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

Reviewer #1

This paper advances the understanding of the global radiative impacts of light absorbing PM organic species (BrC). It appears to be the first research to included BrC in an earth system model (CESM), which goes beyond previous models that only considered BrC direct radiative forcing effects. This more advanced model considers factors such as surface albedo, clouds and various atmospheric dynamic processes. By including the important process of BrC bleaching, Wang et al 2018 made a substantial improvement over previous models of BrC global impacts that assumed it was largely invariant once emitted. This model also considers bleaching, (although only resulting from particle reaction with OH), and with added secondary BrC effects, is likely the most advanced to date.
Reply:
We thank the reviewer for the positive and encouraging comments.

All these models, including the one described in this paper, are still overly simplistic and so the results highly uncertain. The fundamental problem is that not all the processes that influence BrC are known, and there are really no global scale data sets of BrC which can be used to test the model predictions. As has been done in some prior studies, AERONET data are used in this work, but provide only limited validation (inclusion of BrC shows better agreement with AAEs). Because of the advances in modeling BrC over what has previously been done, this paper is a worthwhile contribution, but the specific results are highly uncertain and speculative.
Reply:
We thank the reviewer for the positive comments. We have added more comparisons with observational data in addition to AERONET, and discuss the uncertainty of our results in the revised manuscript. See our responses to the specific comments below.

1. *In addition to (or maybe instead of) using a model to simply assess BrC climate impacts that really can't be verified at this point, additional discussion could be added on what the authors feel could be done to help assess various model performance and move research of BrC radiative impacts forward. For example, are there places where measurements of BrC would be most beneficial?*
Reply:
We appreciate the recommendation. We added some discussion at the end of the paper regarding measurements that would be beneficial to further development of BrC in the model.

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

injection heights 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."

2. *The authors could show more detailed spatial distributions (including vertical profiles) of BrC and BC (maybe include mineral dust too). TOA forcing is highly sensitive to the vertical distribution of light absorbers, and there is evidence that BrC can be enhanced at higher altitudes relative to BC, how confident are the authors of the vertical distribution of BrC in their model, how does the model consider vertical transport of BrC, what is the effect of this uncertainty on radiative forcing?*
If BrC vertical structure is also important for stability, cloud formation etc, (effects other than direct radiative forcing), what are the limitations with the model in this respect?
Reply:
We added a figure looking at vertical profiles over 6 regions with significant BrC aerosol radiation interaction (REari) from the BRC model run. We also added a discussion regarding uncertainty in model vertical distributions of organic aerosols.
In the model, we assume that primary organic aerosols are light-absorbing, depending on the BC/OA ratio in each grid cell. In the model, primary organic aerosols are predicted by treating different processes affecting primary organic aerosols, e.g., vertical injection in the emission, vertical transport by large-scale winds and turbulence mixing, and dry and wet scavenging, determining the vertical profiles of primary organic aerosols.

Added **Figs. S1, S2**

We made a change on **page 7, line 21** - "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 cloud cover and atmospheric stability 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 global observations in Shrivastava et al. (2014) 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. Vertical profiles of aerosols, cloud fraction, and heating rates in CAM are shown over 6 regions with strong BrC radiative effect due to aerosol radiation interaction (Fig. S2)."

3. *Only spatial distributions of POM are shown, similar results for BrC would be of interest.*
Reply:
In the case of this parameterization, the biomass and biofuel POM tracers represent BrC in the model. We have added a clarification in the caption for Figure 1.

We made a change to the **Fig. 1 Caption** - "The column burdens of POM from (a) biomass burning (BB), (b) biofuel (BF), and (c) fossil fuel emissions. Panels (a) and (b) represent BrC in the BRC, BRC_CNST, and BRC_BL model runs. The units are in mg m^{-2} and the values are a 9 year average from 2003-2011."

4. *Another question that may be of interest is how does the model-predicted lifetime of BrC vary geographically? This was alluded to in the paper, but maybe could be expanded more.*
Reply:
We have added a brief analysis of the BrC bleaching effect in different regions of the globe to the supplementary material. We reference this in section 3.1.
As mentioned above, the biomass and biofuel POM tracers represent BrC in the model. Modeled-predicted lifetime of POM is shown in Liu et al. (2016).

Added **Table S1.**

We made a change on **page 10, line 23** - "Table S1 shows [OH] in different regions and the half-life of BrC due to the bleaching effect in these regions, which ranges from 0.37 days (southeast Asia) to 2.09 days (Arctic)."

5. *In summary, maybe the authors could list what are the major uncertainties in their analysis of BrC radiative impacts and what is needed to address them.*
Reply:
We address the main uncertainties in the model parameterizations, the model analysis, and the uncertainties in the vertical distribution of aerosols (addressed in comment 2).

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.

We made a change on **page 8, line 11** - "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."

Minor comments.

6. *P2, L12: Feng et al did not consider BrC bleaching, so this is likely a large over estimation, which should be noted.*
Reply:
We make it clear later in the paper that the BrC estimations by earlier modeling studies are overestimated. This particular line is referring to the burden of OA compared to BC, and so we left it as is.

7. *Don't really understand the layout of the first 3 equations. Eq 1 should be something like RI = ...*

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. *P9 L18, typo BRC_CL ??*
Reply:
Fixed.

9. *Fig 6 and associated discussion and in the sections that follow; be specific about the brown carbon included in the model, ie, was it BRC, CRC_CNST or BRC_BL?*
Reply:
We have paid better attention to how figures and discussion are worded so that the reader can better understand which model simulation is being used.

10. *The model considers BrC bleaching just due to OH. Is this the only route for bleaching? Please justify. What are the limitations with this assumption?*
Reply:
Due to the fact that the BrC bleaching parameterization was designed to match observed BrC lifetimes, inherent in its timing is the inclusion of photochemical oxidation and other effects contributing to BrC aging. However, this is regionally specific to the location of the observations and we discuss some ways the parameterization could be improved (see comment 5).

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