We thank the reviewers for their supportive and thoughtful comments. Our responses to the comments are provided below, with the reviewers' comments italicized.

Review 1:

This paper does a modelling investigation of the light absorbing aerosols in a pristine tropical forest site. The manuscript is extremely well written, figures have good quality and the topic is of interest to the ACP readers. In the attached PDF file I have added comments indicating where further explanation/discussion is needed or could be helpful. Besides these technical remarks, I only have two important comments.

The authors used a modified version of GEOS-Chem which they validated against AERONET, MODIS and CALIOP. However, what happens to aerosol particles in the atmosphere depends on meteorological conditions and you have not shown if your model reproduces the observed climate/weather. Does your model simulates the observed precipitation rates (distribution in time and space)? What about wind direction and speed? Boundary layer height and TKE? Clouds? SW and LW radiation? These are all important for properly accounting for physico-chemical transformations of aerosols in the atmosphere. The model could be getting the ground concentration right (or wrong) by biased PBL heights, for instance. If this modified version of GEOS-Chem was not validated yet, the authors should include some discussion about it, at least in the supplement.

We now explain better in Section 2 that meteorological fields used in GEOS-Chem are from the GEOS-5 FP atmospheric data assimilation system: "We use the GEOS-Chem CTM version 9-02 (http://www.geos-chem.org/), **a global 3-D model of atmospheric composition** driven by assimilated meteorological data **GEOS-5 FP** from the NASA Global Modeling and Assimilation Office (GMAO). **GEOS-5 FP is the current operational meteorological data, which is now produced with version 5.13.0 of the GEOS-Data Assimilation system (Lucchesi, 2013)**." We also acknowledge the source of GEOS-5 FP data in the Acknowledgments: "**The GEOS-5 FP data used in this study/project have been provided by the Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight Center**."

The authors use a mixture of different emission inventories, most of which are widespread used and considered to be reliable in our community. However, looking at the AAOD plot in Fig.14 one can see similar values over the Amazon as over the northeastern coast of Brazil, from Ceara down to Bahia (highly urbanised and lots of anthropogenic emissions). Therefore, it seems that your inventories don't account correctly for urban emissions in South America. Except for SP (which shows up as a red spot at 47W 23S), none of the other large metropolitan areas seems to be represented. If such information was not available for using in GEOS-Chem, the authors should at least discuss the limitations of their results for not considering those emissions.

We now have a more detailed description about the emission inventory in Section 2.2: "**We note that the Bond et al. (2007) emission inventory accounts for urban emissions in NSA (e.g., Ceará and Bahia in the northeastern coast of Brazil and São Paulo) as seen from the distribution of BC emissions in Fig. 1. We also compared the BC fossil fuel and biofuel emissions from Bond et al. (2007) with those from the HTAP emission inventory (http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123), and found that the two have very similar distributions, but that the emissions from Bond et al. (2007) are twice as high as those from HATP over NSA**." We also modified the color scale in Fig. 14 to show the influence of the emissions over the northeastern coast of Brazil and explain better in the text: "During the wet season, AAOD over **the central Amazon including the ATTO site is generally**

lower than 0.0015, reflecting a minimal influence of human activities over this region. High AAOD (up to 0.01) over Colombia is caused by open fires, whereas over Southern Brazil it originates from fossil fuel and biofuel combustion near São Paulo. Fossil fuel and biofuel emissions over the northeastern coast of Brazil, although not as high as those from São Paulo, also result in slightly enhanced AAOD (0.0015-0.003)."

The following responses refer to the comments made by the reviewer into the ACPD pdf file:

Typically, seasonal forecasts use only 1 month spin-up. Please explain why your model needs so much longer time. Do you need to equilibrate the soil moisture? Also tell how many ensemble members are you using, and if you are coupling to an ocean model.

We always spin up the simulation for a sufficient period of time to remove the signature of the restart file from the output. We now explain this better in the text: "We initialize the model **with a 1-year spin-up followed by full chemistry simulation of Jan-Apr 2014. We also run additional simulations with a 6-month spin-up for black carbon and primary organic aerosol to isolate the contributions from different sources by tagging them in the model**".

We are not sure what the reviewer meant by "*how many ensemble members are you using*". We are not coupling to an ocean model. We now explain this better in the text: "We use the GEOS-Chem CTM version 9-02 (http://www.geos-chem.org/), **a global 3-D model of atmospheric composition** driven by assimilated meteorological data GEOS-5 FP from the NASA Global Modeling and Assimilation Office (GMAO)."

As there are other elements, this means you are considering an O:C atomic ratio lower than 0.7. I wonder how much lower, since we expect it to be 0.6-0.8.

Assuming a negligible contribution from H and N, the OA/OC ratio of 1.7 would imply an O:C of about 0.52, which is at the low end of the expected range. However, as mentioned in the text, we adopt the OA/OC ratio of 1.7 from the measurements in the Amazon Basin (Chen et al., 2015; Chen et al., 2009). Since this ratio is lower than the value of 2.1 for aged aerosols suggested by Turpin and Lim (2001), we discussed the uncertainty introduced by this ratio at the end of Section 5.2: "Note that the contribution from BrC is directly affected by the OA/OC ratio assumed for POA in the model. In this work, we adopt a ratio of 1.7 measured in the Amazon basin during the wet season of 2008 (Chen et al., 2009), which is lower than the value of 2.1±0.2 for aged aerosol suggested by Turpin and Lim (2001). Using the latter ratio would increase BrC and total aerosol absorption at 550 nm by 25 % and 6 % at ATTO during the wet season. The corresponding AAE would also be increased to 1.7±0.37."

how are these simulated aerosols distributed in size?

We now modify the text to explain this: "Aerosol types simulated in GEOS-Chem include carbonaceous aerosols, sulfate-nitrate-ammonium aerosols, **fine and coarse mode** sea salt, and mineral dust **in four size classes (aerosols other than sea salt and dust are treated as fine mode aerosols)**."

How well does this hygroscopic growth model represents what has been observed in the Amazon region from HTDMA measurements?

We now add the following sentence in the text: "**The hygroscopic growth factor of OC at 90% RH in the model is 1.25, consistent with the range of 1.0-1.3 for Amazonian fine aerosols at 90% RH reported by Rissler et al. (2006) and Zhou et al. (2002).**"

This value of MAE is much larger than that used by Rizzo et al 2013... and it will scale all your simulated BC aerosol absorptions. Please make a stronger reasoning of why using 12, and what the impact it has on your results.

We now explain it better in the text: "**The MAE value is proportional to the AAOD value, and is a fundamental factor in the estimation of directive radiative forcing. Recent studies have shown that the MAE of BC should be increased from 7.5±1.2 m^2 g^{-1} at 550 nm for freshly generated BC to 9-13 m^2 g^{-1} as BC becomes internally mixed with other aerosols chemical components, which is also supported by ambient measurements and the comparison between models and observations (Bond and Bergstrom, 2006; Bond et al., 2013; Wang et al., 2014a).** In this work, we **thus** scale the MAE at 550 nm to 12 m^2 g^{-1} assuming thick coating due to the abundance of SOA in the Amazon basin (Chen et al., 2009; Pöschl et al., 2010)."

Did you use the AERONET derived values in the model or not? If not, why not?

We didn't use the AERONET derived values and now explain it better in the text: "Our value for the real part is 6 % higher than the retrieved data, **which consequently results in slightly higher dust MEE (0.60±0.026 m^2 g^{-1}) averaged over the dusty region than the AERONET-derived value. However, the value of 1.56 is consistent with the range of 1.51-1.56 in the literature (McConnell et al., 2010)**. … **Considering that the AERONET retrieval may be influenced by aerosols other than dust and that the difference between the model and AERONET retrieved data is not significant, in particular for MAE, we keep the model dust refractive index unchanged for the following analysis in this work**."

Justification is needed here.

We now modify the text: "The results are scaled up by 39 % to cover the range of 70–470 nm **based on an intercomparison campaign where our 4-channel SP2 was compared to a revision-D 8-channel SP2**."

The vertical scale in figure 6 is DOY and it is not possible to identify the dates. I would suggest using horizontal dotted lines in each of the panels in fig. 6 to indicate the start dates of each event.

Done!

Please check. For jan-apr 2014 it should be version 3.30 not 3.01. The 3.01 is only available until Oct/2011.

Thanks! We corrected the error in the text

You should also average the highres caliop data vertically to match the model vertical resolution. Besides, you will have a problem in this comparison because caliop is only probing a very narrow swath, while you are trying to compare with a model grid of 250 x 250 km averages.

We did average the caliop data over the model grid in both horizontal and vertical dimensions. We now explain it better in the text: "For the comparison, observations are averaged over the

model grid **in both horizontal and vertical dimensions**," We also have pointed out later in the text that: "the substantial remaining bias could be due to the narrow swath of CALIOP and the relatively coarse model resolution"

Contributions from different components are very different, as expected. I suggest trying to make a vertical log scale.

The stacked area plot is to display the trend of the contribution of each component as well as the trends of total concentrations to be compared with observations. We tried the log scale as suggested by the reviewer but find it is not an intuitive way to see the relative contribution for each component. Besides, since the log scale has no zero value, stacked bar plots or stacked concentration plots should never be made on log scales. For a discussion, see e.g., http://www.graphpad.com/support/faqid/1477/. So we would like to keep the figure as it is.

From top panel in fig.10 it seems like there should be zero fuel BC at ATTO. Did you try to run the model without that source? Or track the source (geographically) of this spurious fuelBC at ATTO?

Yes, we tagged the BC from NSA fossil fuel and biofuel combustion in the model. We have modified the text: "The overestimate in the background is probably driven by **fossil fuel and biofuel combustion** BC in the model, mainly from NSA **with a mean concentration of 28 ng m^{-3} during the period.**"

Isn't this just because you have too much fuelBC reaching ATTO?

We added this in the text: "**The model bias in the BC background affects the intercept of the line but not the slope**. The difference between observed and simulated slopes could be due to variation in emission ratios from individual fires… "

Besides, aerosol lifetime depends on meteorological conditions and you have not shown any assessment of that. Does your model simulates the observed precipitation rates (in time and space)? What about wind direction and speed? Boundary layer height and TKE? Clouds, SW and LW ?

As stated above, we now explain better in Section 2 that the meteorological fields used in GEOS-Chem are from the GEOS-5 FP atmospheric data assimilation system: "We use the GEOS-Chem CTM version 9-02 (http://www.geos-chem.org/), **a global 3-D model of atmospheric composition** driven by assimilated meteorological data **GEOS-5 FP** from the NASA Global Modeling and Assimilation Office (GMAO). **GEOS-5 FP is the current operational meteorological data, which is now produced with version 5.13.0 of the GEOS-Data Assimilation system (Lucchesi, 2013)**." We acknowledge the source of GEOS-5 FP data in the Acknowledgments: "**The GEOS-5 FP data used in this study/project have been provided by the Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight Center**."

We also add the following sentence in the text to show the reliability of the BC lifetime simulated in the model: "The derived f_e is 7.7±0.76 and 8.3±0.75 ng m^{-3} ppb^{-1} for open fires in NSA and NAF, respectively. **The simulated global mean lifetime of tropospheric BC is 4.3 d, consistent with the range of 4.2-4.4 d that was derived from model simulations constrained by the ensemble of observations (Wang et al., 2014a; Wang et al., 2014b). BC from different sources also has different lifetimes, ranging from 3 d to 6 d depending on the surrounding**

meteorological conditions after it is emitted to the atmosphere."

You can't make that conclusion. Looking at your AAOD in Fig.14 you have similar values over the Amazon as over the NE of Brazil, from Ceara down to Bahia (highly urbanized coast and lots of antropogenic emissions). What you can concluded, hence, is that your emissions don't account correctly for urban emissions in SA except for SP (which shows up as a red spot at 47W 23S).

As mentioned above, we modify the text in Section 2.2 to show that these emissions are included in the model: "**We note that the Bond et al. (2007) emission inventory accounts for urban emissions in NSA (e.g., Ceará and Bahia in the northeastern coast of Brazil and São Paulo) as seen from the distribution of BC emissions in Fig. 1. We also compared the BC fossil fuel and biofuel emissions from Bond et al. (2007) with those from the HTAP emission inventory (http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123), and found that the two have very similar distributions, but that the emissions from Bond et al. (2007) are twice as high as those from HATP over NSA.**" We also modified the color scale in Fig. 14 to show the influence of the emissions over the northeastern coast of Brazil and explain better in the text: "During the wet season, AAOD over **the central Amazon including the ATTO site is generally lower than 0.0015, reflecting a minimal influence of human activities over this region. High AAOD (up to 0.01) over Colombia is caused by open fires, whereas over Southern Brazil it originates from fossil fuel and biofuel combustion near São Paulo. Fossil fuel and biofuel emissions over the northeastern coast of Brazil, although not as high as those from São Paulo, also result in slightly enhanced AAOD (0.0015-0.003).**"

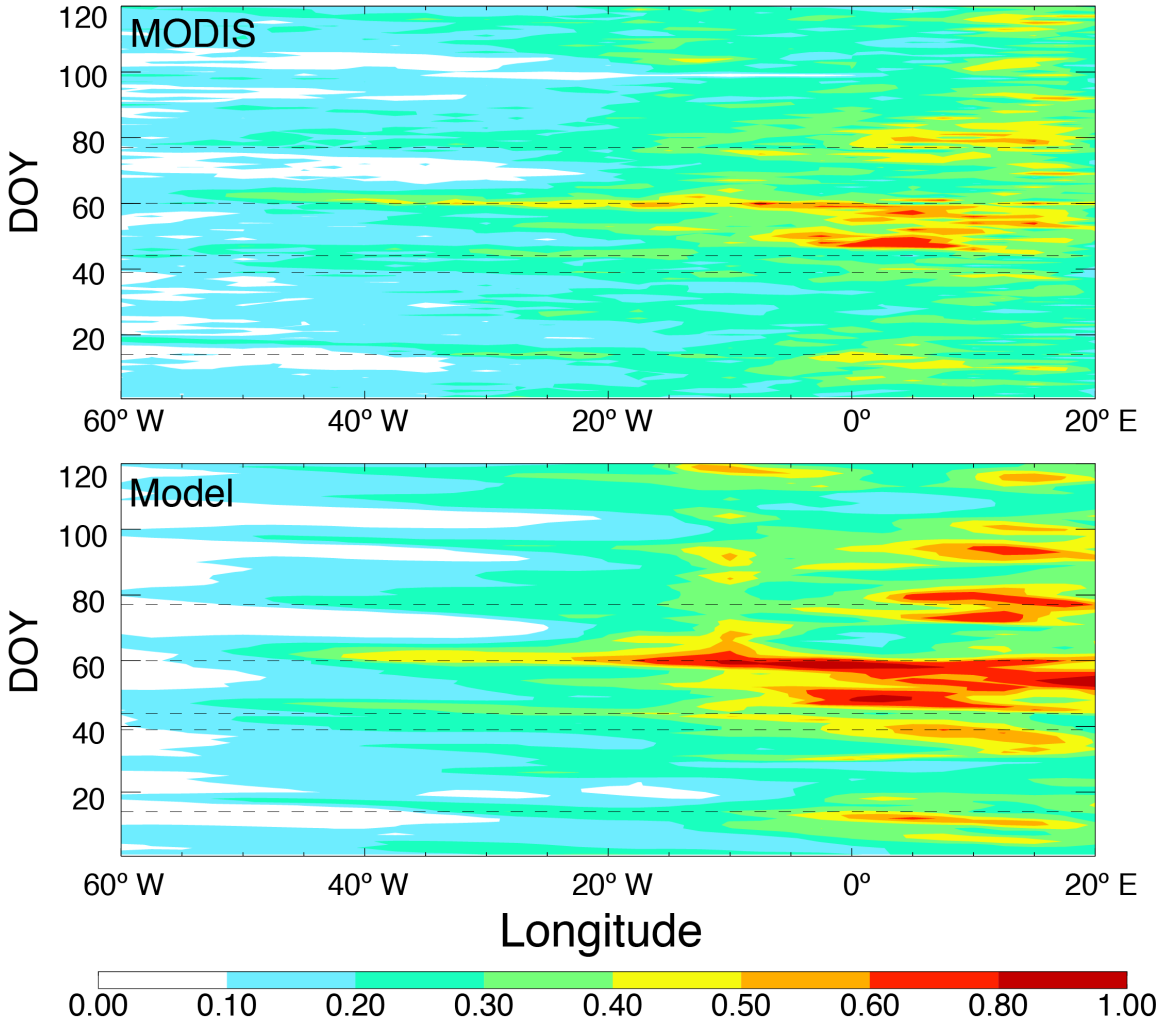

Figure 6: Daily distribution of latitudinally averaged AOD at 550 nm from MODIS and model over the rectangle region between 60° W–20° E and 5° S–25° N from Jan-Apr 2014. The Y-axis is the day of year (DOY), starting with day 1 on 1 January 2014. Dashed lines indicate the starting dates of five trans-Atlantic transport events from the east of the Atlantic (20° W).

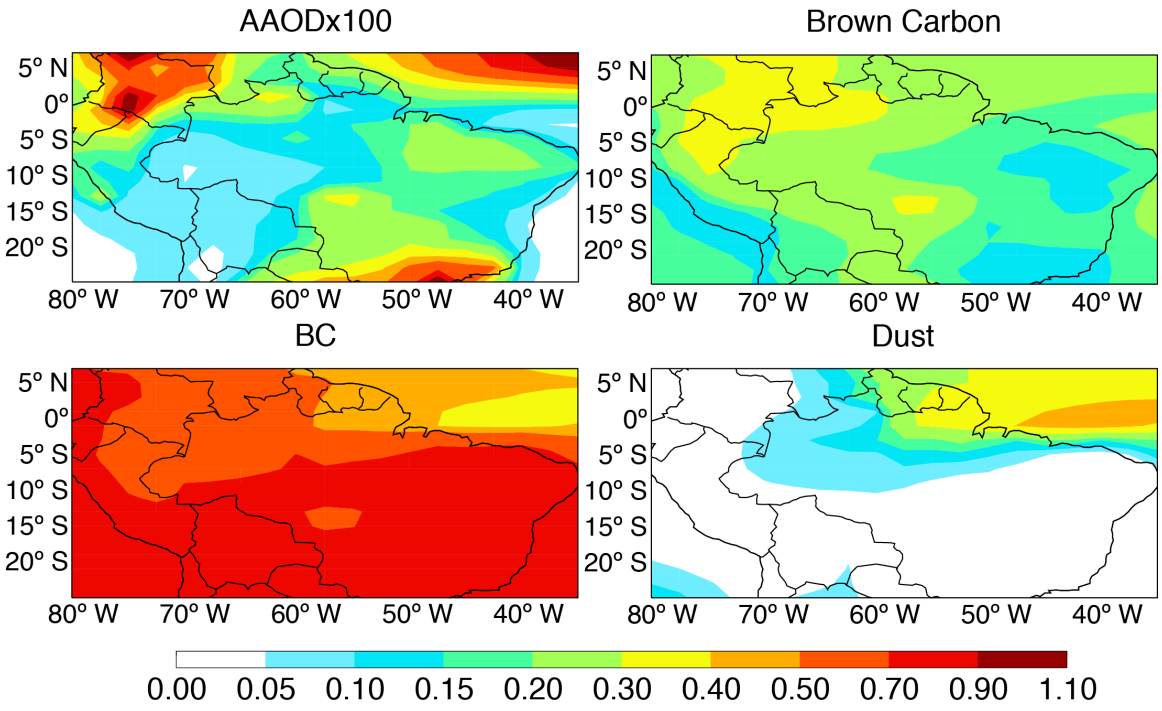

Figure 14: Model AAOD at 550 nm (left top), and the contribution to total AAOD from BC, brown carbon and dust over the Amazon basin averaged over Jan-Apr 2014.