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 aerosol-radiation-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 reviewer 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. Are the authors assuming not much SOA is produced by 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 in Section 3.3 (now called meteorological and aerosol fields):

"Model aerosol fields from the parent 25km domain were evaluated against in-situ 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 WRF-chem, 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. We have added two figures 9 and 10 in the revised paper, as well as discussion in section 4.3 on this topic.

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 at the end of section 2.2 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 1 km horizontal 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 nested domains were driven by model fields from a WRF-Chem simulation at 25km grid spacing over South America, which was run for September 2012 and evaluated by Archer-Nicholls et al. (2015) against in-situ aircraft mea- surements. The Grell-3-D convective parameterisation was used on the 5 km domain, using the recommended subsistence spreading option for running at this scale (Grell and Freitas, 2014). Different scenarios were conducted to probe how effectively the impacts are mod- elled in WRF-Chem and test sensitivity to model resolution and use of convective parame-terisation over the 5 km domain. As a result of the small size of domains, short case-studies, and single model version, the results from this study 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 ± 1.47 Wm^{-2}, which is *countered by a positive semi-direct effect of 6.06±1.46 Wm^{-2}. The shortwave indirect effect is a relatively small 0.266±1.06 Wm^{-2}. Longwave semi- and indirect effects are larger on this case study day, with values of -4.54 ± 0.96 Wm^{-2} and -1.53 ± 0.69 Wm^{-2} 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 -2.67 ± 1.27 Wm^{-2}.*

Further nested simulations at 1km grid spacing were run to explicitly resolve convection. In the finer resolution domain, deep convective clouds have much reduced horizontal spread but higher cloud droplet number within cloud compared to the 5 km domain. The reduction in cloud cover due to the presence of BBA over the 1 km domain therefore has a reduced impact on the net radiative balance and the magnitude of the semi-direct effect is smaller compared to the same region of the 5 km domain. The modelled semi-direct effect is thus 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 5 km domain had reduced daytime convection and precipitation, Comparisons with the TRMM dataset suggest that the 5km simulations without convective parameterisation organise the structure of convective systems better, as isolated cells rather than widespread precipitation. The positive semi- direct effect is lower in the scenarios without convective parameterisation due to the clouds being more cellular, but the negative nighttime longwave semidirect is also diminished. The net forcing from the scenarios with no convective parameterisation on the 18 September case-study is 1.04±0.78 Wm^{-2}. The large sensitivity to use of convective parameterisa- tion highlights the uncertainties with simulating aerosol–radiation–cloud interactions in this regime.

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 the physical significance of the mod- elled droplet number and grid-scale cloud properties of parameterised cloud is question- able. The runs with explicitly resolved convection at 1 km and no cumulus parameterisation at 5 km also show minimal indirect effects, likely due to the deep convective clouds being optically thick and therefor 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 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 with the effects of aerosol included, particularly at hori- zontal 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). The semi-direct effects are impossible to quantify reliably in this WRF-Chem setup due to this high sensitivity to the use of convective parameterisation and model resolu- tion. More coordination between parameterized and explicit treatments of aerosol, cloud and radiation interactions is needed in order to make modelling of these processes at the transition between fully parameterised and fully explicit schemes more consistent. To con- strain the simulation of these interactions, in-situ observations of aerosol size distribution and composition properties, measured before, during and after cloud processing need to be considered alongside remote sensing observations of changes to cloud cover and net radiation in regions of high aerosol loading. 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 cloud-aerosol 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

Artaxo, P., Rizzo, L. V., Brito, J. F., Barbosa, H. M. J., Arana, A., Sena, E. T., … Andreae, M. O. (2013). Atmospheric aerosols in Amazonia and land use change: from natural biogenic to biomass burning conditions. *Faraday Discussions*, *165*, 203. http://doi.org/10.1039/c3fd00052d

Berg, L. K., Shrivastava, M., Easter, R. C., Fast, J. D., Chapman, E. G., Liu, Y., & Ferrare, R. A. (2015). A new WRF-Chem treatment for studying regional-scale impacts of cloud processes on aerosol and trace gases in parameterized cumuli. *Geoscientific Model Development*, (8), 409–429. http://doi.org/10.5194/gmd-8-409-2015

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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 #2
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We thank the referee for their helpful critique of our paper. We appreciate that you found the paper to be well written and documented, agree that there was more scientific value that we could extract from the studies we made and have sought to address the areas of concern.

General/major comments

The authors state that the purpose of the study is to "critically evaluate how regional aerosol-radiation-cloud interactions are captured in WRF-Chem, . . ." (p. 27454, l. 1-9, see also p. 27463, l. 5). Although I realize that comparison with observations has been presented in a companion paper, the present study does not include any comparison with observations, and it is therefore difficult to know which of the experiments are more realistic. In particular, I think the value of what we learn from running with and without convective parameterization at the "grey zone" scales (i.e., <10 km) is limited when there is no idea of which is better. Evaluation against observations of clouds or precipitation, if available, would make this sensitivity experiment of with/without convective parameterization much more useful. At present, I do not agree that the paper is an evaluation paper, it is more a description of what happens when running with different setups.

Both referees have commented on our use of "evaluate" – to address this we have replaced the term with "investigate" instead.

Although I can understand the authors' statement that "The shorter case-studies at high-resolution were prioritized over a longer, low-resolution setup for the purpose and scope of the current investigation" (p. 27472, l. 1-3), the fact that the model region is tiny and the simulated time periods are few and very short, makes it difficult to generalize the results and make broader conclusions. Adding comparison to observations could possibly make up for this, as it may give some idea of which model setup is better.

The authors caution that the calculations of radiative balance should not be seen as robust calculations of radiative forcing (p. 27474, l. 10-11). However, I am not convinced that the method is good enough for drawing conclusions such as on p. 27466, l. 10-11 and p. 27467, l. 18-22 for simulations over such short time and for such a small region. A forcing imposed, e.g., by a reflecting compound such as sulfate, would rapidly lead to a decrease in surface temperature, which again would lead to reduced LW radiation from the Earth's surface, and hence contribute to maintain radiative balance. Supplement Table 4 shows that the near-surface temperature is affected by inclusion of an aerosol layer. In a long global climate model run this is solved by running with fixed sea-surface temperatures. A better, but more complex method would be to include double radiation calls, such as the method of Ghan et al. (2012), to quantify the direct, semi-direct, and indirect aerosol effects. Please justify the method used to calculate the radiative balance.

The authors were unaware of the Ghan et al., paper prior to receiving this review. We thank the reviewer for drawing attention to this article. The methodology described helps deconvolve the different forcings (direct, semi- and indirect) and we have implemented modifications to the WRF-Chem code to repeat the double radiation calls and re-run several of the scenarios over the three case-studies. We repeated the Ghan et al., analysis to generate estimates of direct, semi and indirect effects over the case studies, over the two domains and with and without the convective parameterization on the 5km domain. This has resulted in the development of the figures 3, 4, and 5 included at the end of this document, which will be inserted into the manuscript replacing the previous radiative balance figures 7, 9 and 10 (which have been moved to the supplementary material) as we believe this new analysis is easier to interpret. Details on how we carried out the calculations will be included in the appendix. A description of the methods has been added to new section 3.4, which adds to the previous analysis (which has been moved from the old appendix to this new section).

Our results from this new analysis are entirely consistent with our previous conclusions – namely that, for the case studies considered here, indirect effects are small relative to the dominant semi-direct effects. The semi-direct effect itself is highly sensitive to the convective parameterization and horizontal resolution of domain in question. However, being able to quantify the different effects substantially improves the quality of the study. The newly calculated numbers have also been inserted into to the abstract and conclusion.

We acknowledge that the limited domain size and duration of the study makes drawing general conclusions problematic. We now emphasise that the behaviour observed in this study is only indicative of the current case-studies, and caution should be used before generalizing our findings to other regions and events. The conclusions highlighted in the above comment (at 27466, l. 10-11 and p. 27467, l. 18-22 respectively) have had their language softened accordingly:

"For the limited case-studies considered in this paper, SW↓ Sfc is lower in the FE scenario, but the net forcing is less consistent and of smaller magnitude. The general reduction in cloud cover in the FE scenario adds a semi-direct warming effect which acts counter to the direct cooling of the aerosol, largely cancelling out any net impact."

"Overall, we find that net RB is more sensitive to whether or not a convective parameterisation is used than it is to the presence of aerosol or the horizontal resolution in the current case studies. The diurnally averaged reduction in RB of approximately 20Wm−2 between scenarios with and without convective parameterisation (Table S2 in the Supplement) is largely due a result of the reduction in nighttime clouds in the runs without convective parameterization"

We believe the developments enabling double radiation calls for this further analysis are a valuable tool for analysing impacts of aerosol. We would be interested in feeding these code developments back to be released in future WRF-Chem versions so it may be used in further studies.

While the introduction and model description sections are well referenced, the results section contains very little comparison and reference to other work (with the exception of Zhang et al., 2008). Several papers deal with the impact of biomass burning aerosols on meteorology and radiative forcing so this could easily be added. What about other regions, either of Amazonia, or in other biomass burning regions such as central and southern Africa, or Indonesia? Have similar or different results been found there? E.g., the result that fire aerosols stabilize the atmosphere and inhibits convection and cloud formation (p. 27465, l. 20-22) has also been found before, e.g., recently in tropical Africa (Tosca et al., 2015), and could be mentioned.

We acknowledge this issue and have added the following passage to the end of section 4.2:

"Similar effects have been found by other modeling studies investigating the impact of BBA over continental regions. For example, Zhang et al. (2008) found a peak negative clear-sky forcing of −8 W m−2 over the highest AOD region in the Amazon, but with reductions in cloud cover resulting in localised surface forcings as high as ≈ 22 W m−2 when changes to clouds were included. Kolusu et al., (2015), also show reduced all-sky forcing magnitude compared to clear-sky, show a decrease in precipitation due to BBA over the same SAMBBA period using the Met Office Unified Model (MetUM). Similarly in Africa, BBA has been shown to inhibit convection and cloud formation (Sakaeda et al., 2011, Tosca et al., 2015)."

The paper does not include any estimation of uncertainties in the results (except in the Supplement), but a statement that many of the results are not statistically significant (p. 27471, l. 23-25). In my view, it would still be useful to include some estimation of uncertainties. Including statistical significance based on a Student's t-test or similar could be useful when interpreting the results, and give the reader an idea of which results are robust and which are not.

While the authors agree that an estimation of the statistical uncertainties of results would be useful, the small size of domain and short runtimes, necessitated due to the high cost of running the model, make doing so challenging. From review of the literature, the authors found no standard way to calculate uncertainty for small domains. A common technique is to carry out a student-t test on every grid *point on a difference plot, and only show those results with p values less than 0.05. Using this method for the short runs in our study results in no significant grid-points – we would need to run for much longer periods to have a chance of passing a significance test. Note this does not necessarily mean the results are not important or of consequence, just that we cannot be sure of their robustness and so should avoid drawing general conclusions.*

However, it is still possible to show uncertainty related to domain averaged values. To do this, we have followed a similar methodology to Kolosu et al. (2016). The standard error (SE) is traditionally calculated by dividing the standard deviation by the square root of the number of data points. This method implicitly assumes all data points are independent, which is not the case for the grid points of a model run, where most variables show strong spatial and temporal autocorrelation. Assuming independence results in an erroneously small SE, and therefore too high a significance. We therefore apply a correction factor k (Bence 1995):

$$SE = \frac{SD}{\sqrt{N}} k \qquad 1.$$

where;

$$k = \frac{\sqrt{1+\rho}}{\sqrt{1-\rho}} \qquad 2.$$

and ρ is the autocorrelation factor, varying from -1 (perfect anti-correlation) to 1 (perfect correlation). For all the variables we applied this method to ρ was positive, so the correction acted to increase the SE relative to if we assumed all points were independent. We estimate spatial autocorrelation using the Moran's-I for neighbouring points. While the authors believe this method is valid for averages over the 5km domain, caution is advised over the 1km. The 1km domain region is small, and the region of the 5km domain it covers is not representative of the whole domain. As it is very sensitive to boundary conditions from the 5km domain, chaotic variation in cloud fields can create anomalous strong signals in the 1km domain. The 1km domain would need to be made larger and run for longer to filter out these systematic errors. A description of the statistical methods has been added to the paper in new section 3.5.

For presenting precipitation results, we have shown histograms of the data. In order to compare with TRMM data, the model fields were averaged over 25km boxes (~0.25 degrees) prior to analysis. An example of this analysis is presented in response to the comment on P. 27466, l. 22-26.

P. 27459, l. 28 – p. 27460, l. 2: Do the authors have an idea of how big of an impact this has on the results presented for the 5 km domain?

We tried to investigate this in the study. To test it directly would require two versions of the same convective parameterisation, one with aerosol interactions and one without. As this was not available at the time of study, we ran simulations with and without convective parameterisation and at higher resolution – the sensitivity study presented in the paper. Making these changes caused such a large difference to the cloud fields in the study, indirect effects from aerosol-cloud interactions were buried underneath the resultant noise. This finding is one of the main conclusions of the study. The new double-radiation call analysis further *supports this statement and we emphasise this point more strongly in the abstract and conclusions section. The low magnitude of indirect effects found makes the authors believe that no, the inclusion of cloud-aerosol interactions in the model would not have a large impact to the simulations (in the case studies considered here).*

P. 27466, l. 22-26: I am not sure this assumption and statement can be made without any observations showing that the results are more realistic in the 1 km domain.

Figures of precipitation from the 5km and 1km domain, from the FE and FE_nocu scenarios, during the time of peak precipitation (20:00-21:00 UTC) on 18 September 2012 are included below, with equivalent figure (or close as can be managed) from the TRMM 3B42 satellite product. The small size of the domain and coarse resolution of the TRMM product (0.25 degrees) makes direct comparison difficult. However, it is clear from the TRMM product that precipitation occurs in intense, tight convective cells. In the 5km domain with convective parameterisation, this structure is not well represented, instead there is large areal coverage of light precipitation. In the 1km domain, and in the nocu scenarios, these tight convective cells are seen in the model output, with corresponding lower domain cloud-coverage. The total precipitation in the FE_nocu scenario is substantially reduced.

These figures will be included in the paper in the section on impact of convective parameterisation, along with the following discussion:

"Peak precipitation rates (which occur between 20:00 – 21:00 UTC) in the afternoon of 18 September for the FE and FE_nCU scenarios, with similar figure from the TRMM 3B42 product, are shown in Figure 1. Although the TRMM product is coarse (with a resolution of 0.25 degrees), precipitation can, nonetheless, be seen to occur in small convective cells. In the FE scenario, precipitation less intense and covers a larger area, whereas in the 1km domain and FE_nCU scenario, precipitation follows a more cellular structure with a greater portion of the domain receiving no precipitation. The FE scenario correspondingly has a larger portion of the domain covered by cloud at any one time. However, total precipitation over both domains is greater in the FE scenario than the FE_nCU scenario."

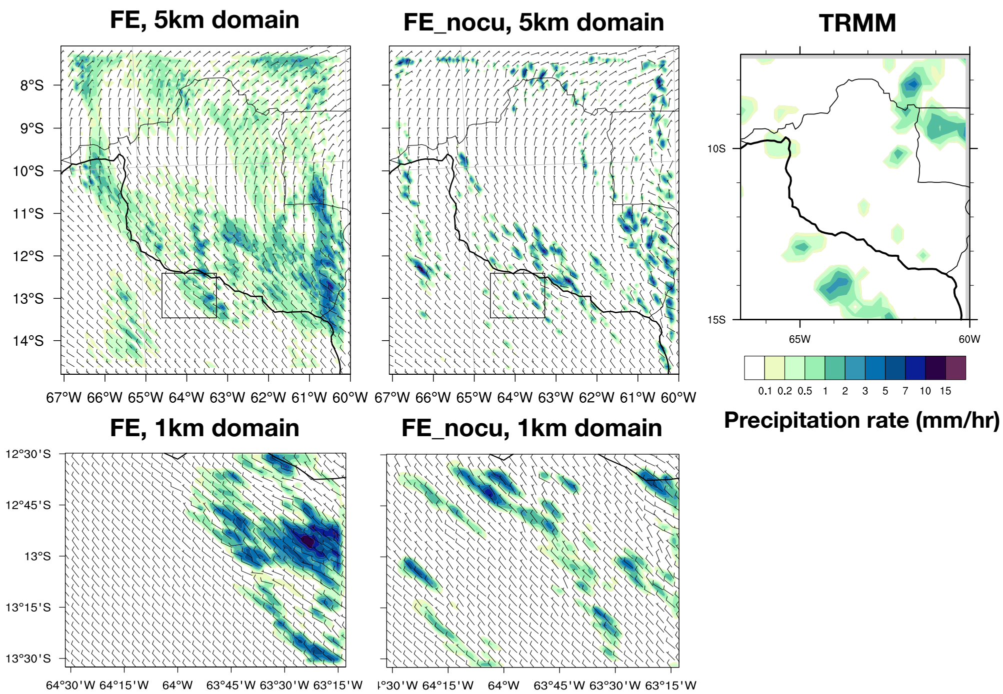

Figure 1. Precipitation rates between 20:00 and 21:00UTC on 18 September, 2012 across 5km (top) and 1km (bottom) domains from FE (left) and FE_nocu scenarios. Precipitation rate from TRMM 3B42 product between 18:00 and 21:00 on 18 September 2012.

We have also presented the precipitation as a bar-chart in Figure 2, which shows how the distribution of precipitation frequency changes between scenarios. For this comparison, the model data was averaged over a 25km grid to be of roughly the same resolution as the TRMM data. In the FE scenario, most grid cells contain at least some precipitation, whereas the TRMM dataset shows most cells with no precipitation. The scenario with no convective parameterization follows a closer distribution to the TRMM dataset, with more cells receiving little to no precipitation and a greater portion of total precipitation from a few cells with high precipitation. The average precipitation over the whole domain on 18 September is 2.30mm, 1.43mm and 1.49mm for the FE, FE_nCU and TRMM datasets respectively. Thus the nocu case preforms better for both precipitation distribution and total magnitude over the region for this case study.

Figures 1 and 2 have replaced the old Figure 8 (which has been moved to supplementary material) in the paper, as we believe this demonstrates the sensitivity to convective parameterisation more simply and effectively.

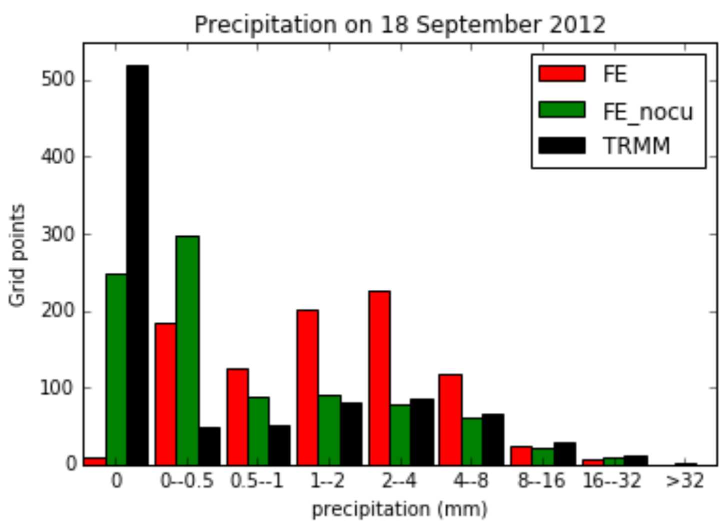

Figure 2. Bar-chart of precipitation on each grid cell over the 5km domain from FE, FE_nocu scenarios and TRMM 3B42 product.

P. 27470, l. 26-28: Again, how can this conclusion be made without comparison to observations?

We believe that our reply to the referee's previous question answers this question too.

Minor comments/corrections:

P. 27454, l. 1: This -> The

Changed accordingly.

P. 27459, l. 11: caries -> carries

Changed.

P. 27462, l. 17-28: The aerosol loadings are mentioned several times, but this is not shown in Fig. 2. Perhaps add reference to Fig. 3 in this paragraph?

Added reference to Fig 3 at end of line 20:

"Extensive fire emissions and minimal precipitation over the region between 10 and 14 September result in high modelled aerosol loadings (see Fig 3.)"

P. 27469, l. 27: subsistence -> subsidence

Changed

P. 27471, l. 20-21: This is an interesting finding and could be made more clear in the abstract?

The authors thank the reviewer for drawing more attention to this. We agree this is an interesting finding, and have emphasized it in the abstract accordingly.

P. 27472, l. 19: Remove "to the" after "includes".

Changed.

P. 27487, l. 3: Scenrios -> Scenarios

Changed

P. 27489, l. 2: averaged -> accumulated

Changed

References:

Bence, J. R.: Analysis of short time series: correcting for autocorrelation, Ecology, 76, 628–639, doi:10.2307/1941218, 1995.

Ghan, S. J., Liu, X., Easter, R. C., Zaveri, R., Rasch, P. J., Yoon, J. H., and Eaton, B.: Toward a Minimal Representation of Aerosols in Climate Models: Comparative Decomposition of Aerosol Direct, Semidirect, and Indirect Radiative Forcing, J. Clim., 25, 19, 6461-6476, 10.1175/jcli-d-11-00650.1, 2012.

Kolusu, S. R., Marsham, J. H., Mulcahy, J., Johnson, B., Dunning, C., Bush, M., & Spracklen, D. V. (2015). Impacts of Amazonia biomass burning aerosols assessed from short-range weather forecasts. *Atmospheric Chemistry and Physics*, *15*(21), 12251–12266. http://doi.org/10.5194/acp-15-12251-2015

Tosca, M. G., Diner, D. J., Garay, M. J., and Kalashnikova, O. V.: Human-caused fires limit convection in tropical Africa: First temporal observations and attribution, Geophys. Res. Lett., 42, 15, 6492-6501, 10.1002/2015gl065063, 2015.

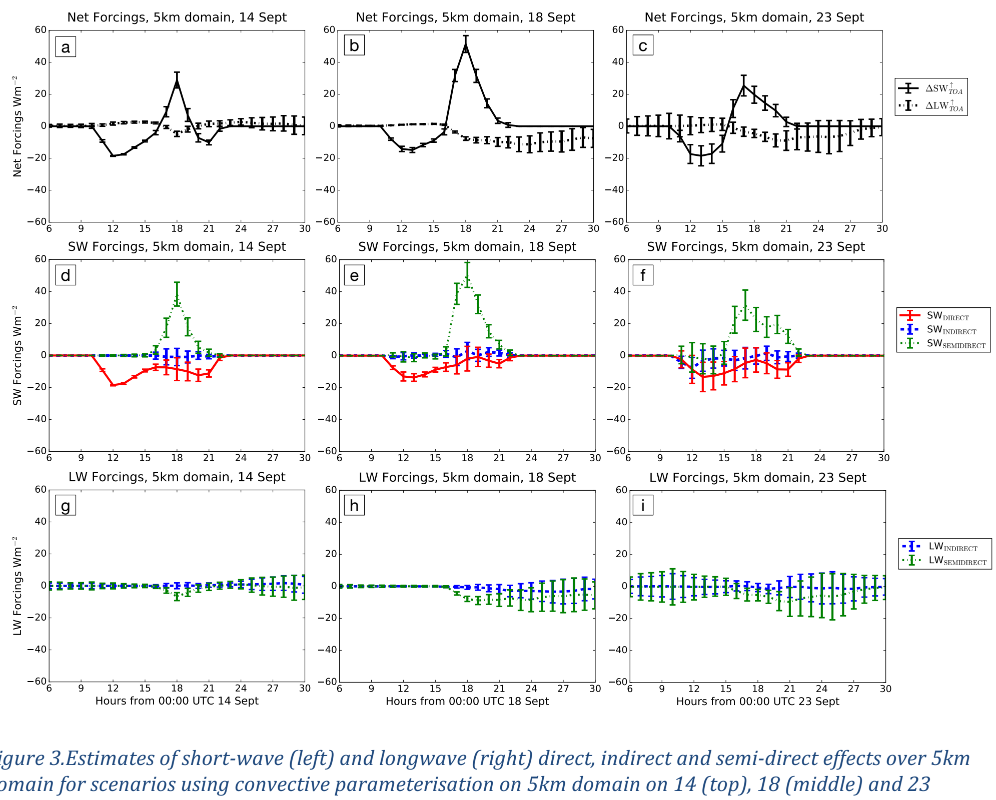

Figure 3.Estimates of short-wave (left) and longwave (right) direct, indirect and semi-direct effects over 5km domain for scenarios using convective parameterisation on 5km domain on 14 (top), 18 (middle) and 23 (bottom) September.

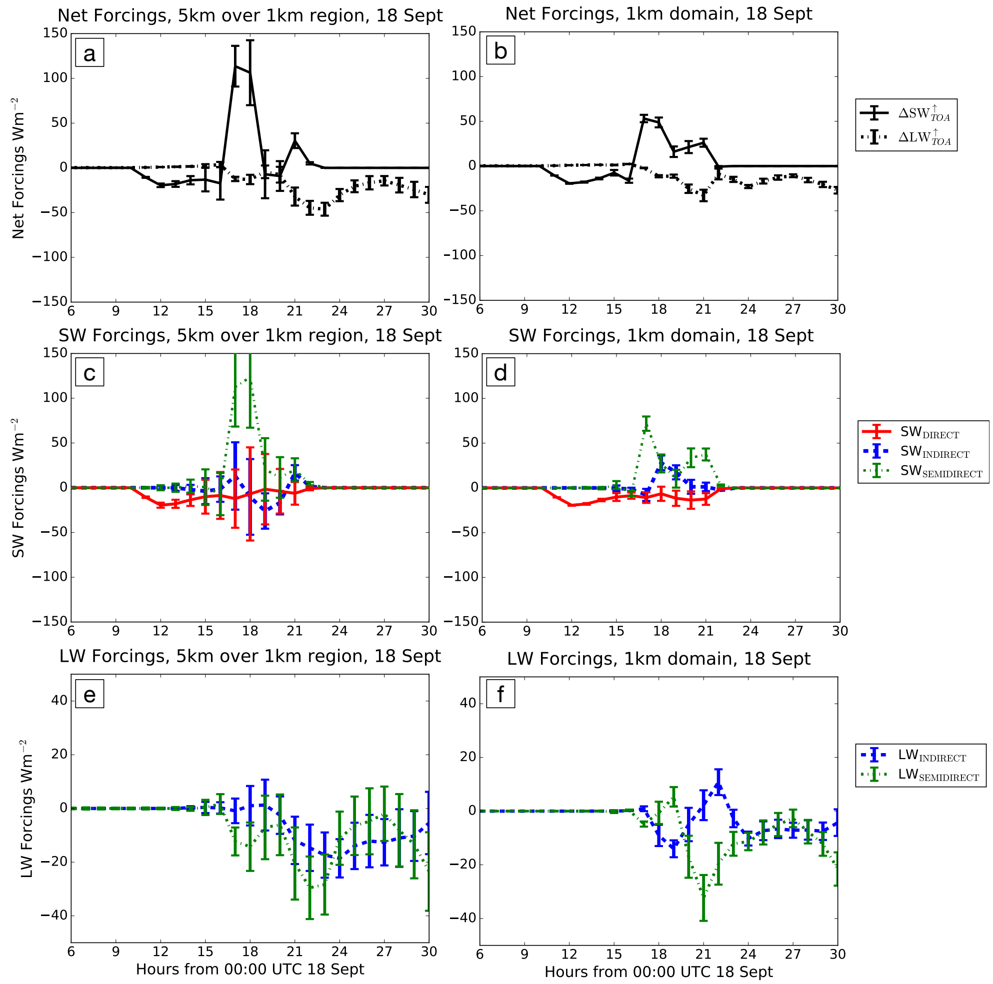

Figure 4. Estimates of direct, indirect and semi-direct effects on 18 September, 2012 from the 5km domain over the 1km domain region (left) and in the 1km domain (right).

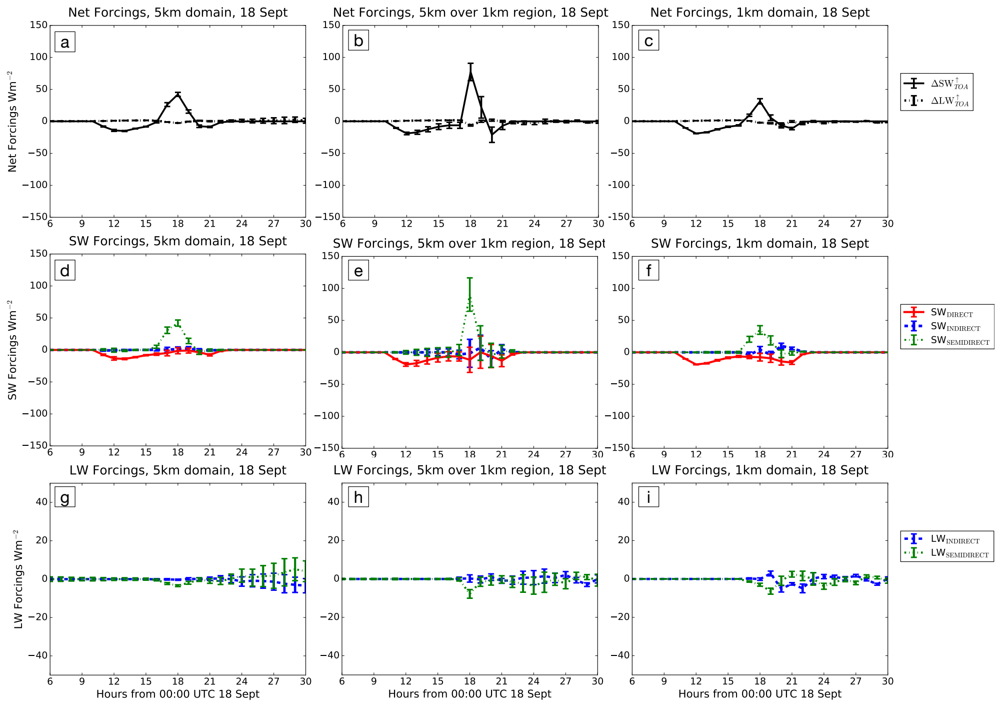

Figure 5. Estimates of direct, indirect and semidirect effect from scenarios with no convective parameterisation on the 5km domain on 18 September 2012. Over whole of 5km domain (left), 1km region of the 5km domain (middle) and 1km domain (right).

Abstract

The Weather Research and Forecasting model with Chemistry (WRF-Chem) has been used to simulate a region of Brazil heavily influenced by biomass burning. Nested simulations were run at 5 km and 1 km horizontal grid spacing for three case studies in September 2012. Simulations were run with and without fire emissions, convective parameterisation on the 5 km domain and aerosol–radiation interactions in order to explore the differences attributable to the parameterisations and to better understand the aerosol direct effects and cloud responses. Direct aerosol–radiation interactions due to biomass burning aerosol resulted in a net cooling, with an average ~~reduction of downwelling shortwave radiation at the surface of -24.7~~short-wave direct effect of -4.08 ± 1.53 $\mathrm{W\,m^{-2}}$~~over the three case studies.~~ . However, around 21.7 $\mathrm{W\,m^{-2}}$ is absorbed by aerosol in the atmospheric column, warming the atmosphere at the aerosol layer height, stabilising the column, inhibiting convection and reducing cloud cover and precipitation. The changes to clouds due to radiatively ~~interacting~~ absorbing aerosol (traditionally known as the semi-direct effects) increase net shortwave radiation reaching the surface by reducing cloud cover, producing a secondary warming that ~~largely~~ counters the direct cooling. However, the magnitude of the semi-direct effect was ~~difficult to quantify, being~~ found to be extremely sensitive to the model resolution and use of convective parameterisation. ~~The 1~~Precipitation became organised in isolated convective cells when not using a convective parameterisation on the 5 km domain~~simulated clouds less horizontally spread, reducing the~~ , reducing both total cloud cover and total precipitation. The SW semi-direct effect varied from 6.06 ± 1.46 $\mathrm{W\,m^{-2}}$ with convective parameterisation to 3.61 ± 0.86 $\mathrm{W\,m^{-2}}$ without. Convective cells within the 1km domain are typically smaller but with greater updraft velocity than equivalent cells in the 5km domain, reducing the proportion of the domain covered by cloud in all scenarios and producing a smaller semi-direct effect. ~~Not having a convective parameterisation on the 5domain reduced total cloud cover, but also total precipitation.~~ BB aerosol particles acted as CCN, increasing the droplet number concentration of clouds. However, the changes to cloud properties had negligible impact on net radiative balance on either do-

main, with or without convective parameterisation. Sensitivity to the uncertainties relating to the semi-direct effect was greater than any other observable cloud adjustments. Although indirect effects. Although the version of WRF-Chem distributed to the community currently lacks aerosol–cloud interactions in parameterised clouds, the results of this study suggest a greater priority for development is to improve the modelling of semi-direct effects by reducing the uncertainties relating to use of convective parameterisation and resolution before WRF-Chem can reliably quantify the regional impacts of aerosols.

1 Introduction

Aerosol particles in the atmosphere have a major impact on global climate, but also contribute some of the greatest uncertainties due to their heterogeneous distribution and complicated interactions with clouds and radiation (IPCC, 2013). The aerosol–radiation interactions, commonly known as the direct effects, tend to result in scattering of solar radiation and cooling of the Earth's surface (Haywood and Boucher, 2000; Zhang et al., 2008; Chand et al., 2009). However, many aerosol particles also contain black carbon (BC), which absorbs radiation across a wide spectrum of wavelengths (Bond et al., 2013). Whether an absorbing aerosol layer has a net cooling or warming effect, as seen from the top-of-atmosphere (TOA), depends greatly on whether it is over a low or high albedo surface (Haywood et al., 1995; Haywood and Boucher, 2000).

As well as their direct interactions with radiation, aerosol particles can perturb the Earth's radiative budget through their impacts on clouds (Lohmann and Feichter, 2005; Rosenfeld et al., 2008; IPCC, 2013; Possner et al., 2015). The absorption and scattering of radiation changes atmospheric stability and circulation, and therefore cloud formation (IPCC, 2013) . These adjustments by the climate system are traditionally known as the semi–direct effects (Hansen et al., 1997; Ackermann et al., 2000). The sign and magnitude of the semi-direct radiative forcings are sensitive to whether the aerosol layer is over land or sea (Allen and Sherwood, 2010), and to the vertical distribution, depending on whether the aerosol layer is below, at or above cloud height (Johnson et al., 2004; Koch and Del Genio, 2010). In addition, aerosol particles act as cloud condensation nuclei (CCN, Andreae et al., 2004; McFiggans et al., 2006; Hennigan et al., 2012). Polluted clouds have increased cloud droplet number, resulting in the first indirect effect whereby brighter clouds reflect more radiation back to space (Twomey, 1974; Lohmann and Feichter, 2005; Possner et al., 2015). Increased droplet number may further perturb cloud lifetime, height and the ability to initiate precipitation (Andreae and Rosenfeld, 2008; Chen et al., 2011). The addition of aerosol particles can either inhibit or enhance cloud formation: a small increase in CCN above pristine conditions in deep convective clouds cause more droplets to reach supercooled levels, increasing the amount of latent heat release and invigorating convection (Rosenfeld et al., 2008; Pöschl et al., 2010; Possner et al., 2015). Rosenfeld et al. (2008) estimate the maximum invigoration point to be at a CCN concentration of 0.4 % supersaturation ($CCN_{0.4}$) of approximately $1200\,cm^{-3}$. Further increases in CCN result in the direct radiative effects dominating, which cool the surface and inhibit convection.

The primary tool for estimating aerosol particles' impact on climate has been the use of global climate models (IPCC, 2013, and references therein). However, horizontal grid spacing is typically in the order of a degree, meaning most clouds are smaller than a grid box and must be parameterised, introducing uncertainties to how the system responds to forcings by aerosol particles (Johnson, 2004; Ghan et al., 2006; Lohmann and Ferrachat, 2010). For example, the magnitude and sign of the semi-direct effects show strong sensitivity to the cloud parameterisation used (Cook et al., 2004).

At the other end of the resolution spectrum, large eddy simulation (LES) models are capable of explicitly resolving clouds with detailed bin microphysics at grid spacings in the order of 10–100 m. Although LES models can only be used over small areas, often with idealised boundary conditions, they are useful to gain insight into how aerosols affect clouds and are known to reproduce more realistic behaviour than the parameterisations used in global models (Romakkaniemi et al., 2009; Chen et al., 2011). Johnson (2004) compared a single-column model, equivalent to a cloud parameterisation used in global models, with a LES model, and found the semi-direct effect over a stratocumulus deck was five times stronger in the LES simulation, implying deficiencies in the ability of global models to parameterise aerosol–cloud interactions.

The need to better understand the impact of aerosol–radiation–cloud interactions on a regional scale has driven the development of "online" models with "full" couplings between the air quality and meteorological components (Baklanov et al., 2011; Grell and Baklanov, 2011; Baklanov et al., 2013). The Weather Research and Forecasting model with Chemistry (WRF-Chem) is one such model (Grell et al., 2005; Fast et al., 2006). Unlike in offline chemical transport models, the gas-phase chemical and aerosol fields are transported using the same timestep and physical parameterisations as the core numerical weather prediction model. By linking aerosol optical properties to the radiation scheme and CCN potential to the microphysics scheme, feedbacks between aerosols and meteorology can be modelled (Chapman et al., 2009; Barnard et al., 2010; Zhao et al., 2011). However, the publicly available version of WRF-Chem is currently limited by having no aerosol–cloud interactions in parameterised convective clouds, and no linkages exist in the model between aerosol particles and ice nuclei (Chapman et al., 2009). Studies into indirect effects with WRF-Chem have therefore tended to focus on marine stratocumulus, which can be resolved at coarser resolutions (e.g. Yang et al., 2011; Saide et al., 2012).

As computing resources have improved, WRF-Chem has been increasingly run at fine resolutions with horizontal grid spacings less than 10 km (e.g. Grell et al., 2011; Wu et al., 2011a, b; Saide et al., 2012; Shrivastava et al., 2013; Fast et al., 2014). These scales (commonly known as the "grey-zone") are challenging to model because the assumptions behind the deep-convective parameterisations begin to break down, but the model cannot be expected to resolve all convection explicitly (Hong and Dudhia, 2012). The Grell-3-D convective parameterisation has in part been developed to be used over these intermediate horizontal resolutions by allowing "subsistence spreading" to neighbouring grid cells (Grell and Freitas, 2014). However, it is currently unclear how effectively cloud responses to aerosol in the "grey-zone" are simulated with this parameterisation. Through further nesting, WRF-Chem can be run at scales where no cumulus parameterisation should be used ($\Delta x \lesssim 4$ km), bridging the gap between global climate and LES models to explicitly resolve aerosol–cloud interactions in warm convective clouds. However, even at these fine scales questions remain as to how well some structures, such as shallow cumulus clouds, are simulated (Hong and Dudhia, 2012).

This purpose of this study is to critically evaluate modelling study investigates how regional aerosol–radiation–cloud interactions are captured in WRF-Chem, using a period during the South American Biomass Burning Analysis (SAMBBA) project as an example. The modelled aerosol direct, semi-direct and indirect effects are calculated as a function of horizontal grid spacing and cumulus parameterisations – both for several different model configurations. These cover two different horizontal grid spacings, and include running with and without fire emissions – illustrating a convective parameterisation, and with and without fire emissions. Using these results, the uncertainties in representing these processes within modelsand , and the difficulties in making accurate predictions. As a result, this study educates , are illustrated. Knowledge about how these processes interact with, and feedback to, each other and the general model configuration, is important for determining the best manner in which to run models such as WRF-Chemusers as to. How these processes, and the strengths and weaknesses of the processes within the model, providing an feedbacks between them, are configured 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 simulations in these challenging regimesthe realism of these simulations, as well as consistency through the different model scales.

The test case used is a region of Brazil known to be heavily polluted by biomass burning aerosol (BBA) during the dry season. The aerosol haze layer is characterised as being highly radiatively absorbing (single scattering albedo between 0.8 and 0.9), optically thick (aerosol optical depths between 0.4 and 1.2), vertically elevated to cloud-level through biomass burning plume processes, and efficient at acting as CCN (Reid et al., 2005a, b; Martin et al., 2010; Archer-Nicholls et al., 2015). The high aerosol concentrations in this region should provide a strong signal for aerosol–radiation and aerosol–cloud interactions for the study.

WRF-Chem has been previously used to investigate the impact of BBA on weather and climate. For example, Grell et al. (2011) found a ~~significant~~ modest improvement to the modelled representation of the vertical temperature profile when biomass burning emissions and aerosol feedbacks were included in runs over Alaska. Zhang et al. (2014) evaluated the direct radiative effects of BBA over Northern Sub-Saharan Africa, and impacts to vary widely depending on the emission inventory used. Wu et al. (2011b) ran simulations over Brazil at 36 and 4 km horizontal grid spacing, with no convective parameterisation on the 4 km domain. They found BBA to inhibit afternoon convection over the domain, reducing daytime precipitation but increasing it night, albeit with a net decrease in precipitation. The 36 and 4 km simulations were qualitatively similar.

This paper follows on from Archer-Nicholls et al. (2015), which aimed to characterise the BBA population in Brazil in the 2012 fire season. The model output was evaluated against remote sensing and in-situ aircraft measurements from the SAMBBA campaign. The ~~initial setup, using the Brazilian Biomass Burning Emissions Model (3BEM) with Freitas et al. (2007) plume-rise parameterisation resulted in injection of fire emissions too high into the atmosphere compared to aircraft measurements. An alternative emissions scenario, using reduced fire size based on remote-sensing measurements of fire radiative products for the 2012 dry season, was developed and compared with measurements, showing an improved vertical distribution but still with some bias towards having too much aerosol between 2–6. The particulate organic matter to BC ratio was lower in model compared to measurement, likely due to uncertainties in biomass burning emission factors and lack of secondary organic aerosol (SOA) formation in the model. However, the single scattering albedo ω_0 was similar to that measured. Aerosol size distribution and CCN concentration were both reasonably well represented.~~

~~The model~~ model fields from Archer-Nicholls et al. (2015) are used to drive initial and boundary conditions for two nested domains with 5 km and 1 km horizontal grid spacing in this study. The 5 km domain was chosen to be within the “grey-zone” in order to probe how the WRF-Chem simulates aerosol interactions and impacts, while the 1 km domain has no need for a convective parameterisation. Several runs were conducted using different emission scenarios and options for aerosol–radiation interactions to separate the instantaneous radiative effects of the aerosol from aerosol–cloud interactions. The sensitivity of the semi-direct and indirect effects to convective parameterisation and horizontal resolution is also investigated. Due to the limited area and duration of the model runs, simulating the full changes to circulation as a result of the forcings are out of the scope of the current study and so only short-term responses are investigated.

2 Model description

This study uses WRF-Chem version 3.4.1 with changes made to use the Model for Simulating Aerosol Interactions with Chemistry (MOSAIC) aerosol scheme (Zaveri et al., 2008) and the updated Carbon Bond Mechanism (CBM-Z) gas phase chemistry scheme (Zaveri and Peters, 1999) with the Brazilian Biomass Burning Model (3BEM) fire emissions (Longo et al., 2010; Freitas et al., 2011), as described by Archer-Nicholls et al. (2015). As an "on-line" coupled model, the meteorological, transport, chemical and aerosol components are integrated at the same time. Forcings from the chemical and aerosol fields can feed-back with the meteorology, and visa-versa (Grell et al., 2005). These feedbacks primarily occur through the aerosol–radiation interactions and aerosol particles acting as CCN to influence cloud properties. A robust approach to describe the aerosol population and their interactions with clouds and radiation is therefore needed.

2.1 The MOSAIC aerosol mechanism

The MOSAIC mechanism is a sectional scheme, whereby the aerosol size distribution is described as a set of discrete size bins (Zaveri et al., 2008). This study uses eight size bins across a range of 39 nm to 10 µm, as shown in Table 1. MOSAIC carries five inorganic ions which can react in the aqueous phase and partition with the gas-phase mechanism, plus three unreactive primary aerosol species: black carbon (BC), particulate organic matter

(POM), and other inorganics (OIN) (Fast et al., 2006; Zaveri et al., 2008). All chemical components within each size bin are assumed to be internally mixed (i.e. evenly mixed within the same particles), whilst different size bins are assumed to be externally mixed (Zaveri et al., 2008).

The version of MOSAIC used in this study does not carry secondary organic aerosol (SOA). Current conventional treatments are unable to capture frequently observed SOA behaviour, such as the formation of sufficient mass from known precursors or the oxygen to carbon ratio (O : C) of the material. Alternative treatments are available, such as the Volatility Basis Set (VBS Donahue et al., 2011; Shrivastava et al., 2011, 2013) (VBS; Donahue et al., 2011; Shrivast but remain unconstrained for the current application. In particular, it is unclear how previously used treatments can capture behaviour such as that summarised in the meta-analysis of Jolleys et al. (2012), which described the lack of increase in organic mass from biomass burning source, but an increase in O : C. Ongoing developments of the VBS are in progress to explore mechanisms by which observed OA behaviour is best captured, but are beyond the scope of the current work. However, it is expected that the current approach will reasonably capture the OA mass and hence POM : BC ratio.

Whilst uncertainties in the model representation of aerosol composition (particularly POM : BC ratio), size distribution and optical properties can result in uncertainties in predicted radiative forcings (Matsui et al., 2013; Kodros et al., 2015), investigation of these uncertainties is beyond the scope of the current study. Notwithstanding the discussed limitations, using a sectional representation of aerosol provides a reasonably robust approach for calculating the aerosol optical properties and interactions with clouds, as described below.

2.2 Calculation of aerosol optical properties

Within MOSAIC, each aerosol chemical component has its own associated complex refractive index, with BC being the most absorbing (Barnard et al., 2010). The overall complex refractive index is calculated for each bin using a mixing rule to approximate the internal structure of the aerosol particles. Assuming an internal mixture of BC with other components can result in an overestimation of the particles absorption cross-section (Bond and Bergstrom, 2006). Describing particles using a spherical BC core with other component shell (a “shell-core” mixing rule) is often regarded as the most robust approach for 3-D model applications (Bond et al., 2006; Bond and Bergstrom, 2006; Barnard et al., 2010; Matsui et al., 2013), but was found to be unstable in WRF-Chem version 3.4.1. In this study, the Maxwell–Garnett mixing rule is used, whereby aerosol particles are assumed to be made up of randomly distributed spheres of BC throughout a mixture of all other components (Bohren and Huffman, 1983, chapter 8). The Maxwell–Garnett rule does not suffer from the anomalous absorption enhancement of the internal mixing rule (Bond and Bergstrom, 2006).

Mie calculations are used to calculate the intermediate optical properties for each bin, which are summed over all size bins to give bulk extinction coefficient (b_{ext}), scattering coefficient (b_{s}), single scattering albedo ($\omega_0 = b_{\text{s}}/b_{\text{ext}}$), and asymmetry factor (g). Each of these variables are functions of the size parameter ($x = 2\pi r/\lambda$), where λ is the wavelength of light and r is the wet radius at the centre of the aerosol bin (Fast et al., 2006). To save on computation, the methodology of Ghan et al. (2001) is employed to carry out the full Mie calculations only on the first call to the subroutine. The net radiative impacts are calculated by passing the bulk optical properties of the aerosol layer to the radiative transfer parameterisation. This study uses the rapid radiative transfer model (RRTMG, Mlawer et al., 1997; Iacono et al., 2000) for both short-wave (SW) and long-wave (LW) radiation following Zhao et al. (2011). Optical properties in the SW are calculated at four wavelengths ($\lambda = 300$, 400, 600, and 1000 nm). For intermediate λ, b_{ext} is estimated using an Ångström coefficient, whereas ω_0 and g are linearly interpolated. A full description of the optical property calculations are given by Fast et al. (2006) and Barnard et al. (2010).

2.3 Calculation of aerosol–cloud interactions

A key process to simulating aerosol–cloud interactions is the activation of CCN to form cloud droplets. Köhler et al. (1936) theory describes the equilibrium state of an aerosol particle, assumed to be an aqueous salt solution, with ambient water vapour. The critical supersaturation (S_{crit}, defined as the supersaturation at which an aerosol particle becomes activated to form a cloud droplet) depends upon both aerosol size and composition. Aerosol particles that are larger and/or more hygroscopic are activated more easily and so have a lower S_{crit} (McFiggans et al., 2006). Within MOSAIC, S_{crit} is calculated for each bin using a mass-weighted average of the associated hygroscopicity of all chemical components within that bin using the methodology of (Abdul-Razzak and Ghan, 2002).

The primary driver of cloud droplet activation is the updraft velocity (w): air parcels with higher w reach higher maximum supersaturations (S_{max}). All particles with $S_{\text{crit}} < S_{\text{max}}$ will be activated, whereas those with $S_{\text{crit}} > S_{\text{max}}$ remain unactivated within clouds and are known as interstitial aerosols (Chapman et al., 2009). Greater CCN concentration increases the total particulate surface area, increasing competition for condensable water and reducing S_{max}. Subgrid variation in updraft velocity (w) is described using a Gaussian distribution function, with a minimum spread of $\sigma_w = 0.1\,\text{m}\,\text{s}^{-1}$ (Ghan et al., 1997). The number and mass fraction of activated CCN in each aerosol bin can then be calculated by comparing S_{max} with S_{crit} at the sectional limits of each bin (Abdul-Razzak and Ghan, 2002). Inversely, this method can also estimate the CCN concentration at given supersaturations. WRF-Chem ~~caries~~ carries six diagnostic variables showing the concentration of particles that can potentially activate at given supersaturations of 0.02, 0.05, 0.1, 0.2, 0.5 and 1 % ($\text{CCN}_{0.02}$, $\text{CCN}_{0.05}$, $\text{CCN}_{0.1}$, $\text{CCN}_{0.2}$, $\text{CCN}_{0.5}$ and $\text{CCN}_{1.0}$ respectively).

Recently, Simpson et al. (2014) have shown the Abdul-Razzak and Ghan (2000) parameterisation produces unrealistic activated fractions of aerosol in some atmospherically relevant conditions when compared with an explicit bin-resolving cloud-parcel model. The scheme was shown to over predict activation when the aerosol population median diameter was $\gtrsim$ 300 nm. However, given the median diameter in BBA populations is generally between 100–150 nm (Janhall et al., 2010), this behaviour should not negatively impact the simulations in this study.

To model the indirect effects the cloud activation scheme needs to be coupled with a double-moment microphysical parameterisation that carries both number and mass loadings for hydrometeors. Following Yang et al. (2011), the double-moment Morrison et al. (2005, 2009) parameterisation has been coupled with MOSAIC aerosol, such that the number concentration of liquid droplets is controlled by activated aerosol. The couplings are currently only for warm-cloud processes, with no direct links between aerosol and ice nuclei (Chapman et al., 2009). A major limitation in using WRF-Chem to assess aerosol–cloud interactions is that the couplings are only computed in explicitly resolved clouds, not convective clouds simulated by the cumulus parameterisation (Chapman et al., 2009; Yang et al., 2011). Work is being conducted to include aerosol interactions with parameterised cloud ~~(e.g. Grell and Freitas, 2014; **?**)~~ (e.g. Grell and Freitas, 2014; Berg et al., 2015) . However, these developments were not available for general WRF-Chem release at the time of this study.

3 ~~Experimental Setup~~

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 W 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) .

In deep convective clouds, secondary activation of aerosol has been observed (Heymsfield et al., 2009) and modeled (e.g. Segal 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 (2002) 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. Use of the aerosol-aware Kain-Fritsch parameterisation (Berg et al., 2015) could enable consideration of this process in parameterized clouds for future studies.

3 Experimental Methods

This section describes the model setup and rationale for the experiments conducted for this study. The objective is to probe the response of the WRF-Chem model to aerosol–radiation and aerosol–cloud interactions across a range of scales and meteorological conditions. The high levels of elevated, highly-absorbing aerosol over Amazonia during the dry-to-wet season transition provide a good test-bed for the experiments by producing a strong signal of aerosol forcings. Several scenarios were constructed to isolate the various aerosol impacts, as described below.

3.1 Domain setup and methods

Archer-Nicholls et al. (2015) described a parent domain run for the whole of September 2012 with 226×196 grid cells at 25 km horizontal grid spacing covering most of South America, 41 vertical levels up to 50 hPa with 18 levels within the lowest 3 km, and a Lambert conformal conic projection. The meteorological input and boundary conditions were driven by the operational, deterministic (high-resolution) 1 day forecasts of the European Centre for Medium-Range Weather Forecasts (ECMWF, http://www.ecmwf.int/). Chemical and aerosol boundary conditions were derived from the MACC-II reanalysis (Monitoring Atmospheric Composition and Climate – Interim Implementation; Hollingsworth et al., 2008; Flemming et al., 2013).

This study focuses on the output of two nested domains, with 5 and 1 km grid spacing respectively. The location of the 5 km nest encompasses a region of high aerosol optical depths (AODs) over Rondônia state. The 1 km domain is positioned over a region with high AODs, flat topography and heavy precipitation on 18 September 2012. A map of all three domains is shown in Fig. 1. The nests were run for three 36 h case-study periods with contrasting meteorological conditions, starting at 00:00 UTC on 14, 18 and 23 September 2012 respectively (where local time = UTC − 4 h). These dates coincide with the SAMBBA flight numbers B731, B734 and B739, against which the model output was evaluated by Archer-Nicholls et al. (2015) . The ndown utility was used to generate hourly offline boundary conditions for the 5 km nests from the 25 km runs. The 5 and 1 km nests were run online without feedback between nests.

Except where otherwise stated, the 5 km domain uses the Grell-3-D convective scheme with subsidence spreading turned on so as to be applicable for use below 10 km grid spacing (Grell and Freitas, 2014). No convective parameterisation is used on the 1 km nest, allowing explicit aerosol–cloud interactions in convective clouds. The differences in model setup between domains are summarised in Table 2. All other physical parameterisations are the same between the nested and parent domains and are described in more detail in Archer-Nicholls et al. (2015).

3.2 Scenarios

Two emission scenarios are considered in this study: fire emissions (FE) and no fire emissions (nFE). FE uses the 3BEM fire emissions with the Freitas et al. (2007) plume-rise parameterisation and modifications for the 2012 biomass burning season described in Archer-Nicholls et al. (2015). The nFE scenario has no fire emissions, but has the same anthropogenic emissions, biogenic emissions and boundary conditions as the FE scenario. Both of these scenarios were run for the entirety of September on the 25 km domain without aerosol–radiation interactions(aer= 0). . The meteorological fields were reinitialised from ECMWF fields at the start of each nested simulation run to minimise synoptic-scale error growth and ensure that any differences within the nested domains were due to processes being investigated within the nests.

To separate the impacts of aerosol–radiation interactions from cloud–aerosol interactions, the ~~model was~~ nested domains were run with aerosol–radiation interactions both turned on and off. Unless otherwise stated, references to the FE and nFE scenarios refer to scenarios with aerosol–radiation interactions on. The scenario with no fire emissions or aerosol–radiation interactions is used as a control simulation (Ctrl), and behaves as a WRF simulation would (i.e. with negligible aerosol effects). Another scenario with fire emissions but no aerosol–radiation interactions (nARI) is used to isolate the impacts of cloud–aerosol interactions. Finally, each scenario was also run with the Grell-3-D convective parameterisation turned off over the 5 km domain (denoted with the suffix "_nCU") for the 18 September 2012 initialisation. The scenarios are summarised in Table 3.

3.3 Meteorological ~~conditions~~and aerosol fields

Figure 2 shows ~~averaged~~ accumulated precipitation and winds at 700 hPa over the three case study periods over the 5 km domain. The meteorological input conditions of each nested simulation case study are derived from the ECMWF data, whereas the chemical and aerosol input conditions are interpolated from the 25 km domain. The first 6 h of integration of each run are discarded as spin-up.

The modelled meteorological conditions differ markedly for each case study. The driest conditions are on 14 September, with only limited convective precipitation. Prevalent winds are easterly or north easterly. Extensive fire emissions and minimal precipitation over the region between 10 and 14 September result in high modelled aerosol loadings ~~.~~ (Fig. 3). By 18 September the transition into the wet season has begun, with widespread precipitation across the 5 km domain and the location of the 1 km nest~~, and prevailing northerly winds.~~ . Aerosol loadings are lower than on the 14 September, but still high. There is heavy precipitation and easterly winds over the northern half of the domain on 23 September, but north to north-westerly winds and little precipitation over the southern half (where the 1 km nest is located). By 23 September, prolonged rainfall has washed out much aerosol. However, the model shows higher aerosol loadings compared to measurements on this date (see Archer-Nicholls et al., 2015, for more details).

The dates of the case studies coincide with the SAMBBA flight numbers B731, B734 and B739. The model output from the parent 25km domain was evaluated against these in-situ flight measurements by Archer-Nicholls et al. (2015) . Modelled POM and $PM_{2.5}$ mass was of similar magnitude to flight measurements on 14 and 18 September, although sufficient aerosol mass was achieved by scaling up emissions to match observed AOD from the MODIS satellite product in the region. On 23 September, 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. Due in part to poorly captured plume-rise, the vertical distribution was biased high in the model between the boundary layer top and 4km above ground. Although there were some discrepancies in POM:BC ratio between model and observations, single scattering albedo compared reasonably well. Overall, the model reproduced aerosol fields well enough to capture the broad impacts of BBA, acknowledging uncertainties due imperfect representation of aerosol vertical distribution and optical properties.

3.4 Radiative flux calculations

The public version of WRF-Chem carries 16 diagnostic variables for assessing simulated radiative fluxes. These are first split into short-wave (SW) and long-wave (LW) portions of the spectrum, and can be calculated at the top of the atmosphere (TOA) or the surface (e.g. SW_{TOA}, SW_{Sfc}), in either the up or down direction ($SW^{\uparrow}_{TOA}$, $SW^{\downarrow}_{TOA}$). Finally, they can be calculated for "all-sky", including the effects of clouds ($SW^{\uparrow}_{TOA}$); or for "clear-sky", ignoring the effects of clouds ($SW^{\uparrow}_{TOA,clr}$). Note that the clear-sky variables are not only calculated in the grid points where there is no cloud, but for every grid point giving the value that would be returned if no cloud existed.

The change to any of these variables due to emission of BBA is calculated by finding the difference between the FE scenario and nFE scenario. For example, the change in downward SW radiation at the surface can be found by:

$$\Delta SW^{\downarrow}_{Sfc} = SW^{\downarrow}_{Sfc,FE} - SW^{\downarrow}_{Sfc,nFE}. \quad (1)$$

Likewise, the difference in upwelling SW radiation at the TOA is given by:

$$\Delta SW^{\uparrow}_{TOA} = SW^{\uparrow}_{TOA,FE} - SW^{\uparrow}_{TOA,nFE}. \quad (2)$$

The radiative balance (RB) is defined as the difference between the radiation going into the system and the out-welling radiation at the TOA:

$$RB = SW^{\downarrow}_{TOA} + LW^{\downarrow}_{TOA} - SW^{\uparrow}_{TOA} - LW^{\uparrow}_{TOA}, \quad (3)$$

with a positive RB indicating a net increase in energy in the system. As such, the RB is generally positive during the day and negative at night. RB can similarly be calculated for clear-sky conditions:

$$RB_{clr} = SW^{\downarrow}_{TOA,clr} + LW^{\downarrow}_{TOA,clr} - SW^{\uparrow}_{TOA,clr} - LW^{\uparrow}_{TOA,clr}. \quad (4)$$

The change in radiative balance (ΔRB) is defined as the difference between a particular scenario and the control simulation (Ctrl) which has no aerosol effects. Given the incoming radiation at TOA is the same for all scenarios, ΔRB is equal to the difference in outgoing radiation, e.g.:

$$\Delta RB_{FE} = RB_{FE} - RB_{Ctrl} = (SW^{\uparrow}_{TOA} + LW^{\uparrow}_{TOA})|_{Ctrl} - (SW^{\uparrow}_{TOA} + LW^{\uparrow}_{TOA})|_{FE}, \quad (5)$$

making ΔRB_{FE} the instantaneous change to the net radiative flux due to the aerosol population. Similar calculations can be made for the clear-sky variables direct aerosol effects from changes to the cloud fields:

$$\Delta RB_{FE,clr} = RB_{FE,clr} - RB_{Ctrl,\,clr}. \quad (6)$$

BBA contains a high proportion of highly absorbing black carbon. The total radiative flux absorbed by the atmosphere can be calculated by finding the difference between fluxes into and out of the atmospheric column:

$$\mathrm{ASW} = \mathrm{SW}^{\downarrow}_{\mathrm{TOA}} + \mathrm{SW}^{\uparrow}_{\mathrm{Sfc}} - \mathrm{SW}^{\uparrow}_{\mathrm{TOA}} - \mathrm{SW}^{\downarrow}_{\mathrm{Sfc}}. \quad (7)$$

The 16 diagnostic radiative flux variables in the public version of WRF-Chem do not, however, provide enough information to fully disentangle the direct, semi- and indirect effects. Following Ghan et al. (2012) , we have added double calls to the radiation driver in each column to calculate an extra set of eight "clean-sky" variables ($\mathrm{SW}_{\mathrm{cln}}$ and $\mathrm{LW}_{\mathrm{cln}}$), which ignore the radiative effects of aerosol by setting the refractive index of all aerosol species to zero. With these extra diagnostics, the influence of aerosol effects on water uptake and absorption can be removed, and giving enough information to calculate the direct, semi- and indirect effects of biomass burning aerosol.

The direct SW radiative forcing ($\mathrm{SW}_{\mathrm{DIRECT}}$) is defined as the difference upwelling SW radiation at the TOA between the FE and nFE scenarios, with the radiative effects of water vapour removed by subtracting the "clean-sky" value:

$$\mathrm{SW}_{\mathrm{DIRECT}} = \Delta\mathrm{SW}^{\uparrow}_{\mathrm{TOA}} - \Delta\mathrm{SW}^{\uparrow}_{\mathrm{TOA,cln}}. \quad (8)$$

The indirect effect is calculated from the scenarios with no aerosol radiative interactions:

$$\mathrm{SW}_{\mathrm{INDIRECT}} = \Delta\mathrm{SW}^{\uparrow}_{\mathrm{TOA,nARI,cln}} = \mathrm{SW}^{\uparrow}_{\mathrm{TOA,nARI,cln}} - \mathrm{SW}^{\uparrow}_{\mathrm{TOA,Ctrl,cln}}. \quad (9)$$

Finally the semidirect effect is the remainder after taking away the direct and indirect effects:

$$\mathrm{SW}_{\mathrm{SEMIDIRECT}} = \Delta\mathrm{SW}^{\uparrow}_{\mathrm{TOA}} - \mathrm{SW}_{\mathrm{DIRECT}} - \mathrm{SW}_{\mathrm{INDIRECT}}. \quad (10)$$

Equivalent variables for LW radiation are also calculated. For more details and discussion, see Ghan et al. (2012) .

3.5 Statistical methods

For the radiative variables defined above, sample means and standard deviations (s), over the domain (ignoring the 5 outermost cells of each domain to avoid boundary issues) are calculated. An estimation of the uncertainty is given using the standard error (SE), following a similar method to Kolusu et al. (2015) . The SE is typically calculated by dividing the standard deviation by the square root of the number of independent data points N. However, the grid points of a model run show strong spatial and temporal autocorrelation. Assuming all grid points are independent results in an erroneously small SE, and therefore too high a significance. We therefore apply a correction factor k (Bence 1995):

$$\mathrm{SE} = \frac{s}{\sqrt{N}}k, \quad (11)$$

where

$$k = \frac{\sqrt{1+\rho}}{\sqrt{1-\rho}}. \quad (12)$$

The autocorrelation factor ρ varies from -1 (perfect anti-correlation) to 1 (perfect correlation). Spatial autocorrelation is estimated using the 2D Moran's-i method for neighbouring points. Thus if ρ is positive the correction acts to increase the SE. For the derived variables defined above, the net SE is estimated by adding the errors for the constituent variables in quadrature. For example, for $\mathrm{SW_{DIRECT}}$:

$$\mathrm{SE(SW_{DIRECT})} = \sqrt{\begin{array}{l}\mathrm{SE(SW^{\uparrow}_{TOA,FE})^2 + SE(SW^{\uparrow}_{TOA,nFE})^2} \\ \mathrm{+\,SE(SW^{\uparrow}_{TOA,FE,cln})^2 + SE(SW^{\uparrow}_{TOA,nFE,cln})^2}\end{array}} \quad (13)$$

This method reasonably estimates the uncertainty associated with domain averages, with uncertainty increasing appropriately as the number of grid cells decreases. However, it does not account for the systematic error associated with the boundary conditions of a nested domain, such as the 1km domain in this study.

4 Results

To assess how the WRF-Chem model simulates the regional impacts of BBA under various model setups and meteorological conditions, the analysis first evaluates the instantaneous direct radiative effects of aerosol–radiation interactions, temporarily ignoring the influence of clouds, in Sect. 4.1. Changes to the atmospheric stability, and how this in turn affects cloud formation and precipitation, are then presented (Sect. 4.2). The radiative balance is evaluated with regard to the cloud response to identify the semi-direct effects, testing the sensitivity of the cloud responses to resolution and convective parameterisation (Sects. 4.2 and Sect. 4.3). Finally in Sect. 4.4, aerosol–cloud interactions in the model are investigated. Output from the 5 km, 1 km domains and runs with no convective parameterisation over the 5 km domain are analysed, testing how much of an impact the lack of aerosol–cloud interactions in parameterised clouds has on the simulations.

4.1 Direct aerosol–radiative interactions and changes to atmospheric stability

Figure 3a–c shows the aerosol optical depth (AOD)

Total column AOD at 550 nm over the 5domain for the FE scenario. AOD in the FE model scenario is highest on the 14 September 2012, case study, with values between 0.8 and 1.2 over most of the domain . The the domain (Figure 3a). AOD on the other two days are is lower, between 0.4 and 1.0 . (Figure 3d–f show the vertical cross-section of extinction, averaged over the 5domain. b–c). The majority of the aerosol layer is in the lower 4 km of the model's atmosphere. Fresh emissions are injected at altitude during the local afternoon of each day (Figure 3d–f). Note that the AOD is non-zero in the nFE scenario, generally between 0.2 and 0.4, owing to contributions from anthropogenic emissions, dust and other long-range transported aerosol.

Figure 4 shows maps of the differences in clear-sky (ignoring cloud effects) radiation fluxes between the FE and nFE scenarios and time-series for the four main scenarios averaged over the 5 km domain for 14 September 2012. Similar figures for 18 and 23 September are included in the Supplement. ~~The calculations of the radiative fluxes are described in Appendix **??**.~~ Downwelling clear-sky SW radiation at the surface ($SW^{\downarrow}_{Sfc,\ clr}$) on 14 September 2012 is reduced by a maximum of $-109.5\,W\,m^{-2}$ compared to the nFE scenario (Fig. 4a and d). The clear-sky radiative effects on the 18 and 23 September case studies are qualitatively similar to 14 September. The difference in clear-sky radiative balance between the FE and nFE scenarios (ΔRB_{clr}) is negative (i.e. the aerosol layer has a net cooling effect at the TOA if cloud effects are ignored; Fig. 4b and e). ~~Although BBA is~~ Although the high BC content of BBA makes it highly absorbing, it has a net negative forcing because ~~it~~ the aerosol layer is predominantly over forest, which has a low albedo of 0.12 in the model. Averaged over 24 h, from dawn to dawn, the difference in RB_{clr} between the FE and nFE scenarios is $-5.0\,W\,m^{-2}$~~, equal to the net direct forcing the aerosol layer would have if there were no clouds in the domain.~~

~~Averaged over 24h~~. Over the same period, around $28\,W\,m^{-2}$ more SW radiation is absorbed by the atmospheric column in the FE scenario than the nFE scenario on 14 September (Fig. 4c and f), compared to 19 and $18\,W\,m^{-2}$ on 18 and 23 September respectively. The full tables of ~~domain averaged~~ domain-averaged radiative budgets are summarised in the Supplement. These results are comparable in magnitude and sign to a similar study conducted over the same case-study using the Met Office Unified Model (Kolusu et al., 2015). Overall, the net direct radiative effects of the aerosol layer are to reduce the total energy in the system, cool the surface and warm the lower troposphere.

4.2 Cloud responses to aerosol forcings

The presence of BBA in the simulations affects the dynamics and stability of the atmosphere, resulting in multiple changes to cloud formation and evolution. Changes can be observed in the vertical profile of the domain averaged potential temperature θ (Fig. 5a–c). On each day after local sunrise (approximately 10:00 UTC), the surface layer and lower

PBL is cooler in the FE scenario, and warmer between 850 and 500 hPa. The changes in θ are indicative of the aerosol layer stabilising the column, inhibiting the initiation of convection and reducing the amount of cloud (Fig. 5d–f). In all three case studies, there is a reduction in cloud formation in the FE scenario during the onset of precipitation around 18:00 to 21:00 UTC (14:00 to 17:00 local time). This change is less dramatic on the 14 September case study, as there was less precipitation on this day compared to the others. On 18 September, the presence of BBA consistently reduces all cloud types into the night. On 23 September, there is some displacement of peak precipitation in the FE scenario, resulting in longer cloud lifetimes and some periods with greater quantities of graupel and snow in the FE scenario. On 18 and 23 September, there is a reduction in nighttime high-altitude ice clouds in the FE scenario.

When the radiative effects of cloud fields ~~are included, the changes to the radiative balance due to~~ are considered for the analysis of model output, the radiative impacts of BBA are dramatically different (Fig. 6). ~~There is little cloud cover in the mornings but large convective clouds form in the afternoon.~~ In the mornings, before convective storms occur, ΔRB_{FE} is negative and ~~qualitatively~~ similar to the ~~behaviour of the~~ clear-sky case in Fig. 4e. In the afternoon, a strong positive forcing is observed in the FE scenario as there is much reduced cloud cover resulting in less SW radiation being reflected to space (see Fig. 3 in Supplement). This difference is greatest on 18 September (the case study with the most precipitation and cloud cover across the domain), peaking at $+70\,\mathrm{W\,m^{-2}}$. This cloud response more than counters the clear-sky direct radiative cooling of the aerosol over the same period.

Similar effects have been found by other modelling studies investigating the impact of BBA over continental regions. For example, Zhang et al. (2008) found a peak negative clear-sky forcing of $-8\ \mathrm{Wm^{-2}}$ over the highest AOD region in the Amazon, but with reductions in cloud cover resulting in localised surface forcings as high as $\approx 22\ \mathrm{Wm^{-2}}$ when changes to clouds were included. Kolusu et al. (2015) also show reduced all-sky forcing magnitude compared to clear-sky, and a decrease in precipitation due to BBA over the same SAMBBA period using the Met Office Unified Model (MetUM). In

Africa, BBA has also been shown to inhibit convection and cloud formation over land (Sakaeda et al., 2011; Tosca et al., 2013) .

At nighttime, there is a net negative forcing of approximately −10−10 W m^{-2} in the FE run on 18 and 23 September, which occurs because there are fewer ice clouds at high altitude in the FE scenario (Fig. 6). Cirrus clouds efficiently trap LW radiation, and so the thinner ice clouds in the FE simulations result in an increase in $LW^{\uparrow}_{TOA}$. Whilst we are unsure of the physical significance of this effect, the forcings due to changes in nighttime ice clouds are comparable in magnitude to the daytime forcings and so have an appreciable impact on the accumulated radiative balance.

To understand the differences in radiative budget between the clear-sky and all-sky variables, we need to first understand how the aerosol layer is affecting cloud formation. Changes can be observed in the vertical profile of the domain averaged potential temperature θ (Fig. 5a–c). On each day after local sunrise (approximately 10:00UTC) the surface layer and lower PBL is cooler in the FE scenario, and warmer between 850 and 500. The changes in θ are indicative of the aerosol layer stabilising the column, resulting in inhibition of the initiation of convection and reducing the amount of cloud (Fig. 5d–f).

On all three case studies, there was a reduction in cloud formation in the FE scenario during the onset of precipitation around 18:00 to 21:00UTC, reducing the average cloud cover. This change is least significant on the 14 September case study, as there was less precipitation on this day compared to the others. On 18 September, the presence of BBA consistently reduces all cloud types into the night. On 23 September, there is some displacement of peak precipitation in the FE scenario, resulting in longer cloud lifetimes and some periods with greater quantities of graupel and snow in the FE scenario. On 18 and 23 September, there is a reduction in nighttime high-altitude ice clouds in the FE scenario.

The 24 h averaged radiative budgets , for each scenario are summarised in Tables S1–S3 in the Supplement, with averages of basic meteorological variables in Tables S4–S6 in the Supplement. Comparing the FE scenario with the nFE and Ctrl scenarios shows the total aerosol impact. Differences between the nARI and Ctrl scenario are indicative of aerosol–cloud interactions. Comparing the 1domain with the same region from the

5domains highlights sensitivity to model resolution. On each of the case studies, more SW radiation is absorbed in the FE scenario over every domain. While $SW^{\downarrow}_{Sfc}$ is lower in the FE scenarioon all days, the net changes to radiative balance (RB) are less consistentand often negligible. The general , but the net forcing is less consistent. The reduction in cloud cover in the FE scenario adds a semi-direct warming effect which acts counter to the direct cooling of the aerosol, largely cancelling out any net impacton the RB. .

To quantify this semi-direct effect, we use the methodology of Ghan et al. (2012) to decompose the radiative forcing into SW and LW direct, semi- and indirect effects. These are presented for each of the case studies in Figure 7. The diurnally averaged SW_{DIRECT} is -5.26 ± 1.26, -3.34 ± 2.68 and -3.65 ± 1.87 Wm^{-2} respectively on the 14, 18 and 23 September case studies respectively. When decomposed, the positive change in radiative balance seen in the afternoon of figure 6d–f is due to the positive $SW_{SEMIDIRECT}$. Diurnally averaged $SW_{SEMIDIRECT}$ is 3.51 ± 1.19, 6.06 ± 1.46 and 5.18 ± 1.82 on the three case studies respectively. At nighttime, the reduction of nighttime cirrus clouds in the FE scenario results in a negative $LW_{SEMIDIRECT}$ on the 18 and 23 September case studies of -4.54 ± 0.96 and -2.80 ± 1.07 Wm^{-2} respectively. In all three case studies, the indirect effects are small relative to the direct and semidirect forcings, with signal typically smaller than the estimated error.

Although the broad conclusions using this extended analysis are similar and roughly equivalent to the analysis of the change in radiative balance, the quantification of different forcings enables greater understanding of the processes and impacts being investigated. However, the results are still specific to the case studies and model setup being studied, and should not be extrapolated due to the small scope of the study.

4.3 Sensitivity to model resolution and a convective parameterisation

There are only major differences to $SW^{\downarrow}_{Sfc}$ and RB the radiative forcings between the 5 km and 1 km domains on the 18 September case study because there is limited cloud cover and precipitation this is the only day with extensive cloud cover over the 1 km domain regionon the other two case studies. In the 1 km domain, convection initiates faster and more energetically in the early afternoon compared to the same runs on the 5 km domain. Clouds are better resolved, covering a smaller portion of the total domain. Therefore, a greater amount of SW radiation reaches the ground in the 1 km domain compared to the same region of the 5 km domain (Fig. **??**a). Over 24h, approximately 18more SW radiation reaches the surface in the Ctrl scenario in the 1S5a in supplement).

The analysis of radiative forcings in the 1km domain is limited by its small size. As the region of the 5km domain the 1km domain covers is not representative of the whole, displacements of clouds in the 5km domain can have a large impact on the net forcing in the 1km domain, and signals are correspondingly noisier. This is highlighted by the large errors of variables calculated over the 1km domain region from the 5km domain (Figure 11). However, sensitivities to the model resolution can be inferred from how forcings differ over the same area between the 1km and 5km simulations. $SW_{SEMIDIRECT}$ is weaker in the 1km domain compared to the same region of the 5domain 5km domain (Figure 11a, c and e), due to the smaller, more cellular structure of convective clouds in the 1km domain. Assuming the representation of convective clouds is more realistic in the 11km domain, the difference between the two domains could be an indication suggests that the Grell-3-D parameterisation, even with subsistence spreading, is not suitable for predicting the semi-direct effect at this resolution. Note "suitability" here is only in regard to the simulation of subsidence spreading, may resolve clouds and their radiative properties too poorly for the accurate simulation of semi-direct effect. The convective parameterisation may still improve the model performance under other metrics, such as precipitation. effects.

4.4 Sensitivity to convective parameterisation

To separate changes due to the aerosol fields from effects due to the convective parameterisation, a set of four scenarios without the Grell-3-D convective parameterisation over the 5 km domain were run for the 18 September case study. Total cloud cover is marginally reduced in the local afternoon without the convective parameterisation. However, the clearest effect is that there is a large reduction in ice clouds at night (Fig. **??**) Peak precipitation rates (which occur between 20:00 and 21:00 UTC on 18 September) for the

FE and FE_nCU scenarios are compared with data from the Tropical Rainfall Measurement Missions (TRMM) 3B42 product (Huffman et al., 2001) in Figure 9. In the FE scenario, precipitation is less intense and covers a larger area, whereas in the 1km domain and FE_nCU scenario, precipitation organises in isolated convective cells with a greater portion of the domain receiving no precipitation. Deep convective towers are smaller and take longer to form without a convective parameterisation, delaying the onset of and reducing total precipitation . The FE scenario correspondingly has a larger portion of the domain covered by cloud at any one time. However, total precipitation over both domains is greater in the FE scenario than the FE_nCU scenario. Although the TRMM product is relatively coarse (with a grid-spacing of 0.25 degrees), precipitation can be seen to occur in small convective cells, suggesting the FE_nCU scenario has slightly increased precipitation at night compared to nFEis more realistic.

The spread of accumulated precipitation in the FE_nCU , whereas the FEscenario has less precipitation over the entire day compared to nFE scenario is closer to that of the TRMM data set than the FE scenario (Fig. ??c). 10), with more grid cells receiving little to no precipitation, and a greater proportion of total precipitation being received from grid cells with high precipitation. The average accumulated precipitation over the 5km domain on 18 September is 2.30mm, 1.43mm and 1.49mm for the FE, FE_nCU and TRMM dataset respectively. Thus, the model scenarios without convective parameterisation perform better for both total accumulated precipitation and distribution over the domain for this case study.

The runs without convective parameterisation have reduced deep convection in the local afternoon, resulting in more downwelling SW radiation at the surface (Fig. ??aS6a in supplement). The change in surface SW radiation at local afternoon is approximately twice as sensitive to the use of convective parameterisation as to the presence of BBA. The reduction of high-altitude nighttime ice clouds Overall, the afternoon peak semidirect effect is weaker when running without the convective parameterisation creates an extremely strong negative forcing at night as more LW radiation is lost to space in the runs without convective parameterisation (Fig. ??b). Overall, RB is more sensitive to whether or not a convective parameterisation is used than it is to the presence of aerosol or the horizontal resolution, with diurnally averaged reduction of approximately 20between scenarios with and without convective parameterisation(Table S2 in the Supplement). This change is largely convective parameterisation in both the 5km and 1km domain (Figure 11). The diurnally averaged value is 3.61 ± 8.55, compared with 6.06 ± 1.46 Wm^{-2} over the same period from the runs with convective parameterisation. There is also no negative nighttime LW semidirect forcing, due to the reduction in lack of high-altitude nighttime clouds in the runs without convective parameterisation.

Changes in cloud cover due to the presence of aerosol have a smaller impact on the net radiative balance on the 1domain, resulting in lower magnitude of changes to the radiative budget (Fig. **??**). The 1domain is sensitive to the boundary conditions from the 5domain, highlighted by the similarity in nighttime radiative balance changes due to ice clouds between the 1domain and same region covered by the 5domain (Fig. **??**a and b). Precipitation is strongly suppressed over the 1region in the 5domain in the scenarios where convective parameterisationis turned off (Fig. **??**c and d). More precipitation is produced in the 1domain for these runs, although still less than in the scenarios with convective parameterisation over the 5domain, highlighting the importance of the boundary conditions from the 5in determining the behaviour of the 1domain. In contrast, Even with aerosol–cloud interactions being present in simulations without convective parameterisation in the 5km domain, the runs with convective parameterisation turned on produce similar amounts of precipitation at both resolutions, implying that a convective parameterisation is needed over the 5domain to produce reasonable levels of precipitationindirect effects are small with no signal above noise. The strong sensitivity of the semidirect effect to use of convective parameterisation, combined with low indirect forcings in this region, highlights the need to better develop parameterisations that can accurately simulate aerosol feedbacks on cloud formation.

4.4 Evidence of aerosol–cloud interactions

To show that BBA are activating to become cloud droplets in the model, we estimate the maximum supersaturation S_{max} in each column of the model with cloud by comparing the maximum droplet number in a vertical column ($N_{\text{d, max}}$) with the CCN concentrations at the base of the cloud. For example, if $N_{\text{d, max}} > \text{CCN}_{0.02}$ but $N_{\text{d, max}} < \text{CCN}_{0.1}$, then S_{max} must be between 0.02 and 0.1 %. This approach implicitly assumes that peak S_{max} is at cloud-base, which is a reasonable assumption given the representation by the Abdul-Razzak and Ghan (2002) activation parameterisation, but not in a parcel model or reality.

Figures 12 and 13 show an increase in $N_{\text{d, max}}$ and corresponding decrease in S_{max} in the FE scenario, consistent with increased CCN activation.

Because the Abdul-Razzak and Ghan (2002) parameterisation estimates the activated fraction based on a Gaussian distribution of the updraft velocity (w), $N_{\text{d, max}}$ and S_{max} are both implicitly sensitive to w. However, most clouds over this period and region were convective and parameterised on the 5 km domain, meaning the subgrid variation in vertical velocities is unresolved. To identify any aerosol–cloud interactions in convective systems, simulations at cloud resolving scales must be run. Comparing clouds in the same region of the 5 km and 1 km domain, S_{max} and $N_{\text{d, max}}$ are both approximately twice as high in the 1 km simulations in both FE and nFE scenarios, but clouds are smaller with less horizontal spread implying higher resolved updraft velocities (Fig. 13). More CCN per unit volume are activated in the 1 km domain due to w being explicitly resolved. However, there is no corresponding increase in scattered radiation, as may be expected from the first indirect effect, because deep convective clouds are already optically thick. Cloud optical depth is most sensitive to an increase in droplet number if the liquid water path is low (Twomey, 1974).

Although CCN are activated in cloud within the model, the net radiative balance was largely not sensitive to aerosol–cloud interactions during the case studies. In Fig. 6d to f, the nARI scenario, with full fire emissions but no aerosol–radiation interactions, closely follows the Ctrl simulation, indicating that the aerosol–cloud interactions by themselves have little impact on the radiative budget. , resulting in small indirect effects (Figures 7 to 11). We believe the small indirect effect is because most of the cloud in the domain is a result of deep convection, which tends to be optically thick even without the inclusion of additional aerosol as described through the Twomey (1974) effect. An exception is on the morning of 23 September 2012 between 11:00 and 14:00 UTC, when there is enhanced cooling in both the FE and nARI scenarios (Fig. 6ca small negative $SW_{INDIRECT}$ forcing (Figure 7f). The large central region in Fig. 12a shows high droplet number in the FE scenario, whereas there is little cloud over the same region of the nFE run (Fig. 12c). This cloud is a ground-level radiation fog, which forms in the high morning humidity of the forest and is enhanced by the added presence of CCN from BBA. This example is the only period of the case studies where BBA aerosol influences the optical properties of resolved clouds in the 5 km domain, simulating the first indirect effect and reducing downwelling SW radiationproducing a $SW_{INDIRECT}$ forcing of greater magnitude than the simultaneous $SW_{SEMIDIRECT}$ forcing.

5 Conclusions

WRF-Chem model simulations for three 36-hour case studies over nested 5 domains at 5km and 1 km nested domains horizontal 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. The These nested domains were driven by model fields from a WRF-Chem simulation at 25 km 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 5 km domain, using the recommended subsistence spreading option for running at this scale (Grell and Freitas, 2014). Different scenarios were conducted to probe how effectively the impacts are modelled in WRF-Chem and test sensitivity to model resolution and use of convective parameterisation over the 5 km domain. As a result of the small size of domains, short case-studies, and single model version, the results from this study 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).

~~The~~

Over the 5km domain, on the 18 September case study, the shortwave direct effects of BBA particles over the region have a ~~negative instantaneous forcing at the top of the atmosphere, despite being radiatively absorbing, due to the aerosol layer being over a~~ negative forcing of -3.34 ± 1.47 ~~low albedo surface. There is a strong cooling at the surface coupled with a warming in the lower troposphere, stabilising the atmospheric column and driving a~~ $\mathrm{Wm^{-2}}$, which is countered by a positive semi-direct ~~warming effect whereby the presence of aerosol inhibits cloud formation, reduces cloud cover and increases the amount of solar radiation reaching the ground. This result is similar to findings by, for example, Zhang et al. (2008) who find that the semi-direct effect in this region tends to be positive, partially or completely cancelling out the negative direct forcing~~ effect of 6.06 ± 1.46 $\mathrm{Wm^{-2}}$. The shortwave indirect effect is a relatively small 0.266 ± 1.06 $\mathrm{Wm^{-2}}$. Longwave semi- and indirect effects are larger on this case study day, with values of -4.54 ± 0.96 $\mathrm{Wm^{-2}}$ and -1.53 ± 0.69 $\mathrm{Wm^{-2}}$ 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 -2.67 ± 1.27 $\mathrm{Wm^{-2}}$.

Further nested simulations at 1 km grid spacing were run to explicitly resolve convection. In the finer resolution domain, deep convective clouds have much reduced horizontal spread but higher cloud droplet number within cloud compared to the 5 km domain. The reduction in cloud cover due to the presence of BBA over the 1 km domain therefore has a reduced impact on the net radiative balance and the magnitude of the semi-direct effect is smaller compared to the same region of the 5 km domain. The modelled semi-direct effect is thus highly sensitive to the model resolution. ~~Any changes to the radiative balance due to~~ Indirect effects from resolved aerosol–cloud interactions in the ~~1domain are masked by this reduction of~~ 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. ~~WRF-Chem (at the time of study) neglects fractional cloud cover within grid cells (Zhang, 2008) , which may be causing an overestimation of a semi-direct effect over~~

the 5domain. A better representation of fractional cloud cover, linked with the radiation and convective parameterisations, is likely needed to better evaluate this forcing at the regional scale.

Simulations run without a convective parameterisation on the 5 km domain had reduced daytime convection and precipitation, indicating some parameterised cloud is likely needed at this scale to produce reasonable precipitation. Turning off the convective parameterisation reduces ice cloud cover over night, allowing more LW radiation to escape resulting in a negative instantaneous forcing of around −50. While this result may be of little practical significance, the large magnitude of the sensitivity Comparisons with the TRMM dataset suggest that the 5km simulations without convective parameterisation organise the structure of convective systems better, as isolated cells rather than widespread precipitation. The positive semi-direct effect is lower in the scenarios without convective parameterisation due to the clouds being more cellular, but the negative nighttime longwave semidirect is also diminished. The net forcing from the scenarios with no convective parameterisation on the 18 September case-study is 1.04 ± 0.78 W m^{-2}. The large sensitivity to use of convective parameterisation highlights the uncertainties with simulating aerosol–radiation–cloud interactions in this regime.

There was evidence that The BBA CCN efficiently activate in the region, modelled as 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. However, there are no aerosol–cloud interactions in parameterised clouds within WRF-Chem v3.4.1 used in this study. 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 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 1 km and no cumulus parameterisation at 5 km also showed show minimal indirect effects, likely due to the deep convective clouds being optically thick and therefor not sensitive to increased droplet number. The model does not produce an aerosol "cloud-invigoration" effect, as described by Rosenfeld et al. (2008) , both in simulations with and without a convective parameterisation. However, seen by Rosenfeld et al. (2008) and Fan et al. (2013) , although this may be because aerosol-ice aerosol–ice nucleation processes are required to reproduce this effect. Overall, we find the these findings suggest that resolving indirect processes in parameterized cloud is of secondary importance for the current case studies. Instead, representation of semi-direct effect has a much aerosol feedbacks has a greater impact on the net radiative balance than any indirect effects.

This case study investigation uses relatively short model simulations that do not have time to fully adjust to the aerosol forcings. The short runs also result in many of the results not being statistically significant according to Student t test criteria or similar, owing to a limited number of independent data points. The innermost 1domain would also benefit from being larger so as to be more consistently representative of the 5domain. A more robust estimate of the aerosol forcings would require long term simulations over multiple months. The high computational expense of running with sufficiently high resolution to resolve the effects investigated in this study would be considered when undertaking studies for this purpose. The shorter case-studies at high-resolution were prioritised over a longer, low-resolution setup for the purpose and scope of the current investigationand associated uncertainties.

Simulating convective systems with the effects of aerosol included, 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; **?**) (e.g. Grell and Freitas, 2014; Berg et al., 2015) . The semi-direct effects are impossible to quantify reliably in this WRF-Chem setup due to this high sensitivity to the use of convective parameterisation and model resolution. More coordinated development of convective parameterisations with aerosoland radiation mechanisms is needed to have more certainty of these impacts and produce reasonable quantitative estimates. coordination between parameterized and explicit treatments of aerosol, cloud and radiation interactions is needed in order to make modelling of these processes at the transition between fully parameterised and fully explicit schemes more consistent. To constrain the simulation of these interactions, in-situ observations of aerosol size distribution and composition properties, measured before, during and after cloud processing need to be considered alongside remote sensing observations of changes to cloud cover and net radiation in regions of high aerosol loading. 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.

6 Calculating the radiative effects of an aerosol layer over WRF-Chem domains

The following is a description of the calculations used to evaluate the changes to the radiative balance due to the aerosol layer. It is related to and builds on other studies, such as Zhang et al. (2014) , but includes to the changes to the long-wave (LW) as well as short-wave (SW) spectrum. WRF-Chem provides a set of 16 diagnostic variables for assessing simulated radiative fluxes, all given in units of . These are first split into SW and LW portions of the spectrum, and can be calculated at the top of the atmosphere (TOA) or at ground level (e.g. SW_{TOA} or SW_{Sfc} respectively) in either the up or down direction ($SW^{\uparrow}_{TOA}$ or $SW^{\downarrow}_{TOA}$). Finally, they can be calculated for "all-sky", including the effects of clouds ($SW^{\uparrow}_{TOA}$); or for "clear-sky", ignoring the effects of clouds ($SW^{\uparrow}_{TOA,clr}$). Note that the clear-sky variables are not only calculated in the grid points where there is no cloud, but rather for every grid point giving the value that would be returned if no cloud existed.

The change to any of these variables due to the aerosol layer can be calculated by finding the difference between the FE scenario and a control scenario. For example, the change in downward SW radiation at the surface can be found by:

$$\Delta SW^{\downarrow}_{Sfc,FE} = SW^{\downarrow}_{Sfc,FE} - SW^{\downarrow}_{Sfc,Ctrl}.$$

The radiative balance (RB) is defined as the difference between the radiation going into the system and the out-welling radiation at the TOA:

$$RB = SW^{\downarrow}_{TOA} + LW^{\downarrow}_{TOA} - SW^{\uparrow}_{TOA} - LW^{\uparrow}_{TOA},$$

with a positive RB indicating a net increase in energy in the system. As such, the RB is generally positive during the day and negative at night. RB can similarly be calculated for clear-sky conditions:

$$\mathrm{RB_{clr}} = \mathrm{SW^{\downarrow}_{TOA,clr}} + \mathrm{LW^{\downarrow}_{TOA,clr}} - \mathrm{SW^{\uparrow}_{TOA,clr}} - \mathrm{LW^{\uparrow}_{TOA,clr}}.$$

The change to the radiative balance due to clouds can then be inferred as the difference between the total radiative balance and the clear-sky case:

$$\mathrm{RB_{cld}} = \mathrm{RB} - \mathrm{RB_{clr}}.$$

The change in radiative balance (ΔRB) is defined as the difference between a particular scenario and the control simulation (Ctrl) which has no aerosol effects. Given the incoming radiation at TOA is the same for all scenarios, ΔRB is equal to the difference in outgoing radiation. For example for the fire emissions (FE) scenario:

$$\Delta\mathrm{RB_{FE}} = \mathrm{RB_{FE}} - \mathrm{RB_{Ctrl}} = (\mathrm{SW^{\uparrow}_{TOA}} + \mathrm{LW^{\uparrow}_{TOA}})|_{\mathrm{Ctrl}} - (\mathrm{SW^{\uparrow}_{TOA}} + \mathrm{LW^{\uparrow}_{TOA}})|_{\mathrm{FE}},$$

making $\Delta\mathrm{RB_{FE}}$ the instantaneous change to the net radiative flux due to the aerosol population. Similar calculations can be made for the clear-sky and cloud-only variables to separate the direct aerosol effects from changes to the cloud fields:

$$\Delta\mathrm{RB_{FE,clr}} = \mathrm{RB_{FE,clr}} - \mathrm{RB_{Ctrl,\,clr}}; \qquad \Delta\mathrm{RB_{FE,cld}} = \mathrm{RB_{FE,cld}} - \mathrm{RB_{Ctl,\,cld}}.$$

The clear-sky variable $\Delta\mathrm{RB_{FE,clr}}$ is somewhat equivalent to the instantaneous forcing due to aerosol–radiation interactions ($\mathrm{RF_{ari}}$) as defined by the IPCC (IPCC, 2013) , whereas $\Delta\mathrm{RB_{FE}}$ gives an indication of the effective radiative forcing with aerosol–radiation aerosol–cloud interactions, after short-term adjustments ($\mathrm{ERF_{ari+aci}}$). However, given the limited spatial and temporal scope of the study and the fact that large scale circulation is unaffected, these calculations should not be seen as robust calculations of the radiative forcing.

BBA contains a high proportion of highly absorbing black carbon. The total radiative flux absorbed by the atmosphere can be calculated by finding the difference between fluxes into and out of the atmospheric column. For example, for SW radiation:

$$\mathrm{ASW} = \mathrm{SW}^{\downarrow}_{\mathrm{TOA}} + \mathrm{SW}^{\uparrow}_{\mathrm{Sfc}} - \mathrm{SW}^{\uparrow}_{\mathrm{TOA}} - \mathrm{SW}^{\downarrow}_{\mathrm{Sfc}}.$$

For all derived radiative variables defined in this appendix, the average effect over a domain can given by calculating a mean across the domain, as well as over a period of time, whilst a measure of the spatial variation can be given by the standard deviation.

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Table 1. 8 bin MOSAIC size grid.

Bin number	particle dry diameter (nm)
1	39.0625–78.125
2	78.125–156.25
3	156.25–312.5
4	312.5–625
5	625–1250
6	1250–2500
7	2500–5000
8	5000–10 000

Table 2. Summary of physical parameterisations and other options used in parent and nested simulations.

Option	d01, 25 km parent	d02, 5 km nest	d03, 1 km nest
Horizontal Grid Cells ($n_i \times n_j$)	226×196	151×171	141×116
Horizontal grid spacing	25 km	5 km	1 km
Cumulus	Grell 3-D	Grell 3-D	None
Subsistance spreading	1	3	NA
Dynamical timestep (s)	120	30	6
Chemistry time-step (min)	2	1	1
Boundary conditions	ECMWF/MACC	offline, ndown	online, no feedback

Table 3. Summary of scenarios: fire Emissions (FE), no Fire Emissions (nFE), fire emissions with no Aerosol–Radiation Interactions (nARI), and a Control simulation with no fire emissions or aerosol–radiation interactions (Ctrl). ~~Scenrios~~ Scenarios without convective parameterisation on the 5 km domain (FE_nCU, nFE_nCU, nARI_nCU and Ctrl_nCU) were run only for the 18 September case study.

Scenario	Fire emissions	Aerosol–radiative feedback	Convective parameterisation on 5 km domain
FE	On	On	On
nFE	Off	On	On
nARI	On	Off	On
Ctrl	Off	Off	On
FE_nCU	On	On	Off
nFE_nCU	Off	On	Off
nARI_nCU	On	Off	Off
Ctrl_nCU	Off	Off	Off

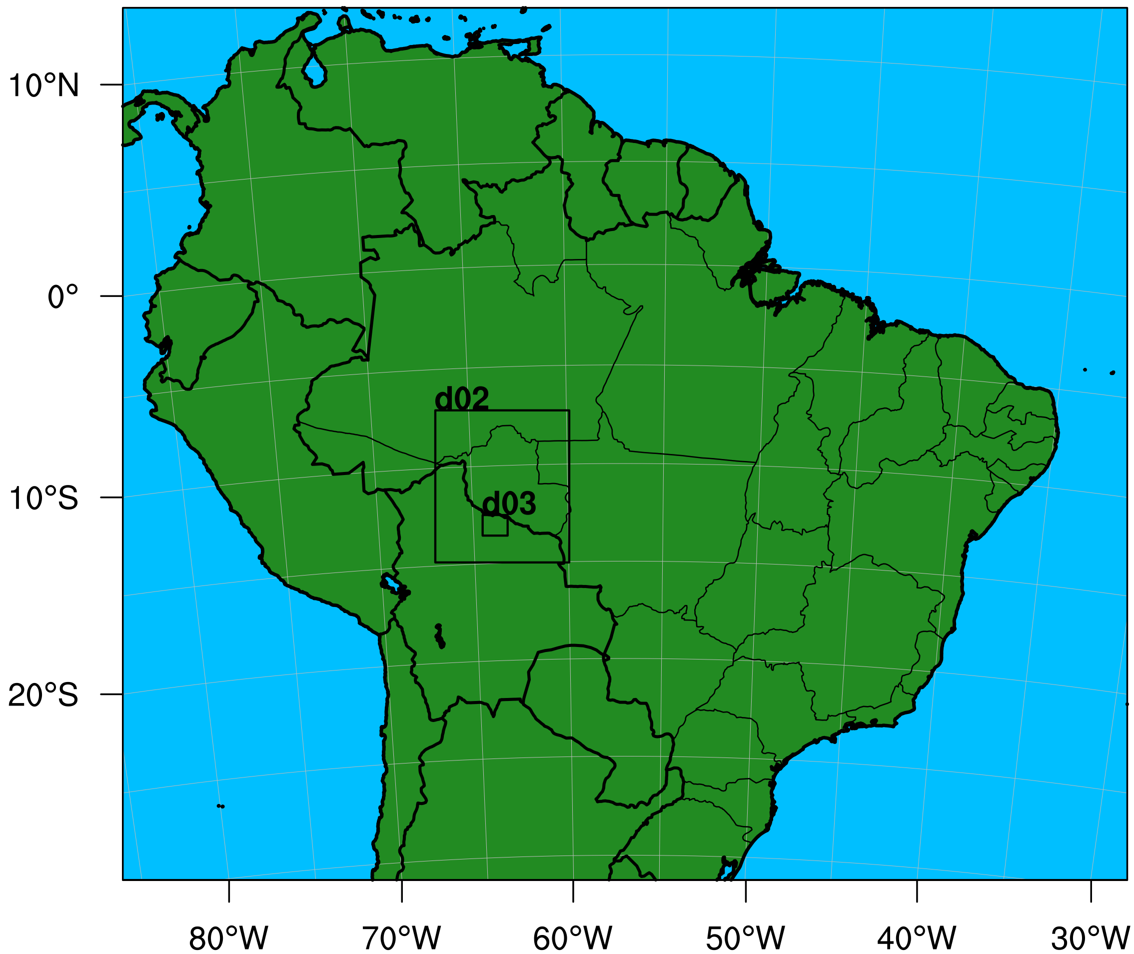

Figure 1. Map of domains used for study. Outer map of parent domain with 25 km horizontal grid spacing, with squares showing extents of 5 km (d02) and 1 km (d03) nests.

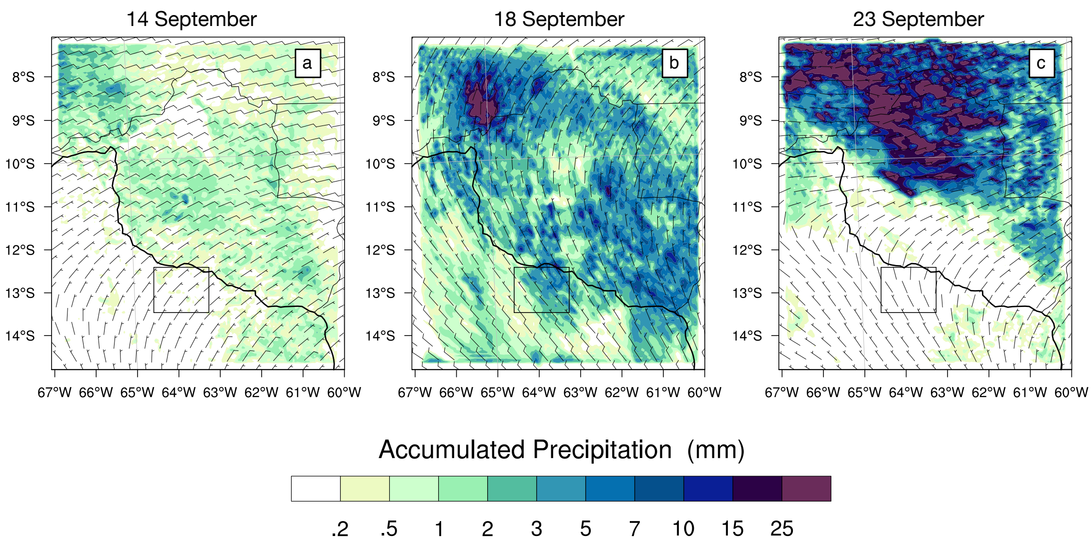

Figure 2. Maps of total precipitation and wind vectors at 700 hPa from the Ctrl scenario, ~~averaged~~ accumulated over 24 h from dawn to dawn for each case study period over the 5 km domain, with black box outlining the 1 km domain. **(a)** from 10:00 UTC 14 September, **(b)** from 10:00 UTC 18 September 2012; and **(c)** from 10:00 UTC 23 September.

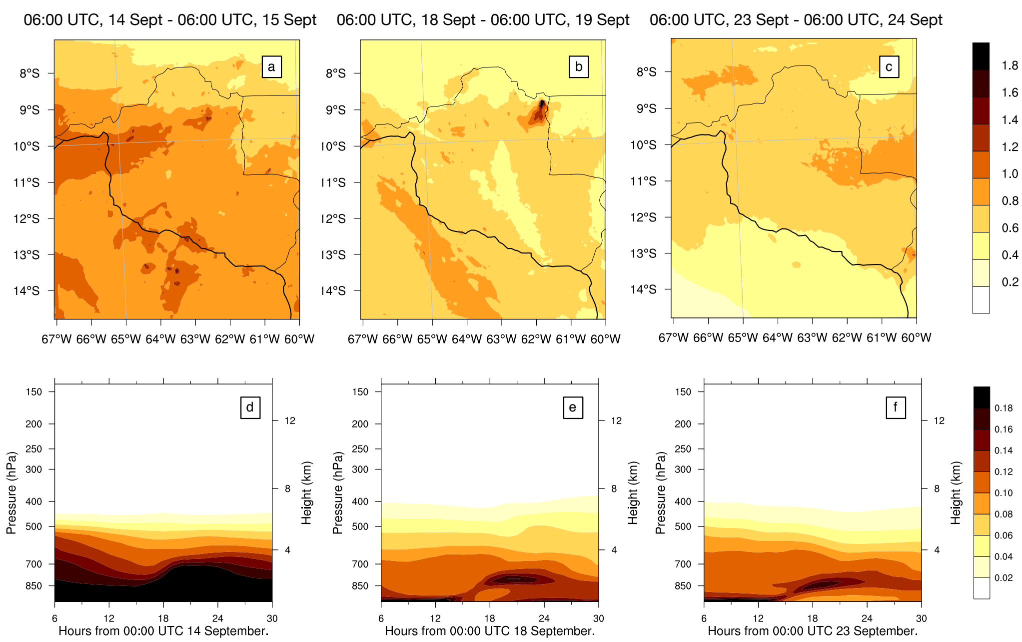

Figure 3. (a–c) Temporally averaged ~~horizontal maps of~~ column AOD at 550 nm from 5 km domain. **(d–f)** Vertical profiles of extinction coefficient b_{ext} at 550 nm (km^{-1}), averaged over interpolated pressure level planes at 25 hPa intervals. All data from FE scenario, **(a)** and **(d)** from 06:00 UTC 14 September; **(b)** and **(e)** from 06:00 UTC 18 September; **(c)** and **(f)** from 06:00 UTC 23 September 2012.

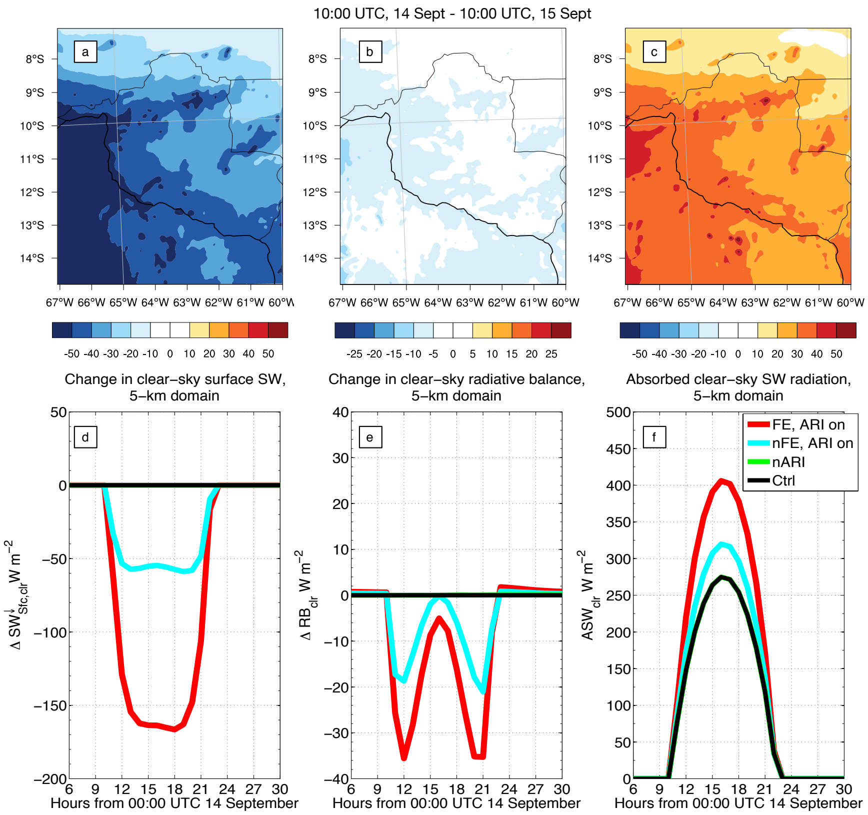

Figure 4. Maps and time-series of changes to clear-sky radiation fields (ignoring the effects of clouds) due to BBA over 14 September 2012. **(a–c)** show maps over 5 km domain of the difference between the FE and nFE scenarios, averaged over 24 h, from dawn to dawn, between 10:00 UTC 14 and 10:00 UTC 15 September. **(d–f)** show model output averaged over the 5 km domain at each hour of simulation for the FE, nFE, nARI and Ctrl scenarios; with **(d)** and **(e)** plotting difference from Ctrl scenario. **(a)** and **(d)** change in downwelling SW radiation at the surface $\Delta SW^{\downarrow}_{Sfc,\ clr}$. **(b)** and **(e)** change in radiative balance (ΔRB_{clr}) at top of the atmosphere (TOA). **(c)** and **(f)** SW radiation absorbed by the atmospheric column (ASW_{clr}). Calculations of derived variables are explained in Appendix**??**. All variables are in units .

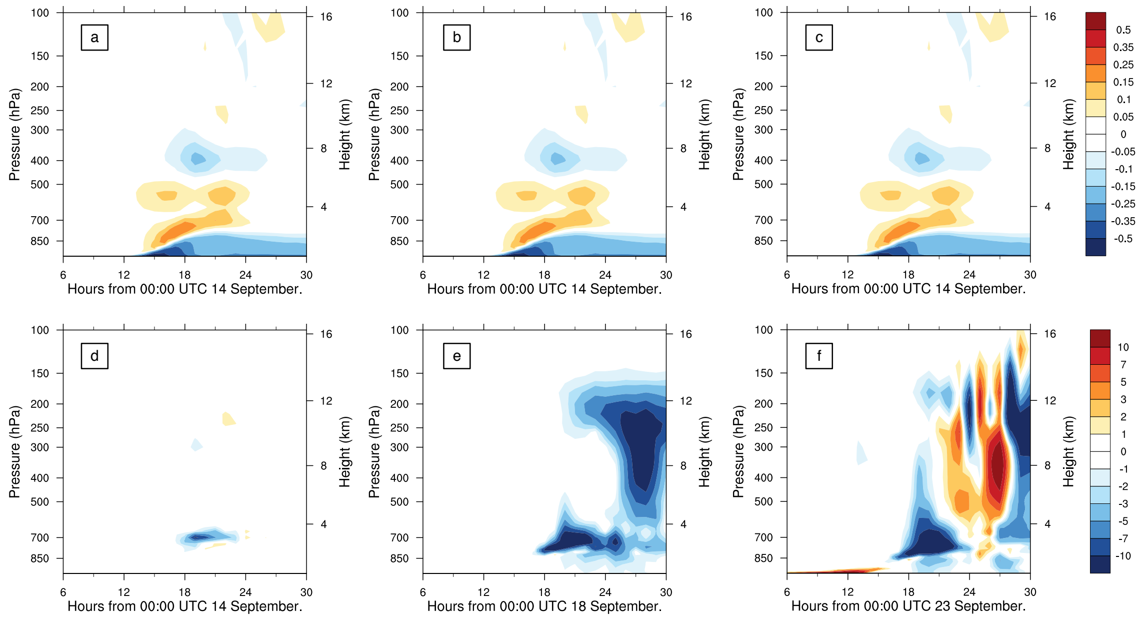

Figure 5. ~~Changes to radiation balance at TOA over the 5domain for each of the three case study days, including the effects of clouds. **(a–c)** show maps over 5domain of the difference between the FE and nFE scenarios ($RB_{FE} - RB_{nFE}$), averaged over 24h, from dawn to dawn, from 10:00UTC for **(a)** 14 September, **(b)** 18 and **(c)** 23 September 2012. **(d–f)** time-series of change in radiative balance from Ctrl scenario (ΔRB) averaged over the 5domain at each hour of simulation. Calculations of derived variables are explained in Appendix **??**.~~Difference plots between the FE and nFE scenarios with data averaged over interpolated pressure levels with 20 hPa spacing~~, excluding the 10 grid cells at each domain border to remove the influence of boundary conditions~~. **(a–c)** difference in potential temperature θ (K), **(d–e)** difference in sum of all cloud variables (QCLOUD + QRAIN + QICE + QGRAUP + QSNOW; mg kg^{-1}). **(a)** and **(d)** from 06:00 UTC 14 September; **(b)** and **(e)** from 06:00 UTC 18 September; **(c)** and **(f)** from 06:00 UTC 23 September 2012.

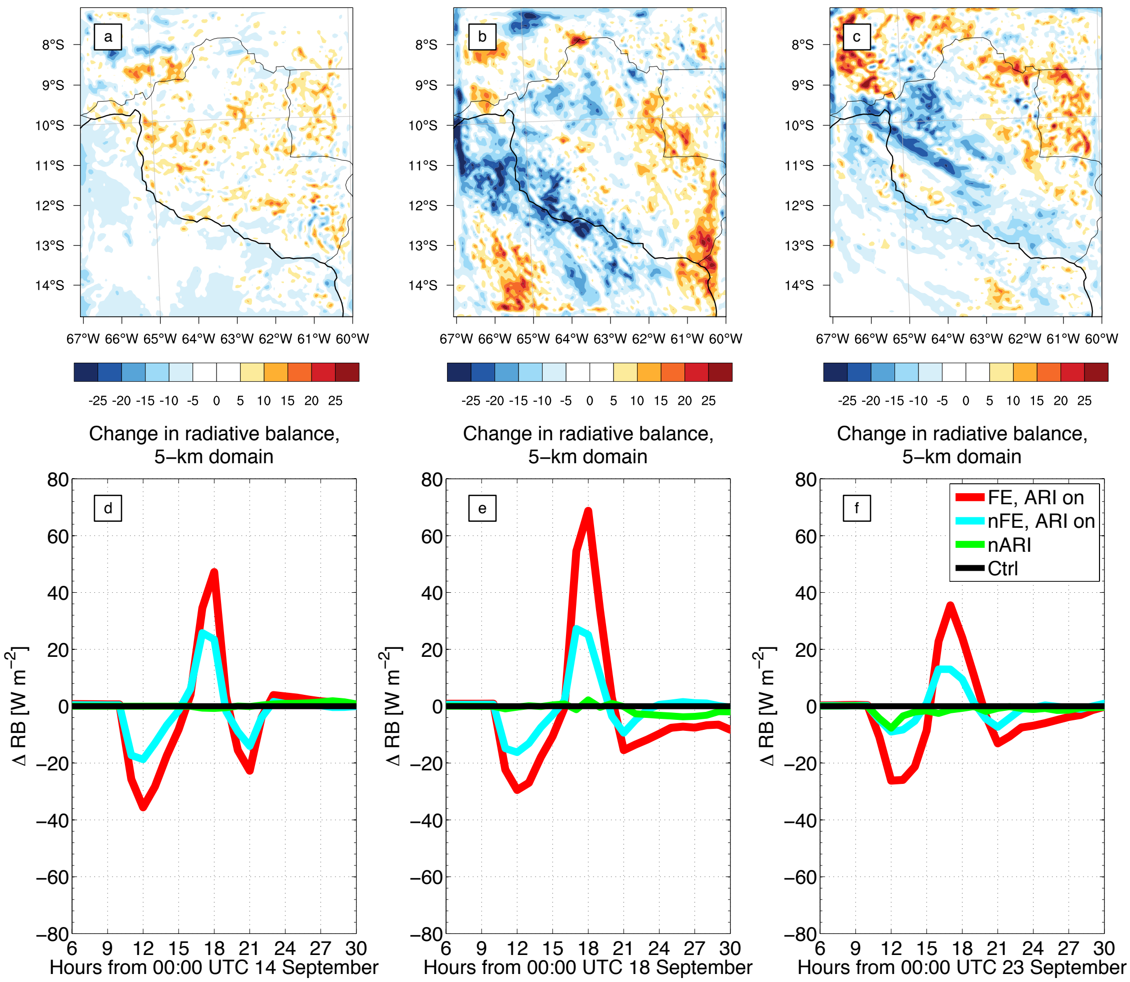

Figure 6. Changes to radiation balance at TOA over the 5 km domain for each of the three case study days, including the effects of clouds. **(a–c)** show maps over 5 km domain of the difference between the FE and nFE scenarios ($RB_{FE} - RB_{nFE}$), averaged over 24 h, from dawn to dawn, from 10:00 UTC for **(a)** 14 September, **(b)** 18 and **(c)** 23 September 2012. **(d–f)** time-series of change in radiative balance from Ctrl scenario (ΔRB) averaged over the 5 km domain at each hour of simulation. Calculations of derived variables are explained in Section 3.4

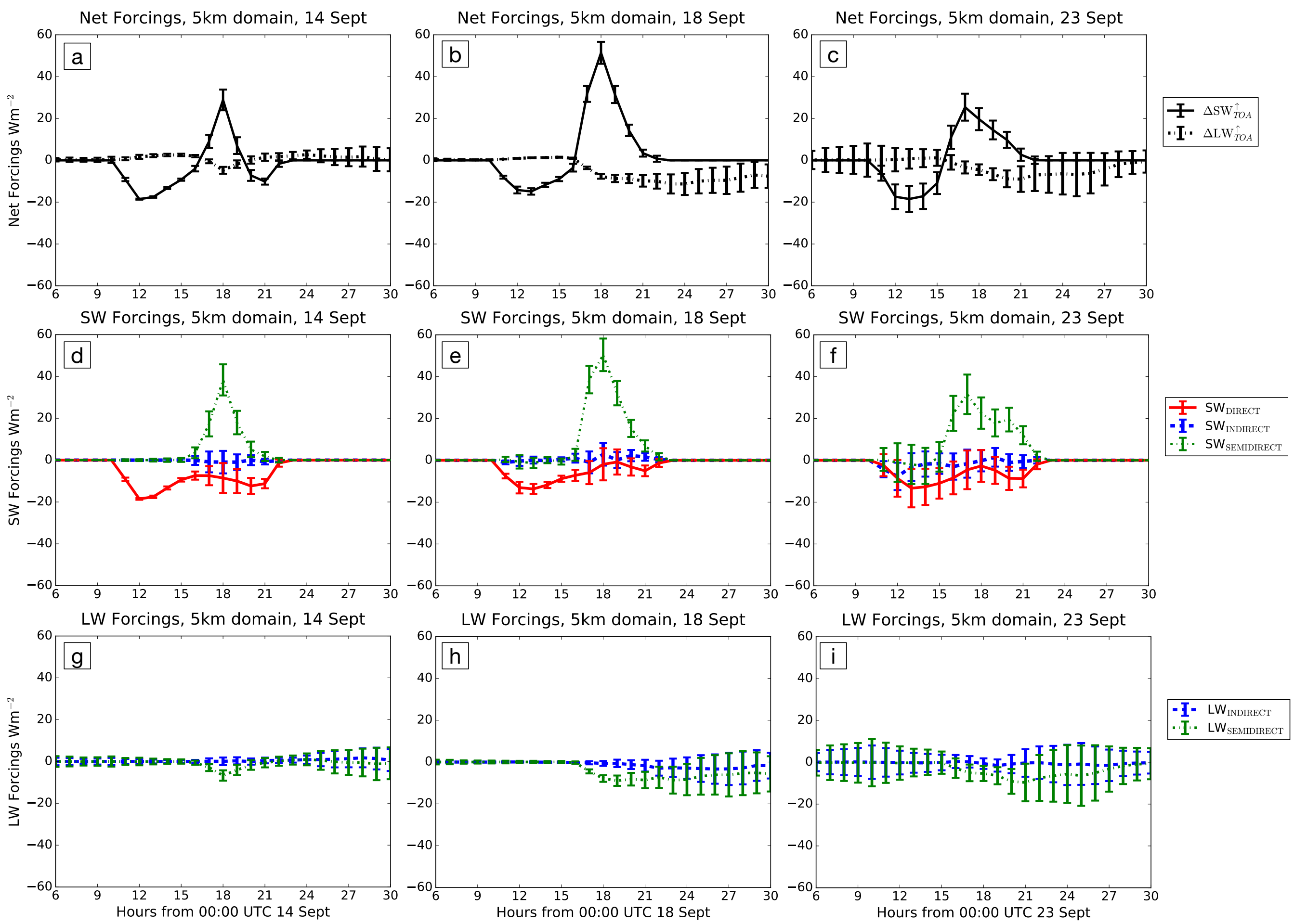

Figure 7. ~~Comparing changes to downwelling SW radiation and radiative balance due to horizontal resolution for~~ Radiative forcings over 5km domain over each case study day: 14 September (left), 18 September ~~2012. **(a)** Difference in downwelling SW radiation at surface between the 1domain~~ (middle) and ~~same region covered by the 5domain~~ 23 September (~~$\Delta SW^{\downarrow}_{Sfc}$~~right). ~~**(b)** Difference in~~ Instantaneous net ~~radiative balance between 1domain~~ forcings (top) and ~~same region covered by 5domain~~ decomposition into shortwave (~~ΔRB~~middle) and longwave (bottom) direct, semidirect and indirect forcings. LW_{DIRECT} is small and so is not plotted.

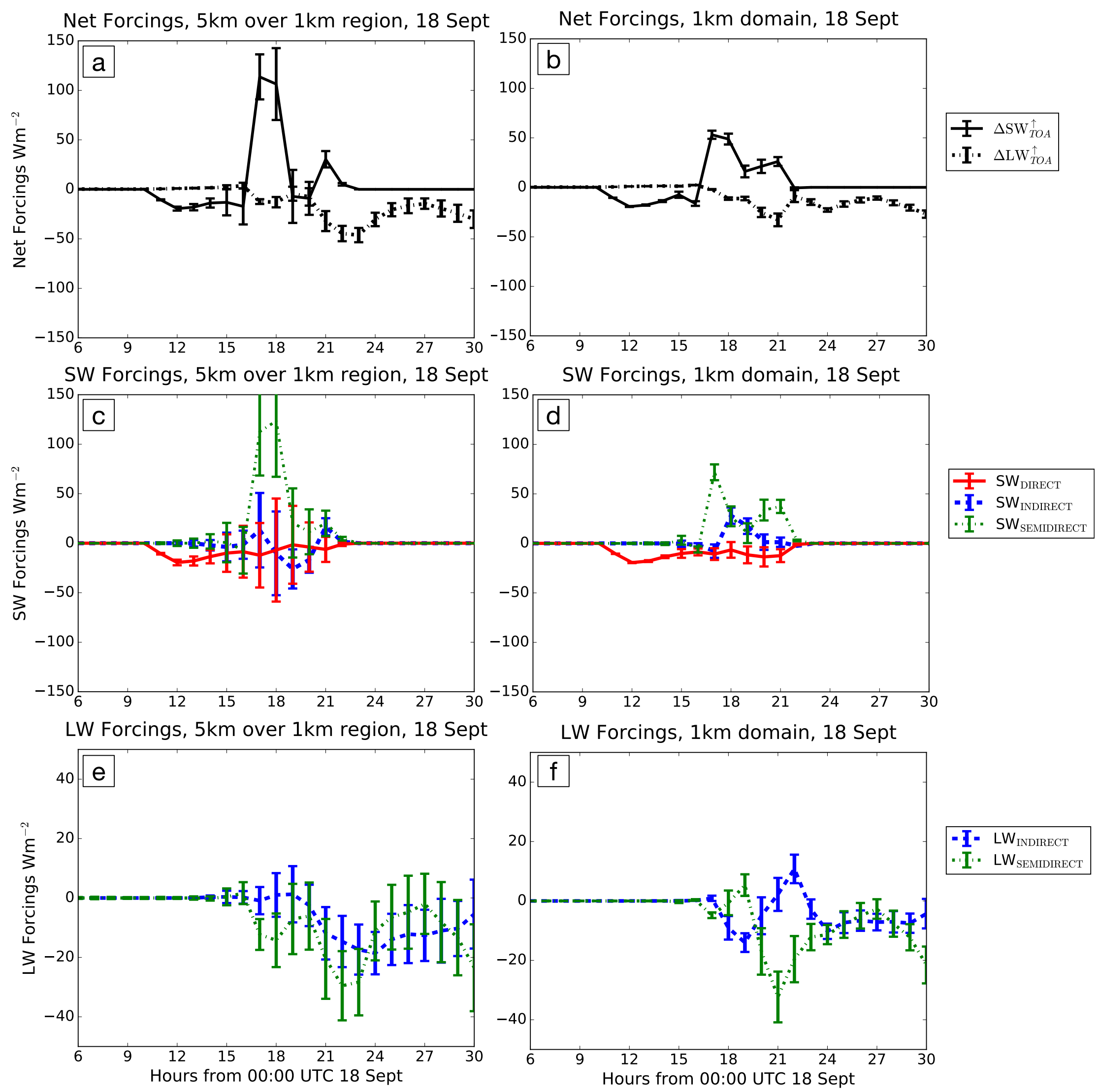
Net Forcings, 5km over 1km region, 18 Sept
Net Forcings, 1km domain, 18 Sept
a
b
Net Forcings Wm⁻²
ΔSW↑TOA
ΔLW↑TOA
SW Forcings, 5km over 1km region, 18 Sept
SW Forcings, 1km domain, 18 Sept
c
d
SW Forcings Wm⁻²
SWDIRECT
SWINDIRECT
SWSEMIDIRECT
LW Forcings, 5km over 1km region, 18 Sept
LW Forcings, 1km domain, 18 Sept
e
f
LW Forcings Wm⁻²
LWINDIRECT
LWSEMIDIRECT
Hours from 00:00 UTC 18 Sept

Figure 8. Cloud cover and precipitation Radiative forcings averaged over 51km region of 5km domain (left) and 1km domain (right) on 18 September 2012, comparing impact of biomass burning aerosol with the use of convective parameterisation. **(a)** shows percentage of domain covered by cloudInstantaneous net forcings (top), **(c)** mean precipitation rate over 5domain. Solid lines show simulations with convective parameterisation ondecomposition into shortwave (middle) and longwave (bottom) direct, dashed line with the convective parameterisation offsemidirect and indirect forcings.

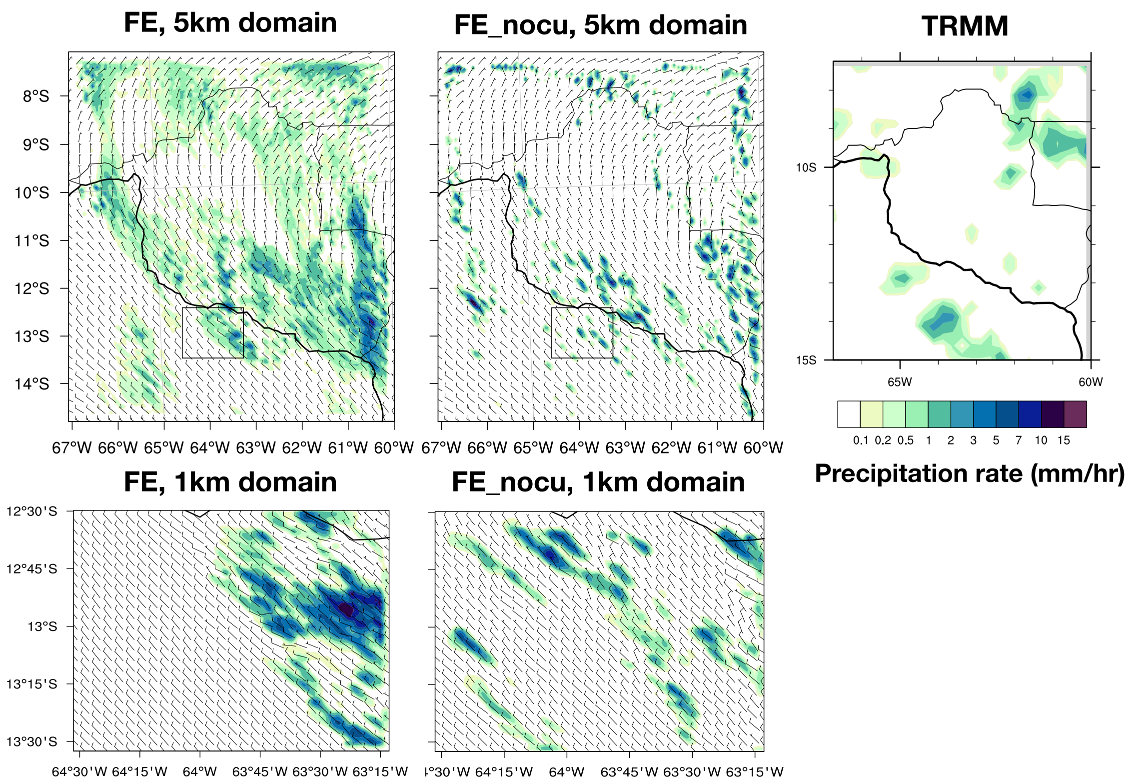

Figure 9. ~~Comparing changes to radiative balance due to aerosol fields and cumulus physics parameterisation~~ Peak precipitation rate over ~~the 5domain for~~ 5km and 1km domains from FE and FE_nCU scenarios, between 20:00 and 21:00 UTC on 18 September ~~,~~2012. ~~**(a)** Difference in downwelling SW radiation at surface to the Ctrl scenario ($\Delta SW^{\downarrow}_{Sfc}$). **(b)** Change in net radiative balance from the Ctrl scenario (ΔRB). Solid lines for runs~~ Compared with ~~cumulus physics parameterisation turned~~ precipitation rate over same region from TRMM data at 21:00 UTC on ~~, dashed lines with cumulus physics parameterisation turned off~~18 September.

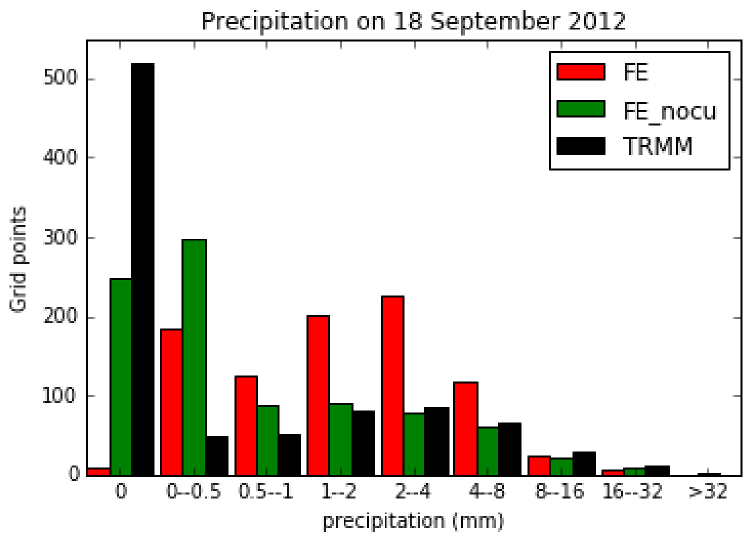

Figure 10. Net radiative balance (ΔRB) and mean Histogram of 24 h accumulated precipitation rate on over 5km domain between 10UTC 18 September – and 10UTC 19 September 2012, over the 1domain region, comparing aerosol effects FE and FE_nCU scenarios with TRMM data. Model output averaged over 5×5 grid (25km) to use be of equivalent resolution to TRMM data ($0.25 \times 0.25°$).

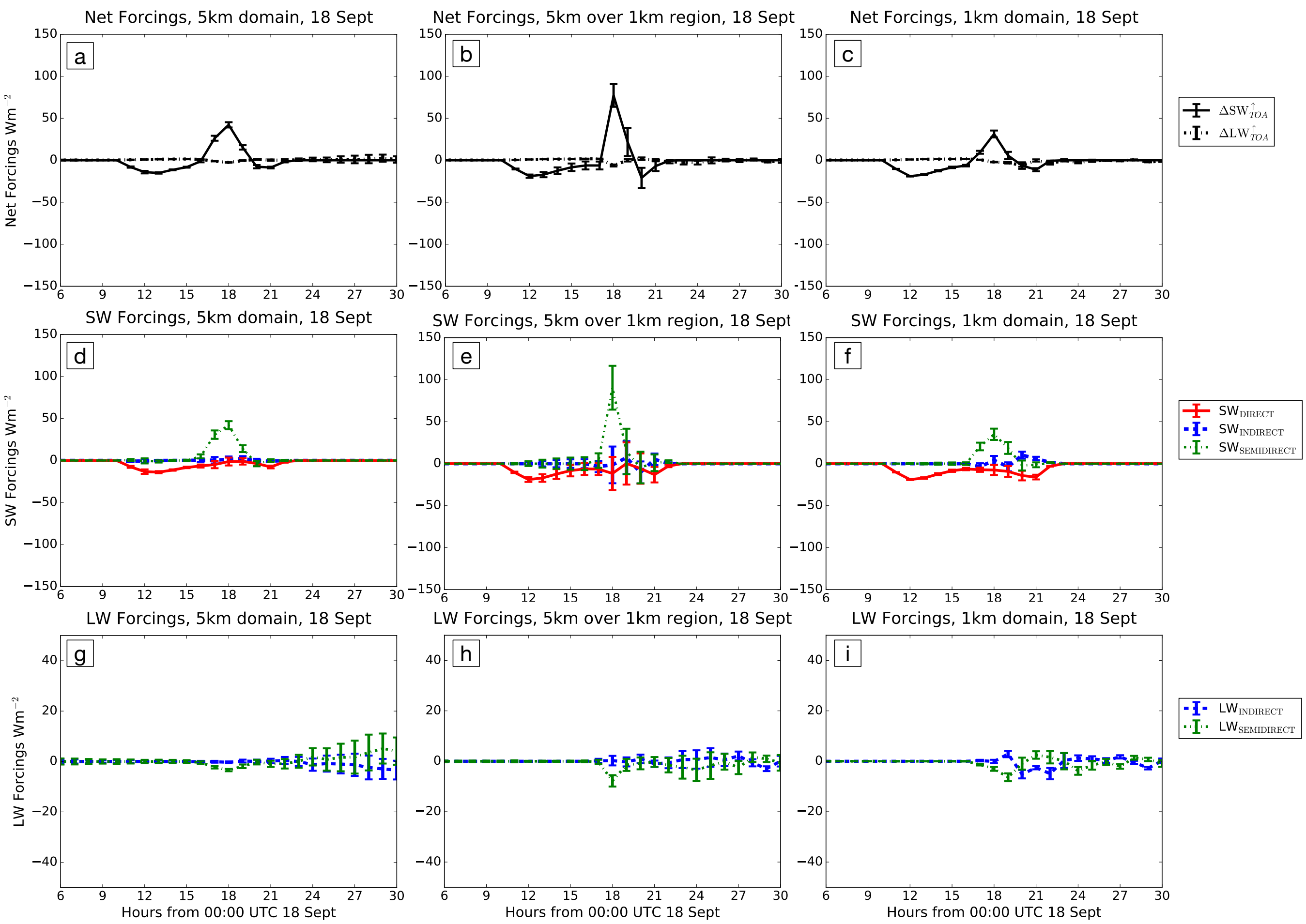
Net Forcings, 5km domain, 18 Sept
Net Forcings, 5km over 1km region, 18 Sept
Net Forcings, 1km domain, 18 Sept
SW Forcings, 5km domain, 18 Sept
SW Forcings, 5km over 1km region, 18 Sept
SW Forcings, 1km domain, 18 Sept
LW Forcings, 5km domain, 18 Sept
LW Forcings, 5km over 1km region, 18 Sept
LW Forcings, 1km domain, 18 Sept
Net Forcings Wm−2
SW Forcings Wm−2
LW Forcings Wm−2
Hours from 00:00 UTC 18 Sept
a
b
c
d
e
f
g
h
i
ΔSW↑TOA
ΔLW↑TOA
SWDIRECT
SWINDIRECT
SWSEMIDIRECT
LWINDIRECT
LWSEMIDIRECT

Figure 11. Radiative forcings from scenarios with no convective parameterisation on ~~the 5~~5km domain for 18 September case study. ~~**(a)** average ΔRB over the 1domain~~Instantaneous net forcings (top), ~~**(b)** average ΔRB from the 5domain over the region covered by the 1domain~~with decomposition into shortwave (middle) and longwave (bottom) direct, semidirect and indirect forcings. ~~**(c)** mean precipitation rate~~ Averaged over ~~the 1~~5km domain (left), ~~**(d)** mean precipitation rate from the 5domain over the~~ 1km region ~~covered by the 1~~of 5km domain ~~. Solid~~ (middle) and ~~dot–dashed lines for runs with convective parameterisation turned on on the 5~~1km domain ~~, dashed lines with convective parameterisation turned off~~(right).

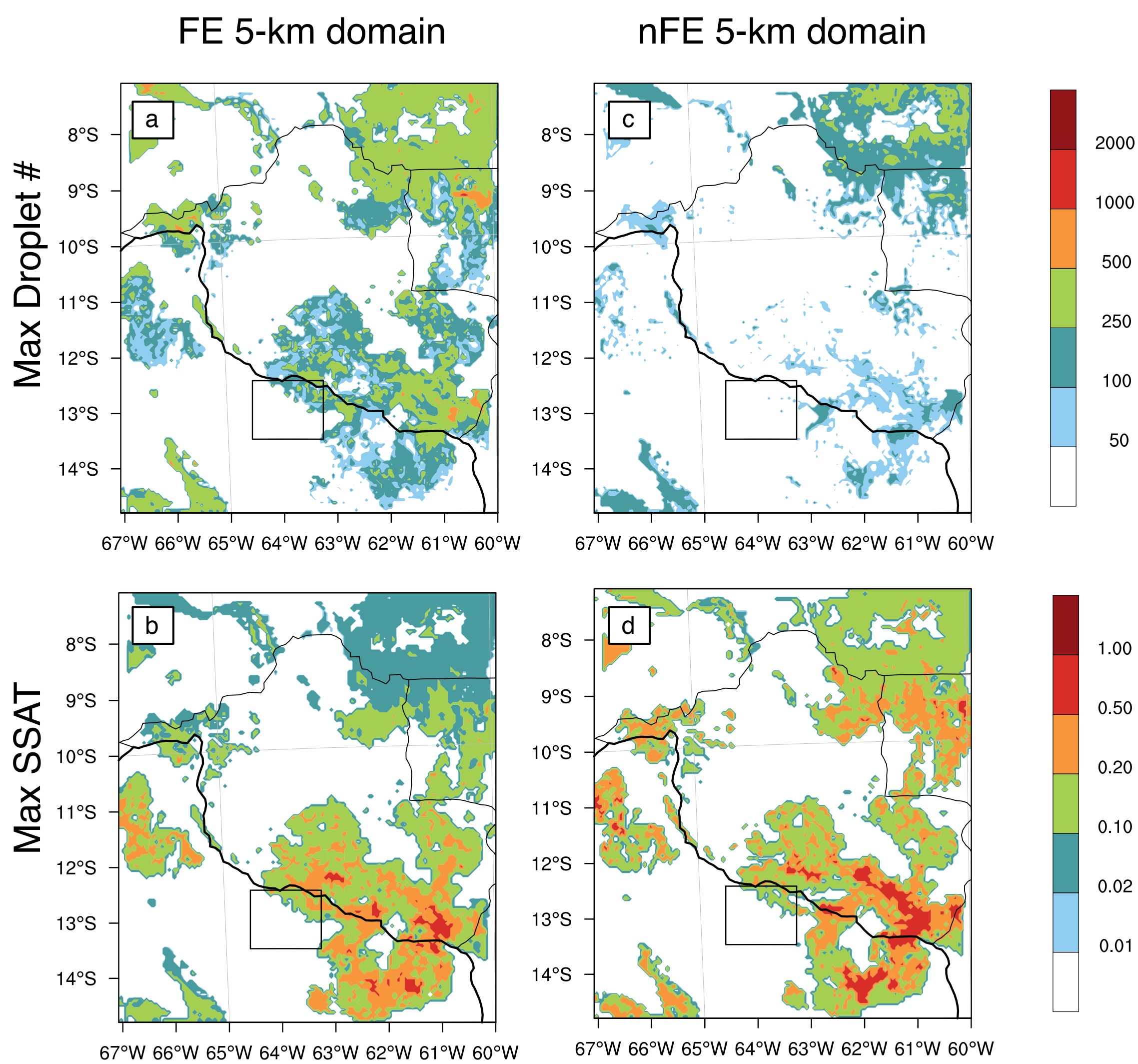

Figure 12. Comparison of maximum droplet number in column $N_{\text{d, max}}$ (cm^{-3}) and estimated maximum cloud supersaturation S_{max} (%) between the FE and nFE scenarios over the 5 km domain on 10:00 UTC (approximately 06:00 LT) 23 September 2012. A and C plots of $N_{\text{d, max}}$; B and D plots of S_{max}. A and B for FE scenario, C and D for nFE scenario.

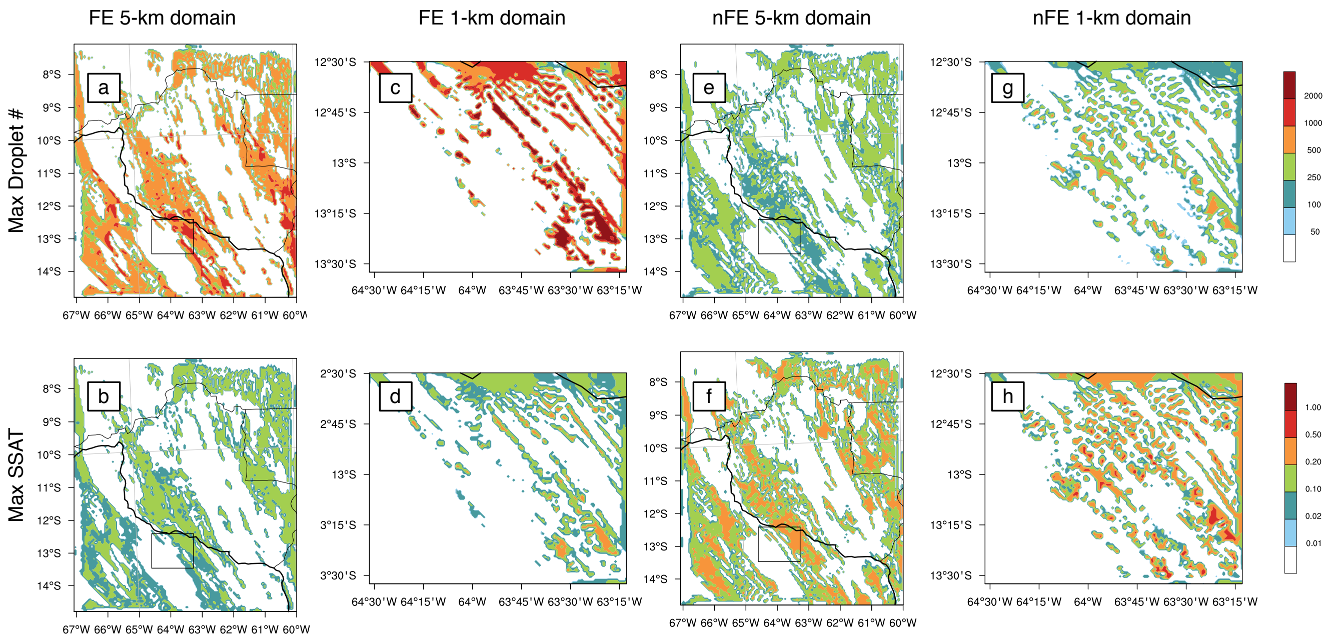

Figure 13. Comparison of maximum droplet number $N_{\text{d, max}}$ (cm^{-3}) and maximum cloud supersaturation S_{max} (%) between the FE and nFE scenarios over the 5 and 1 km domains on 18:00 UTC (approximately 14:00 LT) 18 September 2012. **(a)**, **(c)**, **(e)** and **(g)** plots of $N_{\text{d, max}}$; **(b)**, **(d)**, **(f)** and **(h)** plots of S_{max}. **(a–d)** for FE scenario, **(e–h)** for nFE scenario.