Response to Comments of Referee#2

Dear Reviewer:

We would like to thank you for the valuable and constructive comments/suggestions which helped us to improve our manuscript. We have carefully revised the manuscript accordingly. Please find our point-to-point responses below (line numbers and figure numbers refer to the new version of manuscript; reviewer comments and suggestions are in italics, responses are in plain font; revised sections in the manuscript text in response to the comments are marked with red color).

1. To assess ARI, why not contrasting the experiment PC3_EMISX and PCNR3_EMISX? The current way to obtain ARI has an underlying assumption that that the total aerosol effects are a linear combination of ACI and ARI, which may not be the case because of the complexity of the nonlinear microphysics-dynamics-thermodynamics interactions of the system. Such an uncertainty should be discussed in the paper.

Response: We thank the reviewer for this insightful comment. This paper focuses on assessing the ARI of biomass burning aerosols (BBA), but the PC3_EMISX and PCNR3_EMISX include aerosols from biomass burning origin and other sources such as anthropogenic. Contrasting the experiment PC3_EMISX and PCNR3_EMISX results in the ARI of all aerosols (biomass burning plus other sources). In the studied domain, the contribution of non-BBA to the bulk aerosol optical property, although not dominant, is noticeable, e.g. the black carbon emission rate in the EMIS1 scenario is 1.8 mg m^{-2} s^{-1} for biomass burning emissions and 0.4 mg m^{-2} s^{-1} for anthropogenic emissions, and the non-BBA proportion is even accentuated in the EMIS0.5 case. The method used in this study to assess the ARI of BBA refers to the same method used in Archer-Nicholls et al. (2016) for separating BBA's indirect effect, i.e. ACI in this study, and radiative effect (direct+semi-direct), i.e. ARI in this study. This method assumed a closure relationship between the total aerosol effect and individual effects (ARI and ACI) and calculated the ARI as all the BBA-induced perturbations except those induced by the ACI pathway. This assumption was also found conventionally applied in assessing the radiative forcing of specific aerosols by indirect, direct and semi-direct effects separately and jointly (Ghan et al., 2012). By this method, the ARI of BBA can be obtained without the influence from non-BBA since the ARI from non-BBA (CC3-CCNR3) was deducted from the ARI of all aerosols (PC3_EMISX-PCNR3_EMISX).

On the other hand, the authors acknowledge the reviewer's concern that the nonlinear nature of the cloud system may make ARI assessed in the present way different from the results by contrasting the simulations with and without BBA radiative feedback. We calculated the difference between these two definitions of ARI, based on the EMIS6 scenario, since the non-BBA proportion could be neglected at high biomass burning emission intensity (the black carbon emission rate in the EMIS6 scenario is 10.8 mg m^{-2} s^{-1} for biomass burning emissions and 0.4 mg m^{-2} s^{-1} for anthropogenic emissions). The results (Table S1) show that the difference between these two definitions of ARI is small (within the range of standard error) and does not influence the conclusions about the relative importance of ACI and ARI in this study.

The discussion about this uncertainty has been added (Page 7, Line 199):

'Due to the nonlinear nature of the cloud system, which involves complicated microphysics-dynamics-thermodynamics feedbacks (Stevens and Feingold, 2009), the ARI effect calculated as the residual component of the aerosol total effect aside from the ACI part may be different from directly contrasting the simulations with and without the radiative effect from BB aerosols. To assess this uncertainty, we compared the ARI effect on clouds obtained here with its counterpart, i.e., the difference between PC3_EMIX and PCNR3_EMISX, which directly computes the effect associated with aerosol-radiation interactions from all aerosols, based on the EMIS6 scenario to minimize the influence of aerosols not from BB (Table S1). It shows that the uncertainty in the ARI quantification associated with the cloud nonlinear microphysics-dynamics-thermodynamics feedbacks is very small and would not have a significant influence on the ARI assessment in this study.'

Table S1. Monthly mean perturbations caused by the ARI of BB aerosols for the EMIS6 emission scenario.

	ARI in this study	PC3_EMISX - PCNR3_EMISX	difference
LWP (g m^{-2})	–3.8	–3.9	–0.1 (3%)
IWP (g m^{-2})	0.26	0.24	–0.02 (8%)

2. It is unclear how the model treats the BC aging process. According to the present model description in Section 2.1, it seems the fresh BC are immediately mixed with other types of aerosols after emission. Such a simplified treatment could result in overestimation of the BC absorption and associated radiative forcing [Wang et al., 2018; Peng et al., 2016].

Response: Thanks for pointing out this issue. For calculating the aerosol optical properties, the model uses the Maxwell-Garnett mixing rule, which treats the BC as small particles distributed randomly within a mixture of the other chemical components. The BC aging process has not yet been implemented in the WRF-Chem available to the community. We have added a clarification about the treatment of BC aging in the model:

'Note that the process of BC aging (Peng et al., 2016; Wang et al., 2018) has not been implemented in the model. In the future, it would be desirable to implement BC aging (Peng et al., 2016; Wang et al., 2018) in order to more accurately simulate the mixing state of BC-containing aerosols.'

The immediate mixing of BC with other aerosols after emission did not cause obvious overestimation of BC absorption in the studied domain, as shown from the comparable single scattering albedo (SSA) between the model output and the observation (Table. S3). This could have benefited from the improved mixing rule used here, because the Maxwell-Garnett mixing rule was proven to overcome the unrealistic absorption enhancement of BC by the direct internal mixing to some extent and was found to provide reasonable BC absorption (Bond and Bergstrom, 2006). Besides, the fact that the studied domain is away from the intensive biomass burning source and is impacted by the fire plumes transported there hours after being emitted could also lower the influence of BC aging on the studied domain. The evaluation of simulated SSA and AOD (Table S3) shows that the model can generally capture the aerosol optical features in this region, and therefore is reliable for estimating the aerosol radiative effect.

3. According to Fig. 6, the month-long simulations include a couple of deep convective systems with heavy precipitation (Sept. 9, 17-18). For the precipitation response analyses in Fig. 15, can the authors take a further step to assess the deep convective systems and the rest separately? Maybe a threshold of 3 mm/3hr can be applied to categorize those cases.

Response: Thanks for the insightful suggestion. Accordingly, we have separated the precipitation responses for deep convective systems and the rest as the reviewer suggested and added corresponding figures and discussion to the revised manuscript.

'To examine the precipitation responses at different precipitation intensities (Fig. 10), a threshold of daily maximum 3-hour accumulated precipitation exceeding 3 mm, the upper boundary of the domain averaged amount (Fig. S6), is used to distinguish the intensive precipitation grids from the light precipitation ones. High convective strength indicated by larger CAPE (Fig. 10) corresponds to intensive precipitation, whereas relatively weaker convection is associated with the light precipitation regime. Intensive precipitation shows a significant nonlinear ARI response, whereas light precipitation tends to be reduced monotonically by the ARI. The precipitation reduction by ACI at low aerosol concentration is less prominent in heavy than in light rainfall, due possibly to the dynamic feedbacks in deep convection (Rosenfeld et al., 2008). By contrast, a stronger ACI effect at larger aerosol amounts is shown in heavy precipitation as a result of the larger potential for CCN activation in strong convection (Reutter et al., 2009). The dependence of precipitation change on aerosol concentration is greater for the intensive precipitation than the light precipitation regime, given that the precipitation responses at the EMIS1 and EMIS6 scenarios are –1% and –27 % respectively for the intensive regime and –5% and –17% respectively for the light precipitation. This is consistent with the rainfall sensitivity to increasing aerosol concentration for strong and weak convection in Chang et al. (2015). The dominance role of ARI over ACI at high aerosol loadings is found at both regimes.'

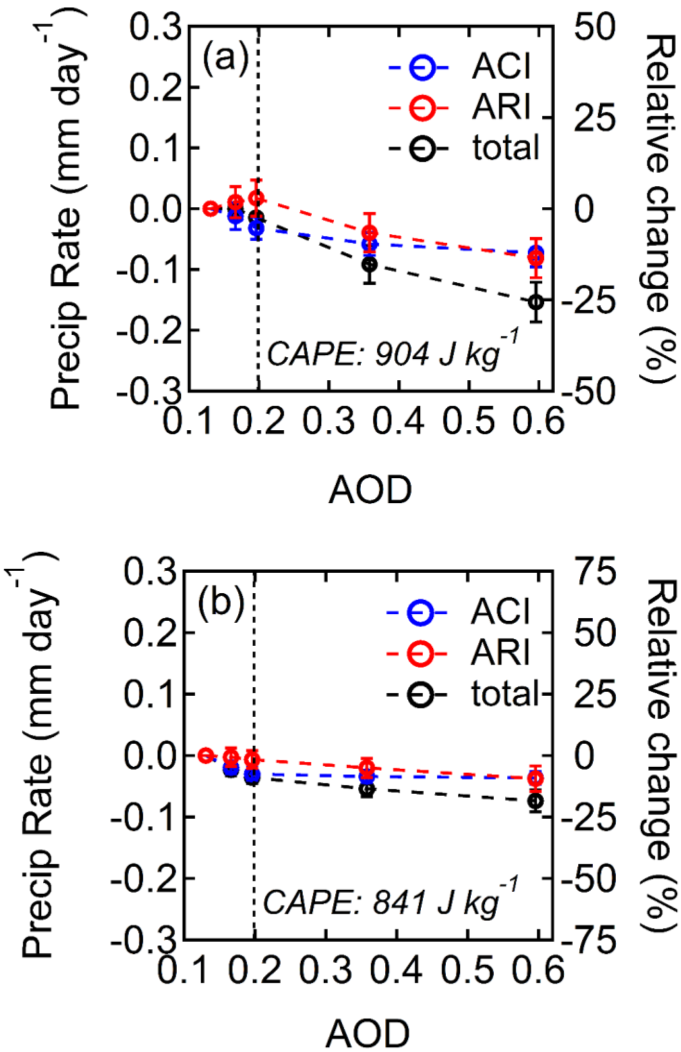

Figure 10. Changes in domain-averaged precipitation rate with increasing BB emission intensity (indicated by domain-averaged AOD in each emission scenario) at intensive precipitation regime (a) and light precipitation regime (b). The vertical dotted line in each plot indicates the EMIS1 scenario. Error bars denote the standard error.

4. For the IWP evaluation, how are the satellite data are averaged spatially? It seems the satellite observations shown in Fig. 5 are averaged over cloud points only. I doubt ice heterogeneous nucleation scheme can explain such a huge discrepancy. Even if the ice production scheme is not a function of INP concentration in this microphysics, it should still exist (most of time as function of temperature).

Response: Thanks for drawing attention to this point. The satellite datasets do have missing data points over the domain due to a combination of both unrecognized cloud ice signals and satellite technical problems such as the orbital gap (Remer et al., 2005). We calculated the countable proportion (the ratio of days when the ice water path data is not missing to the whole number of days in the study period) of each grid cell in the domain throughout the whole month (Figure R1). The countable proportion is approximately 0.5 with 6 out of 30 days having full data coverage. This can basically represent the magnitude of the IWP for the studied period and region. However, as the reviewer correctly points out, the elimination of the unrecognized weak ice signals would bias the observation results towards higher values and thus contribute to the discrepancy between the model and observation.

The ice production terms of the microphysical scheme used in the study include 1) homogeneous nucleation which occurs below -40°C and 2) depositional growth which is a function of temperature in the temperature range from -40°C to 0°C. With such an ice production parameterization, an underestimation of ice water content was found in Baro et al. (2018) by up to 80% and in this study by a similar magnitude (though the data quality contributes to some extent). A recent study by Su et al. (2018) found that introducing the ice nuclei source from dust particles into the microphysical scheme can improve the simulated ice water content by 15%. Analogous to dust particles, the biomass burning aerosols accompanied by biological material, soil dust, or ash particles was identified to efficiently improve the ice heterogeneous formation during the dry season (Seifert, P., et al., 2015). Based on these results, the missing parameterization of heterogeneous ice nucleation was listed in this study as one of the possible reasons for the IWP underestimation in the model.

We have reworded the discussion of the cloud ice comparison (Line 105 in SI). 'The uncertainties inherent in the satellite dataset, e.g., eliminating data points with unrecognized cloud ice, would bias the observation results towards higher values and thus to some extent account for the discrepancy between model and observation. Besides, uncertainties associated with the ice-phase microphysical processes, e.g., the lack of IN parameterization, may also be a potential reason for this discrepancy (Su et al., 2018).'

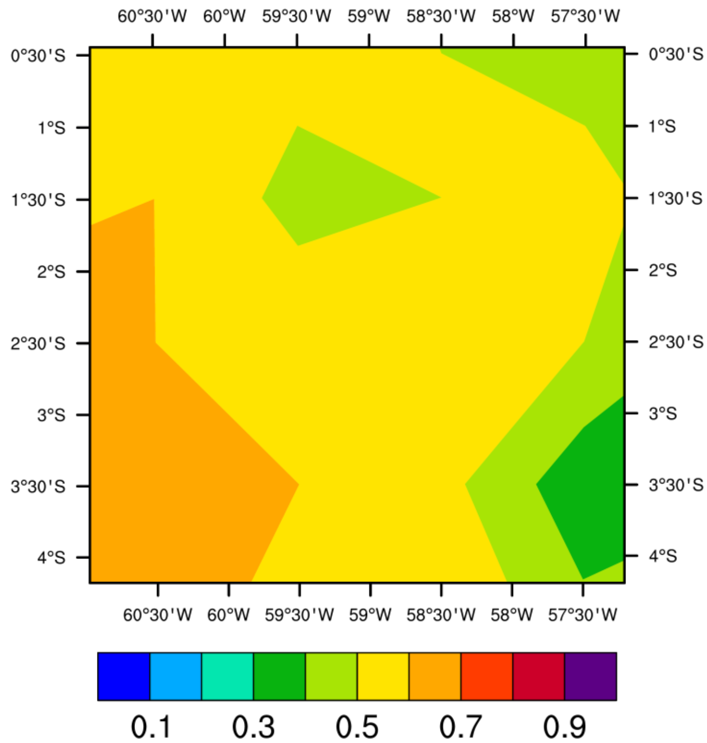

Figure R1. Fraction of countable data at each pixel throughout September 2014.

5. Line 62-65, similarly, a recent study using satellite data shows nonlinear response of deep convective clouds to smoke aerosol in South America [Jiang et al., 2017].

Response: Thanks for recommending the reference. This reference has been added.

6. Out of personal curiosity, to what extent the FDDA can reduce the meteorological biases? If the authors have the model free run available, I like to see a comparison of those two.

Response: In this study, FDDA was used in the outer domains to provide a more accurate meteorological boundary for domain 3. The simulated surface air temperature, relative humidity, and wind speed from the simulation of domain 2 with and without FDDA are compared against the observations at the ATTO site (Fig. R2). A notable improvement can be found in the run with FDDA compared to the free run.

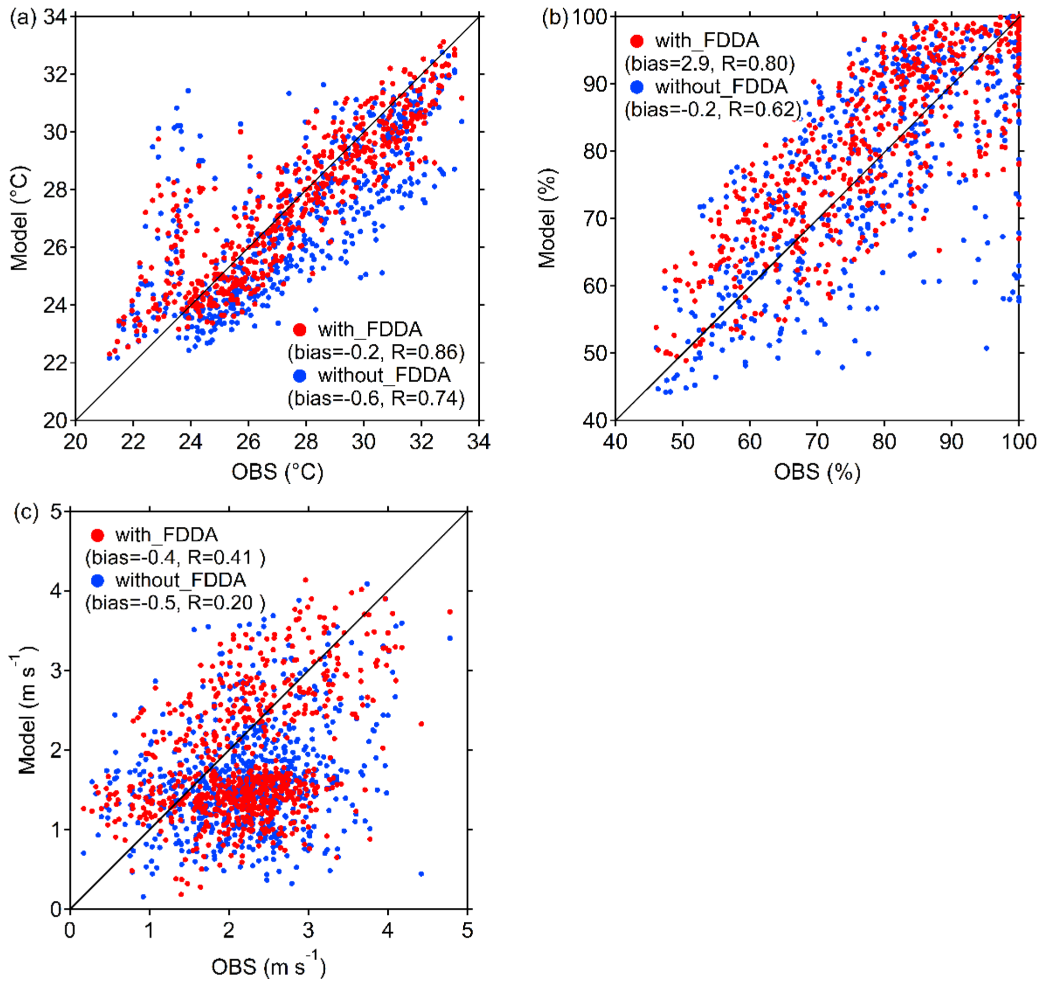

Figure R2. Scatter plot of surface air temperature (a), relative humidity (b) and wind speed (c) from observation and model simulation.

7. Fig. 4, what are the two dash lines in addition to the 1-to-1 line?

Response: The two dashed lines are 1:4 and 4:1 lines. We have clarified this in the caption.

'The dashed lines are 1:4, 1:1, 4:1 from top to bottom, respectively.'

8. Line 326-327, it doesn't make much sense to compare a regional aerosol forcing to the global values.

Response: Thanks, we agree. This comparison has been removed.

9. Fig. 11 is discussed after Figs. 12 and 13. Better to reverse their order.

Response: Accepted. The order has been reversed as the reviewer suggested.

10. In Fig. 11 and 12, the larger updraft velocity and IWP by absorbing aerosols corroborate the thermodynamic invigoration hypothesis by Wang et al. [2013] which suggested larger CAPE above PBL due to the presence of absorbing aerosols in the lower troposphere.

Response: Thanks for pointing out this connection. We have added this discussion at Line 329.

'This positive response of cloud ice and updraft velocity to ARI corresponds to the thermodynamic invigoration mechanism proposed in Wang et al. (2013) which suggested larger convective available potential energy (CAPE) above PBL could be induced by the absorbing aerosols in the lower troposphere.'

*11. Title is too long. Maybe remove "dependence of aerosol-*cloud and aerosol-radiation *interactions on aerosol loading".*

Response: We appreciate the reviewer's suggestion. We intended to use the subtitle to highlight the dependence mechanism studied in the paper and think it would help the readers to catch the key points effectively. We have shortened the title to 'Impact of biomass burning aerosols on radiation, clouds, and precipitation over the Amazon: relative importance of aerosol-cloud and aerosol-radiation interactions'

Reference

Remer, L. A., Kaufman, Y. J., Tanre, D., Mattoo, S., Chu, D. A., Martins, J. V., Li, R. R., Ichoku, C., Levy, R. C., Kleidman, R. G., Eck, T. F., Vermote, E., and Holben, B. N.: The MODIS aerosol algorithm, products, and validation, J. Atmos. Sci., 62, 947–973, 2005.

Seifert, P., Kunz, C., Baars, H., Ansmann, A., Bühl, J., Senf, F., Engelmann, R., Althausen, D., and Artaxo, P.: Seasonal variability of heterogeneous ice formation in stratiform clouds over the Amazon Basin, Geophysical Research Letters, 42, 5587–5593, https://doi.org/10.1002/2015GL064068, 2015.