Responses to all reviewer's comments. The reviewer comments are italicized and our responses are not.

Reviewer #2

This manuscript presents an investigation into the direct and indirect radiative effect form brown carbon using the CESM model. In all, this work is well designed, executed, and of broad interest to the aerosol science community. The writing is clear and thorough. It is worthy to be published on ACP after minor revisions.

Reply: We thank the reviewer for the positive and encouraging comments.

1. *Current knowledge of both brown carbon and how aerosols interact with cloud are with large uncertain, therefore I can imagine the uncertainties in this study may be the subject of discussion. While conducting a very accurate analysis is nearly impossible at this point, I suggest the authors to include a short discussion for the uncertainties. For example, a more reliable range of the radiative effect may be more useful than the global mean numbers.*
Reply:
We appreciate the reviewer's comment about increased discussion regarding model uncertainty. In regards to a more detailed range in BrC RE, sections 3.2 and 3.6 detail regions of significant REari and REaci. We feel that this gives a good representation of the global range in BrC RE. Furthermore, the three model runs NOBRC, BRC_BL, and BRC give a range of 100% immediate bleaching of BrC to no bleaching of BrC. We mention this in the discussion.

We made a change on **page 19, line 20** – "The range in our in BrC REari is from 0 to 0.13 W m^{-2}, representing the effect of emission of 100% immediately bleached BrC (NOBRC) to no bleaching (BRC)."

We added a discussion for the uncertainties in BrC RE (see responses to the reviewer's comment 2 below).

2. *There are also a few questions need to be discussed. How large is the uncertainty? What processes contribute the uncertainty, and what are the most important? What kind of laboratory/field measurements are most useful for reducing the uncertainty?*
Reply:
We have taken special care to include more information regarding the uncertainty in the implemented parameterizations as well as in the model set-up (sections 2.2.2 and 2.2.3). We also included a discussion of information needed to minimize uncertainty in the model at the end of the discussion.

We made a change on **page 6, line 1** - "The GFED 3.1 emissions were used in this study to allow for direct comparison between this study and Jiang et al. (2016). The more recent GFED 4 emission dataset shows an 11% global increase in fire emissions from

GFED 3.1 (Werf et al., 2017), which may result in a slightly stronger climate impact from biomass burning aerosols than that shown in this study."

We made a change on **page 6, line 25** - "Uncertainty in k_{OA} from this parameterization is associated with the lab measurements of the particle mass, the range in assumed complex refractive index for BC, the mixing state of BC and OA, the measured real part of the OA refractive index, and the measured absorption coefficients used in optical closure calculations (Saleh et al., 2014)."

We made a change on **page 7, line 12** - "A few assumptions in this model simulation introduce uncertainty in the representation of BrC in CESM. One of those assumptions is neglecting absorption by BB SOA (Lin et al., 2014; Saleh et al., 2015) or absorbing aromatic SOA (Wang X. et al., 2014; Jo et al., 2016; Wang X. et al., 2018), which is neglected due to the lack of SOA speciation in the model. This assumption, in conjunction with the use of GFED 3.1 instead of GFED 4, may act to underestimate the climate effect due to BrC. Another assumption is the model use of a volume mixing assumption, which may act to overestimate aerosol light absorption (Jacobson, 2000; Adachi et al., 2011). We also assume that the BC-to-OA ratio in transported smoke is similar to BC-to-OA from the source region, allowing for the use of a BC-to-OA ratio at each gridcell at every time step to calculate k_{OA} in each gridcell. The uncertainty in k_{OA} associated with this assumption is small (<10% for BB emissions assuming transport from the Equator to the Arctic (not shown)) and is assumed to be negligible.

Another source of uncertainty when considering an absorbing aerosol in the model is the aerosol's vertical distribution. CAM5.4 uses six vertical injection heights for wildfire emissions described in Detener et al. (2006): 0-100 m, 500-1000 m, 1-2 km, 2-3 km, and 3-6 km. These fire emission heights depend on the geographic location of the fire and the vegetation type derived from GFED, with the highest plumes corresponding to boreal fires. If BrC is lofted over a more reflective surface such as a cloud, its shortwave radiative forcing will be more positive than if it stays below the cloud or remains lower in the atmosphere. A counterbalancing effect is a more negative longwave forcing at higher levels in the atmosphere (Penner et al., 2003). The vertically sensitive semi-direct effects of BrC (i.e., changes in atmospheric stability and cloud cover due to atmospheric heating by BrC) are discussed in more detail in section 3.2. Comparisons between the total OA (POA + SOA) vertical distribution and aircraft observations in Shrivastava et al. (2015) show that the standard CAM5 aerosol treatment largely underestimates Arctic biomass OA, possibly due to the model neglecting important SOA contributions from biomass burning. This could lead to an underestimation of BrC radiative effects due to lower BrC concentrations at all levels of the model."

We made a change on **page 8, line 12** - "While the parameterization depends on OH concentration in the atmosphere, by matching the BrC lifetime to observations the parameterization also includes photochemical oxidation and other bleaching effects that may have been active in the observed smoke plumes. This is true of the regions in which the observations were taken, but may not hold true for global sites or seasons with lower insolation. Uncertainty in this parameterization is associated with the low availability of

observational data, and could be improved with more field measurements of BB smoke aging at different latitudes."

We made a change on **page 20, line 32** - "Also, observational datasets looking at vertical distribution of BrC in the atmosphere would help to determine whether the model is simulating similar processes to observations. This includes more information regarding the transport of BrC to upper levels by deep convection, and the in-cloud aqueous production of BrC (Zhang et al., 2017). GFED emission inventory accuracy is also important because the reported fuel-type and location play a role in the model vertical distribution of carbonaceous aerosols. More observations of BrC bleaching would help refine the bleaching parameterization used in this study by determining if there are geographic differences in bleaching effect due to differences in solar irradiance. Lower BrC bleaching rates in the Arctic suggest important contributions from BrC deposition on snow. Including more measurements of the radiative effects of BrC impurities in snow could help in the validation of future models that include this surface effect. Lastly, measurements of combustion and non-combustion sources of BrC SOA, as well as their composition/evolution, could aid in the development of BrC SOA in CAM."

Specific comments:

3. *Page 4, line 12: I guess it is 25% instead of 0.25%.*
Reply:
We have fixed this value.

4. *Page 5, line 10: Could the authors give more information about the aerosol size distributions? What are the median sizes and standard deviations for each mode? Is there microphysical process changing the size in the model? Please also provide the information of BrC density here even it is discussed later.*
Reply:
We included a better description of the median size distributions and included the standard deviations for each mode. We also mentioned in more detail how the aging process converts Primary Carbon mode to Accumulation mode, increasing the size and hygroscopicity of the primary carbon mode as it ages. We added a mention of BrC density in section 2.2.2 when we discuss the BrC parameterization in the model.

We made a change on **page 5, line 9** - This model also uses the 4-mode version of MAM (MAM4) (Liu X. et al., 2016). MAM4 consists of the following four lognormal modes (shown with their median size ranges and standard deviations): Aitken (0.015 – 0.053 μm, σ = 1.8), accumulation (0.058 – 0.27 μm, σ = 1.6), coarse (0.80 – 3.65 μm, σ = 1.8), and primary carbon (0.039 – 0.13 μm, σ = 1.6). The median sizes of aerosol modes are changed due to the microphysical processes (e.g., condensation and coagulation) while standard deviations for each mode are fixed. The OA from accumulation and primary

carbon modes, which is used to represents BrC depending on its source (mentioned later), has a density in the model of 1 g cm^{-3}.

5. *Page 5, line 18: does it mean the model assumes totally internal mixing everywhere? The influence of mixing assumption worth a discussion in later sections.*
Reply:
The model assumes internal mixing within the mode and external mixing between the modes when calculating the optical properties of each aerosol mode. We mention the impact that this treatment may have on the radiative effects in the model when discussing uncertainties

We made a change on **page 5, line 21** - "The optical calculations in the model consider these aged particles to be internal mixtures of aerosols within the mode, and the RI of the accumulation mode, as well as the other 3 modes, is calculated as a volume-weighted mean of the refractive indices of all of the aerosol's components within the mode (Liu X. et al., 2012; Liu X. et al., 2016)."

We made a change on **page 7, line 16** - "Another assumption is the model use of a volume mixing assumption, which may act to overestimate aerosol light absorption (Jacobson, 2000; Adachi et al., 2011)."

6. *Page 5, line 27: Is there any reason to use GFED3.1 instead of the current version of GFED4?*
Reply:
GFED3.1 was used in order to allow comparison to the Jiang et al. (2016) study which focused on the radiative effects of biomass burning aerosols (neglecting BrC). It is a valid point that using this older version differs in several aspects from the most recent emissions dataset, and we add some discussion regarding the effect of this change on the results.

We made a change on **page 6, line 1** - "The GFED 3.1 emissions were used in this study to allow for direct comparison between this study and Jiang et al. (2016). The more recent GFED 4 emission dataset shows an 11% global increase in fire emissions from GFED 3.1 (Werf et al., 2017), which may result in a slightly stronger climate impact from biomass burning aerosols than that shown in this study."

7. *Page 6 the first equation is not clear.*
Reply:
Equation (1) has been modified to

$$RI = 1.7(\pm 0.2) + k_{OA}i = 1.7(\pm 0.2) + k_{OA,550}\left(\frac{550}{\lambda}\right)^{w} i$$

8. *Page 6, line 24: I cannot understand how the BC/OA ratio is used in this parameterization. Is the BC/OA ratio calculated from the emissions for every grid box at every timestep? Does this mean you assume all the BrC currently simulated in the grid box has the same absorption property as those emitted locally?*
Reply:
That is correct. The BC/OA ratios described in the model run don't change substantially within biomass burning emission plumes, leading us to assume a relatively small effect on the imaginary part of the refractive index. When assuming that high BC/OA emissions from Africa (BC/OA = 0.1) can make it up to the Arctic (BC/OA = 0.06), we find that k_{OA} uncertainty is just under 10%. Given the reduction in aerosol absorption due to wet and dry deposition as well as dilution, the effect of this uncertainty on the radiative effect is further reduced. We address this in our uncertainty discussion and assume that this uncertainty is negligible.

We made a change on **page 7, line 17** - "We also assume that the BC-to-OA ratio in transported smoke is similar to BC-to-OA from the source region, allowing for the use of a BC-to-OA ratio at each gridcell at every time step to calculate k_{OA} in each gridcell. The uncertainty in k_{OA} associated with this assumption is small (<10% for BB emissions assuming transport from the Equator to the Arctic (not shown)) and is assumed to be negligible."

9. *Page 8, line 20: What version of AERONET data is used? level 1.5 or level 2? How good are the AERONET data? How many data points do you have (% per season)?*
Reply:
We use level 2.0 AERONET data and add mention of this on page 9, line 25. There is more available AOD data than SSA data due to the uncertainty involved in the inversion algorithm used to calculate SSA in low aerosol environments (Dubikov et al., 2000). We added percentages of data for the 9-year period for each of the months in the AOD, AAOD, and SSA plots, under the upper x-axis.

Modified **Fig. 3,** and added **Figs. S3, S4.**

10. *Page 8, line 24: I cannot understand why the authors used 550nm data in their analysis. 440nm is a much better choice to evaluate BrC.*
Reply:
Figures were modified to reflect 440 nm AERONET data.

Modified **Figs. 3, S3, S4.**

11. *Figure 3 is not that useful for the readers, maybe combine Figure 3 and 4 to an absorption AOD plot.*
Reply:
We have included an AAOD plot in the supplementary. We did not include this in the main text because the change in AAOD magnitude between the different sites is large, and the large uncertainty in model AAOD makes for a less useful model comparison. We left the SSA plot in the paper and moved the AOD plot to the supplementary.

We added **Fig. S4.**

12. *Page 9, line 28: what wavelengths are λ1 and λ2?*
Reply:
λ1 = 440 nm and λ2 = 675 nm. This was added to the paper.

We made a change on **page 10, line 31** - "AAE is calculated based on the two wavelegths 440 nm and 675 nm (λ_1 and λ_2, respectively) and the measured absorption coefficients at the two different wavelengths ($b_{abs}(\lambda_1)$ and $b_{abs}(\lambda_2)$)."

13. *Before talking about the BrC effect. It may be worthy to describe the model result of BrC absorption briefly. For example, global mean absorption AOD and AAE, spatial and vertical variations of BrC absorption, contribution to total aerosol absorption, etc.*
Reply:
We have added two plots that show the difference in AAOD, AAE, zonally averaged absorption coefficient, and zonally averaged AAE due to the BrC implementation.

We have added **Fig. 5** and **Fig. 6.**

We made a change on **page 12, line 11** - "This can be seen in Fig. 5a which shows the difference in AAOD between the BRC and NOBRC model runs. The max AAOD over southern Africa is about 0.024, which makes up approximately 1/3 of the total AAOD in this same region (Fig. S7a). As with Fig. 4, Fig. 5b shows a global increase in AAE due to BrC. These effects are the strongest over the southern African BB region and in the Arctic, with the strongest AAE increases over the Arctic (Fig. 5b) correlated with weaker AAOD in Fig. 5a. Vertical cross-sections of aerosol absorption coefficient and AAE changes due to BrC (Fig. 6), show the vertical extent of BrC. Zonal BrC absorption is dominated by the African and South American biomass burning regions, with visible aerosol transport to the Arctic from boreal fires (Fig. 6a). While absorption over the Antarctic is nearly zero, upper level transport of dilute BrC to the south can be inferred from the AAE changes in Fig. 6b.."

14. *There are a lot of measurements of BrC absorption in the literature (most at the surface). A brief comparison between model and these observations could provide useful information beyond the limited model validation by AERONET.*
Reply:
We include a table of comparison between three different AAE surface measurements: Ascension Island observations from the LASIC (Layered Atlantic Smoke Interactions With Clouds (LASIC) campaign, the Las Conchas fire in NM, and biomass burning and prescribed burns from SEAC⁴RS (Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys). We also mention biomass burning AAE from three 2018 fires in NM.

We added **Table 2.**

We made a change on **page 11, line 20** - "Other comparisons between the model and observational AAE are shown in Table 2. These observations are from Ascension Island (Zuidema et al., 2018), a southwestern US (New Mexico) wildfire (Liu S. et al., 2014), and continental US BB and prescribed burns from the SEAC⁴RS campaign (Mason et al., 2018). These comparisons are largely qualitative given the coarse resolution of the model (~100 km) compared to the much finer wildfire spatial resolution. Both the Ascension Island and New Mexico observations are from the surface, while the continental US observations are made via aircraft. The Ascension Island and SEAC⁴RS US BB/BF observations were made during different years than represented in the model run, so our comparison operates under the assumption that these regions have consistent seasonal smoke exposure from 2003-2016. The bleaching effect (BRC_BL) drives the model closer to the observations over Ascension Island, but still acts to overestimate the AAE along with BRC and BRC_CNST. This could suggest either too much BrC transport to this region in the model, too weak a bleaching effect, or too much of some other aerosol with a higher wavelength dependence (i.e., dust). The models underestimate AAE over the US, especially in the BRC_BL and NOBRC model runs. This could be due to an influence of fossil fuel emissions in the model grid cell driving the AAE closer to 1.

Other observations of forest fires from Los Alamos National Laboratory (LANL) in New Mexico from 2018 show the variability of measured fire AAE. These include the Buzzard Fire, the Ute Fire, and the San Antonio Fire (with average AAEs of 1.44, 1.42, 2.38, respectively) (Romonosky et al., submitted for publication). Due to their being outside the model run period and their intermittent nature, these fires are not directly compared to the model. However the Las Conchas model comparison from Table 2 is compared to the other fires assuming a similar regional fuel composition in the model. The model with the bleaching effect is closer to the Ute and Buzzard fires than to the Las Conchas and San Antonio fires, and overall the inclusion of BrC brings model AAEs closer to the New Mexico fire measurements."

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