Review replies to "The significant role of biomass burning aerosols in clouds and radiation in the South-eastern Atlantic Ocean" by Haochi Che et al.

We would like to thank all reviewers for their constructive comments and suggestions on the manuscript. The feedback has pointed out important aspects that require additional clarity or information and helped us to improve our paper.

In the following, reviewers' comments are provided in blue, and our responses are in black. Changes to the manuscript made in response to the reviewer are in green.

REVIEWER 1:

In this manuscript, Che et al. run the latest generation of UKESM to investigate aerosol direct, semi-direct, and indirect effects from biomass burning smoke plumes produced by agricultural burning over the southeast Atlantic Ocean. The headline finding (in my read) is that the semi-direct effect of cooling due to increased cloudiness from a stronger cloud-top inversion dominates the overall radiative forcing, offset substantially by the direct effect of smoke absorption and reinforced marginally by indirect effects. The manuscript is well organized and the findings appear sound for the most part. (I do have some questions below, mainly pertaining to the Twomey effect). The text could use some areas of clarification and potentially additional information. I would not anticipate any major new analyses would need to be undertaken to address my comments, so I therefore recommend publication following minor revisions.

Thanks for the positive feedback! Significant reversions have been made to improve the clarity of the manuscript based on the comments from both reviewers.

General comment:

My biggest (really, only) concern with the results is that the indirect effect estimate seems unrealistically small given the change in cloud droplet number concentration you diagnose. From a simple back-of-the-envelope calculation of the Twomey effect, a doubling of cloud droplet concentration (you report 56% of CDNC are from biomass burning) should lead to a radiative forcing of O(10) W/m2, as in Lu et al. (2018). This is before taking into account the small liquid water increases you find. In my own work in the region (Diamond et al., 2020), I've estimated that a 5% increase in climatological CDNC from the influence of shipping produces a radiative forcing of ~-2 W/m2 during austral spring. I thus find it hard to believe that doubling CDNC would produce less than that in austral winter.

We thank the reviewer for pointing this out. This is actually a misunderstanding due to the inaccurate expression. The CDNC increased by the BBA is **up to** 56% only in some specific areas, not the average. We have calculated the mean contribution of BBA to CDNC during July and August in the SEA domain (the area this paper focuses on, ranging from 30° S to 10° N and from 40° W to 30° E), and the result shows the mean CDNC increase by BBA is around 13% and around 1.1% in the cloud box region. Although the indirect radiative cooling associated with the changes of CDNC is smaller than that in Diamond et al., (2020), some differences are to be expected taking significant model differences into account.

To avoid confusion, we have revised the manuscript as follows:

Though BBA can contribute up to 56% of total CDNC in some areas, its average contribution during July to August in the SEA is around 13%, much less than its contribution to the $CCN_{0.2\%}$ budget fraction.

Specific comments:

1. Title: The title is currently rather non-informative. I would recommend highlighting that the semi-direct effects dominate (really the highlight of the paper in my opinion) in the title, and also mentioning that this is a global climate modeling study (as opposed to in situ or satellite observations or high-resolution process modeling).

Agree. Both reviewers have suggested to make the title more specific. According to the comments, the title has now changed to:

Cloud adjustments dominate the overall negative aerosol radiative effects of biomass burning aerosols in UKESM1 climate model simulations over the south-eastern Atlantic

2. Page 2, Line 1: I don't follow why the net sign of the aerosol radiative effect, rather than its magnitude, signifies the importance of aerosol in this region.

The net negative radiative effect of biomass burning aerosol results from the strong cooling of the semi-direct effect. Therefore, this sentence has been deleted, and the previous sentence has been revised as the following to underline the importance of the semi-direct effect.

Among the effects of biomass burning aerosols on the radiation balance, the semi-direct radiative effects (rapid adjustments induced by biomass burning aerosols radiative effects) have a dominant cooling impact over the SEA, which offset the warming direct radiative effect (radiative forcing from biomass burning aerosol-radiation interactions) and lead to overall net cooling radiative effect in the SEA. However, the magnitude and the sign of the semi-direct effects are sensitive to the relative location of biomass burning aerosols and clouds, reflecting the critical task of the accurate modelling of the biomass burning plume and clouds in this region.

3. Page 3, Line 3: I would argue that in situ results showing abundant biomass burning influence within the MBL at Ascension Island from the LASIC campaign (Zuidema et al., 2017) and throughout the SE Atlantic Klein-Hartmann box over the remote ocean from ORACLES (Diamond et al., 2018; Kacarab et al., 2020) are more directly relevant to your point about smoke-cloud interaction.

Thanks for the suggestion! We have adapted the comments and included the papers by Zuidema et al. (2018), Diamond et al. (2018), and Kacarab et al. (2020), in our manuscript.

However, recent studies found abundant biomass burning influence within the marine boundary layer (MBL) at Ascension Island from in-situ observations (Zuidema et al., 2018) and throughout the SEA from flight measurements (Diamond et al., 2018; Kacarab et al., 2020), confirming the interaction of BBA and clouds.

4. Page 3, Line 5: I don't believe the cited literature backs up the claim that "most" of the BBA is entrained into the MBL.

This sentence has been deleted, as the plume-cloud interaction has been described in the above text.

5. Page 3, Line 24: The large eddy simulation results of Yamaguchi et al. (2015) and Zhou et al. (2017) also seem relevant to cite/discuss here.

Agree, the sentence has been revised as:

Hence, related process studies mainly rely on high-resolution limited-area models (Gordon et al., 2018; Lu et al., 2018), as well as idealised large-eddy simulations (Yamaguchi et al., 2015; Zhou et al., 2017)

6. Page 4, Lines 1-2: The BBA plume subsiding and gradually meeting the rising MBL is true in the mean, but the picture is much more nuanced in reality, as instances of smoke-cloud contact were seen to be highly variable between and even with flights during the ORACLES and CLARIFY campaigns. It is also not necessarily the case that smoke-cloud contact corresponds instantaneously with the MBL being polluted, as discussed in Diamond et al. (2018).

Agree, we acknowledge that although the MBL is mostly polluted with BBA at Ascension Island (Zuidema et al., 2018), in other instances, the MBL is relatively clean, as influenced by the recirculation. However, around Ascension Island, both ground-based and flight observations have confirmed the frequently observed BBA (Zuidema et al., 2018, Wu et al., 2020). Therefore, it is reasonable to presume BBA have generally reached the MBL near Ascension Island. We also agree that even with the BBA entering the MBL; cloud properties are not affected by the instantaneous smoke-cloud contact, as discussed by Diamond et al. (2018). Therefore, to make it clearer, we have made the following revision:

These flight campaigns were carried out during the biomass burning seasons, and have provided an ideal dataset covering both BBA above and interacting with clouds, as previous studies have found that the BBA plume layer generally subsides and meets the gradually deepening marine boundary layer in the vicinity of Ascension Island and St Helena (Adebiyi et al., 2015). However, observations also indicate that the entrainment of BBA into the MBL can be intermittent, can require significant contact time (Diamond et al., 2018), and that recirculation patterns can result in clean MBL near Ascension Island.

7. Page 4, Line 13: Is dust included as one of the “five interactive log-normal aerosol modes”? I only count four other components (sulfate, sea salt, black carbon, organic carbon). The phrasing currently is confusing, as it sounds like the dust representation is entirely separate.

In this model configuration, dust is not included in the modal GLOMAP microphysics; it is treated separately in the model using a 6-bin externally mixed scheme(Woodward, 2001), while the interactive log-normal distribution simulates sulfate, sea salt, black carbon, and organic carbon. The “five interactive log-normal aerosol modes” in the manuscript refer to the modal aerosol modes (4 Soluble from nucleation to coarse and one insoluble of Aitken mode), not the aerosol species. The aerosol species are internally mixed in each mode. To avoid confusion, we have made a revision as below:

Aerosol and its interaction with clouds are represented by the UK Chemistry and Aerosol model (UKCA) (Mulcahy et al., 2020; O’Connor et al., 2014), including the modal aerosol microphysics GLOMAP (Mann et al., 2010), with five interactive log-normal aerosol modes (four soluble modes from nucleation to coarse, and one insoluble Aitken mode) comprised of internally-mixed sulfate, sea salt, black carbon, and organic carbon. Mineral dust is represented separately by an externally mixed bin representation (Woodward, 2001).

8. Page 5, Line 9: Somewhere in the manuscript you should discuss the implications of only looking at one part of the biomass burning season (July-August). It is well known that the BB plume properties change over the course of the biomass burning season, influenced in part by meteorological shifts like the strengthening of the southern African Easterly Jet (AEJ-S) in September and October that corresponds with a more elevated plume (Adebiyi & Zuidema, 2016).

Thanks for the suggestion. The corresponding discussion has been added as below:

Two years are simulated in the model (2016 and 2017), however this analysis focuses on July and August, for consistency with the flight campaigns. Note although July and August can be used to represent BBA effects during the African fire season (July-October), this selection will also result in some uncertainties, as the BBA distribution and properties change over the course of the fire season, influenced in part by meteorological shifts, such as the strengthening of the southern African Easterly Jet (AEJ-S) in September and October, corresponding to a more elevated plume (Adebiyi and Zuidema, 2016).

9. Page 5, Lines 17-20: I'm surprised that you do not use any of the new products from MODIS or SEVIRI that account for above-cloud aerosol absorption. I would recommend trying the comparison using one of those products or at least discussing the issues with traditional AOD products that cannot retrieve AOD in the presence of clouds.

The primary model evaluation is done by comparing extinction measured from ORACLES and CLARIFY flights with the model. The MODIS AOD is used to validate the model bias in simulating the BBA plume after the evaluation. The model has been collocated with the MODIS AOD to reduce uncertainties (Schutgens et al., 2016). Although the AOD we used in the manuscript is the traditional product from MODIS, it is comparable with the model, after the collocation.

The main purpose of the AOD comparison is to evaluate the plume simulated in the model. The standard MODIS AOD retrieval is well evaluated and documented, although at Ascension Island, mean AOD (2001–2018) is slightly overestimated (around 0.02) by the MODIS (Gupta et al., 2020). However, the experimental nature of the above-cloud retrievals could be an issue, and require a very careful consistent determination of "above cloud", which is not always trivial in a climate model.

10. Figure 1: What altitude is being shown, or is this a column average? There is a large amount of vertical variability in the plume (as seen in Figure 2) so the 2D picture is a bit difficult to interpret.

This is the collocated model extinction, i.e., the extinction from the model is at the same time, latitude, longitude and altitude as the flight data. Thus, the comparison is point to point. To make it clearer, we have changed the figure caption as below:

Figure 1: Mean along track aerosol extinction coefficient [Mm^{-1}] from the (a) UKESM1 model collocated to the flight tracks, (b) flight observations, and (c) differences between the model and observations. Note that the model extinction is under ambient conditions, whereas the measured extinction is for dry aerosols with relative humidity below 30%.

11. Page 6, Line 4: I do not understand why you only compare September AOD when the analysis focuses on July-August. As discussed earlier, there are known differences in plume location throughout the biomass burning season, in part driven by different meteorological factors between July-August and September-October. Thus, it's entirely possible that the model could represent one part of the season well but the other poorly if it is not representing those meteorological shifts properly. Figure S2 should be replaced with a new version including July and August.

We have changed the figure as illustrated below. The figure now shows the comparison of the mean AOD from model and MODIS (Terra and Aqua) from July to August, 2016-2017.

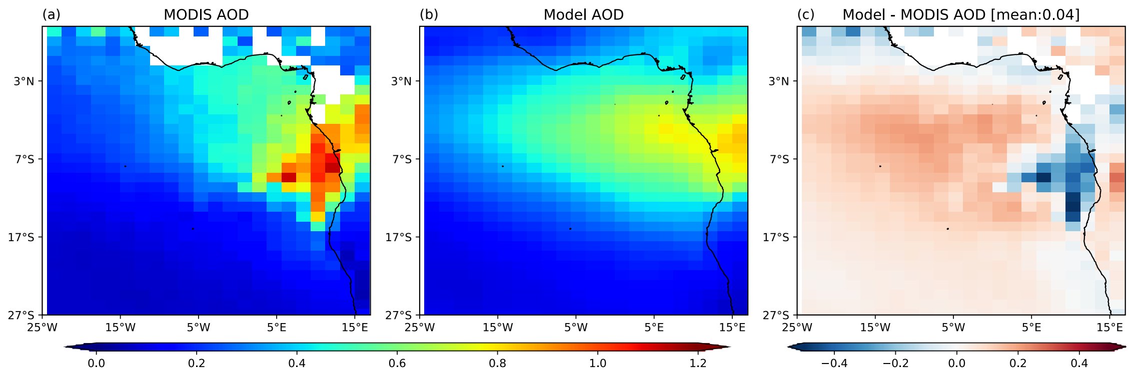


Figure S2. Mean (a) MODIS and (b) UKESM1 simulated AOD during July and August 2016-2017, and the (c) differences between MODIS and the model.

12. Page 6, Lines 12-13: As discussed above, although this description makes sense in the climatology, the picture we found in the field is much more complicated than the mean suggests. For one, much of the smoke in the marine boundary layer at a given location may have been entrained upstream and not necessarily reflect the properties of the plume above-cloud at the time of sampling (Diamond et al., 2018). It may be worth noting that although the mean field shows a plume subsiding from east to west, actual plume distribution and occurrence of plume-cloud contact at any given time is more nuanced.

This is similar to question 6. We agree that even when BBA enter MBL, cloud properties will not be affected by the instantaneous smoke-cloud contact as discussed by Diamond et al. (2018). Therefore, to make it clearer, we have made the following revision:

From east to west, the plume subsides and comes into contact with the clouds. At 5° W, the plume is generally inside the clouds, although the actual plume distribution and occurrence of plume-cloud contact at any given time can be more nuanced (Diamond et al., 2018). Thus, the BBA can interact and modulate cloud properties. This finding is also confirmed by previous studies (Adebiyi et al., 2015; Chand et al., 2009; Deaconu et al., 2019; Gordon et al., 2018).

13. Page 7, Lines 6-7: Are these percentages for the column burden? It may be worth also reporting the values for the marine boundary layer separately (if they differ), as the MBL CCN concentration is what matters most for cloud droplet activation.

Yes, these are percentages of the column burden CCN. We agree that the marine boundary layer CCN is more important in affecting cloud droplet number concentration. The CCN concentration and fraction are indeed quite different in MBL, we have a manuscript under preparation discussing the CCN source attribution, and find the mean number concentration of the BBA $CCN_{0.2\%}$ during biomass burning season is ~75 cm^{-3} in the MBL and ~209 cm^{-3} in the plume layer. The BBA $CCN_{0.2\%}$ fraction is ~40% in the MBL and ~84% in the plume layer, during the BB season. The detail of the CCN distribution will be discussed in the upcoming paper.

14. Figure S3: Figure S3 is an exact copy of Figure 3. I believe the figure the authors meant to include would show the change in CCN due to BBA?

Figure S3 shows the spatial and vertical distribution of total CCN, while figure 3 is the CCN from biomass burning. These two figures share the same pattern, indicating biomass burning is the main source of the CCN. We have changed figure S3 as below, to show the fraction of biomass burning CCN. Although the updated figure still has the same pattern as the CCN from BBA, which highlights the contribution of BBA to CCN.

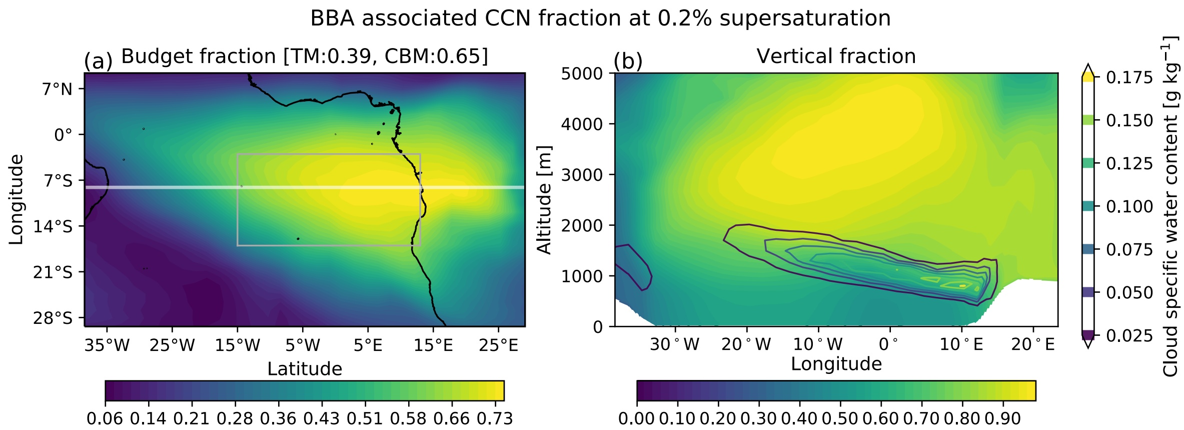


Figure S3. UKESM1 simulated mean fraction of CCN from BBA at 0.2% supersaturation under standard conditions for temperature and pressure (STP) during July and August 2016-2017 as (a) the fraction of vertically integrated burden and (b) fraction of profile along the latitude of Ascension Island, 8.1° S (the white line in Fig. 3a). The domain in Fig. 3a, ranging from 30° S to 10° N and from 40° W to 30° E, is the focus area of this paper. The grey box in the map (cloud box) representing the cloudy areas where the averaged low cloud fraction is above 0.58. The TM is the total mean of the domain, and the CBM is the mean of the cloud box. The contours in Fig. 3b is the cloud specific water content in the baseline simulation.

15. Page 7, Line 27: This should be testable by looking at the average strength of the cloud-top inversion between the different model runs directly.

We have added the variation of the temperature profiles in the supplement to show the strengthening of the cloud-top inversion, as below:

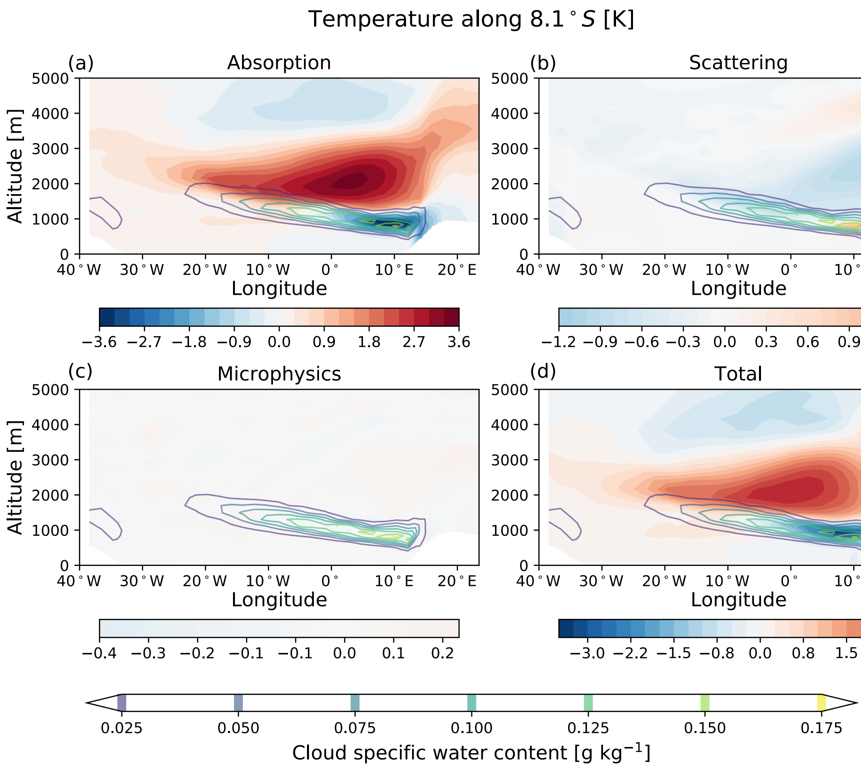


Figure S5. UKESM1 simulated mean vertical profiles of the BBA effects (a) absorption, (b) scattering, (c) microphysical and (d) total on temperature along the latitude of Ascension Island (cf. Fig. 3a) during July and August, 2016-2017. Contour lines represent baseline cloud specific water content. The same colourmap scale is used in each plot to facilitate comparison, but the colourmap ranges differ in each plot, corresponding to the maximum and minimum of temperature at each.

16. Page 8, Line 4: This statement needs qualification, as the SS increases where most of the cloud mass is. Are you only referring to the westernmost region? Or this actually supposed to say that the increase in SS is noticeable in the net (decrease from microphysics is more than compensated by increase from absorption)?

Thanks for the correction. This was a mistake as it should be "increase" not "decrease". The increased SS is noticeable in the net near the continent and at the lower altitude, as the decrease from microphysics effect is totally compensated by the increase from absorption effect. Therefore, the net SS has a similar pattern with the absorption effect in this area. We have changed the sentence as below:

The increase in the maximum supersaturation from the BBA total effect is still quite noticeable.

17. Page 8, Line 20: BBA being 56% of the CDNC is less than the 68% figure quoted above for CCN, but is that for the column or MBL only? It would be more relevant to compare the fraction of MBL CCN that is from BBA to the CDNC change, as the BBA aloft does not activate.

BBA contributes up to 56% of CDNC, while this contribution only happens in some areas. The average contribution of BBA during July-August in the SEA is around 13%. The increased CCN is a column budget. We acknowledge that the CCN in the MBL is more relevant, and it a CCN source and budget analysis is the topic of a manuscript in preparation. We have revised the sentence and made it clearer that the free troposphere CCN would also contribute to the high fraction of CCN. And we also deleted the sentences discussing the supersaturation and particle diameters.

Though BBA can contribute up to 56% of total CDNC in some areas, its average contribution during July-August in the SEA is around 13%, much less than its contribution to the $CCN_{0.2\%}$ budget fraction. This indicates a contribution of BBA above the cloud layer, unable to activate, although they can serve as CCN at 0.2% supersaturation.

18. Page 8, Line 30: The various LES studies cited in this review (Yamaguchi et al., 2015; Zhou et al., 2017) and by the authors (Herbert et al., 2020) seem relevant to reference here in addition to the classic study of Johnson et al. (2004).

Thanks for the suggestion! We have added the corresponding references.

This finding is consistent with the result of large-eddy simulations researches (Herbert et al., 2020; Johnson et al., 2004; Yamaguchi et al., 2015; Zhou et al., 2017) that above cloud BBA can inhibit cloud-top entrainment and increase LWP.

19. Page 8, Line 33: This is due to the absorption effect lowering the relative humidity within the MBL, correct? It would be helpful to be explicit about this.

Yes, when the BBA is inside the MBL, it could reduce cloud droplet numbers by lowering the relative humidity. We have revised this sentence as follows:

When BBA is transported further from the continent, the entrainment of BBA into the cloud layer reduces cloud droplet numbers by lowering the relative humidity through diabatic heating from absorption, which further reduces the increase of LWP, and results in a nearly zero or slightly negative effect on LWP.

20. Page 9, Lines 2-3: The LWP effect of BBA absorption is to increase LWP as one moves from west to east. The text is written to make it sound as if LWP is decreasing from west to east due to BBA absorption. The text should be clarified here.

We have rewritten this sentence in below:

The increased LWP from BBA absorption is mainly located near the continent where the BBA and clouds are well separated, indicating the role of BBA in modulating the cloud distribution.

21. Page 9, Lines 16-17: You should clarify that the BBA in the MBL suppresses CDNC through the semi-direct effect here, not the indirect effect (which actually causes CDNC to increase substantially).

We have made the corresponding change:

However, when more BBA are entrained into the MBL, the BBA decrease the number of cloud droplets through its absorption effect and therefore have a negative impact on the cloud albedo.

22. Page 9, Lines 23-24: As mentioned in the general comment, this result is very surprising given the large increase in CDNC, which should lead to an albedo increase of ~0.05-0.10.

This has been discussed in the response to the general comment. We have calculated the average CDNC in the SEA, and results show that the CDNC increased by BBA is around 13% on average; while in the cloud box region, this number is only around 1.1%.

23. Page 10, Line 3: I don't understand how the indirect effects could have led to a warming given both CDNC and LWP increase. Did cloud fraction decrease anywhere? Or could this just be due to weather noise between different initializations?

We have checked the cloud fraction, and it is increased by the microphysics effect. The warming "indirect effect" occurs mainly in the shortwave, while the cloud fraction and LWP both increase. Therefore, the increase of the indirect effect is likely due to the weather noise between different simulations. We have made the following revision:

In some areas, the indirect effect shows a slight warming effect, which may be caused by the weather noise unconstrained by nudging between different initializations, as the cloud fraction and LWP both increase.

24. Page 10, Lines 20-21: This sentence should be rewritten for clarity. The semi-direct effect is not cooling at cloud top and warming below; rather, above-cloud semi-direct effects lead to a TOA cooling whereas below-cloud semi-direct effects lead to a TOA warming.

As the reviewer suggested, we have revised the sentence as below:

However, in our simulation, the BBA plume is not well separated from the underlying clouds. Thus, when the BBA are closer to the cloud, some BBA may have entered the cloud layer. As a result, the above-cloud semi-direct effects lead to a top-of-atmosphere (TOA) cooling, whereas in-cloud semi-direct effects lead to a TOA warming.

25. Page 10, Line 29: The results of Gordon et al. (2018) are also averaged over a different region that you are using, correct? It would be helpful to compare the values averaged over the same region, as the spatial mismatch could also lead to discrepancies.

Yes, Gordon et al. (2018) used a different domain. We have re-calculated the radiative forcing in the domain studied in their paper and found their direct and semi-direct effects are roughly two times higher than ours, which may be because they only averaged the five most polluted episodes. In contrast, the indirect effect is still quite different and un-proportional, as they have an assumed higher kappa of the OC. The manuscript has been revised as follows:

Comparing the radiative effects in the same domain, the direct and semi-direct effects from their simulations (direct effect: 10.3 Wm^{-2} and semi-direct effects: -16.1 Wm^{-2}) are roughly two times higher than our results (direct effect: 3.3 Wm^{-2} and semi-direct effects: -9.2 Wm^{-2}), as they only sampled the five most polluted days during their simulations. Nevertheless, the indirect effect in their results is -11.4 Wm^{-2}, which is disproportionately higher than our simulation (-0.6 Wm^{-2}). The possible reason behind this discrepancy is that the OC kappa value in their simulation is 0.88, which is much higher than our setting of 0.3.

26. Page 10, Lines 31-33: I would be more believing of this argument (the kappa values in Gordon et al., 2018, do seem unreasonably high) if you did not find a significant increase in CDNC even with your lower (and probably more realistic) kappa values in this study.

Yes, we didn't see a significant increase in the CDNC due to the BBA. The CDNC increased by BBA is ~13% during July to August in the SEA on average, and only 1.1% in the cloud box region (where most BBA are above the cloud layer). The 56% is only for specific areas.

27. Page 11, Lines 4-6: If you're talking about TOA radiation, isn't the relevant effect that less OLR makes it out due to the radiation coming from the relatively cool cloud tops rather than the warmer surface? Zhou et al. (2017) discuss the potentially important role of LW radiative effects in BBA-cloud interactions.

Yes, we agree. The manuscript has revised as follows:

This may result from the semi-direct enhancement of LWP and cloud cover; therefore, the outgoing longwave radiation at the top of the atmosphere is reduced as it comes from the relatively cool cloud tops rather than the warmer ocean surface, as discussed in Zhou et al. (2017).

28. Page 12, Lines 6-7. Increasing the inversion strength, rather than "lowering the temperature inversion"? Or are you talking about lowering the height of the inversion? I'd argue that has more to do with the clouds not being able to grow via entrainment

We have revised the sentence as below, and changed the "lowering the temperature inversion" to 'strengthening the temperature inversion'.

When BBA accumulate above the inversion, the absorbed shortwave radiation warms the air at the bottom of the inversion layer, strengthening the temperature inversion and decreasing the marine boundary layer height.

29. Page 12, Line 12: Cloud top/base is maybe not the most useful shorthand here, as the increase in SS occurs throughout the cloudy layer near the continent, where the cloud deck is most prevalent in general. The base/top difference only shows up further offshore.

Yes, the cloud base and top differences are over the ocean further offshore; near the continent, CDNC generally increases. We have revised the manuscript as follows:

As a consequence, the BBA absorption effect shows a corresponding response: increasing at low altitudes (cloud bottom in baseline simulation) and decreasing at high altitudes (cloud top from baseline) over the ocean further offshore, and generally increasing near the continent. The microphysical effect decreases the maximum supersaturation, as BBA can act as CCN and allow additional water vapour to condense; however, this decrease is comparatively small. The CDNC over SEA is increased especially further offshore due to the BBA microphysical effect, compensating the decreased CDNC at the higher altitude from the absorption effect. In general, BBA absorption and microphysical effects both contribute to the increase of CDNC, although the former is mainly through affecting the maximum supersaturation while the latter is through increasing CCN.

30. Page 13, Lines 4-5: The global and regional indirect effects are “similar” in that they’re both indistinguishable from zero. . . maybe you can argue the global effect is from long range transport and MBL advection, but I wouldn’t necessarily highlight this idea in the very last sentence of your paper.

Thanks for the suggestion. We have deleted this sentence.

REVIEWER 2:

This paper uses UKESM1 simulations to study the contribution of different processes to the radiative effects of biomass burning aerosols. The topic is important, the presentation quality is good and the paper shows interesting results. I don’t have major concerns about the analysis, but I find there is the need for a more detailed description of the methodology and some additional analysis. I also believe that most of the supplementary figures belong to the main body of the paper (see specific comment below). The paper is worth publishing, but given that it needs additional work in a number of areas, I am recommending a major revision.

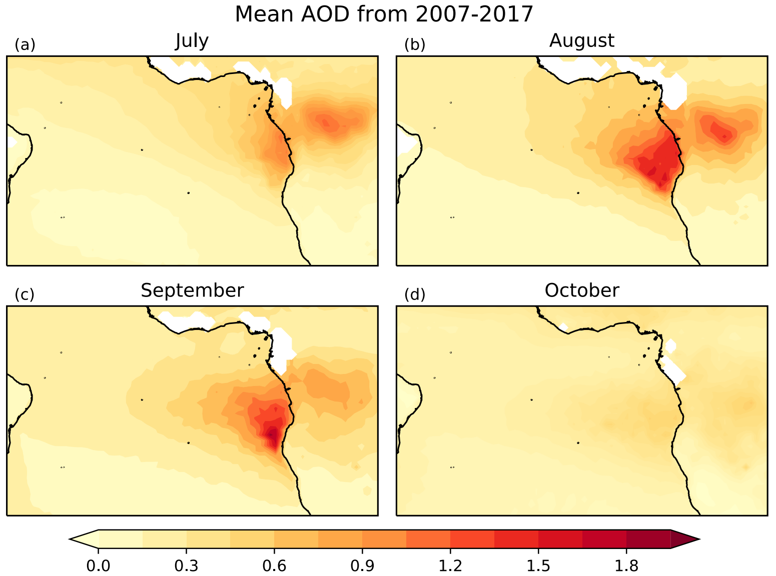
Thanks for the positive feedback! Significant reversions have been made to improve the clarity of the manuscript based on the comments from both reviewers.

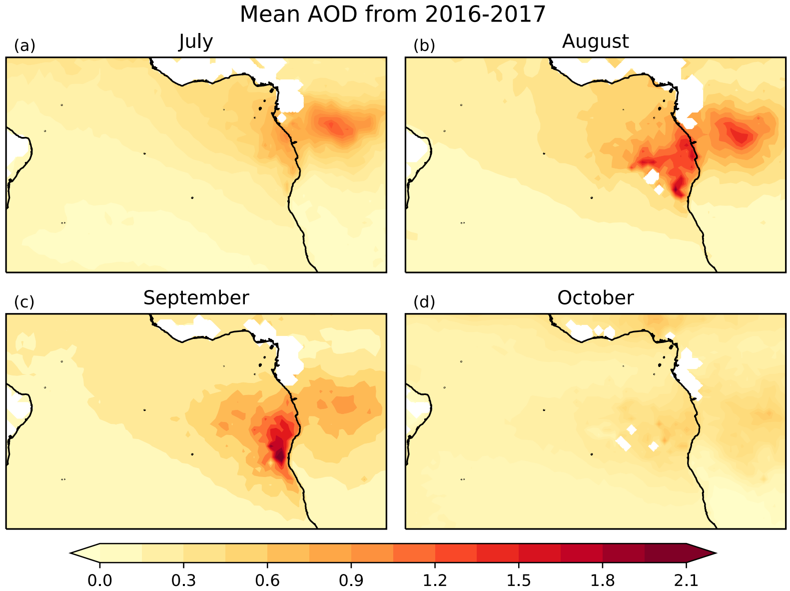
General comments:

1. Only two years of model simulations are used. They are chosen because they coincide with observational campaigns. However, the observational data are only used to perform an initial assessment of the model’s simulations and to justify the use of UKESM1 for the subsequent analysis, which is entirely model-based. Then, why not use a longer simulation period? This will allow to get more robust estimates of the BBA effects in the region, and to quantify the role of interannual variability.

The focus of this manuscript is to investigate the radiative and microphysical effects of BBA in affecting clouds and radiation in the SEA. Those effects are different when BBA are above or below clouds. Therefore, it is critical to simulate the spatial and vertical distribution of the plume. Another crucial task is to have accurate emissions of biomass burning, as it changes interannually. Therefore, we choose to use the GFAS data from satellite measurements to provide the biomass burning emissions. In processing the biomass burning emission data, as we have no information on the vertical distribution of the emission, the emission data was set to emit evenly throughout the boundary layer (representing large fires). However, this method will undoubtedly result in discrepancies of the vertical distribution of BBA. So, we used flight observations to evaluate the model simulation of the BBA. After the evaluation, we confirmed the model did well in simulating the BBA plume in 2016 and 2017, with certain biases regarding the plume distribution. Those evaluations also provide us with the confidence to investigate the BBA effects further, but only for the two evaluated years.

To confirm the biomass burning of the simulated two years is representative, we also calculated the 11 years AOD from the MODIS Terra and Aqua 1-degree product. Then we compared it with the two years AOD from the same MODIS dataset.





By this comparison, we found that the AOD in the two years average is similar to the climatologies in both magnitude and distribution. Therefore, it is reasonable to use the two years average to represent biomass burning and we leave the analysis of the inter-annual variability to future work.

2. The methodological description needs additional work. Especially, I think a brief description of how the different experiments are combined to decompose the BBA effect into individual contributions is lacking. How accurate is this decomposition? What are the caveats?

We have added more detailed information regarding the decomposition of the BBA effect.

To decompose the BBA effect into radiative and microphysical effects, we performed six simulations from 2016 to 2017, one with present GFAS BBA emission as the baseline simulation ($BB_{0.3}$), and one with the same settings but κ_{org} set to 0 (BB_0); two without BBA emission for κ_{org} set to 0.3 and 0 (noBB$_{0.3}$, noBB$_0$), and two with BBA emissions and κ_{org} set to 0.3 and 0 but with the BBA absorption turned off ($noBB_{0.3}^{noABS}$, $noBB_0^{noABS}$) (setting the imaginary part of the refractive index to zero). Radiative and microphysical effects of BBA are separated using the method of Lu et al., (2018), and described by following equations:

Absorption effect = $BB_0 - BB_0^{noABS}$

Scattering effect = $BB_0^{noABS} - noBB_0$

Total effect = $BB_{0.3} - noBB_{0.3}$

Microphysical effect = Total effect – Absorption effect – Scattering effect

This method allows us to decompose the effects of BBA, with some limitations due to inherent assumptions and model structures. For example, our model only allows us to switch off the absorption of BBA, not the total radiative effects. This assumes that the cloud adjustment due to BBA scattering is negligible in our experiments (which excludes fast adjustments to corresponding surface flux changes). Also note that the microphysical effect of BBA decomposed from our setting is driven by the variation of κ_{org}, thus the small fraction (around 10%) of OC from non-biomass burning emission in this region (figure S1) would contribute a small error.

3. Given that this is a model-based study, the accuracy of the results will not only depend on the representation of the BBA plume and the cloud climatology, but also on how good the model is at representing the cloud response to the drivers of changes. For example, the realism of the strong radiative cooling of the semi-direct effect will depend on how well UKESM1 represents the cloud response to a strengthening of the inversion. Figure S4 touches on this, but only in passing. You cite a reference (Adebiyi et al., 2015) that uses radiosondes to look into this. How does the change in the inversion strength in UKESM1 compare to the one observed with radiosondes? I acknowledge that a comprehensive assessment of how well UKESM1 performs in this respect is out of the scope of this paper, but putting the UKESM1 changes into context would be very helpful.

We have preformed the comparison of radiosonde data and model results, and illustrated in the figure A1. The radiosonde data comes from the paper by Adebiyi et al. (2015). Note that the period of the radiosonde soundings is different, as it was averaged in September and November, from 2000 to 2011; while the model is the September mean from 2016 to 2017. Although the averaged biomass burning condition of those two years are similar to the climatologies, the mean meteorological condition may differ, resulting in changes of the temperature profiles. Another major difference is that the model results are averaged over the whole month, decomposing the effect of absorption explicitly, while the radiosonde was averaged by bins of fine-mode aerosol optical depth (AOD$_f$). This means that the different radiosonde bins are also likely to correspond to different meteorological situations. As a result, the changes in the radiosonde profiles are more obvious than the model averages, as illustrated in the figure.

The key difference from the figure1 is the change of the mid-tropospheric temperature profile. Stabilisation in the radiosondes also seems to suppress vertical cloud development, which we do not see in the model. Despite those differences, the temperature responses to the BBA are analogous to the measurement; thus, the inversion and cloud response from the model is reasonable. However, how accurate the model can simulate the temperature responses requires additional working to evaluate the model with detailed observation data, and is out of the scope of this manuscript.

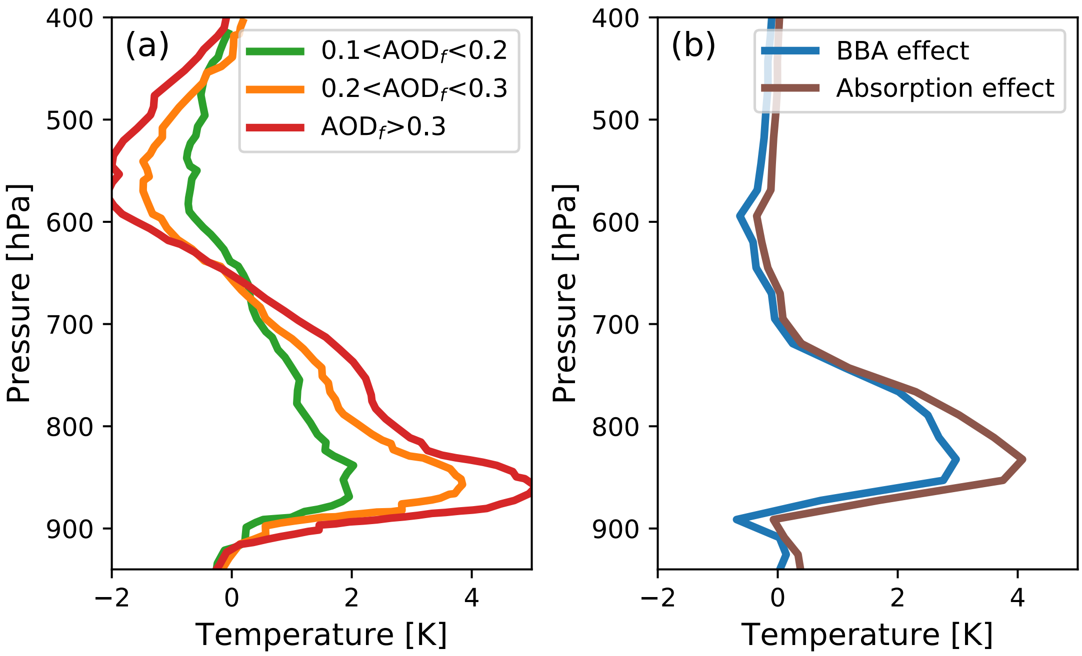


Figure A1. Mean temperature difference profiles in St. Helena Island, calculated from (a) radiosondes (b) UKESM1 model simulation. The differences are constructed by subtracting the mean profiles representing the clean condition, corresponding to (a)$AOD_f < 0.1$ and (b) no BBA simulations. AOD_f is the fine-mode aerosol optical depth, derived from MODIS product. The blue line in (b) is the difference of the temperature profile between simulations with and without BBA, and the brown line is the difference between simulations with absorption on and off. The radiosonde data is from Adebiyi et al. (2015).

The title should be more specific, and should capture the main message of the paper.

Agree. Both reviewers have suggested to make the title more specific. According to the comments, the title has now changed to:

Cloud adjustments dominate the overall negative aerosol radiative effects of biomass burning aerosols in UKESM1 climate model simulations over the south-eastern Atlantic

Specific comments:

1. P3L22-P4L5: Most of this paragraph probably belongs to the methods section. Only the last sentence describes the objectives of the paper. I'd suggest transferring the description of the campaigns to the methods, and expand on what the paper is about.

Thanks for the suggestion, we have made the corresponding revision and moved the description about CLARIFY and ORACLES campaigns into the method section. The last paragraph of the introduction is now revised as below.

The complex interactions between cloud microphysics, radiation, cloud entrainment processes and in particular the small spatial scales involved make the simulation of the stratocumulus clouds deck in the SEA a challenge. Hence, related process studies mainly rely on high-resolution limited-area models (Gordon et al., 2018; Lu et al., 2018) as well as idealized large-eddy simulations (Yamaguchi et al., 2015; Zhou et al., 2017). However, ultimately it is important to represent and constrain the related effects in General Circulation Models (GCM) widely used to investigate climate responses to anthropogenic perturbations, e.g. by the Intergovernmental Panel on Climate Change (IPCC). In this paper, we use the UK Earth System Model (UKESM1), which is also being used in the recent Coupled Model Intercomparison Project Phase 6, to study the BBA effect on the clouds and radiation in the SEA. A detailed description of the model, simulation setup and the data we used for evaluation is provided in section 3. The model is evaluated by observations in section 3.1, and BBA effects on clouds are investigated by decomposition into radiative effects (absorption and scattering) and microphysical effects in section 3.2. The BBA radiative forcing is studied in section 3.3. Section 4 offers conclusions and discussion.

2. Figure 1. The spatial resolution of the model results seems to be much higher than N96. Am I misinterpreting what that figure shows?

Figure 1 shows the measured and collocated model extinction. For each point of the measurement, the model was interpolated to the same point (same coordinate), as described in the method section. By doing this collocation, the model gives point to point data to compare with the measurement; therefore, it’s not on the N96 resolution.

3. Figure 3 caption. "The domain in Fig. 3a, ranging from 30◦ S to 10◦ N and from 40◦ W to 30◦ E, is the areas this paper interested in." I believe the description of the area of interest belongs to the main text.

Thanks for the suggestion. We have now added the description of the area into the main text.

BBA can serve as CCN and further impact the CDNC and cloud optical depth. Meanwhile, it also has a significant impact on the atmospheric thermal structure and therefore, the maximum cloud supersaturation, LWP and cloud albedo. The BBA effects on clouds are decomposed as radiative (absorption and scattering) and microphysical effects (detailed in section 2), and their impact on the clouds is examined in this section. Figure 3 provides the baseline cloud properties from the standard simulation. The domain in Fig. 3a, ranging from 30° S to 10° N and from 40° W to 30° E, is the areas this paper interested in. To get BBA effects on the stratocumulus clouds, a cloud box area is used to represent the stratocumulus cloud deck region (the grey box in Fig. 3a). The mean low cloud fraction is 0.58 in the cloud box region, and its western border reaches the area where stratocumulus to cumulus transition occurs (See Fig. 1 in Gordon et al. (2018)), suggesting the dominant of the stratocumulus clouds in this area.

4. Figure 3 caption. " The grey box in the map (cloud box) representing the cloud areas where the averaged low cloud fraction is above 0.58." That would be the 0.58 isoline, unlikely to have a rectangular shape. Please provide a clearer explanation of what this box is.

This cloud box is selected to represent the area where the cumulus and stratocumulus dominate. By using this cloud box, we can assess the BBA effects on the clouds more specifically. Klein and Hartmann (1993) used a similar method to define the stratocumulus clouds region, while our cloud box region is broader, as we want to encompass stratocumulus and some cumulus clouds. As discussed in Gordon et al. (2018), Ascension Island and nearby is within the area where the stratocumulus to cumulus transition occurs. Therefore, we included Ascension Island inside the west boundary of our cloud box region. The mean cloud fraction of this region is 0.58, which confirms the dominant influence of clouds in this region.

5. P7L18. supersaturation (SS). This is an unfortunate acronym. In general, I don't see the need for using an acronym to compress a single word. For instance, we don't normally use MP for microphysics. Or, what would you use for subsaturation?

Though the acronym of supersaturation is widely used in the field measurements and laboratory studies (Meng et al. (2014), Jung et al. (2018), Zhang et al. (2012), Panicker et al. (2020) and Bhattu and Tripathi (2015)), to avoid unnecessary misunderstanding, we have adopted the reviewer's suggestion, and changed it to supersaturation in the manuscript.

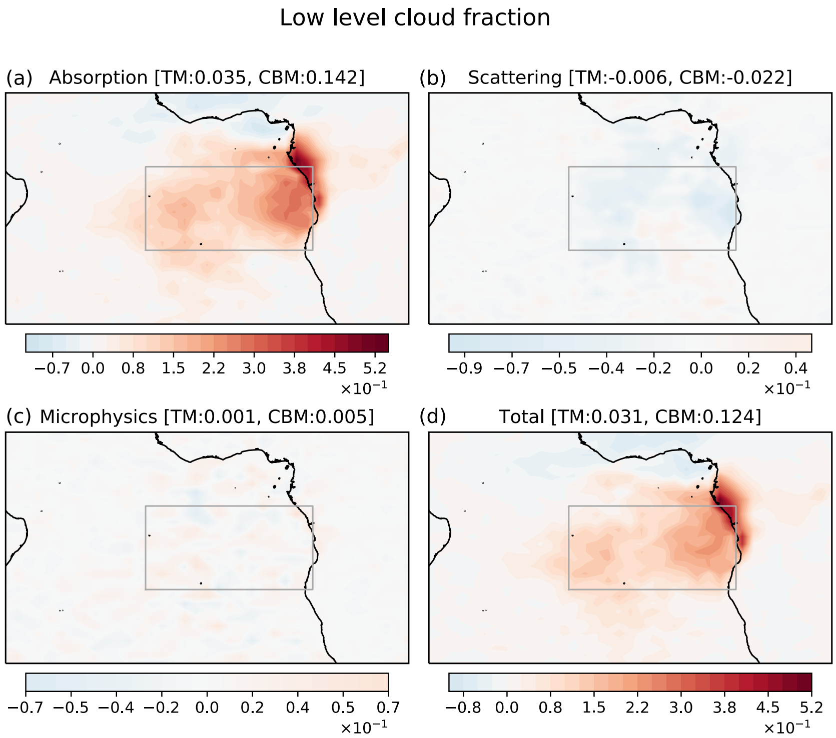
6. P9L2. "LWP from BBA absorption shows a steady negative gradient from west to east". I might be looking at the wrong region, but the gradient in the cloud region looks positive to me. Please clarify.

We changed this sentence as below:

The increased LWP from BBA absorption is mainly located near the continent where the BBA and clouds are well separated, indicating the role of BBA in modulating the cloud distribution.

7. Figure 7. This figure shows changes in cloud albedo using the ISCCP simulator. I might have missed it, but I believe that the use of the ISCCP simulator is not documented in the methodology. Which simulator variables are used? Why is this approach better than looking at changes in cloud fraction and cloud radiative effect?

The object of this manuscript is to study different BBA effects on clouds. The changes of cloud fraction are very similar to the changes of LWP, please see the figure below.



The pattern of the cloud albedo, however, shows a different pattern, as the increased cloud albedo from the absorption effect is in a relatively small area and not so strong. Therefore, we choose to discuss the changes in cloud albedo. Another reason is that cloud albedo is widely used in satellite studies (e.g., Deaconu et al. (2019)); it is an important parameter to assess the cloud radiation properties. Thus, put the cloud albedo in the manuscript allow to compare with satellite studies and understand the role of different BBA properties in affecting the cloud albedo.

Cloud albedo in the UKESM1 is diagnosed by the ISCCP simulator, which is a part of the Cloud Feedback Model Intercomparison Project (CFMIP) Observational Simulator Package (COSP). An additional benefit of using ISCCP output is that it also facilitates model intercomparison by minimizing the impacts of how clouds are defined in different parameterizations. More detailed information and evaluation of the ISCCP are provided by Mace et al. (2011).

In the manuscript, we have now added a brief description of the ISCCP simulator in the context, as below.

In the UKESM1, cloud albedo is diagnosed by the International Satellite Cloud Climatology Project (ISCCP) simulator (Bodas-Salcedo et al., 2011), which can minimize the impacts of how clouds are defined in different parameterizations and facilitate model intercomparison.

8. Discussion of Figure 8c. It would be nice to show a plot with the actual change in the strength of the inversion that drives this strong semi-direct effect.

Thanks for the suggestion. Both reviewers have raised this, and we have added the changes in the temperature profile in the supplement as Fig. S5.

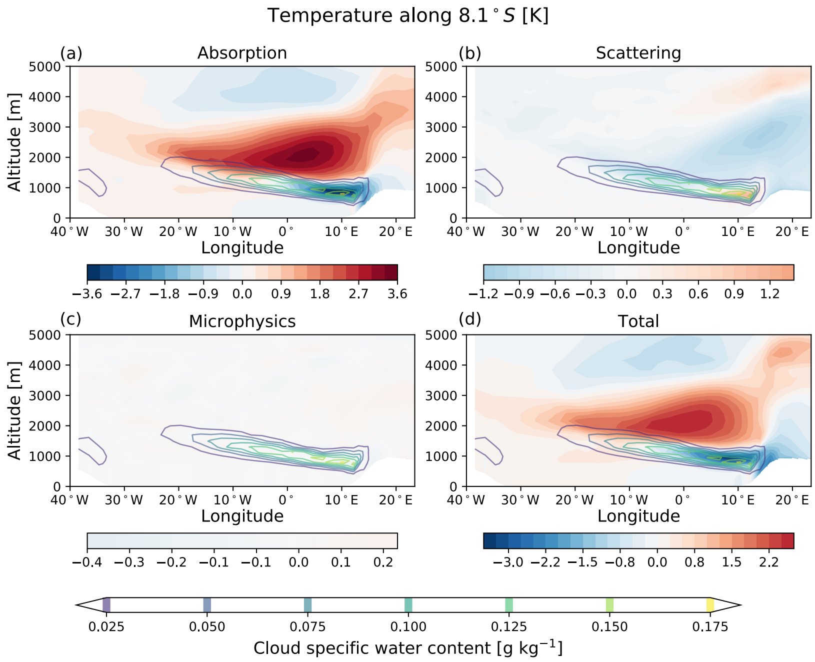


Figure S5. UKESM1 simulated mean vertical profiles of the BBA effects (a) absorption, (b) scattering, (c) microphysical and (d) total on temperature along the latitude of Ascension Island (cf. Fig. 3a) during July and August, 2016-2017. The contour lines are the baseline cloud specific water content. The same colourmap scale is used in each plot to facilitate comparison, but the colourmap ranges differ in each plot, corresponding to the maximum and minimum of temperature at each.

9. Figures S5 to S8. I feel that these figures belong to the main paper, not to the supplementary material. They can be arranged all together in a multi-panel figure with showing the baseline climatologies, complementing the figures showing the changes due to different processes.

Thanks for the suggestion! We have now combined those figures and put them into the main part of the paper.

3.2 Biomass burning aerosol impacts on clouds

BBA can serve as CCN and further impact the CDNC and cloud optical depth. Meanwhile, it also has a significant impact on the atmospheric thermal structure and therefore, the maximum cloud supersaturation, cloud droplets and cloud albedo. The BBA effects on clouds are decomposed into radiative (absorption and scattering) and microphysical effects (detailed in section 2), and their impact on the clouds is examined in this section. Figure 3 provides the baseline cloud properties from the standard simulation. The domain in Fig. 3a, ranging from 30° S to 10° N and from 40° W to 30° E, is the areas this paper interested in. To get BBA effects on the stratocumulus clouds, a cloud box area is used to represent the stratocumulus cloud deck region (the grey box in Fig. 3a). The mean low cloud fraction is 0.58 in the cloud box region, and its western border reaches the area where stratocumulus to cumulus transition occurs (See figure 1 in Gordon et al. (2018)), suggesting the dominance of stratocumulus clouds in this area.

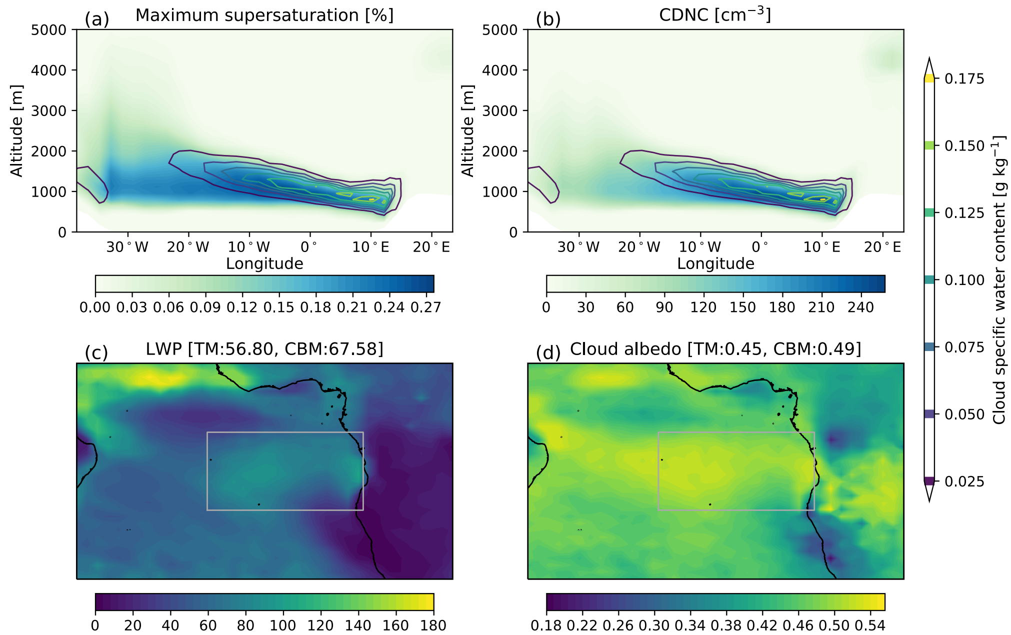


Figure 3. UKESM1 simulated mean (a) vertical profiles of maximum supersaturation and (b) vertical profiles of cloud droplet number concentration along the latitude of Ascension Island; spatial distribution of (c) cloud liquid water path and (d) cloud albedo from the International Satellite Cloud Climatology Project (ISCCP) simulator. These means are averaged during July and August, 2016-2017. The contour lines in (a-b) are the cloud specific water content. The TM in (c-d) is the total mean of the domain, and the CBM is the mean of the cloud box (the grey box on the map) representing areas where the average low cloud fraction is above 0.58.

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