Review of "Radiative Heating Rate Profiles over the Southeast Atlantic Ocean during the 2016 and 2017 Biomass Burning Seasons" by Collow et al.

This manuscript presents a quantitative assessment of the radiative heating rates due to aerosols over Ascension Island based on idealized calculations through a Radiative Transfer Model, using aerosol vertical profiles and optical properties from MERRA-2 reanalysis, thermodynamic profiles, and low-cloud properties from island-based observations during the LASIC field campaign. The authors find shortwave heating within the aerosol layer above Ascension can locally range between 2 and 8 K per day, and shortwave heating due to biomass burning aerosols is not balanced by additional longwave cooling.

The presented assessment of the aerosol radiative heating rates is novel and nicely conducted, which could be informative and insightful to the scientific community if the following concerns and questions are properly addressed and justified. In addition, I find myself having hard times understanding/digesting many of the discussions in the current form of the manuscript. These confusing discussions should be reconstructed and extended with additional details and elaborations before this manuscript can be accepted for publication.

Major concerns/questions:

1. The current introduction section seems a little weak in terms of scientific motivations. I recommend stating a stronger scientific motivation for the study, clarifying the scientific question you want to address with this study, and elaborating more on how is your study going to help us better understand the aerosol-radiation-cloud interactions within the SE Atlantic, rather than saying that the goal is to quantify and report the radiative heating rates over Ascension.

The introduction section has been modified to help clarify the motivation behind the work.

2. You compared vertically integrated aerosol properties from MERRA-2 to LASIC measurements, but not thermodynamical properties, especially thermodynamic profiles, which could have subtle impact on the vertical distribution of biomass burning aerosols.

a. You mentioned in Line 20 on Page 11 that "boundary layer is too deep over Ascension Island in MERRA-2," comparing to LASIC measurements? Could you show the thermodynamic profiles comparison over Ascension?

b. You mentioned potential deficiencies in RH profiles of MERRA-2, can this also be shown as a comparison with LASIC measurements?

Thermodynamic profiles from MERRA-2 are now included in Figures 5 and 6. A discussion on the comparison between the observations and MERRA-2 has been added to the text.

3. You introduced six sets of experiments towards the end of Section 2. Please specify how are they defined/selected?

Additional text and a table have been added to help clarify the sets of experiments.

a. Are you using cloud observations to screen for clear skies?

No, cloud observations are not used to screen for clear skies (see response for comment 3b below).

b. Are you simply setting cloud parameter off in the model for clear sky case, even though the inputting thermodynamic profiles felt the existence of a cloud layer? If this is the case, please discuss how is this artifact affecting your results (i.e. is your heating rates under/over-estimating?).

Clear sky here is simply turning clouds off in the model, and you are correct that the thermodynamic profiles will still reflect the existence of a cloud. This is now noted in the text and the implications are discussed.

c. Not all of these experiments are presented in the following results part, e.g. the clean- cloudy case is not shown.

Actually, the clean-cloudy case is indirectly used for the results presented in panels d-f of figure 6. The calculation shown is Cloudy with aerosol – Clean cloudy. This has been made clearer in the text and Table 2.

4. Section 3 is named as "Results," even though 3.1 and 3.2 are not actual "results" of the radiative transfer calculations. Using "Results" as a section name is vague, and I suggest reconsidering the section organization, one option could be making 3.1 as a new Section 4, and 3.2 as a new Section 5. Currently, all your results are lumped into one section (3.3), which is poorly organized, I suggest making 3.3 as a new Section 6, and break each part into subsections, e.g. SSA sensitivity, SW heating due to black carbon, SW heating enhancement in the presence of clouds, LW cooling, and heating rates along back-trajectories.

The suggested section organization has been implemented.

5. I am a bit lost regarding the purpose of discussing the back-trajectories, how are they (or the origin of aerosol) related to (or affect?) your heating rate calculations? Besides, some of the details regarding the HYSPLIT runs should be introduced in the Data & Methodology section. P7 L15-18 seem abrupt at the end of Section 3.1, they also fit better in the Data & Methodology section. Perhaps these last two paragraphs of Section 3.1 could be moved to Section 2 together.

The back trajectories have been included as they impact the heating rates of the aerosol as the plume approaches Ascension Island. A sentence has been added to the text regarding this. The details on the runs have been moved to the Data and Methodology section as suggested.

a. P7 L2, "determine the origin of the aerosol" is a strong statement, as back-trajectories not necessarily indicate the exact pathways of biomass burning plumes, and one has to combine other sources, e.g. fire emission data, in order to determine the origin of the aerosol observed at Ascension. I suggest rewording.

This sentence has been reworded to indicate we are interested in the trajectory of the aerosol.

b. P7 L5-6, there were aerosol both in the free-troposphere and boundary layer, why 2 km is picked? What is the prior results you are referring to, please provide the reference.

The height of 2 km was selected as that is the middle of the aerosol layer detected by the micropulse lidar. This can also be seen in Figure 4 of Zuidema et al. (2018), which is now cited in the text.

c. P7 L9-10, why the subtropical high over the southern Indian Ocean plays a role here? Monthly mean SLPs could be added to the background of Fig. 3 to base this statement.

The southern Indian Ocean is too far east to nicely include in the figure, but adding monthly mean SLP to the background was a great idea and has been incorporated.

d. P7 L10-14, what are these observations implicating, how are they related to this work, or affecting the interpretation of your results?

The back trajectories have implications on the AOD over the site, as well as the heating within the aerosol layer as it travels towards Ascension Island. Given that this is controlled by the large-scale circulation, the amount of heating on days (and years) not shown can be extrapolated. A sentence regarding this has been added to the text.

6. Regarding Fig. 4, although one can identify an inversion layer from temperature profiles (distinguishing yellow and light orange from your plot), I would argue that potential temperature is a better choice to show the cloud top inversions. I think you could also extend the discussion to the potential role of the RH plot on indicating the biomass burning smoke plumes arriving at Ascension, as RH bursts in the free-troposphere tend to colocate well with the smoke episodes arriving at Ascension.

We investigated showing potential temperature instead however the figures did not prove to show cloud top inversion any better than temperature. As a result, we had elected to stick with temperature for the figure. A discussion on the connection between the smoke plumes and relative humidity has been added to the text.

7. You did not discuss Figure 5b at all, and only mentioning Fig. 5a for clouds that are not visible in the plot. I wonder if this figure is necessary, it doesn't seem to add much useful information, and you barely discussed it in the main text.

The fields shown in Figure 5 are used as an input to the radiation transfer calculations. This is also the first time (to our knowledge) that the microphysical properties of clouds during LASIC have been included in peer reviewed literature. The section of the text has been expanded.

8. The discussion in the first paragraph of Page 9 is very confusing to me. In the first couple of sentences, you mentioned comparing heating due to clouds with heating due to aerosols, and my interpretation of Figure 6d is that this is SW heating due to aerosol under a cloudy sky, and you did not show a case for heating due to clouds alone (no aerosol), so how did you make the comparison? whereas in the last sentence you talked about comparing aerosol heating under cloudy and clear skies (isn't this contradicting to the first couple of sentence? please clarify), and what do you mean by "embellished," I had trouble relating this word to the observations.

This paragraph has been rewritten, with an additional figure and references to the table added. "enhanced" is now used instead of "embellished".

9. I also think the discussion for Figure 6d,e,f could be substantially extended. Currently, you barely discussed them ("embellished" is all you used to describe the comparison), and I will be curious to know why SW heating due to aerosol in the BL is enhanced under cloudy conditions? This is contradicting to my intuition, as the cloud layer reflects SW back to the space, I would expect the SW heating in the BL to decrease instead of increase.

This paragraph has been rewritten and expanded. While clouds reflect SW back to space, the radiation encounters the aerosol layer above the cloud layer before it can leave at the top of the atmosphere. This means that within the aerosol layer, there are two opportunities for absorption.

10. It seems to me that MERRA-2 is not distributing enough BC/OC in the BL based on your Figure 2, 6 and 7 (clear sky condition). I wonder if you have compared MERRA-2 aerosol vertical profiles with extinction profiles from LASIC MPL (when available) or NASA ORACLES HSRL2 profiles or UK CLARIFY EXCALABAR profiles when they become available over Ascension during the 2017 season?

A figure showing backscatter from the LASIC MPL has been added to assist in an evaluation of the vertical profile of aerosol. MERRA-2 actually has more extinction from aerosol in the boundary layer than aloft, however this is almost completely a result of sea salt.

11. Regarding the LW cooling associated with the SW heating, the concern I have is that the observed temperature profiles (from LASIC radiosondes) had already felt that heating, in other words, the temperature increase was already taken into account in the observed profiles. By modifying the observed T profiles, you're artificially increasing the temperatures (artificially boosting the LW cooling). I would recommend just simply turn on the LW calculation using the same observed T profiles, and see if the net radiation budget produce a cooling or a warming.

We agree that by adding the heating rate to the temperature, the temperature profiles had already experienced heating due to aerosols. Accordingly, we have now reworked the LW calculation to account for this.

12. You should state whether this LW experiment is calculated with cloud presence or not.

The calculation was done without clouds which is now noted in the text.

13. The discussion in lines 17-32 on page 10 is particularly hard for me to digest. As we know LW cooling is always happening no matter aerosol presents or not, and the "LW cooling" you are talking about in this paragraph is the additional LW cooling caused by the increase in the temperature profiles due to SW heating (since it is done by subtracting a control run). The following points should be addressed properly in order to make the discussion clear.

a. When you say "LW radiational cooling never offsets the absorption due to aerosols," you should make this clear that you mean the additional LW cooling never offsets the absorption.

This has been clarified.

b. L 25, “magnitude of the LW cooling never reaches ...” same problem as above. LW cooling at inversions can easily reaches 10K/day at night. The LW cooling you are referring to is the difference between the T-modified run and the control run. Please make this clear.

This has been made clearer.

c. L 23, “radiational cooling still occurs...” As mentioned above, LW radiative cooling always occurs no matter the aerosol condition. Since you are showing the difference between the T-modified run and the control run, as long as there is SW heating due to aerosol (no matter how much), T profile will be modified, and difference in LW heating will exist. This cannot be used to demonstrate that additional heating due to aerosol remains in the column, you have to use the real LW heating profile to quantify that, not the difference between two runs. I strongly recommend re-assessing this LW part (see Major comment 11), at least the way you interpret/discuss it.

These points have been clarified in the text and we have changed the methodology for the LW calculations as suggested by Major comment 11.

14. Regarding your case study on the back trajectory, first, please specify reasons for originating at 2 km, second, why the meteorology for HYSPLIT runs are switched to GDAS instead of MERRA-2 as you did for the monthly back-trajectories, is MERRA-2 not capable to do ensemble runs? Please justify. Then, trajectories were forced by GDAS but radiative transfer calculations were using MERRA-2 thermodynamic profiles (why inconsistent)?

A height of 2 km was selected as the origin of the back trajectories based on the central location of the aerosol layer using micropulse lidar back scatter data. There are no ensembles associated with MERRA-2, whether related to the data assimilation process or the actual analysis. This point has been made clearer in the text. The same case study was carried out using the MERRA-2 back trajectories and has now been added to the figure. Given the uncertainty associated with back trajectories, we wanted to make sure there was a measure of the variability for the heating rates. As a result, that is why we ultimately decided to use GDAS. MERRA-2 is one of the few reanalyses that include the assimilation of aerosol optical depth, making it the obvious choice for aerosol properties that are consistent with a thermodynamic profile.

15. Why is the SW heating along the back trajectory limited to below the inversion? I would expect there to be aerosol in the FT along the 7-day back trajectories, and why not showing the aerosol and thermodynamic curtain plots along the trajectories from MERRA-2? In Lines 18-20, you’re saying the aerosol layer is entirely above the inversion along the trajectory, and yet, no heating above the inversion? This is very confusing, please justify.

This comment was extremely helpful. We found a bug in the aerosol input during our investigation, and the figure has been updated accordingly. The results are more reasonable now.

16. In the last paragraph, you mentioned that the ultimate goal is to study how the heating due to aerosols impacts the transition of marine stratocumulus to trade cumulus, I would really love to see more discussions added to the manuscript on how will this study help towards achieving this goal. For instance, how can this study contribute to the understanding of cloud adjustments to aerosols, and what insights can this study provide on the stratocumulus to cumulus transition in

the southeast Atlantic. Such discussions will substantially strengthen the scientific importance of this study.

We have added a paragraph to the conclusions that discusses our results in the context of the stratocumulus to cumulus transition.

Minor issues:

Abstract

• Line 30, you mentioned "stabilization of the lower troposphere," but this is not discussed anywhere in the main text of manuscript. I suggest adding discussion regarding this point you raised in the abstract.

A discussion on this topic has been added to the conclusion section.

Introduction

• P2 L21, you haven't introduced Ascension Island yet, a general reader would have no idea where the island is, near coast? or in the remote ocean? I suggest introducing Ascension Island somewhere in the introduction.

Ascension Island is now described in the introduction on line 21 of page 2.

• P3 L10-15, these information on datasets belong to the Data section, seems to me.

This paragraph has been modified with the information on the datasets getting moved to the Data section.

• P3 L16-18, these sentences seem to belong to the Methodology section. I would suggest adding more motivational statements, clarifying your scientific goals, here in the last paragraph of the introduction, replacing these details of datasets and approaches.

We contemplated moving these sentences but decided to leave them in the current section. As we have noted above based upon your previous comments, we agreed with your assessment about the motivation for the study and have made substantial changes to the manuscript in this regard.

Data and Methodology

• P4 L7-, Because of the location of the AMF1 site, orographically generated clouds frequent present in LASIC AMF1 cloud measurements, please address how will this feature affect your assessment and the general representativeness of your results.

We added a paragraph to the paper that discusses our analysis of the island effects on the cloud observed at the AMF1 site. In short, we analyzed the clouds observed during the entire year and found them to have unequivocally originated from the air near the ocean surface. Their cloud bases correspond to the marine LCL rather than the LCL at the AMF1 site. They are almost certainly modified by the orography given that we observe a systematic updraft of on average 0.5 m s^{-1} near the surface at the AMF1 site to near zero at 600 m. Orographically lifting all near-surface marine parcels to the near their LCL substantially increases the probability of cloud development and we recognize that this enhancement likely impacts our radiation transfer calculations. To adequately address this issue, we note in the revised manuscript that our radiation transfer calculations relative to clouds may serve as an upper bound to these impacts.

Please also specify the temporal resolutions of ARSCL and MICROBASE.

The temporal resolution of 4 seconds for ARSCL and MICROBASE is now noted in the text.

Results

3.1 Evaluation of Aerosols in MERRA-2

• P6 L23, my understanding of the location of AMF1 site is that it is elevated and located at the upwind part of the island, which should be representative of the aerosol condition of a marine boundary layer (minimal island effect). Besides, if indeed there were dust (more scattering) mixed into the AMF1 sampling volume, shouldn't we expect a higher SSA? (Zuidema et al. 2018b's values are lower). Please correct me if this is not the case.

The aeronet site is actually located by the airport, on the opposite side of the island from where the AMF1 was located. We have removed the statement regarding volcanic dust since we cannot prove that is the case.

• P6 L28-29, could be helpful if AODs are overlaid on top of Fig. 2.

This made the figure look too busy but the AODs are now including in the heating rate figures.

• P6 L34, the decrease in BL height is not very evident based on Fig. 2, overlaying some other forms of indication could be helpful.

We have removed to statement regarding the BL height. A decrease in BL height was more obvious in a previous version of the figure that included October as well August and September.

3.2 Thermodynamic Profiles over Ascension Island

• P7 L21, "time-height" should be time-pressure, as you're showing pressure in the vertical.

This has been corrected.

• P7 L22, cloud top inversions at Ascension are not around 700 hPa (~3 km). Please double check the pressure axes in Figure 4.

Yes, you are correct. This was a plotting issue that has been resolved.

• P7 L31, I do not see a "subtle, intermittent sub-layer at ~900 hPa" based on the RH curtain plot, perhaps this will be more visible in a single-profile presentation. Based on Fig. 13 of Zhang and Zuidema (2019), the intermittent layer seems to be located at ~700 m, which is lower than 900 hPa.

A figure has been added to show the monthly mean vertical profile of temperature and RH.

3.3 Heating Rate Profiles over Ascension Island

• P8 L21, it would be easier to visualize this co-variability between the heating rate and the AOD, if you could add MERRA-2 aerosol contours or AOD time series in the background of Fig. 6.

AOD contours have been added to the background of the figure.

• P8 L29, if it is hard to tell with the color bar, could you provide some values to indicate the difference between heating rates calculated from MERRA-2 SSA and RH scaled SSA?

The reader is now referred to Table 3 which shows the heating rates with the different SSAs.

•P9 L18-19, I see heating due to black carbon ~0.5 K/day extending to around 600 hPa just as in Figure 6, please re-state your argument about this observation. Besides, how do you know it is absorption from dust (isn't dust more scattering)? Please justify.

This paragraph has been rewritten with a figure than now shows the percentage of heating due to aerosols that is from black carbon.

• P9 L29, please define "enhancement of heating within the aerosol layer due to clouds" in the text or in the caption, i.e. how did you quantify that, is this cloudy-aerosol run minus clear-aerosol run (Fig. 6f – Fig. 6c = Fig. 8a?)?

A table has been added to clarify the quantities that are shown in the figures.

• P9 L30-32, "a few K per day"? the color bar on Fig. 8 only goes to 0.6, how did you get a few K per day? "...but when all aerosols are considered the majority of the enhancement is located ..." isn't this true for both 'All Aerosols' and 'Black Carbon'? Please check your logic here.

This should have been a few *tenths of a* K and has been corrected.

• P9 L32-34, could you please extend this discussion, especially on why this BL enhancement is not apparent for Black Carbon only case, even though BC is highly absorptive?

There is a minimal amount of black carbon in the boundary layer, which is now noted in the text.

• P10 L1-2, I think you should be careful here and say "...due to the presence of clouds..."

This has been modified as suggested.

• P10 L9 and thereafter, you used the phrase "radiational cooling," while I am more used to seeing "radiative cooling" being used in other literatures.

Radiative cooling is now used throughout.

• P10 L28-29, "some heating occurs above and below the aerosol," we can't tell where the aerosol layer is based on this plot, one option is to put MERRA-2 aerosol contours in the background. Another option is to show a line plot highlighting a single heating profile along with the aerosol profile.

Contours of AOD have been added to the figures.

• P10 L29, please discuss how is this redistribution of heat, as you put it, going to modify the stability of the boundary layer, as you pointed out in the abstract.

A discussion has been added to the conclusion section on this topic.

• P11 L21, please elaborate more on how the depth of BL affects the SW heating. Why minimal SW heating occurs in the last few hours of the back trajectory?

This is not present in the updated version of the figure following updates to the calculation.

• P11 L23, please explain or discuss your speculations on why SW heating maximized at the surface here.

This statement no longer applies to the updated figure.

Summary and Conclusions

• P12 L12, "...greater depth of the boundary layer..." comparing to what?

This statement is not in the revised version.

• P12 L16, "local heating rates are sensitive to the thickness of the aerosol plume" this is not discussed in the results section. Which figure supports this argument? Please mention this argument when you discuss that figure.

This is discussed as part of Figure 8, which is now noted in the text.

• P12 L20, "...most of the SW absorption" please be quantitative here.

It is now noted that up to 80% of the SW absorption can be due to black carbon.

• P12 L21, please be specific about this statement, i.e. which month? over Ascension or the whole SE Atlantic? Zuidema et al. 2018b states smoke often presents in the BL of Ascension Island, more frequent than "at times."

Despite the title of Zuidema et al. (2018), the conclusions presented in the paper are in good agreement with our use of "at times" for the months of August and September as there is a periodicity in aerosol concentrations. We have specified that we are referring to the months of August and September.

From Zuidema et al. (2018):

"1. Near-surface rBC mass concentrations vary significantly at synoptic time scales from June to October at the Ascension Island location….

3. The aerosol loadings within and above the cloudy boundary layer do not necessarily correlate well, with more of the total column aerosol present in the boundary layer early in the BBA season, migrating to predominantly free-tropospheric aerosol in September."

• P12 L23-24, I think what you want to express here is that adding a cloud layer will result in an enhancement of heating. Saying "interaction between SW radiation, clouds, and aerosols" is a bit misleading, as aerosols and clouds are not interacting in your calculations. I suggest a more careful rewording.

This sentence has been reworded.

• P12 L29, I didn't think you were trying to represent the entire southeast Atlantic using Ascension observations until I saw this statement, and I don't think this study should be used to represent the entire region. I suggest stating this clearly in the introduction or data section, that this study only represents the remote SE Atlantic, and cannot be used to represent the entire region. Also, you could change the title to "...remote SE Atlantic..."

This statement has been removed.

• P12 L31, "sensitive the heating ... is," sensitive to what?

This has been clarified to refer to the aerosol optical properties.

•P13 L15-20, the first and third sentences are the same sentence, please double check.

This has been fixed.

Figure/Table issues

1. Table 1, there are no italicized values in this table, please check.

Yes, that caption was for an older version of the table and has since been corrected.

2. Figure 2, please use 10 -5 instead of e-05

This has been modified as suggested.

3. Figure 4, please double check pressure axis, you are showing a 3 km BL.

The pressure axis has been fixed.

4. Figure 5, in my opinion, this figure can be removed, and all the white space above 800 hPa can be minimized.

Rather than removing Figure 5, the discussion pertaining to this figure has been extended. The y axis has been modified to only show below 800 hPa.

5. Figure 6, SSA instead of "SSA albedo." Again, the space above 600 hPa can be minimized, same for Fig. 7, 8 and 9.

This has been fixed.

6. I think for Figure 7 and after, you probably should remind the reader that we should compare these results only with the bottom panel of Fig. 6 (the RH scaled one), by making a note in the caption that SSA is the RH scaled one.

This is now noted in the figure captions as suggested.

7. Most of the results are presented in curtain plots, they are nice in terms of showing the whole month, but rather poorly representing details in vertical. I recommend showing couple plots with single profiles when you discuss details in vertical, especially when you discuss the relative location of heating/cooling to the aerosol layer.

A couple figures have been added that show the mean profile for the month for the thermodynamics and radiative heating.