Response to Interactive comment on "Aerosol-radiation-cloud interactions in a regional coupled model: the effects of convective parameterisation and resolution" by S. Archer-Nicholls et al. Anonymous Referee #1

While the paper does bring up some valid points regarding how to interpret aerosol-radiation-cloud interactions predicted by models such as WRF-Chem, I am not sure what new information is obtained from this model sensitivity exercise. Not enough context is presented regarding the present results and those published previously. Therefore, I do not think the up-front purpose and conclusions derived from this study have not been articulated well enough.

We thank the reviewer for their insightful and helpful comments. We appreciate that the reviewer agrees that a number of the findings of our paper are useful to the community. We agree that the study could be improved though, and have, as detailed below, included extra diagnostics which should provide useful new information and techniques/tools. We have also expanded on the context in which this study sits. We hope these changes will satisfy the referee's concerns about what was lacking in this study.

#### General comments:

Discussion of uncertainties: The authors are correct to point out missing aerosolradiation-cloud processes in models, such as WRF-Chem, and the dilemma of handling aerosol-radiation-cloud interactions using a nesting approach when convective parameterisations are needed on coarser-scale domains while they can be neglected on finer-scale domains. In other words, scale-dependency issues. However, there are other limitations in their approach that warrant more discussion and the uncertainties associated with those could have an impact on the findings from the sensitivity simulations. Some of the processes are briefly acknowledged, like SOA and aerosols in ice-phase clouds, while other processes are not mentioned, such as secondary activation. In general, I think a discussion section (or more discussion in the existing text structure) is needed to place the present results in the context when specific processes are missing or uncertain. Additional experiments could explore the impact of those uncertainties on aerosol-radiation-cloud interactions. For example since SOA is not simulated, biomass burning emissions could be increased or decreased to examine how changing aerosol mass impacts the metrics presented in the figures. I note that once aerosol concentrations get large enough, there are not likely to be further impacts on clouds, but I would expect the largest changes happening in transition regions with low pristine aerosol concentrations and high aerosol concentrations associated with smoke.

# The authors thank the review for highlighting these important points and acknowledge that there are many uncertainties inherent in the current study, and

perhaps more discussion of these is necessary. We believe that the necessary discussion has been added in response to the specific comments below, and in response to the other reviewer's comments. The reviewer raises an interesting point – that we are likely seeing limited aerosol impact on cloud due to the region being largely saturated in aerosol, and that we may see greater impact in transition regions. However, that is the nature of Amazonian troposphere during the dry season, and investigating aerosol-cloud interactions in transition regions is outside of the scope of this study.

Observational evaluation perspective: The authors need to stress that this is a model sensitivity exercise. No observations are presented to support the likelihood that the simulated aerosol-radiation-cloud interactions are realistic or not. The authors use the SAMBBA field campaign period; however, the present modeling study could have been done for any period in the Amazon or elsewhere where biomass burning is important. I understand they are leveraging a previous modeling study, but I have to review the paper as it stands by itself. I have several specific comments below along these lines.

As the reviewer correctly points out, this paper solely presents a modeling study. The scope of the study was not made clear enough from the opening of the original text and we have made changes to the abstract and introduction to make explicitly clear this study relates solely to modeling.

## Specific comments:

Page 27450, lines 18-19: The phrase "The 1 km domain simulated clouds less horizontally spread" is awkward and needs to be revised.

## Changed to:

*"Convective cells within the 1km domain are typically smaller but more energetic than equivalent cells in the 5km domain, ..."* 

Page 27450, line 26: Change to "the publically available version of WRF-Chem" or "the version of WRF-Chem distributed to the community". As indicated by the authors later, there are efforts underway that do include these effects, but are not yet readily available.

Changed to "the version of WRF-Chem distributed to the community"

Page 27451, lines 16-17: Technically it is only the absorption that is included in the semi-direct effect (https://www.ipcc.ch/publications\_and\_data/ar4/wg1/en/ch7s7-5- 2.html), and not scattering

Acknowledged; "and scattering" has been removed from the text and a

citation to IPCC working group 1 added.

Page 27453, line 8: same comment about wording of WRF-Chem as comment on page 27450, line 26.

Changed to "the publically available version of WRF-Chem".

Page 27454, line 1: The authors state that the purpose of the paper is to "evaluate" how aerosol-radiation-cloud interactions are captured in WRF-Chem. To me "evaluate" means comparison with observed quantities, which are not presented in this study. I think a better word is "illustrate", since this is a model sensitivity study only. While the study may be illustrative for WRF-Chem users, it does not provide any quantitative information on performance. This needs to be made clear.

The authors apologise for the confusion resulting from the use of the term "evaluate". We used it to mean critically assess the behavior of the model, not necessarily against measurements. However, as both reviewers have cited issue with this term, it has been changed to "investigate" accordingly and more effort has been made to emphasize that this is a modeling study in the abstract and introduction.

Page 27454, line 5: I think "cumulus parameterisations" needs to be changed to something about "with and without the use of a cumulus parameterisation". I got the impression that multiple cumulus parmeterisations were to be tested, but instead found out later that was not the case and the investigators simply turned on and off a single cumulus parameterisation.

Changed to:

"... with and without the use of a convective parameterization and at 5 and 1km horizontal resolution..."

Page 27454, line 7: I agree this is a true statement, but the authors can make this statement much stronger. Knowing the details on how feedbacks are handled is important for ALL models, including climate models. The number and type of feedbacks various from model to model, making comparisons between models problematic. Also, some aspects of aerosol-cloud interactions are highly uncertain and poorly constrained by data (i.e. heterogenous ice nucleation). Therefore, I think a little more discussion is needed here to justify this aspect of the paper.

Changed to:

"Knowledge about how these processes interact with, and feedback to, each other and the general model setup, is important for determining the best manner in which to run models such as WRF-Chem. The manner in which these processes, and the feedbacks between them, are setup and coded varies between different limited area coupled models or global climate models. This kind of detailed analysis therefore has to be done for each model (rather than assuming that certain interactions between processes will all behave in the same manner in every model). This study is intended to show how these processes interact within WRF-Chem and provide impetus for further developments to improve the realism of these simulations, as well as consistency through the different model scales."

Page 27454, line 19: I think "significant improvement" is an overstatement of the results from that paper. The authors of that paper do note "some improvement", but it is really difficult to see in their figure that modest improvement.

The term "significant" is used in the passage in question from the paper. However, the authors do agree that it is a small change. Text changed to "modest improvement" accordingly.

Page 27457, lines 7-8: The authors note that no SOA treatment is used in this study and then provide a few sentences noting the uncertainties in parameterising SOA. It is true that SOA is still uncertain in models; however, I do not agree that the present model is capable to represent OA mass. If I understand correctly, all OA in their simulation originates from POM emissions, anthropogenic and biomass burning. I'm assuming biomass burning dominates in this region. But I would expect that OA mass is dominated by biogenic SOA, in the absence of biomass burning? There is debate in the literature on this subject, with some models including a SOA from biomass burning (e.g. Shrivastava et al., JGR 2015). If there were comparisons of observed and simulated OA in the Archer-Nicholls (2015) paper, some discussion of that is warranted in the paper. Is the model too high or too low in simulated OA? OA will be the largest fraction of aerosol mass, and thus influence CCN. So SOA is a critical point in these simulations when assessing cloud-aerosol interactions.

In Archer-Nicholls et al., (2015), we do show that the model produces enough OA in the simulations, although this has been achieved by scaling of the base fire emissions. The greater difficulty we have found is in representing the vertical structure of the aerosol layer. We add the following passage summarising the findings from the previous paper at this point in the text:

"Model aerosol fields from the parent 25km domain were evaluated against insitu flight measurements in Archer-Nicholls et al., (2015). Net mass of POM and  $PM_{2.5}$  was of similar magnitude to that measured by flights on 14 and 18 September. Note that sufficient aerosol mass was achieved in part by scaling up emissions to match observed AOD from the MODIS satellite product in the region. However, due in part to issues relating to the plume-rise parameterisation, the vertical distribution had some errors, with a bias towards too much aerosol in the model between the boundary layer top and 4km above ground. On 23 September, the aerosol mass was overestimated in the model compared to flights, attributed to a combination of emission fields not decreasing commensurately with the transition into wet-season meteorological conditions and insufficient wet deposition of aerosol mass. Although there were some discrepancies in POM:BC ratio between model and observations, SSA compared well."

There is further discussion warranted on the influence of SOA. First, although in this region biogenic SOA is the dominant source of fine aerosol during the wet season, it is negligible relative to that from biomass burning in the dry season (Artaxo et al., 2013). More importantly, whether there is a significant contribution of SOA to OA mass from biomass burning is subject to intense debate in the literature (for example, the meta-analysis of Jolleys et al., 2012 shows no clear evidence for any SOA contribution along diluting and ageing BB plumes). In the absence of consensus relating to a SOA contribution and resulting total lack of quantitative mechanistic understanding, approximating the organic aerosol as primary emissions scaled to produce sufficient aerosol mass is completely reasonable if close to emission sources.

While Shrivistava and others have worked to implement a VBS scheme in WRFchem, at the time of the study this was still experimental, with many associated uncertainties and important aerosol processes (such as aerosol-radiation interactions) yet to be implemented. Running with a VBS scheme to investigate SOA processes over the region has formed a large part of follow up work for the current study.

Page 27459, line 27: As far as warm-cloud only processes, Yang et al. (JGR, 2015) describe a version that now includes ice-borne aerosol.

We thank the reviewer for pointing out this reference, which has been added to the text. However, these changes were not available to us at the time of the study so only warm-cloud processes could be reported.

Page 27460, Section 2. Wet removal is not described in any way. This is an important process that seems to warrant some discussion on how it is handled for the various simulations (25, 5, 1 km).

We acknowledge that this is an important process for modeling accurate aerosol loadings. We have added this line describing wet-removal in WRF-Chem:

"Wet removal is one of the main sinks of particulate mass. Wet scavenging of interstitial and activated aerosol, both in and below cloud, are parameterised following scavenging efficiencies described by Slinn (1984). Wet deposition of MOSAIC aerosol species is handled for explicitly resolved clouds, but not parameterized convective precipitation (although this has been implemented with the Kain-Fritsch parameterisation in later versions of WRF-Chem; Berg et al., 2015). Once aerosol particles are attached to hydrometeors, they are assumed to be immediately deposited out of the atmosphere, without possibility of re-suspension following evaporation (for more details see Yang et al., 2015)."

Page 27460, line 3: The Berg et al. paper is now published so the reference should be updated.

Reference updated.

Page 27461, line 7: It would be useful to include, somewhere in the manuscript, a short summery of the performance of the model in the paper cited here.

A summary of the findings of Archer-Nicholls et al., (2015) has been written above in response to comment on Page 27457, lines 7-8.

Page 27461, line 24: aer\_rad\_feedback=0 may be familiar to WRF-Chem users, but is not very useful for a wider audience. This could easily deleted.

Deleted as suggested.

Page 27462, line12: Would it be possible to include TRMM precipitation estimates over the domain for these periods? Or was precipitation evaluated in the previous paper?

Precipitation was evaluated in the previous paper. The general magnitude and form of storms were well simulated, although individual storms were often displaced. Some examples of comparisons between the model scenarios and TRMM precipitation are included in response to the other reviewer's comment on P. 27466, l. 22-26, specifically relating to how the structure of precipitation is represented. We found that for the 5km domain with convective parameterisation precipitation was spread over too wide an area, without the small cells of intense precipitation seen in the TRMM observations.

Page 27464, line 11: For the absorbing BBA, I assume the authors mean the BC emitted by the primary emissions rather than the OC. Does the model include a treatment of absorbing brown carbon? It would be useful to clarify this point in the model description section.

Yes, only the BC component of the aerosol is absorbing. Changed to: "Although the high BC content of BBA makes it highly absorbing, …"

Page 27464, lines 25-27: I assume the authors are talking about the model results here, but sometimes it is not clear whether they are talking about observed or simulated values. Here and elsewhere, it would be useful to include

"simulated" (or some other words) to let the reader know what they are talking about would be useful.

This sentence specifically refers to whether radiative effects of clouds are considered for the analysis (by using the all-sky radiation variables, see Appendix). Language has been changed here and elsewhere to make it clear we are referring to simulated values:

"When the radiative effects of cloud are considered for the analysis of model output, ..."

Page 27466, line 25: I don't understand the logic of connecting the Grell 3-D scheme and its ability to predict the semi-direct effect. The semi-direct effect would result from the radiation parameterisation. I think this must be a poorly worded sentence.

We acknowledge that this sentence is poorly worded. The point we are trying to convey is that, whilst the semi-direct effect obviously results from the radiation scheme, it is also highly dependent on the simulation of clouds within the model. If cloud representation is poor, due in part to the convective parameterization, then the model will have difficulty accurately simulating the semi-direct effect. The sentence has been reworded to:

"Assuming the representation of convective clouds is more realistic in the 1km domain, the difference between the two domains suggests that the Grell-3-D parameterisation, even with subsistence spreading, resolves clouds and their radiative properties too poorly for the accurate simulation of semi-direct effects."

Page 27467, line 1: Change the title of this section to "Sensitivity to a convective parameterisation". The authors are only looking at one parameterisation here, and their results will likely vary if other cumulus parameterisations are used.

Changed accordingly.

Page 27468, line 16: Secondary activation is likely to be important for deep convection (see Yang et al, JGR, 2015). The authors should discuss the implications of neglecting this process in the present simulations.

Although not the focus of the current study, we agree that secondary activation could have important consequences for the current simulations. However, there are other uncertainties related to the representation of in-cloud aerosol processes in deep-convective clouds which also bear consideration. The following paragraph has been added to discuss this point: "In deep convective clouds secondary activation of aerosol has been observed (e.g. Hetmsfield et al., 2009) and modeled (e.g. Segan et al., 2003, Yang et al., 2015), whereby further interstitial aerosol particles are activated above cloud base due to supersaturation not being fully offset by droplet growth, as hydrometeors are scavenged in the cloud column. This is a process unrepresented in the current model setup, as the Abdul Razzak and Ghan (2001 etc.) parameterisation assumes all activation at cloud base. If secondary activation were included in the model it would, primarily, act to increase the efficiency with which aerosol is scavenged from cloud and reduce the amount of aerosol transported to the mid/upper-troposphere (Yang et al., 2015). However, representing this process is challenging in this scale of model, without bin microphysics or fully-resolved updraft velocities. In future studies, we plan to use the aerosol-aware Kain-Fritsch parameterization (Berg et al., 2015) to enable this functionality in parameterized clouds."

Page 27469, line 19: This section is largely a "summary" section. There are very few conclusions. Either change the section name or re-write the text in this section.

A conclusions section is required by the Copernicus journal standards, so cannot be changed to "summary". Changes have been made to accentuate conclusions from the study, whilst removing unnecessary repetition. The new version of the conclusion has been included in its entirety here. Note this version assimilates suggestions and changes made to accommodate the second reviewer.

### Conclusions

WRF-Chem model simulations for three 36 hour case studies over nested domains at 5km and 1km grid spacing were conducted over a region of Brazil heavily influenced by biomass burning aerosol (BBA) to evaluate the regional impact of aerosol-radiation and aerosol- cloud interactions. These domains were one-way nested in a WRF-Chem simulation at 25km horizontal grid spacing over South America, which was run for September 2012 and evaluated by Archer-Nicholls et al. (2015) against in-situ aircraft measurements. The Grell-3-D convective parameterisation was used on the 5km domain, using the recommended subsistence spreading option for running at this scale (Grell and Freitas, 2014). Different scenarios were conducted to illustrate how aerosol-radiation-cloud interactions are modelled in WRF-Chem and test sensitivity to model resolution and use of convective parameterisation over the 5km domain. Due to the small size of domains, short case-studies, and single model version, the results from this study should be seen to apply to the specific case studies and model setup presented. *Caution should be used when extrapolating from the results of these case studies to* make more general conclusions about aerosol-cloud interactions (especially if applying these findings to other limited area or global climate models).

Over the 5km domain, on the 18 September case study, the shortwave direct effects of BBA particles over the region have a negative forcing of  $-3.34\pm2.68$  (standard error= $\pm0.043$ )Wm<sup>-2</sup>, which is largely cancelled out by a positive semi-direct effect

of 6.06±14.29(0.13)Wm<sup>-2</sup>. The shortwave indirect effect is a relatively small 0.266±9.47(0.079) Wm<sup>-2</sup>. In the 1km domain, deep convective clouds consisted of smaller cells, covering less of the total domain compared to the same region of the 5km domain. Longwave semi- and indirect effects are significant, with values of - 4.54±7.60(0.20)Wm<sup>-2</sup> and -1.53±4.52(0.10) Wm<sup>-2</sup> respectively. These are largely a result of decreases in nighttime cirrus clouds in the runs with BBA. Overall, there is a net negative forcing of -3.08±1.24(0.177) Wm<sup>-2</sup>. Note that due to the small size of the domain and short runs, these results only apply to the current model and case studies and caution should be taken before generalizing the findings.

The semi-direct effect was thus much lower in the 1km domain compared with the same region of the 5km domain, showing it is highly sensitive to the model resolution. Indirect effects from resolved aerosol-cloud interactions in the 1km domain were smaller than the semi-direct effect, although the small size of the 1km domain and sensitivity to boundary conditions from the 5km domain results in a noisy signal.

Simulations run without a convective parameterisation on the 5km domain had reduced daytime convection and precipitation. Comparisons with the TRMM dataset suggest that the 5km simulation without convective parameterisation simulates the structure of convective systems better, as more localized cells rather than an areal spread of precipitation, and a total precipitation rate closer to that measured in the TRMM dataset over the region for this testcase. The semi-direct effect is lower in the scenarios without convective parameterization due to the clouds being more cellular and there being reduced cirrus cloud cover over night. The net forcing from the scenarios with no convective parameterization on the 18 Septmeber is  $0.327 \pm 1.28(0.127)Wm^{-2}$ , a largely neutral result compared to the negative forcing from the scenario with convective parameterization. Although this result is unlikely to be physically realistic, the large magnitude of the sensitivity highlights the uncertainties with simulating aerosol-radiation-cloud interactions in this regime. WRF-Chem (at the time of study) neglects fractional cloud cover within grid cells (Zhang, 2008), which may be causing an overestimation of a semidirect effect over the 5km domain. A better representation of fractional cloud cover, linked with the radiation and convective parameterisations, is needed to estimate this forcing at the regional scale.

The BBA CCN efficiently activate in the model, as shown by an increase in droplet number and decrease in maximum supersaturation in clouds. With the exception of an enhanced fog formation event on the morning of 23 September, aerosol-cloud interactions did not cause a noticeable change to the radiative balance. More CCN are activated in deep convective clouds in runs with fire emissions and convective parameterisation on, but without resolving the high in-cloud updraft velocities or secondary activation, the physical significance of the modelled droplet number and grid-scale cloud properties of parameterised cloud is questionable. The runs with explicitly resolved convection at 1km and no cumulus parameterisation at 5km also showed minimal indirect effects, likely due to the deep convective clouds being optically thick and therefor their radiative properties are not sensitive to increased droplet number. The model does not produce an aerosol "cloud-invigoration" effect (as seen by Rosenfeld et al. 2008 and Fan et al., 2013), although this may be because aerosol-ice nucleation processes (not included in the version of WRF-Chem used here) are required to reproduce this effect. Overall, these findings suggest that resolving indirect processes in parameterized cloud is of secondary importance for the current case studies. Instead, representation of semi-direct aerosol feedbacks has a greater impact on the net radiative balance and associated uncertainties.

Simulating convective systems while including the effects of aerosol, particularly at horizontal grid spacings of less than 10 km, is a challenging task and work is being conducted to develop new parameterisations for this purpose (e.g. Grell and Freitas, 2014; Berg et al., 2015). More coordination between parameterized and explicit treatments of aerosol, cloud and radiation interactions is needed in order to make modeling of these processes at the transition between fully parameterized and fully explicit schemes more coherent. To constrain the simulation of these interactions, the latest in-situ and remote observations of aerosol interactions in deep-convective clouds need to be considered. Without a consistent methodology for simulating aerosol-radiation-cloud interactions across scales, it is impossible to be sure how much of an impact the aerosol should be having on cloud properties and lifetime."

Page 27471, line 17: As far as convective invigoration, I suggest the authors read Fan et al., PNAS, 2013. I believe that paper had a similar conclusion; however, they found that the most important part was that aerosols lead to a larger and longer lasting anvil. So, I am wondering if the authors could look at their results to determine whether simulations with and without fires changed cirrus amount detrained from convection. As the authors speculate, the current model formulation may not be complete. The PNAS paper also used spectral-bin microphysics that may behave differently than two- moment schemes, in terms of cloud-aerosol interactions.

From our study, we see the opposite effect – namely that there is little invigoration of cirrus cloud from BBA. The more dominant factor is the greater amount of convection in the simulations without BBA increase the level of cirrus clouds, presumably from outflow of anvils from deep convection. This is highlighted by the small and inconsistent changes to LW indirect forcings in the case study (see extra figures in reply to reviewer #2). Its important to note that there are substantially fewer nighttime cirrus clouds in the runs with no convective parameterization. On reflection, the content of this text has been left the same but with the Fan et al., 2013 reference added.

Page 27472, lines 9-14: While I don't disagree with these statements, what is really missing here are means to evaluate whether parameterisations for cloudaerosol interactions in deep convection are producing the right results for the right reasons. In other words, some observational and theoretical work is needed as well. Parameterisation development needs to be constrained by observations. Shallow cloud systems are far simpler and it has been easier to have confidence in how aerosol-cloud interactions are treated in those systems and in situ measurements of aerosols, cloud droplet number, etc. can be made within clouds. Such sampling is more problematic for deep convection.

The authors acknowledge that observations will absolutely be required to constrain future convective parameterisation development. Changed to:

"More coordinated development of convective parameterisations with aerosol and radiation mechanisms is needed. To constrain the simulation of these interactions, the latest in-situ and remote observations of aerosol interactions in deep-convective clouds need to be considered. Without a consistent methodology ... "

Figure 2: Add the date and time at the top of each panel.

Changed accordingly.

Figure 3: The first phrase is awkward, change the first phrase to "Temporally averaged column AOD at 550 nm over the 5 km domain". Add date and time at the top of each panel.

Changed accordingly.

Figure 4: Add the date and time at the top of a) – c).

Changed accordingly.

### References

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