes, as shown in Figs. 6-7 using data from von Randow et al. (2004). On a local scale, at least during the dry season, these changes decrease moisture content and increase surface temperature and the depth of the convective boundary layer over pasture areas (Fisch et al., 2004). Wang et al. (2009) found that deep convection was stronger over forested areas due to the greater humidity, but that shallow convection was enhanced over pasture areas.

The PROVEG dataset (years 2000-2001) was the most recent available for use in regional models at the time of this study. However, deforestation from 2000-2009 (see figure below) was minimal in the BARCA flight regions. Recently an updated vegetation map based on MODIS observations in 2012 was produced for regional models, and will be used in modeling studies going forward.

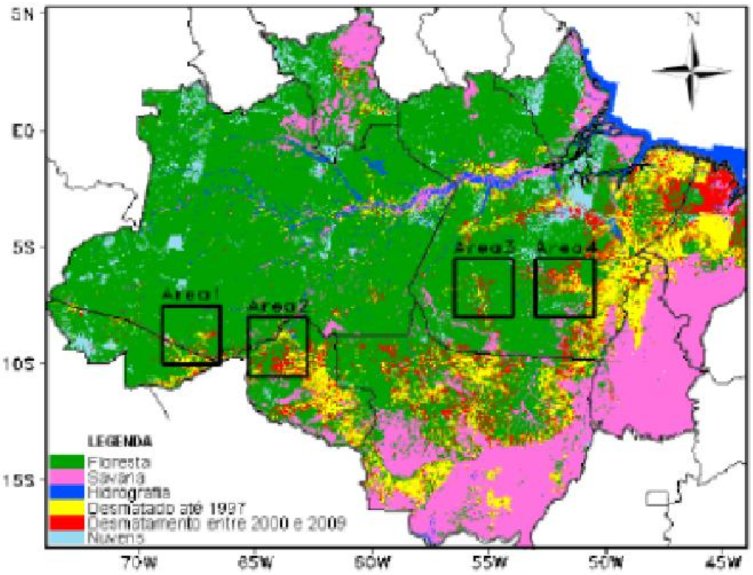

Figure 3.4 from Oliveira (2009): red areas were deforested from 2000-2009 according to data from PRODES (Satellite Monitoring of the Brazilian Amazon) Project (2010, http://www.obt.inpe.br/prodes/)

References:

Fisch, G., Tota, J., Machado, L. A. T., Silva Dias, M. A. F., Lyra, R. F. da F., Nobre, C. A., Dolman, A. J., and Gash, J. H. C.: The convective boundary layer over pasture and forest in Amazonia, Theor. Appl. Climatol. 78, 47–59, DOI 10.1007/s00704-004-0043-x, 2004.

Oliveira, R. A., Análise das Tendências da Precipitação sobre o Brasil e Impactos do Desmatamento no Regime de Chuvas na Amazônia Legal, Master's Thesis in Meteorology, National Institute for Space Research (INPE), São José dos Campos, Brazil, sid.inpe.br/mtc-m18/2011/12.08.10.56-TDI, 2009.

Wang, J., Chagnon, F. J. F., Williams, E. R., Betts, A. K., Renno, N. O., Machado, L. A. T., Bisht, G., Knox, R., and Bras, R. L.: Impact of deforestation in the Amazon basin on cloud climatology, P. Natl. Acad. Sci. USA, 106(10), 3670-3674, doi:10.1073/pnas.0810156106, 2009.

4. Line 18, page 14022: Other modeling groups will, I hope, find the observations very useful for model evaluation. As it may prove problematic to sample other models in the manner the authors have could the authors comment on the biases from averaging the observed O3 in large areas compared to the sampling they perform in the current manuscript (i.e. if they were to average the model O3 from -3N to 4N, -58E to -68E

(roughly speaking the clean sector in Figure 2 (a), how would that compare to the results presented in Figure 2(a)?).

Following the reviewer's suggestion, to facilitate other modeling groups' comparisons with the data and modeling results of this study, a new figure has been added (Fig. 16) which compares the mean observed profiles with large averaged area from the models for: clean (West, North and around Manaus regions) and polluted (East and South regions) regions during BARCA A and all regions during BARCA B.

The following text has been added (lines 350-356) explaining the methodology:

"To facilitate comparison of other models with the data presented in Fig. 2, mean profiles from the large regions corresponding to clean (West, North and around Manaus regions) and polluted (East and South regions) regions during BARCA A and all regions during BARCA B are presented in Fig. 16. From the models, all horizontal grid points falling within the corresponding region's longitude and latitude bounds for each flight day (Table 6) and the closest model output times (12-18 UTC / 8-14 LT) were averaged into 500 m vertical bins."

The following text has been added (lines 538-542) presenting the results:

"A similar model behavior is seen as in the mean profiles for individual regions. All simulations over-estimate O_3 throughout the PBL and lower troposphere during clean conditions in BARCA A, but under-estimate O_3 in polluted conditions. This is especially true from 2-4 km where biomass burning plumes detrain O_3 precursors. During BARCA B all simulations show good agreement."

5. Line 5, page 14023: The authors need to include the geographic extent that "west, north etc." refer to in Figure 2 (and Figures 18-21).

A table has been added to include the geographic extents and dates encompassed by the regions (lines 430-431):

"The longitude and latitude bounds and flight dates included in each geographic region from BARCA A and BARCA B are listed in Table 6."

6. Technical corrections (line number, page and comment):

Line 24, page 14013: Typo. "increased" should have "be" inserted before it.

Line 24, page 14015: Typo. "northem" should be "northern".

Line 18, page 14030: Typo. Amazonia needs correcting.

We apologize for these errors, and we have made the suggested corrections in the text.

Reviewer #2

The paper describes an analysis of the temporal and spatial variability in ozone concentrations, fluxes and controlling processes as observed during the BARCA campaigns. This analysis is supported by model simulations done with the regional chemistry transport modelling systems CCATT-BRAMS and WRF-CHEM. I deem this being a very interesting analysis that aims to identify the role of chemical versus physical and dynamical processes in O3 over the Amazon forest for the contrasting meteorological and chemical conditions of the wet and dry seasons. This analysis combines the information gained from both detailed observations as well as model analysis. As such it fits in very well with the scope of ACP but there are, according to me, a number of major issues that must be resolved. For example, in the model application there have been some processes not being considered/not well described (anthrogenic emissions) but that are of potentially large relevance for O3/photo-chemistry over the Amazon forest (see detailed comments below). My most serious concern is about the model application being used too much in a "black box" mode. There are many statements including the term "may" expressing that the models are somewhat being applied as a black box not really being able to really nail down the reasons for the found discrepancies between model simulated and observed chemical and meteorological properties. By the way, from the evaluation of the meteorological parameters is becomes obvious that the model representation of the meteorology for the Amazon region still poses a large limitation to properly simulate the atmospheric chemistry being largely driven by these meteorological (and hydrological) drivers.

We thank the Reviewer for his/her general comments. Numerous modifications were made that are detailed in the responses to the specific comments below.

1. Abstract: "However, O3 simulated by the models was lower than both BARCA observations in mid-levels and total tropospheric O3 retrieved from OMI/MLS, suggesting that the satellites are dominated by middle troposphere and long-range processes and are not a good indication of O3 conditions in the PBL."; Satellites are dominated?? This is apparently a very weird sentence that requires re-writing and re-thinking. The observations should be all right but apparently the models do a relatively poor job on representing the free troposphere-BL gradient in O3.

We thank the reviewer for his/her comment. We did not intend to indicate that the model-satellite discrepancy indicates an error in the satellite retrieval, and have altered the text to specify that the models do a relatively poor job of representing the free troposphere-BL gradient in O_3 compared with aircraft and satellite observations. The new sentence (lines 33-36) now reads:

"O_3 simulated by the models was lower than both BARCA observations in mid-levels and total tropospheric O_3 retrieved from OMI/MLS, which is primarily comprised of middle troposphere O_3 and thus reflects long-range transport processes. Therefore, the models do a relatively poor job of representing the free troposphere-BL gradient in O_3 compared with aircraft and satellite observations, which could be due to missing long-range and convective transport of O_3 at mid-levels."

2. Introduction; the paper starts straight away on the research questions to be addressed in this paper but where it seems that first indicating why an improved understanding/quantification of ozone temporal and spatial variability in the tropical rainforest environment is important.

We agree with the reviewer that explaining the motivation for the study is important before presenting the specific science questions. The introduction has been revised to start with an explanation of why an improved understanding/quantification of ozone temporal and spatial variability in the tropical rainforest environment is important, followed by the statement of the scientific questions.

2. Introduction, line 65: "high availability of solar radiation"; rephrase to high solar radiation levels

The sentence has been revised to include the reviewer's suggestion.

The new sentence (lines 58-61) now reads:

"The Amazon Basin continues to rapidly urbanize, and urban emissions of O_3 precursors are also expected to grow. Emissions from cities in the tropics may have a larger impact on the upper troposphere due to high solar radiation levels and intense convective transport (Gallardo et al., 2010)."

3. Line 78; here it is stated that in-situ observations of cloud properties and chemical species are the reason that we cannot constrain this system well ; I think it is much more than only cloud properties and chemical species measurements; we need information on

many additional parameters; land use changes, boundary layer dynamics, cloud aerosol interactions at the larger scale, etc.

We agree with the reviewer that many parameters/processes affect atmospheric chemistry over Amazonia. The sentence has been revised to include the reviewer's suggestion.

The new sentence (lines 97-99) now reads:

"In-situ data on cloud properties and chemical species, as well as observations of land use changes, boundary layer dynamics and larger-scale cloud-aerosol interactions, are scant in this region."

4. Line 90: "It is interesting to compare BARCA data to observations from the NASA Amazon Boundary Layer Experiments ABLE campaigns (ABLE-2A and -2B), which took place during the dry season of 1985 and wet-to-dry transition of 1987". I also think this is interesting to do but then it should be stated what is expected from such a comparison with these data from the 80's.

We agree that it is important to explain the purpose of comparing data from the current campaign with one which took place three decades ago.

The following text has been added (lines 213-221) to reflect this suggestion from the reviewer:

"Andreae et al. (2012) showed that CO mixing ratios were about 10 ppb higher during ABLE-2B than in BARCA B everywhere except the southern region, reflecting the global trend towards decreasing CO emissions since the 1980s, particularly in the Northern Hemisphere. The CO comparison also showed a similar enhancement of 10–20 ppb in the lowest 1 km above the surface, attributed to diffuse biogenic sources, and also indicated that the much higher enhancements during the dry season in BARCA A must be due to anthropogenic or biomass burning inputs. The O_3 comparison is expected to yield information in long-term trends in O_3 production in the Amazon Basin, as well as the relative importance of biogenic, urban and fire sources."

5. Line 134: "During BARCA A, coarse model aerosols were predominantly from biogenic emissions and biomass burning, while fine mode aerosols consisted of biomass smoke and some Secondary Organic Aerosol (SOA) from biogenic Volatile Organic Compounds (VOCs)". I guess you refer here to coarse mode aerosols but how do you know what the sources are of these coarse mode aerosols?

Numerous studies have focused on aerosol composition and origin in the Amazon (Martin et al., 2010 provides a review). These studies show that the dominant coarse mode source is primary biogenic emissions, while the main fine mode source is biomass burning in the dry season and Secondary Organic Aerosol (SOA) from biogenic Volatile Organic Compounds (bVOCs) in clean conditions. However, the aerosol size distribution was not measured during BARCA, so the following sentence was removed:

"During BARCA A, coarse model aerosols were predominantly from biogenic emissions and biomass burning, while fine mode aerosols consisted of biomass smoke and some Secondary Organic Aerosol (SOA) from biogenic Volatile Organic Compounds (VOCs)."

Reference:

Martin, S. T., Andreae, M. O., Artaxo, P., Baumgardner, D., Chen, Q., Goldstein, A. H., Guenther, A., Heald, C. L., Mayol-Bracero, O. L., McMurry, P. H., Pauliquevis, T., Pöschl, U., Prather, K. A., Roberts, G. C., Saleska, S. R., Dias, M. A. S., Spracklen, D., Swietlicki, E., and Trebs, I.: Sources and properties of Amazonian aerosol particles, Rev. Geophys., 48, RG2002, doi:10.1029/2008RG000280, 2010.

6. Line 142: "The mean contribution from biomass burning to total CO during BARCA-A was about 31%, with a contribution from background (110 ppb) of about 61%"

First of all refer to all flights in a consistent way; BARCA-A (previously it was BARCA A); Furthermore, the second part of the sentence reads weird; rephrase.

The sentence has been corrected to refer to the field campaigns in a consistent way (BARCA A) and to explain the data more clearly.

The revised sentence (lines 159-161) now reads:

"According to analysis of tracer simulations, during BARCA A biomass burning contributed on average about 56 ppb (31%) to the total CO of around 180 ppb, while the background was 110 ppb (61%)."

7. Line 150: "Small boundary layer enhancements were attributed to a source from the oxidation of biogenic VOCs". Would be good to see some reference here.

The reference (Andreae et al., 2012) has been included at the end of this sentence (lines 168-170).

8. Line 152: “The simulated vertical CO profiles matched mean observed values, but were overly vertical (too low near the surface and too high above 3 km), suggesting that the models were overly diffusive or had too much convective transport”. Here you already discuss a model result, one that is indicating a quite essential problem with the models relevant for the presented analysis before you have even introduced in more detail these models and their set-up.

The following sentence was added at the beginning of the paragraph (lines 171-174) to indicate that the model results being discussed are from Andreae et al. (2012), not the current study:

“Andreae et al. (2012) also showed simulated vertical CO profiles from CCATT-BRAMS and WRF-Chem simulations, as well as the Stochastic Time Inverted Lagrangian Transport (STILT) model with two different meteorological field inputs and the WRF Greenhouse Gas Module (WRF-GHG).”

9. In the overview of the O3 observations and role of different mechanisms explaining this behavior I miss the references to studies that have demonstrated/explained the behavior, e.g., line 181 on the role of convection in lofting O3 and a chemical production of 15 ppbv d-1 over Brazil but also already at the beginning of the section on the role of NO/BVOC emissions versus transport, on the observations collected in Rondonia, etc.

We thank the reviewer for his/her suggestion on how to clarify the references in the text. This portion of the background section has been condensed as follows (lines 77-80):

“Previous analyses of satellite ozone data have noted early-year O_3 maximums in the tropical Southern Hemisphere primarily associated with cross-Atlantic transport of biomass burning emissions from Africa (Fishman and Larson, 1987; Thompson et al., 1996), Northern Hemisphere fires and lightning NO_x (Edwards et al., 2003).”

10. Lines 186-193; this is one example of extreme long sentences that make the paper difficult to read; there are many more of those long sentences that require editing.

Numerous sentences were edited to make them shorter and easier to read.

11. Lines 218: “dry deposition in the region was a globally significant O3 sink”, dry deposition in the region provides a significant sink in the global O3 budget.

Line 228; “aboard”, onboard (?)

The suggested changes have been made in the text.

12. Line 230; where the measurements collected at 1.5m above the soil surface or above the canopy top? and what was the vertical extent over which the profiles were sample? In the forest canopy there are large gradients especially during nighttime and then the reference height becomes very important.

The sentence has been revised to clarify the height and vertical extent at which the measurements were collected. The sentence (lines 206-211) now reads:

"As part of ABLE-2, near-continuous O_3 surface measurements (1.5 m above the soil surface) showed daytime maximums of 3.7 ppb inside a forest and 5.7 ppb in a clearing (typical standard deviations of 0.3 ppbv). Additionally, tower measurements at the clearing site showed higher O_3 values of 6.7 ppb at 7 m above the soil surface and 6.9 ppb at 15 m above the soil surface (Kirchhoff et al., 1990)."

13. Line 281; I appreciate the overview of all the measurements that have informed us about the typical features of O3 and the photochemical and mixing/transport regimes over the Amazon but at the end what can be concluded from this?? Because of the vast amount of information it would be optimal to draw some conclusions about the main findings.

Different O_3 measurement methods enable the observation of different physical and chemical processes affecting O_3 variability in the Amazon, with satellites identifying fire and lightning sources of precursors, ground measurements observing surface processes, and aircraft in the location of convective transport. The following paragraph was added to the Introduction (lines 91-97):

"Thus, satellite observations enable the attribution of tropical O_3 maxima to biomass burning and lightning NO_x sources, while ground-based measurements allow the identification of key surface processes in the Amazon Basin affecting O_3 amounts. These processes include O_3 production from soil NO_x emissions and removal via dry deposition to the forest canopy. Aircraft campaigns complete the suite of observations, allowing the examination of convective lofting of surface emissions, with biomass burning emissions of particular importance on the regional scale."

14. Line 328; I think that indicating the location with 2 numbers behind the comma suffices.

We have corrected the latitude/longitude locations to use two decimal places (lines 401-402).

15. Line 387: "Anthropogenic emissions were estimated from the RETRO, GOCART and EDGAR v4.0 global databases updated with South American inventories (Alonso et al., 2010)". It is rather easy to read over this quite essential part of the analysis. The emissions, especially those of NOx, will ultimately determine to a large extent the photochemistry over the Amazon basin. Than having an estimate of the emissions based on a selection of different emission inventories might introduce a large range in results. I think it is essential to provide the emission inventory as used in this analysis and also show how the numbers compare to the different alternatives; e.g., how do the RETRO and EDGAR v4.0 compare for this domain and how does the actually applied inventory compare to those global inventories for the domain?

In PREP-CHEM-SRC, the emissions are obtained from RETRO if available for that species, then from EDGAR v4.0, otherwise from GOCART. The purpose of this is to use the most consistent emissions inventory possible. The following sentence has been added in order to clarify this point (lines 282-288):

"Emissions are obtained from RETRO if available for that species (CO, NO_x, chlorinated hydrocarbons, acids, esters, alcohols, ethers, benzene, ketones, methanal, other alkanals, other aromatics, C_2H_2, C_2H_4, C_2H_6, C_3H_6, C_3H_8, C_4H_{10}, C_5H_{12}, C_6H_{14} plus higher alkanes, other VOCs, toluene, trimethylbenzenes, xylene), then from EDGAR v4.0 (NMVOC, SO_4, CO_2, SF_6, N_2O), otherwise from GOCART (BC, OC, SO_2, DMS), in order to use the most consistent emissions inventory possible."

As the differences between the RETRO emissions and PREP-CHEM-SRC emissions are documented and illustrated in Alonso et al. (2010), we do not feel it is necessary to include another figure in the present paper.

16. I also realized, reading through the rest of the paper, that there is not reference at all to how the atmosphere-biosphere NOx exchange is treated, a component that is essential for the analysis in all the areas without substantial anthropogenic influences.

Biogenic NO emissions were not included in these simulations as NO was not available for the MEGAN 2000 climatology. Future simulations will include online MEGAN emissions of NO and other biogenic species. The following sentence was added to the model description section (lines 290-293) to make this clearer:

“The MEGAN 2000 climatology includes numerous biogenic species (acetaldehyde, formaldehyde, other ketones, acetone, isoprene, propane, methane, propene, ethane, methanol, sesquiterpenes, ethene, monoterpenes and toluene), but not soil NO emissions.”

17. Line 410: “while in WRF-Chem, wet deposition and lightning production of NOx were not considered.”. Why?? I think this should be explained and then later on it will be important to demonstrate/discuss the consequences of ignoring these quite essential features in the presented analysis

The text was modified to explain why these processes were not included in the WRF-Chem simulation:

(lines 315-317) On the other hand, no wet scavenging is included for cloud water and precipitation resolved by the microphysics scheme, because this option is not currently available in WRF-Chem for the RACM chemical mechanism.

(lines 326-328) “In WRF-Chem, lightning production of NO_x was not included, because these parameterizations have not yet been evaluated for the Amazon region.”

The following text discusses the consequences of ignoring these processes:

(lines 317-324): “O_3 production in the upper troposphere is affected by the net convective transport of soluble HO_x precursors (including hydrogen peroxide (H_2O_2), methyl hydroperoxide (CH_3OOH) and formaldehyde (CH_2O)). However, uncertainties remain about the scavenging efficiencies of these and other soluble species by deep convective storms.”

(lines 328-321): “In the tropics, over continents, lightning production is comparable to other sources of NO_x, including biomass burning and soil release, and it is the primary source over oceans (Bond et al. 2002). Since lightning NO_x production peaks in the upper troposphere, it could be an important contributor to ozone production.”

18. Line 480: “Especially in the case of WRF Chem, the excessive precipitation rate may be due to a too sensitive deep convective trigger function or underestimated shallow convection, leading to a more unstable atmosphere”; Would there be a way that you could indeed confirm this explanation doing some sensitivity experiments?

We thank the reviewer for their suggestion. Sensitivity experiments on parameters of the convective parameterization such as the trigger function would indeed be interesting and provide useful information for tuning the convective parameterization for Amazonia. However, we feel these tests fall outside the scope of this study. Simulations for subsequent field campaigns will use updated versions of the convective schemes and at that point it may be appropriate to tune the parameterizations.

19. In the discussion about the meteorological conditions I think it is essential to start with the analysis of the shortwave radiation terms since if this parameter is off in the models, then you would also not expect the latent and sensible heat fluxes to be correctly simulated.

We agree that the shortwave radiation will affect the heat fluxes, and we have reordered the text to reflect the reviewer's suggestion (Section 4.2, lines 476-490).

20. Line 513; "The overestimated moisture in CCATT-BRAMS may be due to overactive convective detrainment at midlevels, and could be associated with over-active O3 production"

Here you suggest with this sentence that O3 is somehow responsible for the overestimation of moisture in the model. I guess that you would like to express that the issues on moisture representation in the model coincide with issues on the O3 simulations due to issues on the convective transport.

We did not intend to suggest that overestimated O_3 production causes high moisture bias in the models. Therefore we have altered this sentence to clarify that excessive moisture may stimulate O_3 production (lines 495-497):

"The models generally show good agreement with soundings at Manaus (Figs. 8-9), but excess moisture (positive dewpoint bias of 10 K) in CCATT-BRAMS above 500 hPa may lead to increased photochemical production of O_3."

21. Line 534; Overall the analysis of the meteorological parameters (measurements and models) does not give a lot of confidence in this feature essential to a fair evaluation of the chemistry. There appear to be substantial issues on the representation of some of the key drivers of chemistry (solar radiation), tracer transport and removal processes. I also think that the analysis is not very well structured going back and forth between all the relevant meteorological parameters. Is there a more optimal way to structure this description of the analysis of the meteorology?

We agree that accurate simulation of meteorological parameters in the Amazon continues to be a challenge, and that these parameters will drive some of the main processes that affect O_3 production and transport. We now state this at the beginning of Section 3.2 (lines 444-446):

"Tropospheric O_3 distributions are driven by both chemical processes, including chemistry and emissions of O_3 precursors, and meteorological ones, such as solar radiation, tracer transport and removal."

We also added a new paragraph (lines 467-475) that summarizes the key findings of the model-data meteorological comparison and their implications for chemistry:

"Now we summarize the key findings of the model-data meteorological comparison and their implications for the chemistry simulations. The models capture the seasonal spatial distribution of precipitation over northern South America (Fig. 4), and the signs of NE-SE differences are correctly modeled by both models during both seasons, i.e., the NE is drier than the SE during November and vice-versa during May. For the Amazon, CCATT-BRAMS slightly underestimates the precipitation rates in both seasons, but the rate in WRF-Chem is about twice that of TRMM 3B43 (Table 2). This may lead to errors in the strength and vertical distribution of convective transport and the amount of convective wet removal."

22. Line 551: "Typical model anthropogenic NOx emissions values over the Amazon, primarily from biofuel source...... (Garcia-Montiel et al., 2003)." Another example of a way too long sentence.

This sentence was divided into four sentences to increase readability (lines 569-577):

"Typical model anthropogenic NO_x emissions values over the Amazon, primarily from biofuel sources, were 0.008-13 μg N m^{-2} hr^{-1} N. These NO_x emissions included in the models were less than one third of the mean values of 44 ± 14 μg N m^{-2} h^{-1} NO measured by Kaplan et al. (1988) during ABLE-2A. This is considered a threshold value for NO_x-driven O_3 production to be the dominant O_3 source in the PBL. The model emissions were also much lower than the mean emissions from forest of 35.8 μg N m^{-2} h^{-1} NO measured in the 1998 dry season (Garcia-Montiel et al., 2003)."

23. Line 727: "These discrepancies of models with observations may result from an overly mixed (constant with altitude) profile due to overly active turbulent mixing from 1-2 km or too much downward convective transport of O3 from 2 km to the surface, as observed by Betts et al. (2002)."

This statement is an example of where I think that this analysis would benefit from more in depth analysis of what really explains the observed discrepancies between the models and the measurements. There are many statements including the term “may” expressing that the models are somewhat being applied as a black box not really being able to really nail down the reasons for the misrepresentations. On this particular topic I think it would be very useful to see some analysis of the boundary layer (BL) depth, how this compares to observations of the BL depth over tropical rainforest and also to see, if the BL depth would be different, to what extent this is due to issues on the surface energy balance representation, model representation of entrainment/detrainment processes, etc.

We agree that we would like to better understand what explain the discrepancies between the model and observations. For complex coupled meteorology-chemistry models, with many feedbacks among processes, physical and chemical parameters and input data sources, it is difficult to attribute an error to specific processes. In response to the reviewer’s suggestion, we have included an analysis comparing the maximum CBL depth from Fisch et al. (2004) with the models at forest and pasture sites for both seasons. The text describing this analysis is as follows (lines 414-424):

“Fisch et al. (2004) found that in the dry season (14-25 August, 1994), higher sensible heat fluxes over pasture increase the maximum height at 21 UTC (17 LT) of the Convective Boundary Layer (CBL) from around 1100 m for forest (Rebio Jarú) to 1650 m over pasture (FNS). On the other hand, during the wet season (Jan.-Feb. 1999) the height of the CBL is similar for both land types, around 1000 m. The simulated height of the PBL at 21Z above the forest and pasture sites (Table 4) was analyzed from model output using two different methods: *TKE*, the first level above the surface where the Turbulent Kinetic Energy (TKE) from the PBL schemes dropped below 0.01 m^2 s^{-1} and *Theta*, the first level above the surface where theta exceeded theta of the level below by 0.6 K. In addition, *WRF MYNN* is the diagnostic from the WRF PBL scheme which takes into account TKE as well as stability.”

Reviewer #3

1. Overall, the paper presents valuable results but would benefit from better organization around the main science questions. For example, the introduction (Section 1) and Previous studies (Section 1.3) could be combined and condensed so that they lead directly into the questions this study will address. Stronger links between the model evaluation and the science questions would also be helpful.

We agree that the previous studies should be presented in order to justify the science questions of this current study. Following the reviewer's suggestion, the introduction and previous studies sections have been combined and condensed, followed by the science questions.

Specific Comments:

2. Abstract Line 15-18: There are a number of reasons ozone might be higher in OMI/MLS than the model besides lack of PBL sensitivity in the satellite data.

We agree that in the scope of this study it is difficult to assess the accuracy of the OMI/MLS data. The too-vertical nature of the model profiles could be due to missing inputs from the boundary conditions and errors in the convective transport. This sentence in the abstract has been modified to:

"O_3 simulated by the models was lower than both BARCA observations in mid-levels and total tropospheric O_3 retrieved from OMI/MLS, which is primarily comprised of middle troposphere O_3 and thus reflects long-range transport processes. Therefore, the models do a relatively poor job of representing the free troposphere-BL gradient in O_3 compared with aircraft and satellite observations, which could be due to missing long-range and convective transport of O_3 at mid-levels."

3. P14010 Line 16: Please explain "The flights consisted of quasi-Lagrangian measurement"

Lagrangian measurements involve following an air parcel as it moves through the atmosphere in order to be able to constrain sources and sinks of chemical species found within the parcel. As it is nearly impossible to do this with an aircraft, the term "quasi-Lagrangian" is used to refer to sampling a parcel, then intercepting what is thought to be the same parcel at a later time and location. The following paragraph (lines 130-138) has been rephrased in order to clarify this terminology:

"In-situ measurements were made of carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), ozone (O_3), and aerosol number concentration and optical properties. Flask samples were collected to determine CO_2, CH_4, sulfur hexafluoride (SF_6), CO, nitrous oxide (N_2O), hydrogen, and the oxygen-nitrogen ratio (O_2/N_2).The flights consisted of quasi-Lagrangian measurements, which attempt to sample an air parcel at multiple locations along its path in order to constrain regional and basin-wide fluxes of these species. The aircraft had a ceiling of 4500 m, and flights usually consisted of ascending and descending vertical profiles separated by short (5–30 min) horizontal legs."

4. P14022 Line 26-28: What is the advantage of using the 16 boxes instead of just sampling the model at the location of the observation?

The following sentence was inserted to explain this reasoning (lines 337-339):

"As the model output has a much coarser spatial and temporal resolution than the aircraft measurements, the model value is interpolated to the observation time and location."

5. Section 3.3 1st Paragraph: Is this background information or findings of this study? If it is background, please include citations.

This is background information to set up the analysis of the impact of seasonal variations in meteorological and emissions conditions on the chemistry. The paragraph has been edited to include references as follows:

(lines 446-448) "During the dry-to-wet transition season, increased actinic fluxes stimulate the production of OH radicals from O_3 photolysis that can lead to net O_3 production (Seinfeld and Pandis, 2006)."

(lines 452-457) "Decreased surface temperatures and incident solar radiation due to cloudiness suppress emissions of biogenic VOCs such as isoprene (Fall and Wildermuth, 1998). In addition, higher surface humidity and precipitation decrease the occurrence of fires (Morton et al., 2013; Chen et al., 2013) that emit NO_x and VOCs (Freitas et al., 2007). O_3 precursors are further decreased by wet removal within the storms (Barth et al., 2007a). On the other hand, during the dry-to-wet transition season, increased actinic fluxes stimulate the production of OH radicals from O_3 photolysis that can stimulate net O_3 production (Seinfeld and Pandis, 2006).

References:

Barth, M. C., Kim, S.-W., Skamarock, W. C., Stuart, A. L., Pickering, K. E., and Ott, L. E.: Simulations of the redistribution of formaldehyde, formic acid, and peroxides in the July 10, 1996 STERAO deep convection storm, J. Geophys. Res., 112, D13310, doi:10.1029/2006JD008046, 2007.

Chen, Y., Velicogna, I., Famiglietti, J. S., and Randerson, J. T.: Satellite observations of terrestrial water storage provide early warning information about drought and fire season severity in the Amazon, J. Geophys. Res. Biogeosci., 118, 495–504, doi: 10.1002/jgrg.20046, 2013.

Fall, R., and Wildermuth, M. C.: Isoprene Synthase: From Biochemical Mechanism to Emission Algorithm, J. Geophys. Res., 103(D19), 25599-25609, doi: 10.1029/98jd00808, 1998.

Morton, D. C., Le Page, Y., DeFries, R., Collatz, G. J., and Hurtt, G. C.: Understorey fire frequency and the fate of burned forests in southern Amazonia, Phil. Trans. R. Soc. B, 368(1619), doi: 10.1098/rstb.2012.0163, 2013.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 2nd edition, J. Wiley, New York, 2006.

6. Page 14030 Lines 3-5: The second part of the sentence does not necessarily follow from the first, since there could be errors in the model's vertical distribution of ozone.

We agree with the reviewer that we cannot conclude that the satellite data is missing PBL O_3. The tropospheric O_3 may be lower in the models than satellite due to missing mid-level inflow and sources, as is also indicated by the comparison with the aircraft observations and SHADOZ. This sentence has been revised as follows (lines 743-746):

"In addition, simulated O_3 was lower than both the OMI/MLS total tropospheric O_3 and the BARCA observations in mid-levels, indicating that the models are missing sources at mid-levels from long-range and convective transport."

7. P14034 Lines 6-7: Better agreement than what?

Both sensitivity simulations agreed better with observations than the original simulation. The sentence was revised and now reads (lines 715-718):

"Additional simulations with WRF-Chem showed that O_3 in the lower boundary layer was about twice as sensitive to increases in O_3 deposition velocity as reductions in NO_x emissions, but both simulations achieved better agreement with observations than the base case simulation."

8. P14034 Lines 9-10: Are there other possible sources of model error?

Yes, in clean (NO_x-sensitive) conditions, low ozone deposition and NO_x emissions can contribute to the O_3 overestimate, while in polluted (VOC-sensitive) conditions these errors may be compensated for by insufficient VOC reactivity. We have added the following sentence to clarify this reasoning (lines 736-741):

"In polluted, VOC-sensitive conditions, approximately the correct net amount of O_3 is generated in the PBL. This suggests there is insufficient VOC reactivity in the models, since the correct amounts of O_3 deposition velocities and NO_x emissions would both decrease O_3 production. Additionally, in clean, NO_x-sensitive conditions, proportionally more O_3 is produced per unit NO_x emissions and the O_3 deposition velocities are still too low, resulting in an overestimate."

9. P14034 Line 24: Could insufficient ozone deposition also contribute?

Yes. See response to Comment #8.

10. P14035 Lines 1-4: While the lack of surface sensitivity in the satellite data is known and is a potential factor in the model/obs mismatch, there can be many sources of model error. This statement, here and in the abstract, needs to be re-worded; one cannot conclude simply from the fact that simulated ozone was lower than OMI/MLS at mid-levels that the O3 observed by satellites is dominated by the mid-troposphere and long-range transport.

These statements were revised as follows in the Abstract (lines 33-39):

"O_3 simulated by the models was lower than both BARCA observations in mid-levels and total tropospheric O_3 retrieved from OMI/MLS, which is primarily comprised of middle troposphere O_3 and thus reflects long-range transport processes. Therefore, the models do a relatively poor job of representing the free troposphere-BL gradient in O_3 compared with aircraft and satellite observations, which could be due to missing long-range and convective transport of O_3 at mid-levels."

And in the Conclusions (lines 743-746):

"In addition, simulated O_3 was lower than both the OMI/MLS total tropospheric O_3 and the BARCA observations in mid-levels, indicating that the models are missing sources at mid-levels from long-range and convective transport."

11. P14025 Lines 5-8: This sentence is confusing. Please re-word.

Following the suggestion of the reviewer, the sentence was re-worded to be clearer as follows (lines 477-480): “However, for the southern Amazon forest and pasture sites peak shortwave may be overestimated (underestimated) by 50-100 W m^{-2} by CCATT-BRAMS (WRF-Chem) (Figs. 6-7), suggesting that there is insufficient (excessive) cloudiness in the models.”

12. Figure 2 Caption: What statistical test does Matlab use to determine outliers?

The following text was added to the Fig. 2 caption to include this information:

“the whiskers extend to the most extreme data points not considered outliers and outliers are plotted individually as red plusses. Values are drawn as outliers if their values exceed q3 + w(q3 - q1) or are less than q1 - w(q3 - q1), where q1 and q3 are the 25th and 75th percentiles, respectively, and w is the maximum whisker length with the default value of 1.5. For normally distributed data, the whiskers encompass from approximately the 2.7 to 99.3 percentiles.”

Comments about organization:

13. P14008 Lines 8-13: This seems like a separate paragraph and should be moved elsewhere.

A new paragraph was created after the Lelieveld et al. (2008) citation and the remainder of the paragraph was reordered as follows (lines 58-69):

“The Amazon Basin continues to rapidly urbanize, and urban emissions of O_3 precursors are also expected to grow. Emissions from cities in the tropics may have a larger impact on the upper troposphere due to high solar radiation levels and intense convective transport (Gallardo, et al., 2010). In the upper troposphere, O_3 acts as a greenhouse gas, increasing surface radiative forcing (IPCC, 2001). Inhalation of elevated levels of ozone can irritate the lungs; aggravate asthma and cause emphysema, bronchitis, and premature death (Schwela, 2000). High ozone concentrations can also inhibit photosynthesis in plants and damage leaf tissue, harming wild ecosystems and reducing crop productivity (Reich and Amundson, 1985). Thus, an improved understanding/quantification of O_3 temporal and spatial variability in the tropical rainforest environment is important for projecting future impacts of land use and climate change in the Amazon Basin and other tropical rainforest regions worldwide on their expanding human populations and significant biodiversity.”

14. P14009 Line 15: Description of BARCA seems like it should be a separate paragraph

A separate section was created for the BARCA description (Section 2, lines 122-221)

15. Are sections 1.1-1.3 all subsections of the introduction? Section 1.3: This section could potentially be combined with the introduction. It contains a lot of detail on past studies, but it would be helpful to relate this information more strongly to the goals of the current study and how the current study will advance our understanding.

Subsection 1.3 was condensed and integrated into the main body of the introduction to create a more coherent justification of the current study.

16. Section 3.2: There is a lot of detail in this section that is difficult for the reader to keep track of and relate to the main chemical processes. The last paragraph provides a nice summary, so perhaps other portions of the text and the number of figures could be reduced. Another possibility would be to combine sections 3.2 and 3.3 but discuss each portion of the campaign separately.

Following the reviewer’s suggestion, this section (now 4.2, lines 444-508) has been condensed and reframed in terms of impacts on the chemical processes.