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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ozone production and transport over the Amazon Basin during the dry-to-wet and wet-to-dry transition seasons" *by* M. M. Bela et al.

Anonymous Referee #2

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Review of paper acp-2014-228: Ozone production and transport over the Amazon Basin during the dry-to-wet and wet-to-dry transition seasons by Bela et al.

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 infor-

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mation 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.

Below you can find my more specific comments.

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 allright but apparently the models do a relatively poor job on representing the free troposphere-BL gradient in O3.

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.

Introduction, line 65: "high availability of solar radiation"; rephrase to high solar radia-

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tion levels

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.

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.

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?

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.

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.

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

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models relevant for the presented analysis before you have even introduced in more detail these models and their set-up.

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.

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.

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 (?)

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.

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.

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

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 pho-

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tochemistry 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? 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.

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

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?

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.

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.

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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?

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.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 14005, 2014.