Review replies to " Seasonal variations in fire conditions are important drivers to the trend of aerosol optical properties over the south-eastern Atlantic" by Haochi Che et al.

We would like to thank both reviewers for their comments and suggestions on the manuscript. The feedback has allowed us to clarify and improve our manuscript.

The reviewer's comments are provided in **blue in the following**, and our responses are in **black**. Changes to the manuscripts made in response to the reviewer are in **green**. In addition, numerous corrections in the manuscript are not shown in this response but are highlighted in the revised version.

Reviewer #1:

Che et al. investigated the effects of seasonal biomass burning in south central Africa on optical properties observed downwind at Ascension Island using observations from the LASIC campaign during two biomass burning seasons (June – October 2016, 2017). The authors leveraged the comprehensive set of LASIC optical measurements in combination with satellite products and reanalysis to show that changes in cloud processing, fuel source and type, and regional circulation influence the absorbance properties of the aerosol that reach Ascension Island. The paper highlights that the observations show a much more absorbing Southeast Atlantic boundary layer than is simulated by most climate models. This is an exceptionally thorough and convincing paper that is well written and presented. I only have a few minor comments that need to be addressed.

Minor Comments:

1. Line 57: add “the”; “during [the] CLARIFY (...) campaign”

We thank the reviewer for this correction. The manuscript has been corrected accordingly.

2. Line 127: (Pennypacker et al., 2020) described an Ascension Island biomass burning background BC median concentration of 20 ng m-3 (ultra clean median of 51 ng m-3) with some monthly variability during the LASIC campaign. Was this/these value(s) also considered for the background calculation of BC/dCO as was done for dCO? If not, can you speak to how you think these might impact the calculation and subsequent interpretation of results?

We thank the reviewer for this comment. The background condition is slightly different in our manuscript than Pennypacker et al. (2020). The background value in their paper is calculated from the non-burning season from December 2016 to April 2017, which represents the clean state during the period when Ascension Island is not affected by the African BB aerosols. However, due to the limitations of the instrument and signal noise, even under very clean conditions, SP2 gives a signal of rBC mass, which is interpolated as this 20 ng/m^3 background value in their study. In our work, we have removed all data with rBC < 20 ng/m^3 to reduce the influence of noise when the atmosphere is clean. We set the rBC value to 0 ng/m^3 as a background, which is an ideal state indicating that BC aerosols are completely removed and the atmosphere is currently unaffected by biomass burning. For the background CO, our values generally vary within the range between 50-

60 ppb, consistent with the previous observations that reported background CO concentrations between 50-60 ppb (Allen et al., 2008, 2011; Shank et al., 2012).

In a previous version of our manuscript, the BC background concentration was also set to the lowest monthly 5th percentiles, the same as the background CO. Then the derived BC background value varied between 22 and 45 ng/m^3. This did not affect the conclusions of our article, and ΔBC/ΔCO still showed a good linear relationship with SSA and MAC_{BC}. (Figure R1)

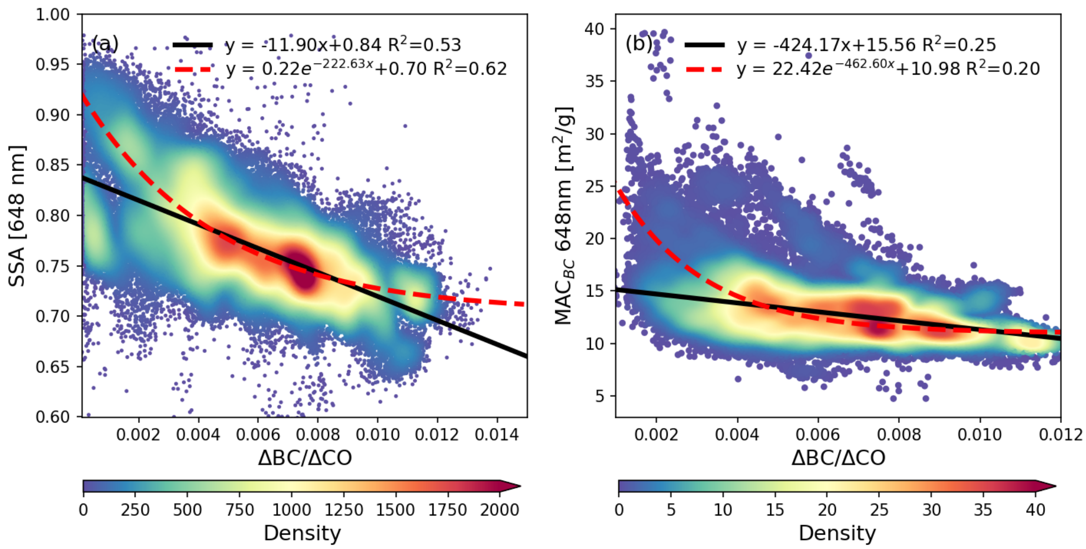

Figure R1. Relations of ΔBC/ΔCO with (a) SSA and (b) MAC_{BC} at 648 nm wavelength. The black lines represent the linear regressions, and the red dash lines are the curves fitting with the function displayed in the legend. The colour scale indicates the density of the data in each plot.

To make this clearer, we have revised the manuscript as the follows.

The background concentration of BC was considered to be zero, representing the ideal state where BC aerosols are completely removed and the atmosphere is free from BB. Whereas the background concentration of CO was taken as the lowest 5th percentiles of the observations per month, to approximate the clean state during the BB season. The background CO values ranged from 50 to 60 ppb (Fig. S1 in the supplement), consistent with the previous observations in the Southern Hemisphere (Allen et al., 2008, 2011; Shank et al., 2012). Hence, ΔBC/ΔCO is equal to BC/ΔCO, and this ratio will be used in the ensuing analysis. Note the CO was converted to the same mass unit as the BC; therefore, BC/ΔCO is a unitless parameter. Note that BC values less than 20 ng/m^3 are removed to reduce the effect of instrument noise signals in a clean atmosphere.

3. Line 208: delete “the”; “the July-September averaged MACBC measured in 2016 on [the] ASI”

We have corrected this sentence.

4. Line 235: change “consisting” to “consistent”

Thank the reviewer, we have corrected this sentence.

5. Meteorological factors (Section 4.3.2): Although this section discuss the impact of winds on combustion conditions, the authors should also make note of the effects of the Southern African easterly jet and it's potential influence on the seasonal optical properties at Ascension. Specifically, (Adebiyi and Zuidema, 2018) described a peak in the AEJ-S during the September – October period when the transport of BB aerosol can be very efficient. Were jet-level winds/trajectories (~600 hPa) also examined as a part of this study?

We thank the reviewer for this comment. Here we show the monthly mean winds at 600 hPa in the figure R2. The wind is calculated from ERA5 data. From June to October, the average easterly wind speed increases within the main BB transport area (the grey area in Fig. R2). Thus, from June to October, the strengthening of the easterly winds is favourable for the transport of BB aerosols. From June to August, BB aerosol concentration observed at ASI increases, consistent with changes in the easterly jet, but BB aerosol concentration decreases from September to October, in contrast to changes in the easterly jet.

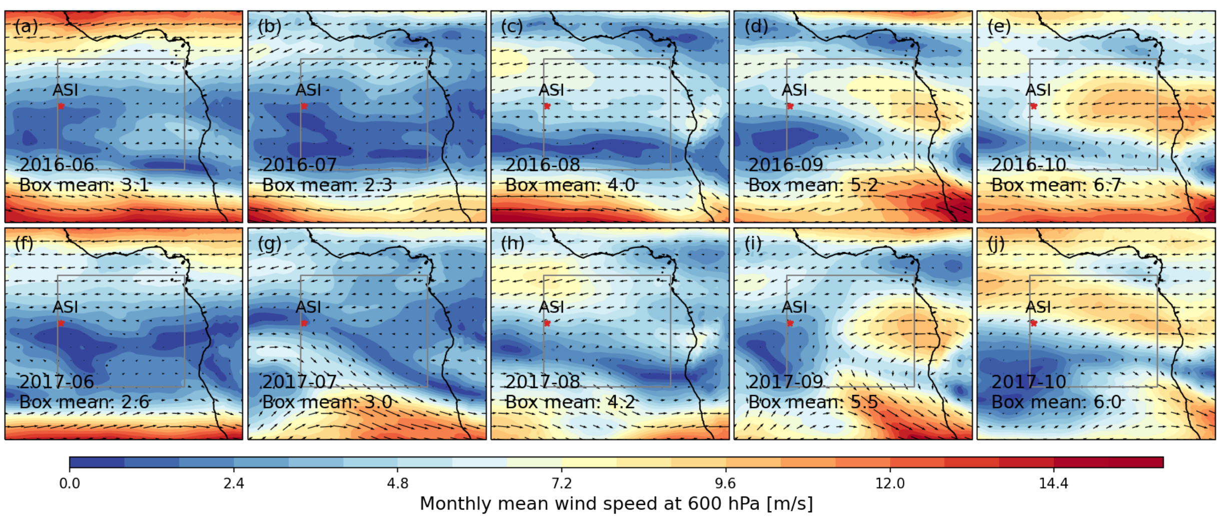

Figure R2. Monthly mean wind at 600 hPa was calculated from the ERA5 reanalysis data. The location of ASI is marked as the red star. The grey boxes represent the range of the main transport paths of BB aerosols observed on ASI. The box means are the mean wind speed in the grey box region.

In our manuscript, we firstly used BC/ΔCO to proxy the optical parameters of the aerosol (SSA and MAC_{BC}) observed at Ascension Island. Then we discussed the monthly variation in BC/ ΔCO and, in turn, the causes of the variation in those aerosol optical properties. The magnitude of BC/ΔCO depends primarily on two factors, one being the condition of the BB combustion, and the other being the scavenging of aerosols during the transport of the plume. Although the increase in BB aerosol concentrations observed on the Ascension Island from June to August is consistent with the easterly jet, the easterly jet does not directly affect the BC/ΔCO. This is because the easterly jet does not directly remove BC aerosols, and therefore it does not directly change the magnitude of BC/ΔCO. However, when easterly winds strengthen, BB aerosols can be transported further away from the African coast and over warmer oceans. The cloud top height in these regions may be higher, which in turn may produce a cloud removal effect on BB aerosols. The monthly contribution of cloud to the BB aerosol removal is discussed in the manuscript.

We have revised the manuscript to acknowledge the variation of the easterly jet as follows.

The increase in aerosol absorption coefficients observed at ASI from June to August indicates an increase in BC loading. This most likely reflects an increase in burning, which has a climatological peak in August (Scholes et al., 1996). Upper-level zonal winds are still weak in June and July, and are strongest in September and October (Adebiyi and Zuidema, 2016). During August, the developing heat low over land strengthens the free-tropospheric zonal winds, with a maximum at approximately 700 hPa (Ryoo et al., 2021). This will facilitate the transport of aerosol to the Ascension boundary layer, in addition to the increased burning (see, e.g., example in Zuidema et al., 2018). From August to September-October, the aerosol absorption coefficient decreases, in contrast to the continued strengthening of the easterly jet. This may occur in part because the strongest zonal winds are now at a higher altitude (~600 hPa), discouraging entrainment of the aerosol into the boundary layer. The burning on land also diminishes swiftly in October as moist convection moves southward, generating less available aerosol for transport (Ryoo et al., 2021; Adebiyi et al., 2015).

Reviewer #2:

The authors analysed the seasonal variations in the optical properties of aerosols observed at Ascension island as well as the causes of these seasonal variations. One significant contribution is that they found the lower aerosol single scattering albedo during biomass burning season in Africa and suggested that the burning conditions are the major factor in the variation of aerosol optical properties. The paper is well written with comprehensive analysis and interesting findings, which should be published. However, I believe that some definitions and explanations need to be clearer and I therefore recommend that this paper be accepted with minor revision.

Specific comments

1. Line 24: change 'The absorption enhancement Eabs' to 'The absorption enhancement (Eabs)'

Thanks to the reviewer, we have corrected it in the manuscript.

2. Line 23 to 25 and other places: Please be consistent with if there is a space needed between the symbol '~' and numbers.

Thanks to the reviewer. We have corrected the spacing throughout the manuscript.

3. Line 27: '(BC/ΔCO)' looks like should be 'the enhanced ratio of BC to changes in CO (BC/ΔCO)'.

The enhanced ratio of BC to CO is actually ΔBC/ΔCO. Since we consider the background BC concentration to be 0 , ΔBC/ΔCO is essentially equal to BC/ΔCO. We have revised the manuscript to make it clearer (L).

We find the enhanced ratio of BC to CO (ΔBC/ΔCO, equal to BC/ΔCO as the BC background concentration is considered to be 0) is well correlated with SSA and MAC_{BC}.

4. Line 29: 'better capture' compared to what?

We thank the reviewers for their comments. In our manuscript, we present a linear function and an exponential function to represent the relationship between aerosol optical properties (MACBC, SSA) and BC/ΔCO. We compared the two methods and found that the exponential function better captures the variation in aerosol optical properties when the BC/ΔCO is small. As we do not mention linear functions in the abstract, the word "better" may lead to misunderstandings. We have therefore revised the sentence as follows:

The exponential function we proposed can approximate SSA and MAC_{BC} with BC/ΔCO, and when BC/ΔCO is small it can capture the rapid growth of SSA as BC/ΔCO decreases.

5. Line 34: Maybe specify 'these period' refers to which period. Probably also in line 18 specify the month-year period of the 17-month campaign.

Thanks for the suggestion. We have revised the manuscript to make it clearer.

The reduction in the water content of fuels may be responsible for the change in the burning conditions from June to August.

From June 2016 to October 2017, a 17-month in-situ observation campaign on ASI found a low single-scattering albedo (SSA) as well as a high mass absorption cross-section of black carbon (MAC_{BC}), demonstrating the strong absorbing marine boundary layer in the south-eastern Atlantic.

6. Line 44 and other places: 'the savannah region' to 'the Savannah region'

We have corrected this in the manuscript.

7. Line 71: any explanations to why the simulation of absorption in this region is bad in climate models? Are the simulations in other regions, by any chance, better?

This is mainly because our observed SSA on ASI is small during the BB season, with a very low monthly mean value of 0.78 (at 550 nm) in August. In addition, the E_{abs} of BB aerosols are about 2 during the BB season and up to 2.4 in October, whereas many models typically have E_{abs} of 1.5 (Wang et al., 2014), which can lead to an underestimation of the absorption of aged BC aerosols.

This bias is mainly due to the thick coating of BC particles. We only evaluated the southeastern Atlantic region, as this is our primary interested area.

8. Line 72: maybe worth mention which months are the BB season here.

Thanks for the suggestion, we have revised the manuscript.

During the BB season (from June to October), the physical and chemical properties of BB aerosols change with variations in combustion conditions, source fuel, and meteorological conditions, resulting in variations in the optical characteristics of BB aerosols (Pokhrel et al., 2021).

9. Line 88: 'MACBC' should be MAC_{BC}

Thanks for the correction. We have corrected this in the manuscript.

10. Line 95: latitude and longitude should be in degree and indicating which hemisphere, e.g. 7.97 °N.

Thanks for the correction. We have corrected this in the manuscript.

LASIC campaign was carried out on the Atmospheric Radiation Measurement (ARM) Mobile Facility 1 site at Ascension Island, located at the latitude of 7.97 °S, the longitude of 14.35 °W and the altitude of 340.8 m

11. Line 101: either be '464nm, 529 nm, and 648 nm' or '464, 529, and 648 nm'.

Thanks for the correction. We have revised the manuscript to be consistent.

Absorptions were measured at 464, 529, and 648 nm wavelengths, while scattering was at 450, 550, and 700 nm.

12. Line 101 to 105 and hereafter: why switch to past tense in these sentences, while you used present tense in most other places.

Thanks to the reviewer for the correction. We have changed the past tense to the present tense for consistency.

13. Line 103: change 5 Mm^{-1} to 5 M m^{-1}.

We apologise for the misunderstanding. Mm is a single unit, representing megameters. 1Mm = 10^6 m. The most common unit for the aerosol absorption/scattering coefficients is Mm^{-1}.

14. Line 107: 'represent' should be represents

Thanks for the correction. We have corrected this in the manuscript.

15. Line 137 and Figure 3 caption: '9:43-9:49' refers to am or pm?

Thanks for the correction. The time is indeed UTC time. UTC uses 24-hour time notation. Thus 9:43-9:49 refers to AM.

We have corrected the manuscript to make it clearer.

sampled at 2017/08/24 9:43-9:49 UTC, at 319.4 m

16. Figure 1. The 10th percentile for aerosol absorption in July 2017 is less than 0. Aerosol absorption should not be less than 0. The authors might need to double check.

We are grateful to the reviewers for pointing this out. We have removed all negative data and replotted the figure.

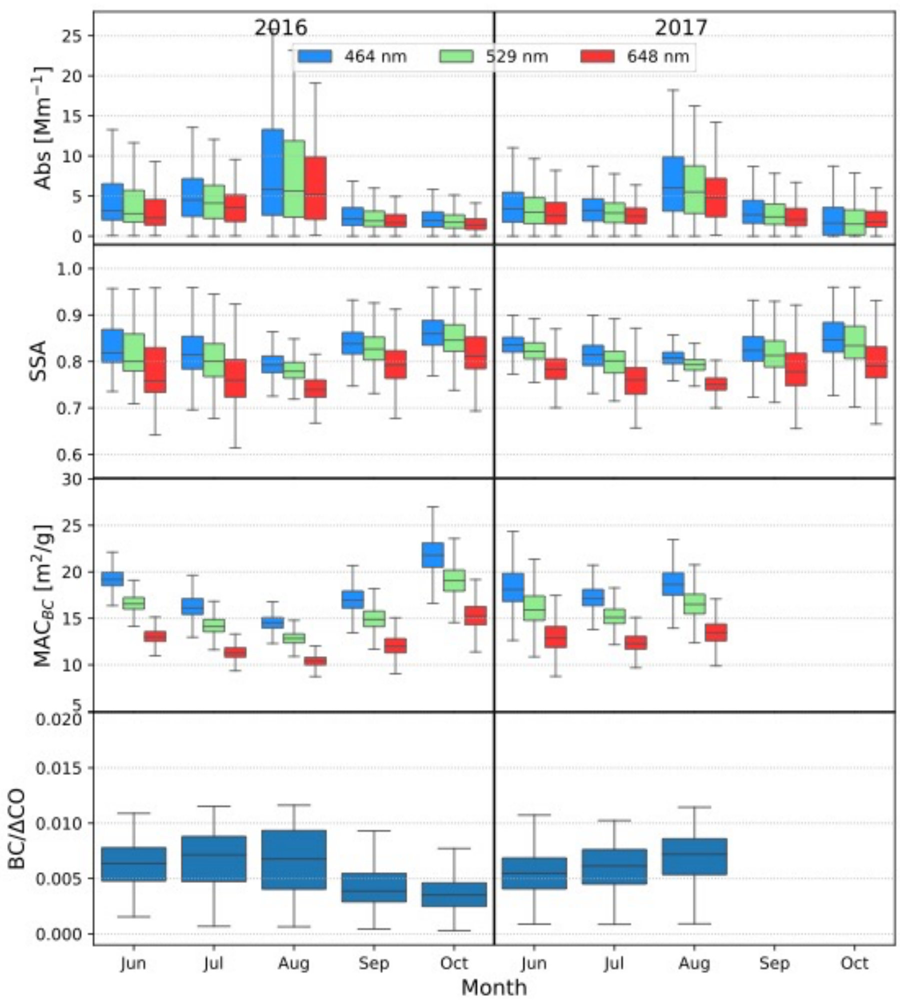

Figure 1. Monthly percentiles (10%, 25%, 50%, 75%, and 90%) of aerosol absorption coefficient, SSA, MAC_{BC}, and BC/ΔCO as box-whisker plots spanning the fire season in 2016 and 2017. The light blue, green and red colours of absorption, SSA, and MAC_{BC} indicate observations at wavelengths of 464, 529, and 648 nm, respectively.

17. Line 205. 'Taylor et al. (2020) found an averaged Eabs around 1.85..'. Is this average for the whole campaign or is it an average at a certain height? Is it within a boundary layer?

This value is the campaign average, not at a certain altitude. It includes observations in the marine boundary layer and free troposphere.

To make this clearer, we have revised the manuscript as follows.

Taylor et al. (2020) found an average E_{abs} around 1.85 around ASI (campaign average, including boundary layer and free troposphere).

18. Line 207: 'Zuidema et al. (2018) reported the July-September averaged MACBC measured in 2016 on the ASI as 15.1, 13.3, and 10.7 m2 g−1 at 464, 529 and 648 nm, which corresponds to Eabs as 1.70, 1.71, and 1.68 respectively". The authors state earlier that the errors in the Eabs for 464 and 648 nm are large (because of the uncertainty in the AAE), and only the Eabs for 529 nm are compared. Then why caculate and compare the Eabs at 464 and 648 nm from Zuidema' results?

We thank the reviewer for this comment. Yes, only the E_{abs} at 529 nm were used for comparison. Therefore, we removed the E_{abs} values at the other wavelengths and revised the manuscript.

Zuidema et al. (2018) reported the July-September averaged MAC_{BC} measured in 2016 on ASI as 13.3 m^2 g^{-1} at 529 nm, which corresponds to E_{abs} as 1.71. The relatively low value they observed is due to the lower enhancement between July and September, with August having the lowest average E_{abs} of around 1.64.

19. Line 218. 'suggesting BrC have a minimal influence on the ASI'. Not sure I understand what does it mean here.

We have revised the manuscript to make it clearer.

Fig. 2 shows the evaluated contribution from BrC on total absorption at 464 nm is generally around 1-2%, suggesting BrC have a minimal influence on the aerosol absorption measured on ASI.

20. Line 235: 'consisting' should be consistent.

Thanks for the correction. We have corrected this in the manuscript.

21. Line 261: Do the authors have data (or other literatures) from actual measurements of coating thickness to support their conclusions?

The LASIC data we used in the manuscript is from the ARM archive: https://adc.arm.gov/discovery/. The database does not currently contain the coating thickness of BC particles. However, Taylor et al. (2020) calculated a shell-core ratio of about 2.3 for BC particles in the marine boundary layer from CLARIFY campaign, which is very close to our estimate of 2.2.

We have revised the paper to include the findings of Taylor et al. (2020) to support our results.

We estimate the shell-core ratio (spherical equivalent particle diameter divided by BC core diameter) is ~ 2.2 on average, suggesting that the coating thickness is 1.2 times the BC core radius. This result is consistent with that of Taylor et al. (2020), who found the shell-core ratio of BC particles around 2.3 in the marine boundary layer from CLARIFY aircraft campaign.

22. Line 276: 'curves fitting' shoud be curve fittings

Thanks for the correction. We have corrected this in the manuscript.

23. Line 330: 'Our measured BC/ΔCO values are generally smaller than 0.012, suggesting this linear relation is applicable to our data'. For fresh plume, MEC and BC/ΔCO have a linear relationship when BC/ΔCO is less than 0.015. But the BC/ΔCO observed by the authors are for aged smoke, does the threshold of 0.015 still be applicable?

We thank the reviewer for this comment. Yes, May et al. (2014) found the growth of MCE becomes much slower for BC/ΔCO greater than 0.015, suggesting the relationship between BC/ΔCO and MCE is not linear across the entire range. However, in Fig. 5 (in the manuscript), we can clearly see that overall, MCE increases with BC/ΔCO. This relationship suggests that BC/ΔCO can be used to represent the combustion state of the fuel.

Here we are trying to establish that conclusion. But the reviewer is right, 0.015 may not be applicable to our aged BB plume. We therefore have revised the manuscript as follows.

The result shows a reasonable linear relationship of MCE and BC/ΔCO, with the R^2 value = 0.44. Therefore, this result suggests that with the increasing fraction of flaming combustion, BC/ΔCO also increases. The relationship of MCE with BC/ΔCO therefore suggests that one of the major factors contributing to the variation in the observed BC/ΔCO is the change in combustion state from the BB region.

24. Line 347: 'with the relationship between BC/CO and SSA'. Do the authors mean BC/ΔCO?

Yes, we thank the reviewer for the correction. We have corrected it in the manuscript.

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