Response to the comments of Reviewer #2

First of all, we would like to thank the two anonymous reviewers for their thoughtful reviews and valuable comments on the manuscript. In the revision, we have accommodated all the suggested changes into consideration and revised the manuscript accordingly. All changes are highlighted in the revised manuscript in **BLUE** in the revision. In this response, the questions and comments of reviewers are in **BLACK** font, and responses are highlighted in **BLUE.** The changes made in the revised manuscript are marked in **RED** font.

General comments:

Materials presented in the manuscript are interesting and well suited to the scope of the current journal. The authors seem to have successfully operated a chain of models to present the simulation results, but their argument needs improvements and additional calculations may be required. Thus, the manuscript should be accepted in the journal, only after the authors revise the manuscript by reflecting the following general and specific comments.

Response: Thanks very much for your comments. The specific responses and revisions are shown in follows.

Comments: The title says "regional impacts of ARI". The authors only presented ARI at several grid points, but the regional mean ARI should be presented and discussed. Without the presentation of regional ARI, the current study is not comparing US and China, but comparing a few fire grids in US and a few urban grids in China.

Response: Thanks very much for your comments. In the revised manuscript, the regional ARI is presented. Due to huge computations, the regional ARI was calculated using the mean aerosol optical properties, mean cloud properties, and mean albedo, etc.

We must clarify that the calculation of ARI using mean optical properties would result in some differences from those using temporary optical properties, as the aerosol optical properties would show temporary variations. However, as an estimation for the impacts on the BC morphology, it is still reasonable to make some simplifications as the mean optical properties can also represent the spatial distribution of ARI to some extent. Besides, and similar methods were also used in previous studies (eg. Saleh et al. (2015); Tuccella et al. (2020a)).

Comments: There are several types of BC-ARI, but the differences are not clearly stated. For example, the authors mentioned 1.1 W m^{-2} for global mean BC-ARI (Bond et al., 2013), but actually the value is “total climate forcing”. It is totally different from the authors’ BC-ARI, that is “clear-sky direct affect”. For every ARI value in the manuscript, please be aware which types of ARI you are siting.

Response: Thanks very much for pointing it out. We have corrected it in the revised manuscript.

Comments: There are clear-sky and all-sky ARI. All-sky ARI is more popular and meaningful. There are also instantaneous ARI and ARI with rapid adjustment. The reviewer understands that the authors’ simulation setup cannot derive ARI with rapid adjustment (though they are using WRF-Chem), but all-sky instantaneous ARI can be readily calculated. Impacts of morphology on all-sky ARI should be of interest for readers, too.

Response: Thanks very much for your comments. The all-sky ARI was calculated in the revised manuscript. The cloud properties were not simulated in our WRF-Chem simulations. We use the daily-mean cloud optical thickness, cloud effective radius, and cloud cover products from MODIS for the all-sky ARI calculations.

Comments: The reviewer does not understand why impacts of morphology on EAE and AAE are important. Presentations of impacts of morphology on regional mean (or regional maximum) clear-sky and all-sky ARI should be much more meaningful.

Response: Thanks very much for your comments. The all-sky ARI was calculated in the revised manuscript, and the regional ARI was also calculated using the mean optical properties. The ARI in typical sites are still shown for comparison with previous studies, but the deviations of ARI between non-spherical BC and spherical BC were deleted, as we agree that presentations of impacts of morphology on regional mean clear-sky and all-sky ARI are much more representative and meaningful.

Specific comments:

Comments: Abstract: many important information is missing: “simulation period: season and duration”, “clear-sky”, “external mixture assumption”.

Response: Thanks very much for your comments. We have added the information in the revised manuscript, we have re-written the abstract:

“Black carbon (BC) is one of the dominant absorbing aerosol species in the atmosphere. It normally has complex fractal-like structures due to the aggregation process during combustion. A wide range of aerosol-radiation interactions (ARI) of BC has been reported throughout experimental and modeling studies. One reason for the large discrepancies among multiple studies is the application of the over-simplified spherical morphology for BC in ARI estimates. In current climate models, the Mie theory is commonly used to calculate the optical properties of spherical BC aerosols. Here, we employ a regional chemical transport model coupled with a radiative transfer code that utilizes the non-spherical BC optical simulations to re-evaluate the effects of particles' morphologies on BC shortwave ARI, and the wavelength range of 0.3 - 4.0 μm was considered. Anthropogenic activities and wildfires are two major sources of BC emissions. Therefore, we choose the typical polluted area in eastern China which is dominated by anthropogenic emissions, and the fire region in the northwest US which is dominated by fire emissions in this study. A one month-simulation in eastern China and seven-days simulation in the fire region in northwest US was performed. Compared to the spherical BC model, the fractal BC model generally presents a larger clear-sky ARI. Assuming BC particles are externally mixed with other aerosols, the relative differences in the time-averaged clear-sky ARI between the fractal model with a fractal dimension (D_f) of 1.8 and the spherical model are 12.1% - 20.6% and 10.5% - 14.9% for typical polluted urban cities in China and fire sites in northwest US, respectively. Furthermore, the regional-mean clear-sky ARI is also significantly affected by the BC morphology, and relative differences of 17.1% and 38.7% between the fractal model with a D_f of 1.8 and the spherical model were observed in eastern China and the fire region in northwest US, respectively. However, the existence of clouds would weaken the BC morphological effects. The time-averaged all-sky ARI relative differences between the fractal model with a D_f of 1.8 and the spherical model are 4.9% - 6.4% and 9.0% - 11.3% in typical urban polluted cities in eastern China and typical fire sites in northwest US, respectively. Besides, for the regional-mean all-sky ARI, the relative differences between the fractal model and the spherical model are less than 7.3% and 16.8% in the polluted urban area in eastern China and the fire region in northwest US, respectively. The results imply that current climate modeling may significantly underestimate the BC ARI uncertainties as the morphological effects on BC ARI are ignored in most climate models. ”

Comments: Lns. 53-56, WRF-Chem, FlexAOD, and libRadtran: reference is missing.

Response: Thanks very much for your comments. We have added the references in the revised manuscript.

Comments: Section 2 and 3 should be combined to one section, “Method”.

Response: Thanks very much for your comments. We have combined Section 2 and 3 into one section, “Method”, in the revised manuscript.

Comments: Sect. 2: Which meteorological analysis used for the simulation of China?

Response: Thanks very much for your comments. We are very sorry for without clearly clarifying meteorological analysis. For both the simulations in East China and North America, the National Center for Environmental Prediction (NCEP) Global Forecast System's final gridded analysis data set was used to provide the meteorological initial and boundary conditions. The chemical initial and boundary conditions were obtained from the Model for Ozone and Related Tracer, version 4 (MOZART-4). We have clarified it in the revised manuscript.

Comments: Lns. 125-128: The reviewer does not fully understand why size distribution of WRF-Chem is not directly used for the optical and radiative transfer calculations.

Response: Thanks for your comments. In this revised manuscript, the internally mixing assumption was assumed. Thus, the size distributions of aerosols in WRF-Chem were for the total mixed aerosols, but not BC. In this work, as the first step for using the non-spherical BC in estimating the ARI, we just consider the externally mixed BC, and the internally mixed BC would be considered in the future. Thus, we didn't use the size distributions in WRF-Chem.

Comments: Sect. 3: Equations 4-10 are too general and thus you don't need to describe them in the paper. Rather, descriptions or equations describing how to directly calculate the optical properties of fractal agglomerates by MSTM should be elaborated in this section.

Response: Thanks for your comments. With the refractive index, wavelength, input position file, which includes the positions and radius of spheres, the MSTM can output the extinction efficiency (Q_{ext}), scattering efficiency (Q_{sca}), and phase function (P), and the extinction cross-section (C_{ext}) and scattering cross-section (C_{sca}) were further calculated using Equations 4-5. We have added some descriptions:

"The MSTM can efficiently calculate the optical properties of spheres without intersecting surfaces. The MSTM has high computational efficiency because it theoretically calculates the optical properties of randomly oriented particles without numerically averaging them over different particle orientations. the MSTM can output the extinction efficiency (Q_{ext}), scattering efficiency (Q_{sca}), and phase function (P) with the refractive index, wavelength, input shape file."

We did not delete Equations 4 -10, as it show how the bulk optical properties of non-spherical BC were calculated.

Comments: Ln. 143: What are "the pmom code"? Avoid model-specific terms in a paper.

Response: Thanks for your comments. The pmom is a tool available in Libradtran for calculating the Legendre moments. The inputs of pmom are the aerosol phase function and the desired number of Legendre moments. In the revised manuscript, we have added some descriptions of this tool:

"In this work, we used the pmom tool which is available in libRadtran software for calculating the Legendre expansion coefficients. With the inputs of the aerosol bulk phase function and the desired number of Legendre expansion coefficients, the pmom tool can calculate the Legendre expansion coefficients."

Comments: Ln. 158: FlexAOD

Response: Thanks for your comments. We have corrected it in the revised manuscript.

Comments: Ln. 200: What do you mean by "standard atmosphere background"? Instead of using standard atmosphere, the authors should use the atmospheric conditions predicted by WRF.

Response: Thanks for your comments. In principle, we should use the atmospheric conditions predicted by WRF. However, this work mainly aims to investigate the effects of BC morphology on ARI, so we use a representative atmospheric profile to eliminate the perturbs of other factors. As the ARI was calculated by the difference between the fluxes with aerosols and without aerosols, the effects of atmospheric conditions should have small impacts on ARI. Thus, we just use the standard atmosphere background. However, after carefully checking the calculations, we found that we have made a mistake in the previous study (we have mistake the aerosol optical properties at the top layer with those at bottom layer). Thus, we have re-conducted the calculations.

Comments: Ln. 201: Double periods ".."

Response: Thanks for your comments. We have corrected it in the revised manuscript.

Comments: Ln. 212, "PM2.5": 2.5 is lowercase here and elsewhere.

Response: Thanks for your comments. We have corrected it in the revised manuscript.

Comments: Ln. 217, It is a very good idea to compare simulated AOD and AAOD against AERONET in Beijing. Why not other sites in China and US, rather than to compare surface $PM_{2.5}$ only?

Response: Thanks for your comments. AERONET sites in our simulation area are rather limited, and other AERONET data for the other site is not available, so we just compare the simulated AOD and AAOD against AERONET in Beijing, and we used the $PM_{2.5}$ comparison for the supplements to show the reasonable predictions. We have clarified it in the revised manuscript.

Comments: Ln. 224, "400 ug/m3": it seems the nighttime concentration which does not affect ARI. Please show shortwave and longwave ARI, separately. Longwave ARI could be negligibly small. You may see the phrase "which should have a strong impact on the aerosol radiative effects" is totally wrong. Also, it is just a surface concentration, but the column amount matters for ARI. Reorganize the discussion here.

Response: Thanks for your comments. In this work, just shortwave ARI was considered, and the wavelength range was in the range of 0.3 um – 4 um. However, Longwave ARI should be negligibly small. We have clarified it and re-written the sentence in the revised manuscript:

"As shown in Figure 3, the temporal BC concentrations at fire sites can even exceed approximately 400 $\mu g/m^3$ when the fire occurs, while the BC concentrations are extremely low in other days"

Comments: Acknowledgement: please remove FlexAOD here because code availability is in different section.

Response: Thanks for your comments. We have removed FlexAOD here.

Comments: Figures: please clearly state if the authors use UTC or local time for all time series panels.

Response: Thanks for your comments. We have clarified the UTC times in the revised manuscript.

Comments: Caption of Fig. 12: probably EAE, not AAE. Probably not lambda=450-850 nm, 850 nm pair but lambda = 450 nm, 850 nm pair.

Response: Thanks for pointing it out. We have corrected it in the revised manuscript.

Comments: Code availability: code availability should be also stated for WRF-Chem, libRadtran, and MSTM.

Response: Thanks for your comments. We have stated the code availability for WRF-Chem, libRadtran, and MSTM.

Comments: Data availability: "athour"-> "author". The statement "the data can be requested from the corresponding author" may not be allowed by ACP.

Response: Thanks for your comments. We have corrected "author" in the revised manuscript. Besides, we have made the ARI data available in the revised manuscript.

Comments: Table S1: please remove (mp_physics), …, (bl_pbl), as those are model-specific terms. Explain acronyms, RRTMG and YSU. Better to include references of each option.

Response: Thanks for your comments. We have corrected it and added related references in the revised manuscript.

Comments: Table S2: avoid model specific terms. What do a01, a02, a03, and a04 indicate? If it indicates size bins, define the sizes. Same for Table S3. What are those acronyms, for example, orgalk1j?

Response: Thanks for your comments. We have revised Table S2 and Table S3 in the revised manuscript.

Comments: Fig. S1: boarders (national, province, land/ocean) and symbols are hardly legible. Probably, better to use "white" color for tiny values, instead of "blue".

Response: Thanks for your comments. We have replotted the figures, as shown in the following:

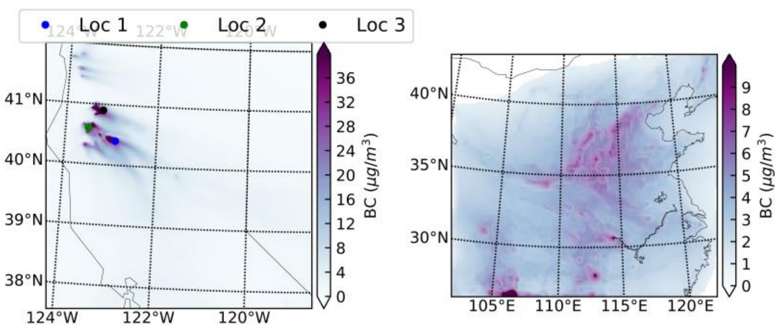

Figure 1 the BC concentrations in different regions.