We thank the reviewer for his/her careful reviews and helpful comments. The manuscript has been revised accordingly and our point-by-point responses are provided below. (Reviewer's comments are in italic and the responses in standard font).

Reviewer #1

This paper examines the global and regional radiative forcings by black carbon and organic carbon aerosols from open fires. The authors use the NCAR Community Atmosphere Model version 5.3 (CAM5) with the four-mode version of the modal aerosol module (MAM4) and employ two methods to calculate forcing. In one method, they follow Ghan et al. (2013), which may produce a more robust estimate of forcing. In the second method, they follow a more traditional approach. The authors find that top-of-atmosphere (TOA) forcing from aerosol-cloud interactions dominates the total global forcing (-0.70 W m^{-2}). When aerosol-radiation interactions and aerosol effects on snow are also considered, the global annual mean forcing from open fire aerosols is -0.55 W m^{-2}. The authors also estimate the climate impacts of fire aerosols.

The paper leads to no startling new conclusions, but may provide a more accurate estimate of the global and regional climate impacts of aerosols from open fires. The paper should be revised in response to the major criticisms and resubmitted.

Reply: We thank the reviewer for helpful comments. The manuscript is revised following the comments and criticisms from the reviewer.

Major criticisms.

1. The paper needs to make more clear what is new in the results, or why this approach represents a substantial improvement over previous results. Central to this paper should be the answer to this question: Why does this research give us greater confidence in our knowledge of the effects of fire aerosols on climate?

In Lines 147-150, the text lists a few improvements, but supplies little elaboration. The improvements are: (a) higher spatial resolution, (b) use of the latest CAM5 model with updated MAM4, (c) calculation of daily instead of monthly fire emissions, and (d) use of an alternative methodology to calculate radiative forcings of aerosols (Ghan 2013). It's not clear why the relatively small increase in spatial resolution would lead to better results, or why calculation of daily instead of monthly fires matters. Almost no information on the updates in MAM4 is given or what difference they make for forcing calculations. A detailed explanation of the benefits of the Ghan (2013) method over other methods is absent.

→Reply: We thank the reviewer for the comments. We now make it more clear what is new in our results, and why our approach represents a substantial improvement over previous results in the revised manuscript.

Specially, following the reviewer's comment, we elaborate more on the improvements of our approach and model configuration in the revised manuscript:

(a) *higher spatial resolution*. A model resolution change from 2 degree (used in previous studies) to 1 degree (in this study) represents a resolution increase by 4 times. A higher resolution allows more efficient transport of aerosols from the sources to remote regions due to reduced wet scavenging of aerosols as a result of less frequent collocation between aerosols and clouds at higher resolutions (Ma et al., 2013; 2014). Model resolution has also been shown to be important for aerosol radiative forcing due to aerosol-cloud interactions (Ma et al., 2015).

(b) *use of the latest CAM5 model with updated MAM4*. Compared to the 3-mode version of MAM (MAM3) used in previous studies, MAM4 includes a primary carbon mode to explicitly treat the microphysical ageing of primary carbonaceous aerosols (POM/BC) in the atmosphere. Primary carbonaceous aerosols are emitted in the primary carbon mode and transferred to the accumulation mode due to aerosol condensation and coagulation. Because of a lack of primary carbon mode, MAM3 assumes that primary carbonaceous aerosols are emitted in the accumulation mode and thus instantaneously mixed with other soluble aerosol species (e.g., sulfate), subject to wet scavenging in the accumulation mode. As a result, MAM4 has higher BC and POM burdens over MAM3 in the remote regions by ~30%.

(c) *calculation of daily instead of monthly fire emissions*. Using daily emissions will allow the model to consider the effect of fast changes in fire emission flux on the local atmospheric conditions. It is expected that using the monthly mean emission flux the model can't consider the effect of the extremely strong fires, thus it might underestimate the fire forcing for such cases. Considering that the aerosol effect is often non-linear, using higher temporal resolution emission data will make a difference, at least for the effect on daily extremes.

(d) *use of an alternative methodology to calculate radiative forcings of aerosols (Ghan 2013).* Ghan (2013) provides a more accurate method to calculate the radiative forcing of aerosols. Central to this method is that the radiative forcing due to aerosol-radiation interactions must be calculated in the presence of clouds (i.e., under all-sky condition, $\Delta(F - F_{clean})$), and the radiative forcing due to aerosol-cloud interactions be calculated under the condition of no aerosol effects on radiation (i.e., $\Delta(F_{clean} - F_{clean,clear})$). F_{clean} is calculated from the diagnostic radiation call with aerosol scattering and absorption neglected, and $F_{clean,clear}$ from the diagnostic radiation call with both aerosol and cloud scattering and absorption neglected. With the radiative forcing decomposition of this method, the impact of aerosols on surface albedo is also quantified (i.e., $\Delta F_{clean,clear}$).

In addition to the above improvements in model configuration and approach of

calculating radiative forcings, we validate the model performance through a comparison of our modeled AOD and SSA with the AERONET data; modeled radiative forcing due to aerosol-radiation interactions compared with satellite-derived estimations, and modeled BC-in-snow concentrations with observations in Northern China and the Arctic. These model improvements and evaluations give us greater confidence in our knowledge of the effects of fire aerosols on climate.

Some notable key findings from this study are highlighted in the conclusion section:

a) Fire aerosol radiative effect due to ARI in the Arctic regions (0.428±0.028 W m^{-2}) is larger than that in the tropical regions (0.172±0.017 W m^{-2}), although the fire aerosol burden is largest in the tropics, which results from the larger amount of low clouds in the Arctic.
b) The large cloud liquid water path over land areas and low solar zenith angle of the Arctic favor the strong fire aerosol radiative effect due to ACI (up to -15 W m^{-2}) during the Arctic summer.
c) The global annual mean surface albedo effect (SAE) of fire aerosols over land areas (0.03±0.10 W m^{-2}) is relatively small and insignificant.
d) The fire aerosols reduce the global mean surface air temperature (T_s) by 0.03± 0.03 K and precipitation by 0.01±0.002 mm day^{-1}. Significant reductions of precipitation in southern Africa and NH high-latitudes are noticed.

2. The paper uses outdated terms to describe radiative forcing by aerosol, and does not adequately describe what adjustments to the model meteorology have been allowed in the forcing calculations. Following IPCC AR5, the authors should use the terms aerosol-radiation interactions (AR1), aerosol-cloud interactions (ACI), and forcings due to surface albedo changes (Boucher et al., 2014; Myhre et al., 2014). ACI in the IPCC framework includes the effects of aerosols on cloud droplet number, cloud lifetime and takes into account the "semi-direct effect" of absorbing aerosols. The ACI category of forcings is useful as it makes it unnecessary to distinguish between the sometimes competing effects of aerosols on clouds.

→Reply: Thank for the suggestion. Following the reviewer's comment, we now use the terminology of the radiative forcings by aerosol from IPCC AR5 in the revised manuscript. In our results, the cloud radiative effect (CRE), i.e., radiative effect due to aerosol-cloud interactions (ACI) includes the effects of aerosols on cloud droplet number and cloud lifetime through acting as CCN, and the semi-direct effect of absorbing aerosols.

All the atmospheric variables (including temperature, precipitation, and circulation) are allowed to adjust. However, with sea surface temperatures (SST) and sea ice are prescribed in the simulations, only the rapid adjustments are taken into account. We have made it clearer in the revised manuscript.

The authors should further state whether they calculated radiative forcings (RF) or effective radiative forcings (ERF), which take into account the rapid adjustments to a

range of meteorological variables. If these are ERFs (and they seem to be), the authors need to make clear what meteorological variables they allowed to adjust. The authors should emphasize in the abstract and conclusions that the forcings they report are relative to the case of no fires, and not to conditions in 1750s.

→Reply: Yes, with the method of Ghan (2013), the effective radiative forcings (ERF) are calculated in this study. All the atmospheric variables (including temperature, precipitation, and circulation) are allowed to adjust. However, with sea surface temperatures (SST) and sea ice are prescribed in the simulations, only the rapid adjustments are taken into account. We also emphasize in the abstract and conclusions that the radiative effects we report are relative to the case of no fires. We now use the term "radiative effect" instead of "radiative forcing" of fire aerosols throughout the text.

3. It's not clear why the paper does not consider the effects of fire aerosols on sea ice albedo. Is this not an important forcing term? Also the authors neglect the issue of brown carbon, which has recently been suggested as a main component of primary organic matter (POM) in fire plumes (Feng et al., 2013). MAM4 may not be capable of simulating brown carbon, and this should be acknowledged.

→Reply: In our simulations with the stand-alone CAM5, sea surface temperatures and sea ice are prescribed, and thus the effects of fire aerosols on sea ice albedo are not considered. The effects of fire aerosols on sea surface temperatures and sea ice albedo will be presented in our future study using a slab ocean model coupled with CAM5.

The effects of POM as brown carbon are not considered in MAM4, and we acknowledge this in the revised manuscript.

4. The authors report a large number of changes in global mean variables without giving uncertainty ranges or stating which changes are statistically significant. Given that many of the variables have been calculated using an ensemble of simulations, uncertainties should be easy to calculate.

→Reply: Following the reviewer's comment, we added the uncertainty ranges ($\pm 1\sigma$ uncertainty) for changes in global mean variables in the revised manuscript.

Other criticisms.

Title: Given the distribution of fires in Figure 2, it looks like the authors include agricultural fires in their analysis, and so the term "wildfire" should be changed to "open fires."

→Reply: Yes, the agricultural fires are included. We changed the term "wildfire" to

"open fires".

Abstract. The abstract should state the time period under investigation. Also large regional forcings should be quantified, as they could have importance for regional climate.

→Reply: We added the time period (2003-2011) in the abstract. Also the following sentence is added in the abstract for large regional forcings: "REs due to fire ARI and ACI in the Arctic (0.43±0.03 and -1.38±0.23 W m^{-2}, respectively) are stronger than those in the tropics (0.17±0.02 and -0.82±0.09 W m^{-2}, respectively), although the fire aerosol burden is higher in the tropics."

Introduction. The introduction is too long. The first paragraph should make clear exactly what problem is being considered, and it should succinctly explain why this investigation represents a major improvement over past research. Throughout the introduction, many old references brought up – e.g., Chuang et al. (2002) or IPCC AR4. The authors should condense the introduction and focus on Chapters 7 and 8 in AR5 and subsequent papers – e.g., Myhre and Samset (2015), Chakrabarty et al. (2014), and many others. Missing from the introduction is a discussion of the radiative effects of organic vs black carbon.

→Reply: Thanks for the suggestions. Following the reviewer's comment, we made it clear in the first paragraph what problem is being considered in this study by adding the sentence: "A qualification of radiative forcing of fire aerosols is the first step to reduce these uncertainties [*Ward et al.,* 2012]".
We added the explanation why this investigation represents a major improvement over past research (see our response to the reviewer's major criticism #1).
We condensed the introduction and focused on Chapters 7 and 8 in AR5 and subsequent papers. We removed the old references, e.g., Chuang et al. (2002) or IPCC AR4 in the revised manuscript.

The following dicussion of BC and POM's radiative effects are added: "Although there are many studies quantifying the RE of fire aerosols, a further investigation is still needed, as current estimations of the RE of fire aerosols from climate models are still associated with large uncertainties [*Myhre and Samset*, 2015; *Chakrabarty et al.*, 2014], and the REs of fire POM versus BC are even less clear."

Line 174. The authors state that MAM4 "significantly increases (and improves) the BC concentrations in the Arctic…." Why does inclusion of the primary carbon mode in MAM4 improve the treatment of microphysical aging of BC? How did the authors decide that inclusion of this mode "significantly" improves the BC simulation? By what measure? Elsewhere the authors state that MAM4 "realistically represents the external/internal mixing of BC" (Line 578). But no detail is given about these improvements.

→Reply: In the 3-mode version of MAM (MAM3), due to a lack of primary carbon mode, BC is emitted directly into the accumulation mode, and thus is instantaneously mixed with other soluble aerosol species (e.g., sulfate), subject to wet removal by clouds and precipitation. MAM4 includes an additional primary carbon mode on top of MAM3. BC is emitted in this primary carbon mode, and is gradually transferred to the accumulation mode due to the microphysical aging (condensation and coagulation). Aerosol in the primary carbon mode is less hygroscopic than that in the accumulation mode, and thus is less susceptible to the wet scavenging by clouds. Therefore, BC concentration from MAM4 is increased, especially in the Arctic, which improves the agreement with observations. The details of MAM4 and comparison with MAM3 are given in Liu et al. (2016). Please see also our reply to the major criticism #1 for the description of BC representation in MAM4 versus in MAM3.

We added the following details in the introduction of the revised manuscript: "MAM4 includes an additional primary carbon mode on the top of MAM3 to explicitly treat the microphysical ageing of primary carbonaceous aerosols (POM and BC) in the atmosphere. POM and BC in MAM4 are emitted in the primary carbon mode instead of the accumulation mode as in MAM3. Thus MAM4 increases the BC and POM concentrations over MAM3 due to reduced wet scavenging of POM and BC in the primary carbon mode with a lower hygroscopicity than that in the accumulation mode."

Section 2.3. See major criticism #2 above. Please rewrite using IPCC AR5 convention for describing forcings.

→Reply: Done. See our reply above to the major criticism #2.

Results. The results section rambles. The authors should decide which are the key results and provide more detailed explanations of the mechanisms driving these results. Also, the statistical significance of results should be given, where possible. Since the authors performed an ensemble of simulations, many results can be reported with one standard deviation uncertainty. For example, what is the uncertainty of the forcings calculated following Ghan 2013? Is the -0.03°C temperature effect of fire aerosols statistically significant?

→Reply: Thanks for the suggestions. We revised the results section and emphasized the key results. Please see our response above to the major criticism #1 for the key results. We have provided more detailed explanations of the mechanisms driving these results.

Following the reviewer's comment, we added the statistical significance of results with one standard deviation uncertainty. This is done for the uncertainty of the forcing calculated following Ghan (2013) as well as the temperature and precipitation

changes due to fire aerosols.

Finally, the forcings calculated for specific regions should be compared to recently published estimates – e.g. Brieder et al. (2014) for the Arctic and Sena and Artaxo (2015) for South America.

→Reply: Thanks for the suggestion. We tried to compare our forcings with those estimated from Brieder et al. (2014) for the Arctic. However, we found that this study reported the distribution, aerosol optical depth, and absorption of Arctic aerosol components and source contributions calculated using the GEOS-Chem model, and did not present the forcing estimates.

Following the reviewer's comment, we added the following comparison of our forcing estimates with those from Sena and Artaxo (2015) for South America in the revised manuscript: "The fire aerosol RE due to ARI over South America for the period of 2000 to 2009 is estimated with the TOA shortwave flux from CERES (Clouds and Earth's Radiant Energy System) and AOD from MODIS by Sena and Artaxo (2015). The clear-sky RE during the fire season (August to September) is estimated to be -5.2 W m^{-2}, which is larger than our result (-2.1 W m^{-2}). This is consistent with the underestimation of our modeled AOD in South America when compared to the AERONET data (Figure 3)."

Line 241. Here and elsewhere. It is not clear whether the fires examined in this study include agricultural fires such as those in Equatorial Asia and South America.

→Reply: Yes, the agricultural fire is included. We made it clear in the revised manuscript.

Lines 276-on. The text should state whether the modeled AOD includes aerosol from all sources, not just fires.

→Reply: The modeled AOD includes aerosol from all sources. We made it clear in the revised manuscript.

Line 311. The text states, "Although MAM4 increases the column burdens of POM and BC by up to 40% in many remote regions compared to MAM3…." Why does this large increase occur?

→Reply: see our response above for the explanation of MAM4 and MAM3 simulated BC differences.

Line 338. Text should be more clear about how clouds amplify the forcing of BC.

→Reply: We added the following explanation in the revised manuscript: "When BC resides above clouds, its absorption of solar radiation is significantly enhanced due to the reflection of solar radiation by clouds [*Abel et al.*, 2005; *Zhang et al.*, 2015]".

Line 343. Why is the forcing estimated from Terra different from that of Aqua?

→Reply: First of all, we notice that we had a wrong subtitle in Figure 7b and Figure 7c. Figure 7b should be for Aqua/MODIS, and Figure 7c should be for Terra/MODIS. The figure caption is accurate in the text.

Over southeastern Atlantic, smoke aerosols usually reside above the stratocumulus clouds. Therefore, the direct radiative forcing strongly depends on the underlying cloud fraction. If the cloud fraction is higher, for the same amount of smoke aerosols at exact the same altitude, smoke aerosols can exert stronger direct radiative forcing. Since stratocumulus clouds over this region exist the diurnal cycle, the forcing estimated from Terra (morning time, with larger amount of clouds) is different from the one estimated from Aqua (afternoon time, with smaller amount of cloud). For more detail, we recommend the reviewer to check Figure 3 in the reference:
Min M., and Zhang Z. (2014), On the influence of cloud fraction diurnal cycle and sub-grid cloud optical thickness variability on all-sky direct aerosol radiative forcing, J. Quant. Spectros. Radiat. Transfer, doi:10.1016/j.jqsrt.2014.03.014.

Line 346. There is no mention here or elsewhere about the effect of solar zenith angle on radiative forcing at high latitudes, particularly the Arctic.

→Reply: We agree with the reviewer that the cloud radiative forcing due to fire aerosols at high latitudes can be affected by the solar zenith angle (Shupe et al., 2004). In the boreal summer, the lower solar zenith angle favors the larger DRE in the Arctic. We added this effect in the revised manuscript.

Line 349. Here and elsewhere, the authors should take care with the terms "summer" and "autumn" when referring to the Southern Hemisphere.

→Reply: Thanks. We made it clearer in the revised manuscript. All terms were changed to "boreal summer" or "boreal autumn".

Line 354. "noises" Please fix English.
→Reply: Thanks. We changed to "…, and there is much less noise".

Line 364. The text states: It is not clear why removal of POM in the simulation affects BC concentrations. If indeed this is what happens, then the Ghan method for calculating forcing should not be used for individual fire components.

→Reply: Because fire POM and fire BC are co-emitted and assumed to be internally

mixed. The burden of fire POM is about a few times larger than that of fire BC, especially in Arctic. With the removal of fire POM emission and thus fire POM in the NOFIREPOM experiment, fire BC will be impacted due to changed properties (e.g., size) of aerosol particles within which co-emitted fire BC is internally mixed with fire POM. Our results show that the fire BC burden in the Arctic is reduced in NOFIREPOM with the exact mechanism warranty of a detailed budget analysis. We added an explanation in the revised manuscript.
We would like to keep the Ghan method for calculating the radiative effects of individual fire components (POM and BC). The reason is that the Ghan method only introduces the relatively large bias for fire POM radiative effect (due to aerosol-radiation interactions), and the bias for fire BC radiative effect is small (comparing the Ghan and the BBFFBF methods). By using the two different methods we will be able to examine the uncertainty range of radiative effects of individual fire components. Also the Ghan method allows us to calculate the radiative effects of individual fire components due to aerosol-cloud interactions.

Line 379. See above comment.
Cloud radiative forcing section. Please see major criticism #2. Also, this section should provide discussion of why the forcing due to ACI is stronger in some regions compared to others.

→Reply: Please see our responses to the major criticism #2 above.

We added the following discussion of why the forcing due to ACI is stronger in some regions compared to others in the revision: "The different spatial distributions of fire aerosol radiative effect (RE) due to ACI in the NH high latitudes and in the tropics result from the difference in cloud distributions between the two regions. During the fire season the cloud LWP over the land areas in the NH high latitudes is three times larger than that over the ocean areas in the tropics. Larger cloud LWP favors the stronger RE due to ACI, because the larger LWP associated with the warm cloud and rain processes favors the aerosol effect on slowing down the autoconversion of cloud water to rain [*Ghan et al.*, 2012; *Jiang et al.*, 2015]. Meanwhile, in the Arctic, the low solar zenith angle in summer favors the large fire aerosol RE due to ACI."

Line 411. The text should state why larger cloud liquid water path leads to stronger forcing due to ACI.

→Reply: We added the following explanation: "Larger cloud LWP favors the stronger RE due to ACI, because the larger LWP associated with the warm cloud and rain processes favors the aerosol indirect effect via slowing down the autoconversion of cloud water to rain [*Ghan et al.*, 2012; *Jiang et al.*, 2015]."

Section on surface snow albedo forcing. Why are forcings due to BC deposition on sea ice not considered? The section seems misnamed, since forcings on all light colored

surfaces are seen in Figure 12.

→Reply: In our simulation, the sea surface temperature and sea ice is prescribed, and thus the radiative effect due to fire BC deposition on sea ice is not estimated.

We rename the title of the section to "Surface albedo effect". The surface albedo change not only results from the radiative effect of fire BC deposition on snow albedo, but also from atmospheric feedbacks (e.g., snow depth change and snow melting) due to fire aerosols.

The forcings on surface albedo calculated with the Ghan 2013 method look suspiciously high over low latitudes (Figure 12). The authors should comment on these high values – e.g., +0.5 Wm^{-2} over parts of the U.S. south. Are these results comparable to those from SNICAR?

→Reply: The SAE of fire aerosols is also noticed over low latitudes, which includes the surface albedo changes from atmospheric feedbacks (e.g., snow depth change and snow melting) [*Ghan,* 2013]. These high values over low latitudes are not evident in those from SNICAR, which are diagnosed in the standard model simulation and don't include atmospheric feedbacks. We added a comment on these high values in the revised manuscript.

Figure 12b reveals no significant differences in forcings for the fire vs no-fire cases over the Arctic or north China. The authors should acknowledge this. Given the results from SNICAR, it seems that the only region that might show a significant impact of fire aerosols on surface albedo is Greenland and the very northern reaches of Canada.

→Reply: The annual mean fire BC forcing in the Arctic and North China (~ 0.01 W m^{-2}) is much smaller than that in Greenland and the very northern reaches of Canada. It is because the snow-covered time of Arctic and North China is shorter. The forcing in these two regions (Greenland and the very northern reaches) can reach up to 0.5 W m^{-2}. We acknowledged this in the revised manuscript.

Line 458. It sounds like snow melting is one of the rapid meteorological adjustments allowed to occur in the forcing calculation. Is this correct?

→Reply: Yes, the snow melting is allowed when calculating the surface albedo effect of fire aerosols.

Section on the fire aerosol effects on shortwave radiation, global temperature and precipitation. Here the statistically significance and the uncertainties of global results should be stated. If the global mean changes of some variables are not statistically significant, then that should be made clear.

→Reply: We added the significant information (e.g., one-standard deviations) in the text and in Table 2. The global mean changes not statistically significant are acknowledged in the revised manuscript.

Discussion section. Again the authors should stress the key points and put them in context of other new studies besides just Ward 2012 and Tosca 2013. What exactly is new in this study? Limitations and uncertainties of the study should be discussed – i.e., what are the shortcomings of the approach used here?

→Reply: We have included a discussion of the key points of this study as summarized as follows:

a) Fire aerosol RE due to ARI in the Arctic regions (0.43 ± 0.028 W m^{-2}) is larger than that in the tropical regions (0.17 ± 0.017 W m^{-2}), although the fire aerosol burden is higher in the tropics. This results from the larger low cloud amount in the Arctic;
b) The large cloud liquid water path over land areas, and low solar zenith angle of the Arctic favor the strong fire aerosol RE due to ACI (up to -15 W m^{-2}) during the Arctic summer;
c) The global annual mean surface albedo effect (SAE) over land areas (0.03 ± 0.10 W m^{-2}) is relatively small and insignificant;
d) The fire aerosols reduce the global mean surface air temperature (T_s) by 0.03 ± 0.03 K and precipitation by 0.01 ± 0.002 mm day^{-1}. Especially, significant reductions of precipitation in southern Africa and in the NH high-latitudes are noticed.

Following the reviewer's comment, we added a discussion of limitations and uncertainties of this study:

1) The RE estimate of co-emitted fire POM with the Ghan (2013) approach is not accurate due to the assumption of internal mixing of individual fire components (POM and BC);
2) There is large noise associated with the surface albedo effects of fire aerosols with the Ghan (2013) approach due to the snow melting and atmospheric feedbacks;
3) There are uncertainties with the model simulation and configuration. For example, the model still underestimates observed AODs (mostly within a factor of 2) at the sites predominantly influenced by biomass burning aerosols during the fire season. It implies that the fire aerosol radiative effects can be stronger than those estimated in this study. In our simulation, the sea surface temperature and sea ice is prescribed, and the fire BC effects on sea ice is not considered. The brown carbon component of POM [Feng et al., 2013] is not considered in our current simulations, which may result in an underestimation of atmospheric absorption of fire aerosols."

Tables and Figures.

There are too many Figures. Decide what is important and put rest in a supplement.
→Reply: We moved the original Figure 2 (POM and BC burdens from different sources) and Figure 7 (fire aerosol radiative effect due to ARI at four seasons) to the supplement. We now have 12 figures.

Captions should be stand-alone so that the browsing reader can understand what is being shown. Unusual acronyms should be explained.
→Reply: We added the standing-alone captions of all figure and tables at the end of the manuscript. We removed some unusual acronyms and added explanations for the others in the revised manuscript.

Units in Table 2 should be within the table, not in the caption.
→Reply: Done.

Uncertainty ranges should be included in Table 2, and significant changes shown in boldface.
→Reply: We revised Table 2 to include the uncertainty ranges and those significant changes are shown in boldface.

Text on all legends should be large enough to read. The latitude and longitude labels on the global maps can be eliminated for a cleaner, less cluttered appearance.
→Reply: We enlarged the text on legends of the figures. The duplicated latitude and longitude labels on the global maps were eliminated.

Global mean values should be reported to 2-3 significant digits.
→Reply: The global mean values were now reported to 3 significant digits.

Figures 4 and 5 should include error bars.
→Reply: Done.

Figure 7. What does white space represent?
→Reply: White space represents the missing values. As we mentioned in the figure caption, the radiative effect is estimated for above-cloud aerosols only. During the fire season, cloud fractions over the land, especially below 10°S, are extremely low, and close to 0. No above-cloud smoke aerosols were detected by satellites over these regions; therefore, no radiative effect due to above-cloud aerosols is estimated.

Figure 14. Replace acronyms above the panels with standard English terms.
→Reply: Done.

We thank the reviewer for his/her careful reviews and helpful comments. The manuscript has been revised accordingly and our point-by-point responses are provided below. (Reviewer's comments are in italic and the responses in standard font).

Reviewer #2

General comments

My only somewhat major comment is that the climate responses explored (i.e. temperature and precipitation) are based on atmosphere-only experiments, and therefore are somewhat incomplete. That does not mean that it is not worth showing the results, but it should be clearly stated that these results come from fixed-SST simulations, and therefore more work will be needed in the future in a coupled framework to understand the role of fire aerosols on climate in a more complete fashion. Adding a few sentences in the abstract, the corresponding section and in the conclusions would be sufficient for clarifying this better

Reply: Following the reviewer's comment, we have added the statement in the abstract, the corresponding section and in the conclusions that these results (i.e., climate responses) are based on atmosphere-only experiments with fixed-SSTs, and more work using a coupled atmosphere-ocean model will be needed in the future to understand the role of fire aerosols on climate in a more complete fashion.

Specific Comments:

Page 2, Line 32: Please remove "the".
Reply: Done.

Page 2, Line 38: Not sure whey a range is indicated both by two numbers and by the +/-
Reply: We changed to "-0.05 W m^{-2} and 0.04±0.01 W m^{-2}, respectively based on two calculation methods". The first number does not have an uncertainty range, since it is derived from a clean calculation.

Page 2, Line 39: South Africa -> southern Africa (here and elsewhere in the text).
Reply: Done here and elsewhere in the text.

Page 2, Line 48: Suggest stressing that this effect is small and insignificant.
Reply: Done. We added this information to the abstract.

Page 2, Lines 45-47: Need to clearly mention here that this is inferred from atmosphere-only simulations (and not from full coupled climate simulations).
Reply: We added this information in the abstract.

Page 3, Lines 55-56: Worth citing the review paper by Voulgarakis and Field (2015) here, as it is very relevant.
Reply: Done.

Page 3, Lines 59-60: Worth citing the paper of Bistinas et al. (2014) here.
Reply: Done.

Page 3, Lines 61-63: This reads as if this manuscript will fill the gap of knowledge of how fires will change in the future, which is not the case. Please rephrase to something that aligns better with the focus of the manuscript (or you could remove the second part of the sentence entirely).
Reply: Following the reviewer' comment, we removed the second part of the sentence.

Page 3, Line 72: Suggest changing "indirect effect" to "indirect effects"
Reply: Done.

Page 4, Line 76: Suggest removing "the" before "climate change".
Reply: Done.

Page 4, Lines 76-78: Well, it depends. RE is not always for both anthropogenic and natural. Sometimes we just study anthropogenic or natural RE individually. I suggest rephrasing to "RE represents the instantaneous radiative impact of atmospheric particles on the Earth's energy balance"

Reply: Thanks. We revised the sentence as the reviewer suggests.

Page 4, Lines 78-81: Similarly, RF does not have to always be pre-industrial to presentday. I suggest rephrasing to ". . .as the change of RE between two different periods, e.g. the pre-industrial and the present-day. . .", and then change the second half of the sentence accordingly.

Reply: Thanks. We have revised the sentence accordingly.

Page 5, Line 98: many -> some
Reply: Done.

Page 7, Lines 147-148: It is mentioned that two methods are presented – worth briefly mentioning them here.

Reply: Thanks for the suggestion. We added the following sentence to briefly mention the two methods: "One method estimates the DRE with different model simulations

[*Ghan*, 2013], and the other one calculates the DRE directly by multiple diagnostic radiation calls in a single simulation."

Section 2.1: Is the aerosol interactive with the model's chemistry?

Reply: The secondary aerosol, e.g., sulfate is produced from the model's gas and aqueous sulfur chemistry. The version of the model we are using in this study does not include a full-chemistry mechanism, and the oxidants (e.g., OH, HO_2 and O_3) are prescribed [*Liu et al.*, 2012].

Page 8, Lines 179-180: Any other performance features apart from the Arctic? What about over key biomass burning regions, and what about OC?

Reply: Compared to MAM3, MAM4 increases the concentrations of BC and POM in most global regions. The increase is the strongest over the remote regions (e.g., oceans and Arctic) and relatively small over the land source regions [*Liu et al.*, 2016]. We modified the sentence to include more discussion of performance features.

Page 9, Lines 194: Suggest changing "climate" to "atmospheric" or "short-term climate" as the SSTs/sea ice are prescribed.

Reply: Following the reviewer's comment, we changed "climate" to "atmospheric".

Page 10, Lines 227-228: I may be missing something here, but how can the difference between F in two simulations that do not involve any aerosols ("clean" and "clean, clear") tell you something about the aerosol-induced cloud radiative effect (CRE)?

Reply: Typically, the aerosol-induced CRE is estimated by the difference of the shortwave cloud forcings (Δ SWCF, or Δ (F- F_{clear})) between two simulations. With this method, however, the absorbing aerosols above clouds will produce a positive direct forcing and induce a bias in estimated CRE (Ghan, 2013). Ghan (2013) indicates that CRE be calculated under the clean conditions (i.e., no aerosol *direct* effects). The clean conditions are not meant to have no aerosols in the control simulation, but to have no aerosols in the diagnostic radiation call in the same control simulation. So the SWCF in clean conditions (Δ $SWCF_{clean}$, or Δ ($F_{clean} - F_{clean,clear}$)) is used to estimate the CRE.

As we define in the text, "F_{clean} is the radiative flux at TOA calculated from a *diagnostic radiation call* in the same control simulations, but neglecting the scattering and absorption of solar radiation by aerosols."

Page 11, Line 238: Could add "each time" before "neglecting the. . .".
Reply: Done.

Page 11, Lines 239: Suggest adding "more direct" between "This" and "method".
Reply: Done.

Page 12, Line 257: topics -> tropics
Reply: Done. Thanks.

Page 13, Line 284: activities -> activity
Reply: Done.

Page 13, Lines 286-288: If scaling is not applied here, mention it clearly (e.g. ". . .whereas here we do not apply any such scaling").

Reply: Done. We added the following words: ", whereas here we do not apply any such scaling."

Page 13, Line 294: trend -> seasonal cycle
Reply: Done.

Figure 3: Do the selected AERONET sites have data for exactly the same years as the simulation? Not entirely necessary, but needs to be mentioned. Also: Worth mentioning in the caption (also in Fig. 4) that the first row shows sites in southern Africa, the second row sites in South America, and the third row sites in the Arctic.

Reply: We downloaded the AERONET data at the selected sites for exactly the same years as the simulation. However, the selected AERONET sites have missing data for some periods of the model simulation, as shown in Figures 3 and 4. We mentioned this in the revision.
We now added the site information to the figure caption.

Page 14, Lines 303-304: There is also a notable early peak. Worth mentioning and perhaps commenting on.

Reply: Yes, the modeled AOD shows a notable early peak before the fire season, especially for Alta Floresta and Rio Branco, which could be due to the model overestimation of fire emission in this period. We mentioned this in the revision.

Page 14, Line 306: However, there is too strong a seasonality, it seems? Any explanation?

Reply: Yes, the modeled SSA is too low during the fire season and exhibits too strong a seasonality. It implies that the model underestimation of scattering aerosols (e.g., POM) may be more severe than that of BC during the fire season.

Page 15, Line 321: Sulfate and OC, right?

Reply: Yes. We changed to "e.g., sulfate and POM".

Page 15, Line 327: No need for "respectively" here.
Reply: Thanks. We removed "respectively" here.

Figure 5: There are too many significant figures in the global mean values shown on each panel (also in later figures). Also: With respect to what is statistical significance estimated for the right panels? Interannual variability or ensemble member diversity? Needs to be mentioned here and also in later figures. And why is significance not shown for the left hand panels?

Reply: Following the reviewer's comment, we reduced the number of figures in the global mean values shown on each panel (also in later figures) in the revised manuscript. The statistical significance test is applied to the results using the Ghan (2013) method, because DRE from this method is calculated as the radiative flux difference between two model simulations. Therefore, the difference is not only from the DRE of fire aerosols, but also from the model internal variability which includes both the interannual variability (2003-2011) and the ensemble member diversity (10 members). We mentioned this in the revised manuscript.

The statistical significance test is not applied to the BBFFBF method (shown in the left hand panels). The reason is that DRE using this method is calculated as the radiative flux difference between the control run and diagnostic radiation calls in each model time step, which ensures that the climate background (e.g., clouds) is exactly the same between the control run and diagnostic calls.

Page 15, Line 332: Why have you chosen to report only the global mean from the BBFFBF method in the text, and not from the one based on Ghan (2013)?

Reply: Actually here the global mean (0.155±0.01 W m^{-2}) is from the Ghan (2013) method. With the Ghan (2013) method, the radiative effects including DRE, CRE and SAE of fire aerosols can be estimated, while the BBFFBF method only estimates DRE. We added a note in the revised manuscript that the two methods give very similar results for DRE of all fire aerosols, and thus we will report the DRE of all fire aerosols with the Ghan [2013] method.

Page 15, Line 336: "The" is not needed.
Reply: removed.

Page 16, Line 340: "of the tropical regions" -> "of the SH tropical regions"
Reply: Done.

Figure 6: Is the model panel (a) produced with all-sky values? In fact, was that the case for Figure 5 too?
Reply: It is DRE in the all-sky condition. This is also the case for Figure 5.

Figure 7: Which method was used for those maps to be made?
Reply: It is from the method of Ghan (2013). After a comparison with method BBFFBF, the DRE due to all fire aerosols estimated with Ghan (2013) is used in the rest of the paper. We added a note in the revised manuscript.

Page 16, Line 354: Define "high latitudes" here. Is it the same definition as the Arctic?
Reply: We changed the "high latitudes" to "Arctic regions".

Page 16, Lines 359-360: "there are much less noises from" -> "there is much less noise with"
Reply: Done.

Page 17, Lines 371-373: Why would it affect BC? Not clear. Explain better.

Reply: Because fire POM and fire BC are co-emitted and assumed to be internally mixed. The burden of fire POM is about a few times higher than that of fire BC, especially in Arctic. With the removal of fire POM emission and thus fire POM in the NOFIREPOM experiment, fire BC will be impacted due to changed properties (e.g., size and hygroscopicity) of aerosol particles within which fire BC and POM are internally mixed. Our results show that the fire BC burden in the Arctic is reduced in NOFIREPOM with the mechanism worthy a detailed budget analysis. We added an explanation in the revised manuscript.

Page 17, Line 373: it -> one
Reply: Done.

Page 17, Lines 375-376: global regions -> globe
Reply: Done.

Page 17, Lines 378-382: Could the authors provide a reference for this mechanism?

Reply: We added the following reference:
Zhang, Z., Meyer, K., Yu, H., Platnick, S., Colarco, P., Liu, Z., and Oreopoulos, L.: Shortwave direct radiative effects of above-cloud aerosols over global oceans derived from 8 years of CALIOP and MODIS observations, Atmos. Chem. Phys., 16, 2877-2900, 10.5194/acp-16-2877-2016, 2016.

Sect. 3.3: Can's some of the cloud changes that lead to indirect effects be a result of dynamical changes due to fire aerosols?

Reply: Yes, the cloud changes as a result of dynamical changes due to fire aerosols is also considered as a part of aerosol induced cloud radiative effect (CRE) with the

Ghan (2013) method. Since the same sea surface temperatures (SSTs) are used in these simulations, CRE as a result of dynamical changes due to fire aerosols should be small.

Page 19, Line 418: Please provide reference to support this statement ("Larger. . .").

Reply: We added the two following references:

Ghan, S. J., Liu, X., Easter, R. C., Zaveri, R., Rasch, P. J., Yoon, J.-H., and Eaton, B.: Toward a Minimal Representation of Aerosols in Climate Models: Comparative Decomposition of Aerosol Direct, Semidirect, and Indirect Radiative Forcing, Journal of Climate, 25, 6461-6476, doi:10.1175/JCLI-D-11-00650.1, 2012.

Jiang, Y., Yang, X.-Q., and Liu, X.: Seasonality in anthropogenic aerosol effects on East Asian climate simulated with CAM5, Journal of Geophysical Research: Atmospheres, 120, 2015JD023451, 10.1002/2015JD023451, 2015.

Page 19, Lines 420-421: What does "low-level" mean here?

Reply: The low-level clouds mean "vertically-integrated low clouds (from surface to 750 hPa)" as defined in CESM. We revised the sentence in the manuscript to make it clear.

Page 20, Lines 434-435: The higher OC/BC ratio does not seem like a good explanation, as it is mentioned a bit earlier that POM and BC are comparable in the NH and SH.

Reply: we agree with the reviewer, and removed "higher fire OC/BC ratios" in the revised manuscript.

Page 21, Line 449: I suggest adding "slightly" between "agree" and "better".

Reply: Done.

Page 21, Line 452: It reads as if you take values from Ghan (2013). Suggest rephrasing.

Reply: Thanks. We rephrased the words to "estimated with *Ghan* [2013]" in the revised manuscript.

Page 21, Lines 469-470: Even in tropical areas? Please discuss.

Reply: We re-wrote the sentence as:

"The negative SAE over land is a result of the surface albedo change (including snow depth change) caused by fire aerosols."

Page 22, Line 484: Instead of "The shortwave flux change in the atmosphere ", I suggest writing "The shortwave atmospheric absorption change", as it is more conventional.

Reply: Done. Thanks for the suggestions.

Figure 13a: Clarify to the reader why the values in Fig. 8a are somewhat different to those in Fig. 13a.

Reply: Figure 13a shows the net shortwave flux change at TOA due to fire aerosols, which is a sum of fire aerosol DRE, CRE and SAE. The CRE (-0.70±0.05) is larger than the DRE (0.155±0.01) and SAE (0.03±0.10). Thus, the TOA solar flux change is dominant by the CRE and similar to distribution of the CRE (Figure 8a). These values are also listed and compared in Table 2.

Page 23, Lines 505-506: There are also substantial differences with Tosca et al. (2013), especially over tropical oceans, therefore I would add "partly" before "consistent". Also the results over southern Africa are consistent with the recent findings of Hodnebrog et al. (2016), which the authors can mention.

Reply: Thanks for the suggestions. We added the word "partly" and also the sentence that "The precipitation reduction in southern Africa is consistent with the recent findings of *Hodnebrog et al.* [2016]" in the revised manuscript.

Page 23, Line 511: After this line, I suggest that you add a statement clearly stating that these results do not represent the complete impact of fire emitted aerosols on temperature and (especially) precipitation, since the climate system has not been allowed to fully respond (SSTs are fixed).

Reply: Thanks for the suggestion. We added a statement in the revised manuscript: "We note that the temperature and (especially) precipitation changes reported here do not represent the complete impact of fire aerosols, since the SSTs are fixed in our simulations. Fully-coupled atmosphere and ocean model will be used to further investigate the impact of fire aerosols."

Page 24, Line 519: effect -> effective
Reply: Done.

Page 26, Lines 575-579: Again, I suggest reminding the reader that these do not represent the full climate responses, given the atmosphere-only nature of the experiments.
Reply: following the reviewer's comment, we added a statement here in the revised manuscript "These results are based on the simulations with fixed SSTs and may not represent the full climate responses."

Page 27, Lines 596-597: Is the difference in emitted POM between the two studies equivalent (in size) to the difference in the CRE?

Reply: The CRE is strongest over southern Africa, South America and the Arctic. The emission scaling factors used in *Ward et al.* [2012] for these three regions are 3, 2 and 3, respectively. The CRE of their study is about 2.4 times of our study (-1.64 versus -0.70 W m^{-2}). So the difference in CRE between the two studies is approximately equivalent (in size) to the emission difference.

Impacts of Global Open Fire Aerosols on Direct Radiative, Cloud and Surface-Albedo Effects Simulated with CAM5

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Abstract

Aerosols from open-land fires could significantly perturb the global radiation balance and induce the climate change. In this study, Community Atmospheric Model version 5 (CAM5) with prescribed daily fire aerosol emissions is used to investigate the spatial and seasonal characteristics of radiative effects (REs, relative to the case of no fires) of open fire aerosols including black carbon (BC) and particulate organic matter (POM) from 2003 to 2011. The global annual mean RE due to aerosol-radiation interactions (REari) of all fire aerosols is 0.16±0.01 W m^{-2} (1σ uncertainty), mainly due to the absorption of fire BC (0.25±0.01 W m^{-2}), while fire POM induces a small overall effect (-0.05 W m^{-2} and 0.04±0.01 W m^{-2}, respectively based on two methods). Strong positive REari is found in the Arctic and in the oceanic regions west of southern Africa and South America as a result of amplified absorption of fire BC above low-level clouds, in general agreement with satellite observations. The global annual mean RE due to aerosol-cloud interactions (REaci) of all fire aerosols is -0.70±0.05 W m^{-2}, resulting mainly from the fire POM effect (-0.59±0.03 W m^{-2}). REari (0.43±0.03 W m^{-2}) and REaci (-1.38±0.23 W m^{-2}) in the Arctic are stronger than those in the tropics (0.17±0.02 and -0.82±0.09 W m^{-2}, respectively for REari and REaci), although the fire aerosol burden is higher in the tropics. The large cloud liquid water path over land areas and low solar zenith angle of the Arctic favor the strong fire aerosol REaci (up to -15 W m^{-2}) during the Arctic summer. Significant surface cooling, precipitation reduction and low-level cloud amount increase are also found in the Arctic summer as a result of the fire aerosol REaci based on the atmosphere-only simulations. The global annual mean RE due to

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surface albedo changes (REsac) over land areas (0.03 ± 0.10 W m^{-2}) is small and statistically insignificant, and is mainly due to the fire BC-in-snow effect (0.02 W m^{-2}) with the maximum albedo effect occurring in spring (0.12 W m^{-2}) when snow starts to melt.

1. Introduction

Open fires or biomass burning of living and dead vegetation are an integral component of the Earth system, and have significant impacts on the carbon cycle [*Ciais et al.*, 2013] and the climate [*Bowman et al.*, 2009; *Keywood et al.*, 2011; *Liu et al.*, 2014; *Sommers et al.*, 2014; *Voulgarakis and Field*, 2015]. On one hand, open fires can perturb the climate system by emitting greenhouse gases and aerosols [*Kaiser et al.*, 2012; *Wiedinmyer et al.*, 2011]. On the other hand, climate states and variabilities can play a critical role in determining the occurrence frequency and intensity of open fires [*Marlon et al.*, 2009; *van der Werf et al.*, 2008; *Westerling et al.*, 2006; *Bistinas et al.*, 2014]. However, there are still large unknowns regarding the feedback mechanisms between open fire and climate interactions [*Carslaw et al.*, 2010; *Liu et al.*, 2014]. A qualification of radiative forcing of fire aerosols as conducted in this study is the first step to reduce these uncertainties.

Particles emitted from open fires can exert significant perturbations to the climate system by scattering and absorbing the solar radiation in the atmosphere (i.e., direct effect) [*Carslaw et al.*, 2010] and by changing the surface albedo when they are deposited on the snow and ice (i.e., surface albedo effect) [*Flanner et al.*, 2007; *Quinn et al.*, 2008; *Randerson et al.*, 2006; *Qian et al.*, 2011, 2015]. In addition, open fire or

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smoke particles can modify the cloud properties, precipitation efficiency, and the hydrological cycle by changing the atmospheric thermal structure (i.e., semi-direct effect) [*Koch and Del Genio*, 2010; *Andreae et al.*, 2004b] or acting as cloud condensation nuclei (CCN) (i.e., indirect effects) [*Andreae and Rosenfeld*, 2008; *Qian et al.*, 2009; *Lu and Sokolik*, 2013].

The radiative effect (RE) [*Boucher and Tanre*, 2000] and radiative forcing (RF) [*Forster et al.*, 2007; *Myhre et al.*, 2013a] are typical metrics used to assess and compare anthropogenic and natural drivers of climate change. The aerosol RE represents the instantaneous radiative impact of atmospheric particles on the Earth's energy balance [*Heald et al.*, 2014]. RF is calculated as the change of RE between two different periods, e.g., the pre-industrial and the present-day times [*Heald et al.*, 2014; *Liu et al.*, 2007], based on the aerosol and precursor gas emissions in the two periods [*Dentener et al.*, 2006; *Lamarque et al.*, 2010].

RF due to aerosol and radiation interactions (RFari) of biomass burning aerosols has been estimated since the IPCC second Assessment Report (AR2). Based on the Aerosol Comparisons between Observations and Models (AeroCom) Phase II simulations [*Bond et al.*, 2013; *Myhre et al.*, 2013b], RFari of biomass burning aerosols in the IPCC Fifth Assessment Report (AR5) is estimated to be 0.0 W m^2 (ranging from -0.20 to 0.20 W m^{-2}), and RFari of biomass burning black carbon (BC) and primary organic matter (POM) are of the opposite sign (i.e., 0.10 and -0.10 W m^{-2}, respectively).

There are also some studies that estimated the RE due to aerosol and radiation

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interactions (REari) of fire aerosols by comparing the simulation with fire emissions against the simulation with no fire emissions. For example, using the NCAR Community Atmosphere Model version 4 (CAM4) with a bulk aerosol module, *Tosca et al.* [2013] reported that the top-of-atmosphere (TOA) REari from global biomass burning aerosols is 0.18±0.10 W m^{-2} averaged for the period of 1997-2009. *Ward et al.* [2012] estimated the REari from biomass burning aerosols in the pre-industrial (for the year 1850), present-day (for the year 2000), and future time periods (for the year 2100), and found that the biomass burning aerosol REari for the year 2000 is 0.13 W m^{-2} and -0.27 W m^{-2} in all-sky and clear-sky conditions, respectively.

RE due to aerosol and cloud interactions (REaci) of biomass burning aerosols can be comparable in magnitude to or even stronger than the REari [*Liu et al.*, 2014]. With a global aerosol-climate model, the REaci of biomass burning aerosols was estimated to range from -1.74 to -1.00 W m^{-2} for the year 2000 in *Ward et al.* [2012]. The semi-direct radiative effect of biomass burning aerosols is not independently assessed in IPCC reports. The magnitude was reported to be about 7.0 W m^{-2} in the Southern American biomass burning regions by examining the radiative flux difference with and without the biomass burning aerosol effect on clouds [*Liu*, 2005].

The RF or RE due to surface albedo changes (RFsac or REsac) of BC from open fires and other sources has been estimated in previous studies. For biomass burning emissions with a strong (1998) and weak (2001) boreal fire year, RE of fire BC-in-snow was estimated to be 0.011 and 0.006 W m^{-2}, respectively [*Flanner et al.*, 2007]. *Randerson et al.* [2006] reported that BC from a boreal forest fire deposited on

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snow and sea ice introduced a global annual mean RE of 8±5 W per m^2 of burned area in the first year when the fire happened. A summary of BC-in-snow forcing/effect can be found in *Bond et al*. [2013]. They reported that the present-day RE of fire BC-in-snow ranges from 0.006 to 0.02 W m^{-2} based on previous studies [*Jacobson*, 2004; *Rypdal et al.*, 2009; *Skeie et al.*, 2011; *Hansen et al.*, 2005; *Flanner et al.*, 2007, 2009; *Koch et al.*, 2009].

Biomass burning aerosols can have significant impacts on global and regional precipitation and atmospheric circulation. With the change of fire emissions from year 1860 to 2000, *Jones et al.* [2007] found that biomass burning aerosols decrease the global near-surface air temperature by about 0.25°C, when considering the feedbacks of sea surface temperature (SST) in the model. As shown in *Tosca et al.* [2013], the direct and semi-direct effects of biomass burning aerosols reduce the precipitation near the equator and weaken the Hadley circulation. With a regional climate model, *Zhang et al.* [2009] found that biomass burning aerosols may warm and stabilize the lower troposphere and thus reinforce the dry season rainfall pattern in the Southern Amazonia. The absorption of shortwave radiation by biomass burning BC could increase the vertical stratification and inhibit both the cloud formation and precipitation [*Ackerman et al.*, 2000; *Tosca et al.*, 2014]. In contrast, biomass burning aerosols could invigorate the convective clouds [*Andreae et al.*, 2004a; *Koren et al.*, 2005] through suppressing warm rain processes in the convection, and enhance the latent heat release at higher levels [*Andreae and Rosenfeld*, 2008].

Although there have been many studies quantifying the RE of fire aerosols, a

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further investigation is still needed, as the current estimations of fire aerosol RE are still associated with large uncertainties [e.g., *Myhre and Samset*, 2015; *Chakrabarty et al.*, 2014]. The REs of co-emitted fire POM versus BC are even less clear. In this study, we estimate the present day (from year 2003 to 2011) open fire aerosol REs (including REari, REaci and REsac) using the NCAR Community Atmosphere Model version 5.3 (CAM5) with the four-mode version of the modal aerosol module (MAM4). We use two methods to calculate the REari of fire aerosols (total, BC-only, and POM-only). One method estimates the REari based on different model simulations [*Ghan*, 2013], and the other one calculates the REari directly through multiple diagnostic radiation calls in a single simulation. The spatial and seasonal characteristics of fire aerosol REs, and the impacts on the global precipitation and temperature are discussed.

Compared to earlier studies of fire aerosol REs [*Tosca et al.*, 2013; *Ward et al.*, 2012], a number of improvements are made in this study. First, a higher model horizontal resolution at 0.9° by 1.25° is used versus 1.9° by 2.5°. The higher resolution allows more efficient transport of aerosols from the sources to remote regions [*Ma et al.*, 2013; 2014]. Model resolution has also been shown to be important for aerosol REaci [*Ma et al.*, 2015]. Second, the latest CAM5 model with MAM4 is used. MAM4 with an additional primary carbon mode explicitly treats the microphysical ageing of primary carbonaceous aerosols (POM/BC) in the atmosphere. MAM4 has higher BC and POM burdens over the earlier three-mode version of MAM (MAM3) in the remote regions by ~30% [*Liu et al.*, 2016]. Third, daily instead

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of monthly fire emissions are used, which allows the model to consider the effect of fast changes in the fire emission flux on local atmospheric conditions. It is expected that using the monthly mean emission flux the model can not consider the effect of extremely strong fires, thus it might underestimate the fire aerosol REs for such cases. Finally, a new methodology [*Ghan*, 2013] is used to more accurately diagnose the REs of fire aerosols. Central to this method is that the REari must be calculated in the presence of clouds (i.e., under the all-sky condition), and the REaci be calculated under the condition of no aerosol effects on radiation. With the radiative forcing decomposition of this method, REsac can also be quantified.

The paper is organized as follows. Section 2 introduces the model and experiments. Section 3 describes the methods to diagnose the fire aerosol REs. Section 4 presents the model results of fire aerosol REs, and impacts on global and regional surface temperature and precipitation. Conclusions and discussion are given in Section 5.

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2. Model, Experiment Design and Aerosol Radiative Effect Method

2.1 Model

In our study, we use the Community Earth System Model (CESM) version 1.2, with the Community Atmosphere Model version 5.3 (CAM5.3) [*Neale et al.*, 2010] coupled with the Community Land Model version 4 (CLM4) [*Oleson et al.*, 2010]. The SNow, ICe, and Aerosol Radiative model (SNICAR) [*Flanner and Zender*, 2005] is turned on in the simulations to diagnose the biomass burning BC-in-snow effect. CAM5 includes several major updates in its physics parameterizations compared to

previous CAM versions. A two-moment stratiform cloud microphysics scheme is included in CAM5 to predict both the mass and number mixing ratios of cloud liquid and cloud ice [*Morrison and Gettelman*, 2008]. MAM4, which was updated from MAM3 [*Liu et al.*, 2012], includes aerosol mass and number mixing ratios in four lognormal modes: Aitken, accumulation, coarse, and primary carbon mode [*Liu et al.*, 2016]. An additional primary carbon mode is included in MAM4 on the top of MAM3 to explicitly treat the microphysical ageing of primary carbonaceous aerosols (POM and BC) in the atmosphere. POM and BC in MAM4 are emitted in the primary carbon mode instead of directly in the accumulation mode as in MAM3. MAM4 significantly increases the BC and POM concentrations in the remote regions (e.g., over oceans and Arctic) due to reduced wet scavenging of POM and BC in the primary carbon mode with a lower hygroscopicity than that in the accumulation mode. The increase is relatively small in the land source regions [*Liu et al.*, 2016].

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2.2 Experiment design

CAM5 was run with the finite volume dynamics core in a resolution of 0.9° latitude by 1.25° longitude and 30 vertical levels. The model was run for the time period of year 2003 to 2011 (i.e., for 9 years) with prescribed monthly SST and sea ice. The year 2003 was run twice and the first year simulation was used as a model spin-up. Global Fire Emissions Database version 3.1 (GFED 3.1) daily emissions [*Giglio et al.*, 2013] for BC, POM and sulfur dioxide (SO_2) from 2003 to 2011 are prescribed, and the vertical distribution of fire emissions is based on the AeroCom

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protocol [*Dentener et al.*, 2006]. Anthropogenic aerosol and precursor gas emissions are from the IPCC AR5 dataset [*Lamarque et al.*, 2010]. We performed our control experiment (FIRE) with the GFED fire emissions turned on and a sensitivity experiment (NOFIRE) with the fire emissions turned off. Differences between FIRE and NOFIRE experiments are used to calculate the REs and atmospheric effects of biomass burning aerosols on temperature and precipitation. Two additional experiments (NOFIREBC and NOFIREPOM) were performed with fire BC and POM emissions turned off, respectively. Differences between the control (FIRE) and these two experiments represent the contribution from biomass burning BC and POM, respectively. Other forcings (e.g., SST, greenhouse gases) of all these experiments are kept the same. We performed ten ensemble members for each of these experiments. Furthermore, we performed the other experiment (FIRE_BBFFBF) using the modified CAM5 model that separately predicts the BC and POM from biomass burning (BB), fossil fuel (FF) and biofuel (BF) sources, while other model features are kept the same as the FIRE experiment. A summary of all the experiments in this study can be found in Table 1.

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2.3 Methods of calculating fire aerosol radiative effects

The REs of all fire aerosols, fire BC, and fire POM are calculated from the differences of TOA shortwave fluxes (ΔF) between the FIRE experiment and the three other experiments (NOFIRE, NOFIREBC and NOFIREPOM), respectively. All the atmospheric variables (including temperature, precipitation, and circulation) are

allowed to adjust in the experiments. However, with SST and sea ice prescribed in these experiments, only the rapid adjustments are taken into account. Thus the *effective* radiative effects are actually calculated in this study.

$$\Delta F_{\text{fire aero}} = F_{\text{fire}} - F_{\text{nofire}} \quad (1)$$

$$\Delta F_{\text{fire bc}} = F_{\text{fire}} - F_{\text{nofirebc}} \quad (2)$$

$$\Delta F_{\text{fire pom}} = F_{\text{fire}} - F_{\text{nofirepom}} \quad (3)$$

The total TOA shortwave flux change can be broken into the REari, REaci, and REsac. The aerosol REaci results from both the aerosol effect on clouds via acting as CCN and the aerosol semi-direct effect on clouds via affecting the atmospheric states due to absorbing aerosols. We adopt the method of *Ghan* [2013] to separate the REari, REaci, and REsac from the total effects of all fire aerosols, fire BC and fire POM, respectively. The method is summarized as follows. F_{clean} is the radiative flux at TOA calculated from a *diagnostic radiation call* in the same control simulations, but neglecting the scattering and absorption of solar radiation by aerosols. $F_{\text{clean,clear}}$ is the clear-sky radiative flux at TOA calculated from the same *diagnostic radiation call*, but neglecting scattering and absorption by both clouds and aerosols.

$$\Delta F = \underbrace{\Delta(F - F_{\text{clean}})}_{\text{(REari)}} + \underbrace{\Delta(F_{\text{clean}} - F_{\text{clean,clear}})}_{\text{(REaci)}} + \underbrace{\Delta F_{\text{clean,clear}}}_{\text{(REsac)}} \quad (4)$$

In the method above, REaci includes both aerosol indirect and semi-direct effects. The fire BC has a much weaker indirect effect due to its lower mass burden and lower hygroscopicity compared to fire POM [*Koch et al.*, 2011]. Thus the fire aerosol semi-direct effect can be approximately represented by the REaci of fire BC. The fire

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aerosol indirect effect can be estimated as the difference of fire aerosol REaci and semi-direct effect. With the sea ice prescribed in these experiments, the radiative effect of fire aerosols on sea ice albedo is not considered in REsac.

We undertake another method to estimate the fire aerosol REari from the experiment (FIRE_BBFFBF). With explicit predictions of fire POM and fire BC in FIRE_BBFFBF, the REari of fire BC and fire POM are estimated by two diagnostic radiation calls, each time neglecting the scattering and absorption of solar radiation of fire BC and fire POM, respectively. This more direct method is named as BBFFBF, and the REari of fire BC and fire POM will be compared with those from the method of *Ghan* [2013]. The fire BC-in-snow effect is calculated from SNICAR, and compared with the REsac estimated from *Ghan* [2013].

3. Results

3.1 Simulation of biomass burning aerosols

The biomass burning BC and POM from forest, grass and agriculture fires are significant contributors to the total BC and POM emissions. Figure 1 shows the seasonal variation of GFED fire emissions (including forest, grass and agriculture fires) in the global, tropical (25°S to 25°N), and Arctic (60°N to 90°N) regions. Global fire emission is the largest during the boreal summer as well as in the boreal autumn (September/October), when it is the fire season in the tropical regions of the Southern Hemisphere (SH). The tropical fire emission contributes the most to the annual global fire emission (80% for BC and 85% for OC, respectively), compared to

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other regions. Arctic is the other important fire region, where the emission maximum is found during the boreal summer. In the boreal summer, the OC emission in the Arctic regions is about 50% of that in the tropical region. The BC emission in the Arctic is much smaller than that of the tropical regions even in the boreal summer fire season. The dominant fire type in the SH tropics is deforestation, savanna and grassland fires, while that in the Arctic is the forest fires. The OC to BC ratio (OC/BC) of forest fires is almost three times higher than that of deforestation, savanna and grassland fires [*van der Werf et al.*, 2010].

Figure S1 in the supplemental materials shows the latitudinal and longitudinal distributions of vertically integrated concentrations (column burdens) of BC and POM from BB, FF, and BF sources based on the FIRE_BBFFBF experiment. The BC and POM from BB source are mainly distributed in the tropical and sub-tropical regions (southern Africa, South America and Southeast Asia) and in the mid- to high latitudes (North of 45°N) of the Northern Hemisphere (NH) (Northeast Asia, Alaska and Canada). The largest column burdens of biomass burning aerosols are located in southern Africa and adjacent oceanic areas (1.5 and 20 mg m^{-2} for BC and POM, respectively). The biomass burning aerosols are important aerosol species in the Arctic regions, and contribute up to 53% and 86% to the total burden of BC and POM in the Arctic (from 60° N to 90°N), respectively. In comparison, the maximum column burdens of fossil fuel BC and POM are found in East Asia, South Asia, Western Europe and North America. The maximum column burdens of biofuel BC and POM occur in East Asia, South Asia and Central Africa. The biofuel and fossil

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fuel sources are dominant contributors to BC and POM in East Asia and South Asia. In other regions of the world, biomass burning is the primary source of BC and POM. Globally, the biomass burning contributes 41% and 70% to the total burdens of BC and POM, respectively. Biomass burning can also emit SO_2. However, it only contributes ~3% to the total global sulfate burden (figure not shown), so only radiative effects of biomass burning POM and BC are discussed in this study.

The simulated aerosol optical depth (AOD) and single scattering albedo (SSA) (including aerosols from all sources) are validated with observations from the AErosol RObotic NETwork (AERONET, http://aeronet.gsfc.nasa.gov) at sites significantly affected by biomass burning activity in southern Africa, South America and the Arctic regions, as shown in Figures 2 and 3 (see Figure S2 in the supplemental materials for the site locations). The AERONET AOD and SSA data are averaged for the years from 2003 to 2011 to match the simulation period, although there are missing AERONET data for some periods. We note that *Tosca et al.* [2013] and *Ward et al.* [2012] applied scaling factors (from 1 to 3 varying by regions) to fire emissions to improve modeled AOD magnitudes, whereas here we do not apply any such scaling. In southern Africa, modeled monthly AOD agrees with observations within a factor of 2 for the three sites (Figure 2a-2c). The underestimation of AOD is found in the tropical site (Mongu) (Figure 2a) during the boreal autumn (the fire season). The simulated AOD in the two other sites (Skukuza and Ascension Island) is generally consistent with observations in both the magnitude and seasonal trend. The simulated SSA in southern Africa ranges between 0.75 and 0.95 and generally

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matches the observed SSA magnitude and seasonal cycle in the two land sites (Mongu and Skukuza) (Figure 3a-3b). However, an overestimation of SSA is found in the oceanic site (Ascension Island) (Figure 3c). The reason for this overestimation of SSA and thus the underestimation of absorption AOD (AAOD) is unclear and could be due to that the model has not treated the absorption enhancement of aged fire BC during its transport.

The simulated AOD in South America is generally consistent with observations within a factor of 2 (Figure 2d-2f). The seasonal variation of simulated AOD generally matches the observations. The underestimation of AOD in Alta Floresta and Cuiaba-Miranda is most obvious in September and October (the fire season), which may be attributed to the underestimation of fire emissions. However, the modeled AOD is higher than observations before the fire season for Alta Floresta and Rio Branco, which could be due to the overestimation of fire emission in this period. The simulated SSA in South America ranges mostly between 0.87–0.95 and matches the observations reasonably well (Figure 3d-3f). The modeled SSA is too low during the fire season and exhibits too strong a seasonality. It implies that the model underestimation of scattering aerosols (e.g., POM) may be more severe than that of BC during the fire season.

In the Arctic, small AOD (less than 0.3) and large SSA (larger than 0.9) are observed for the three sites. The observed large SSA in the fire season (boreal summer) is consistent with the high OC/BC ratio of fire emissions in the Arctic (Figure 1). The model significantly underestimates the observed AOD in the Arctic in

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both fire and non-fire seasons. The underestimation of AOD can be due to (1) the underestimation of fire emissions in the NH high latitudes [e.g., *Stohl et al.*, 2013] and/or fossil fuel emissions in Asia [e.g., *Cohen and Wang*, 2014], (2) the excessive scavenging of aerosols during their transport from the NH mid-latitude industrial regions by liquid-phase clouds [*Wang et al.*, 2013a], and (3) the coarse horizontal resolution (~100 km) of the model [*Ma et al.*, 2014]. Although MAM4 increases the column burdens of POM and BC by up to 40 % in many remote regions compared to MAM3, it still underestimates the surface BC concentrations in the Arctic [*Liu et al.*, 2016]. The modeled SSA in the Arctic is lower than observations, which implies that the simulation of AAOD is better than that of AOD and the underestimation of non-absorbing aerosols (e.g., sulfate and POM) in the Arctic may be more severe than that of BC.

3.2 Radiative effect due to aerosol-radiation interactions

The annual mean REari of all fire aerosols (including BC, POM and sulfate) estimated with the method of BBFFBF and with the method of *Ghan* [2013] is shown in Figure 4a-4b. The fire sulfate is not included in the calculation of REari of all fire aerosols with the method of BBFFBF. Its effect is minor since the global annual mean burden of fire sulfate (0.09 mg m^{-2}) is much smaller than that of fire POM (1.25 mg m^{-2}), both of which are light-scattering. The statistical significance of REari estimated with the *Ghan* [2013] method over the interannual variability and ensemble member diversity is shown in Figure 4 (and also later figures). The REari of all fire aerosols

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from the two methods agree with each other very well. Thus, we will report the REari of all fire aerosols with the *Ghan* [2013] method below. The global annual mean REari of all fire aerosols is positive (0.16±0.01 W m^{-2}), which indicates a warming effect from all fire aerosols. The REari is positive on the globe except in some land areas (e.g., southern Africa, South America, Great Lakes, North Canada, and East Siberia). The maximum positive REari is located in ocean areas west of southern Africa (~5.0 W m^{-2}) and South America (~1.5 W m^{-2}). Positive REari up to 1 W m^{-2} is found in the Arctic (60°N to 90°N). The different signs of REari between land and ocean areas of southern Africa and South America result from the differences in cloud fraction and cloud liquid water path (LWP) between land and ocean regions. In the fire season (August-September-October) of the SH tropical regions, cloud fraction and cloud LWP over the land areas (10% and 20 g m^{-2}, respectively) are much smaller than those over the adjacent ocean areas (70% and 70 g m^{-2}, respectively). The biomass burning aerosols are transported above the low-level stratocumulus clouds, and when biomass burning BC resides above clouds, its absorption of solar radiation is significantly enhanced due to the reflection of solar radiation by underlying clouds [*Abel et al.*, 2005; *Zhang et al.*, 2016].

A comparison of modeled REari in the boreal autumn (September-October-November) over the South Atlantic Ocean with satellite observations is shown in Figure 5. The observed above-cloud aerosol REari is calculated with the method of *Zhang et al.* [2014] using the Aqua/MODIS and Terra/MODIS products, respectively. The observed above-cloud aerosol REari over

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southeastern Atlantic Ocean is 3-12 W m^{-2}, with higher values near the coasts. The simulated REari agrees better with Aqua/MODIS observed REari than with Terra/MODIS in both the magnitude and spatial pattern. REari estimated from Terra/MODIS (morning time) is stronger than the one estimated from Aqua/MODIS (afternoon time) due to the larger amount of underlying clouds in the morning [*Min and Zhang*, 2014]. Over South America during the fire season (August to September), the clear-sky fire aerosol REari is estimated to be -5.2 W m^{-2} by *Sena and Artaxo* [2015], which is larger than our model result (-2.1 W m^{-2}). This is consistent with the underestimation of modeled AOD in South America compared to the AERONET data (Figure 2).

The seasonal variation of REari of all fire aerosols with the *Ghan* [2013] method is shown in the supplemental Figure S3. The REari has a maximum (1.13 W m^{-2}) in the boreal summer (June-July-August, JJA) over the Arctic regions, partially due to the low solar zenith angles there. The maximum positive REari in the tropical regions occurs in the boreal summer and autumn (September, October and November, SON) during the fire season of southern Africa and South America. The REari reaches a positive maximum in Southeast Asia during the fire season in March, April and May (MAM).

The REari of fire BC is shown in Figure 4c-4d. The fire BC REari calculated from the two methods are similar in magnitudes and spatial patterns, and there is much less noise with the BBFFBF method. The global annual mean fire BC REari is about 0.25±0.01 W m^{-2} and positive over the globe (the regions with negative values

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in Figure 4d are in general not statistically significant). Unlike all fire aerosols, fire BC generates a positive forcing in the land regions of southern Africa and South America, and the amplification effect of low-level clouds on fire BC positive forcing can be clearly seen in southern Africa and adjacent Atlantic Ocean.

The global annual mean REari of fire POM from the two methods somewhat differs from each other (Figure 4e-4f). The BBFFBF method gives a small negative value (-0.05 W m^{-2}), while the *Ghan* [2013] method shows a small positive value (0.04 ± 0.01 W m^{-2}). The difference is mainly in the Arctic regions where the positive forcing from *Ghan* [2013] is larger than that from the BBFFBF method. This is because the removal of fire POM emissions in the NOFIREPOM experiment affects the properties of aerosol particles within which co-emitted fire BC is internally mixed with fire POM, causing a decrease of BC burden in the Arctic (by ~0.05 mg m^{-2}) compared to the FIRE experiment. Thus, one should be careful in using the *Ghan* [2013] method to diagnose the radiative forcing of a single component within co-emitted aerosols. The REari of fire POM is negative in most of the globe. However, positive forcing can be found over oceanic regions west of southern Africa and South America, North Pacific Ocean and the Polar regions where large amount of low-level clouds, sea ice or land ice exist. The multiple scatterings between the above-cloud fire POM and low-level clouds or between the fire POM and the Earth's bright surface with high albedos could reduce the amount of solar radiation reflected by these low-level clouds and bright surface in the case without the fire POM [*Zhang et al.*, 2016]. With the BBFFBF method the sum of REari from fire POM and fire BC (i.e.,

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0.20 W m^{-2}) is larger than that of all fire aerosols (0.15 W m^{-2}). It reflects the nonlinear interactions among different aerosol components [*Ghan* et al., 2012]. The nonlinearity is stronger with the *Ghan* [2013] method.

3.3 Radiative effect due to aerosol-cloud interactions

The annual mean REaci due to all fire aerosols, fire BC, and fire POM are shown in Figure 6. The REaci diagnosed with the *Ghan* [2013] method includes both aerosol indirect and semi-direct effects. The fire aerosol semi-direct effect (to be discussed below) is much smaller (-0.04±0.03 W m^{-2} on the global mean) than the indirect effect, and the REaci is mostly from the fire aerosol indirect effect. The global annual mean REaci of all fire aerosols is -0.70±0.05 W m^{-2} (Figure 6a). In the tropical regions, the strong negative REaci is located in the adjacent ocean areas of southern Africa, South America and Australia, with the maximum REaci of -8.0 W m^{-2} over the South Atlantic Ocean. The strong negative REaci also occurs in the Arctic (60°N to 90°N). The REaci in East Siberia, Alaska and Canada is as large as -6.0 W m^{-2}.

The fire BC has a weak indirect effect by acting as CCN, but can reduce the cloud amount through its semi-direct effect. The REaci of fire BC (Figure 6b) can approximate the fire BC semi-direct effect with a small global annual mean value of -0.04±0.03 W m^{-2}. However, stronger positive effect can be found in the western Pacific (3.0 W m^{-2}) and Arctic regions (1.0 W m^{-2}). The global annual mean REaci of fire POM is -0.59±0.03 W m^{-2} (Figure 6c), and dominates the cloud effect of all fire aerosols. The sum of REaci from fire BC and POM (-0.62±0.03 W m^{-2}) is smaller

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than that of all fire aerosols (-0.70±0.05 W m^{-2}) due to the non-linear interactions of fire BC and fire POM [*Jiang et al.*, 2013] as well as the negative REaci of fire sulfate.

The seasonal variation of all fire aerosol REaci is shown in Figure 7. The maximum of fire aerosol REaci is in the boreal summer (i.e., the fire season in NH) located in the NH high latitudes (60°N to 90°N). The largest summer REaci is found in the land areas and is as large as -15 W m^{-2}. The fire aerosol REaci in the tropical regions is most significant in the boreal summer (up to -15 W m^{-2}) and autumn (up to -10 W m^{-2}) over the ocean areas. The different spatial distributions of fire aerosol REaci in the NH high latitudes and in the tropics result from the difference in cloud distributions between the two regions. During the fire season the cloud LWP over the land areas in the NH mid- and high latitudes is three times larger than that over the ocean areas in the tropics. Larger cloud LWP favors the stronger REaci, because the larger LWP associated with the warm cloud and rain processes favors the aerosol indirect effect via slowing down the autoconversion of cloud water to rain [*Ghan et al.*, 2012; *Jiang et al.*, 2015]. Meanwhile, in the NH high latitudes, the lower solar zenith angle in the boreal summer favors the stronger REaci. Like the fire aerosol REari, the smallest fire aerosol REaci occurs in the boreal spring.

Seasonal variations of zonal mean fire aerosol REari, REaci, cloud LWP, low-level (from surface to 750 hPa) cloud amount, and vertically-integrated (burden) concentrations of fire POM and fire BC are shown in Figure 8. The seasonal variation of fire BC and fire POM burdens is largest in the SH low latitudes (from 30°S to 0°N) and NH mid- and high latitudes (50°N to 90°N). Distinct features of these two areas

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816 can also be noticed that the maximum fire BC burden in NH (0.3 mg m^{-2}) is much
817 lower than that in SH (0.8 mg m^{-2}), while the maximum POM burdens in these two
818 areas are comparable. Interestingly, the REari is larger in the boreal summer in NH
819 than that in the boreal autumn in SH, although the fire BC burden is much lower in
820 the NH summer. It is mainly due to the larger amount of low clouds in the NH high
821 latitudes, which enhances the absorption of fire BC. The maximum REari in the NH
822 summer is found near the North Pole (70 °N to 90 °N), and not around 60 °N where
823 the fire aerosol burden is highest. The REaci of fire aerosols is about 3 times larger in
824 the boreal summer in NH than that in the boreal autumn in SH, although the burden of
825 fire POM in NH is comparable to that in SH. The larger cloud LWP in the NH
826 summer around 40-70°N favors the stronger REaci there.
827
828 3.4 Surface albedo effect
829 Here we compare the modeled BC-in-snow (BCS) concentrations with
830 observation data collected from multiple field campaigns over the Arctic [*Doherty et*
831 *al.*, 2010] and Northern China [*Wang et al.*, 2013b; *Qian et al.*, 2014]. Figure 9a
832 shows the simulated (from FIRE and NOFIRE experiments) and observed BCS
833 concentrations as a function of latitude. The range of observed BCS concentrations is
834 between 1 and 200 ng g^{-1} in the Arctic and between 50 and 2000 ng g^{-1} in Northern
835 China, respectively. Both FIRE and NOFIRE experiments capture the meridional
836 gradient in BCS concentrations between the mid-latitudes (Northern China) and high
837 latitudes (Arctic). The mean and median concentrations of BCS are both

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overestimated in Northern China, implying the high biases from the anthropogenic emissions and/or model physics (Figure 9b). The mean and median BCS concentrations from the FIRE experiment agree slightly better with observations than those from the NOFIRE experiment in the Arctic (Figure 9b). This suggests that fire emissions are important for BCS concentrations in the Arctic.

The annual mean REsac of all fire aerosols estimated with *Ghan* [2013] and the fire BCS effect diagnosed from SNICAR are shown in Figure 10a. We note that the radiative effect due to BC deposition on sea ice is not considered since sea ice is prescribed in the simulations. The global annual mean REsac (0.03 ± 0.10 W m^{-2}) is much smaller compared to the REari and REaci. The REsac over land is maximum in spring (0.12 ± 0.27 W m^{-2}) and winter (0.06 ± 0.16 W m^{-2}). The REsac over land in summer and autumn is very small (less than 0.01 W m^{-2}). We note that the mean REsac calculated with *Ghan* [2013] is much smaller than the standard deviation resulted from the internal variability.

The annual mean fire BCS effect calculated from SNICAR is shown in Figure 10b and 10c. The spatial distribution of the fire BCS effect is similar to the fire REsac implying that the fire REsac has a significant contribution from the fire BCS effect. Averaged when only snow is present, the fire BCS effect is larger (0.048 W m^{-2}). The global mean fire BCS effect (with the presence of snow) can be as large as 0.06 W m^{-2} in spring. The maximum fire BCS effect (up to 1 W m^{-2}) is located in Greenland and the very northern reaches of Canada, while that in the other Arctic regions and North China is smaller.

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The positive REsac in Siberia, North America and Canada can be a result of BCS effect. However, the REsac in these regions is larger than the BCS effect especially in spring. The snow melting and snow depth change due to the BCS warming may induce a larger positive REsac than the albedo change due to BCS itself. The negative REsac over land can be a result of atmospheric feedbacks caused by fire aerosols [*Ghan*, 2013].

3.5 Fire aerosol effects on shortwave radiation, global temperature and precipitation

Here, we show the annual mean net shortwave flux change at TOA (i.e., total radiative effect), in the atmosphere and at surface, and changes in surface air temperature, convective and large-scale precipitation due to all fire aerosols in Figure 11 and Table 2. The global mean net shortwave flux change at TOA due to all fire aerosols is -0.55±0.07 W m^{-2}, which indicates that fire aerosols lead to the reduction of shortwave flux into the Earth's system. The zonal mean TOA shortwave flux reduction in the Arctic regions (-1.35±1.03 W m^{-2}) is much larger than that in the tropical regions (-0.66±0.09 W m^{-2}). The cooling at TOA is mostly from fire aerosol REaci. The maximum negative RE is located in the land areas of the Arctic and ocean areas of the tropics. Although the global mean total radiative effect is negative, positive effect is found in some land areas (e.g., Africa, Greenland).

The shortwave atmospheric absorption change in the tropical regions is larger than that in the Arctic regions. It is because BC burden in the tropics (0.17 mg m^{-2}) is larger than that in the Arctic (0.09 mg m^{-2}). Strong absorption (~8 W m^{-2}) in the

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atmosphere is found in the land areas of southern Africa and South America and in the Southeast Atlantic. The surface shortwave flux change in the Arctic is mostly from the TOA shortwave flux reduction due to the fire aerosol REaci, while the surface shortwave flux change in the tropics is mostly due to the fire BC absorption in the atmosphere.

The fire aerosols lead to the reduction of the global mean surface air temperature (T_s) by 0.03±0.03 K, consistent with the reduction of shortwave fluxes at TOA and at surface. The largest surface cooling is found in the Arctic and tropical regions by up to 0.6 K. The cooling of the Arctic is related to the strong fire aerosol REaci, while the cooling in the tropics is mainly from the surface shortwave flux reduction due to the fire BC absorption. The T_s change in the ocean areas is very small since the SST is prescribed in our simulations.

The global mean total precipitation is reduced by 0.010±0.002 mm day^{-1} due to all fire aerosols (Table 2). Unlike the T_s change, the precipitation reduction in the tropics (0.016±0.01 mm day^{-1}) is much larger than that in the Arctic (0.001±0.02 mm day^{-1}, not statistically significant). The reduction in the tropics is mainly from the large-scale precipitation decrease (0.015±0.003 mm day^{-1}). The net decrease in the convective precipitation is very small in the tropics (0.001±0.009 mm day^{-1}, not statistically significant), as the convective precipitation is significantly decreased near the equator and increased in the regions away from the equator, partly consistent with the results of *Tosca et al.* [2013]. The precipitation reduction in southern Africa is consistent with the recent findings of *Hodnebrog et al.* [2016]. The shortwave flux

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reduction at surface leads to a stabilization of the atmospheric boundary layer and a suppression of the convection near the equator. The strong atmospheric absorption by fire BC leads to the reduction of low-level clouds and large-scale precipitation in the tropics. Both effects lead to a significant reduction of total precipitation near the equator. The precipitation decrease in the NH high latitudes is mainly from the reduction of convective precipitation. We note that the temperature and (especially) precipitation changes reported here do not represent the complete impact of fire aerosols, since the SSTs are fixed in our simulations. Fully-coupled atmosphere and ocean models will be used to further investigate the impact of fire aerosols.

Figure 12 shows the changes of T_s, total precipitation, cloud LWP, and low-level cloud cover in the summer due to all fire aerosols. The T_s is reduced by more than 1 K in most of land areas around 60°N. The maximum cooling (larger than 1.5 K) is found in East Siberia, Alaska and Canada. A decrease of total precipitation (by about 0.2 mm day^{-1}) is found in these regions. Accompanying the surface cooling and precipitation reduction, a significant increase of cloud LWP and low-level cloud cover is found there. This is a result of the indirect effect of fire aerosols in the land areas of the Arctic (60°N to 90°N). The fire POM leads to the reduction of cloud droplet effective radius and the increase of cloud droplet number concentration, consistent with observed fire effects on clouds in Canada and the United States [*Peng et al.*, 2002].

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4. Discussion and Conclusions

Although many studies have been conducted on the fire aerosol RE and RF [e.g., *Bond et al.*, 2013; *Myhre et al.*, 2013b; *Ward et al.*, 2012; *Tosca et al.*, 2013], the current estimations are still associated with large uncertainties. In this study, the fire aerosol RE (including REari, REaci and REsac) is calculated based on a new method from *Ghan* [2013]. In addition, the fire aerosol REari and fire BC-in-snow effect are diagnosed from an experiment of CESM which tracks the open fire BC and POM separately from fossil fuel and biofuel sources and compared with the estimates from the *Ghan* [2013] method.

The BC and POM burdens from open fires are largest in the tropical regions (southern Africa, South America and Southeast Asia) and in the NH mid- to high latitudes (North of 45°N) (Northeast Asia, Alaska and Canada). Fire aerosols contribute 41% and 70% to the global burden of BC and POM, respectively. When comparing with the AERONET AOD and SSA data, modeled monthly AOD agrees with observations within a factor of 2 for most of the southern African and South American sites. The model underestimation of AOD is found in the South American sites near fire source regions, which is most obvious in the fire season (September and October). The model underestimates the observed AOD in the Arctic regions in both fire and non-fire seasons. The modeled SSA in southern Africa and South America is generally in agreement with observations, while the modeled SSA in the Arctic is lower.

The annual mean REari of all fire aerosols is 0.16±0.01 W m^{-2} and positive over most areas except in some land areas (e.g., southern Africa, North Canada, and East

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Siberia). The annual maximum REari is found in the oceanic areas west of southern Africa (5 W m^{-2}) and South America (1.5 W m^{-2}). The positive REari over the land regions of southern Africa and South America is smaller, although the fire aerosol burdens are higher. The annual zonal mean REari in the Arctic regions can reach 0.43 ±0.028 W m^{-2}, and is larger than that in the tropical regions (0.17±0.017 W m^{-2}), although the fire aerosol burden is higher in the tropics. The annual mean REari of fire BC is about 0.25±0.01 W m^{-2} and positive over the globe. Fire POM induces a weak negative REari globally (-0.05 W m^{-2}) with the BBFFBF method and a small positive value (0.04±0.01 W m^{-2}) with the *Ghan* [2013] method. The positive REari of fire POM is found over oceanic areas west of southern Africa and South America, North Pacific, and polar regions where the low-level cloud coverage is large or the surface albedo is higher.

The global annual mean REaci of all fire aerosols is -0.70±0.05 W m^{-2} and the maximum effect is located in the ocean areas west of southern Africa and South America and land areas of the NH high latitudes. The maximum fire aerosol REaci occurs in the NH high latitudes in the boreal summer, which results from the large cloud LWP over the land areas and the low solar zenith angle. Associated with the strong indirect effects of fire aerosols in the Arctic summer, significant surface cooling, precipitation reduction, and low-level cloud cover increase are found in these regions.

Modeled BCS concentrations from the FIRE experiment are evaluated against observations in Northern China and in the Arctic, and generally agree with the

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observations for the mean and median values in the Arctic regions. The high bias of modeled BCS concentrations in Northern China may not result from the fire BC because differences in BCS concentrations between FIRE and NOFIRE experiments are very small in North China. The global annual mean REsac is 0.03±0.10 W m^{-2} (statistically insignificant) with the maximum effect in spring (0.12 W m^{-2}). The REsac is mainly due to the effect of fire BC deposit on snow (0.02 W m^{-2}) diagnosed from SNICAR with the maximum effect as large as 0.06 W m^{-2} (when snow is present) in spring.

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The fire aerosols reduce the global mean surface air temperature (T_s) by 0.03± 0.03 K and precipitation by 0.01±0.002 mm day^{-1}. The maximum cooling (~1 K) due to fire aerosols occurs around 60°N in summer, and a suppression of precipitation (~0.1 mm day^{-1}) is also found there. The strong cooling is a result of the strong indirect effects (-15 W m^{-2}) in the land areas of the Arctic regions (60°N to 90°N). A significant reduction of precipitation in southern Africa is also noticed. We note that these results are based on the simulations with fixed SSTs and may not represent the full climate responses.

In our study, the global radiative effect of fire aerosols is estimated from simulations performed with the 4-mode version Modal aerosol module (MAM4) [*Liu et al.*, 2016], daily fire emissions with prescribed vertical emission profiles, and higher model resolution (0.9° by 1.25°) compared to earlier modeling studies of fire aerosols [*Tosca et al.*, 2013; *Ward et al.*, 2012]. In their studies, the GFED fire aerosol emissions were increased by a factor of 1-3 depending on regions to match the

observed AOD. In our study, we do not apply the scaling factor to the fire aerosol emissions. Our global annual mean REari of fire aerosols (0.16±0.01 W m^{-2}) is, however, close to 0.18 W m^{-2} in *Tosca et al.* [2013] and 0.13 W m^{-2} in *Ward et al.* [2012]. The similar fire aerosol REari from our study but with smaller fire emissions than these previous studies can result from (1) the use of MAM4 in our study which more realistically represents the external/internal mixing of BC with other soluble aerosol species; (2) the more accurate estimation of REari of fire aerosols in the presence of low-level clouds with the method of *Ghan* [2013]; and (3) the inclusion of vertical emissions of fire aerosols, which allows more efficient transport of fire aerosols from sources. The REaci due to fire aerosols in our study (-0.70±0.05 W m^{-2}) is smaller than -1.64 W m^{-2} in *Ward et al.* [2012] due to the lower fire POM emissions used in this study compared to *Ward et al.* [2012].

We note that there are limitations and uncertainties with our study. The model still underestimates observed AODs (mostly within a factor of 2) at the sites predominantly influenced by biomass burning aerosols during the fire season, which implies that the fire aerosol radiative forcing can be stronger than estimated in this study. The RE estimates of fire POM and fire BC with the *Ghan* [2013] approach may not be accurate due to the internal mixing of co-emitted fire components (POM and BC). In our simulations, sea ice is prescribed, and thus the fire BC effect on sea ice albedo is not considered. The brown carbon component of POM [*Feng et al.*, 2013] is not treated in the current CESM model, which may result in an underestimation of atmospheric absorption of fire aerosols.

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Figure Captions

Figure 1. Seasonal variation of GFED monthly fire (a) organic carbon (OC) and (b) black carbon (BC) emissions (Tg C $month^{-1}$) averaged for the period of year 2003 to 2011 in the global, tropical (25°S to 25°N) and Arctic (60°N to 90°N) regions.

Figure 2. Comparison of modeled seasonal variations of aerosol optical depth (AOD) for the period of 2003-2011 with observations for the same period from the AERONET sites. The upper, middle, and bottom panels represent the sites in southern Africa, South America, and the Arctic, respectively.

Figure 3. Same as Figure 2, but for the comparison of single scattering albedo (SSA) at 550 nm.

Figure 4. Annual mean radiative effect due to aerosol-radiation interactions (REari) (W m^{-2}) averaged over the period of 2003-2011 due to (a) all fire aerosols, (c) fire BC, and (e) fire POM estimated with the method of BBFFBF (left panels), and with the method of Ghan (2013) ((b), (d), and (f) in the right panels). The plus signs in Figure 4(b), (d) and (f) denote the regions where the radiative effect estimated with Ghan [2013] is statistically significant at the 0.05 level.

Figure 5. (a) September-October-November (SON) mean fire aerosol radiative effect due to aerosol-radiation interactions (REari) (W m^{-2}) for the period of 2003-2011 over the Southeast Atlantic Ocean due to all fire aerosols. (b) and (c) are the same as (a), but for the above-cloud aerosol REari for the period of 2007-2011 estimated using Aqua/MODIS and Terra/MODIS products [*Zhang et al.*, 2014], respectively.

Figure 6. Annual mean radiative effect due to aerosol-cloud interactions (REaci) (W m^{-2}) averaged over the period of 2003-2011 due to (a) all fire aerosols, (b) fire BC, and (c) fire POM. The plus signs denote the regions where the radiative effect is statistically significant at the 0.1 level.

Figure 7. Seasonal variation of radiative effect of all fire aerosols due to aerosol-cloud interactions (REaci) (W m^{-2}) for the period of 2003-2011 for (a) December-January-February (DJF), (b) March-April-May (MAM), (c) June-July-August (JJA), and (d) September-October-November (SON). The plus signs denote the regions where the radiative effect is statistically significant at the 0.05 level.

Figure 8. Month-latitude cross sections of zonal mean and monthly (a) vertically-integrated concentrations (mg m^{-2}) of fire BC and (b) fire POM, (c) cloud liquid water path (LWP, in g m^{-2}), (d) low-level cloud cover (CLDLOW, in %), (e) radiative effect due to aerosol-radiation interactions (REari, in W m^{-2}), and (f) radiative effect due to aerosol-cloud interactions (REaci, in W m^{-2}) of all fire aerosols.

Figure 9. Evaluation of CAM5 simulated black carbon (BC) concentration for the period of 2003-2011 (in ng g^{-1}) in the top snow layer against observations in the Arctic [*Doherty et al.*, 2010] and Northern China [*Wang et al.*, 2013b]. The top snow layer ranges in thickness from 1 to 3 cm. Configuration of the two CAM5 simulations (FIRE and NOFIRE) is summarized in Table 1. Panel (a) shows the comparisons at different latitudes. The box and whisker plot in panel (b) shows the minimum and maximum value with the bar, the 25th and 75th percentiles with the box, the 50th percentile (i.e., median) by the bar within the box, and the mean value with the dot.

Figure 10. (a) Annual mean radiative effect due to surface albedo changes (REsac, W m^{-2}) averaged over the period of 2003-2011 of all fire aerosols over land regions, and annual mean surface effect of fire BC-in-snow calculated from SNICAR averaged (b) over all times and (c) only when snow is present. The plus signs in (a) denote the regions where the radiative effect is statistically significant at the 0.1 level.

Figure 11. Annual mean net shortwave flux changes (W m^{-2}) over the period of 2003-2011 (a) at top of the atmosphere, (b) in the atmosphere, (c) at surface, and changes of (d) surface air temperature (TS, K), (e) convective precipitation (mm d^{-1}), and (f) large-scale precipitation (mm d^{-1}) due to all fire aerosols. The plus signs denote the regions where the change is statistically significant at the 0.1 level.

Figure 12. Changes in (a) surface air temperature (K), (b) total precipitation (mm d^{-1}), (c) cloud liquid water path (g m^{-2}), and (d) low-level cloud cover (%) due to all fire aerosols in the boreal summer (JJA) averaged for the period of 2003-2011. The plus signs denote the regions where the change is statistically significant at the 0.1 level.