Response to the comments of Reviewer #1

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.

The authors investigated ARI of BC and some other radiative properties with a modified BC-model, as the fractal-like structure is more realistic. The authors found some biases in the traditional sphere BC models in AOD, AAOD and AAE. For ARI, the more realistic BC model produces higher ARI forcing compared with sphere model. The method of this study is solid and the results are well presented, providing some new insights of BC ARI to the community.

However, this paper needs some revisions before the acceptance for publication.

Major comments:

Comments: It looks like a *discussion* section is missing. A more detailed discussion is needed (e.g., limitations, interpretation of the results, comparison with previous studies).

Response: Thanks very much for your comments. In the revised manuscript, a discussion section is added, and the contents are shown in follows:

"In current climate models, such as CESM, MIROC-SPRINTARS, and WRF-Chem, the Mie theory was commonly used to calculate the optical properties of BC aerosols. However, fractal-like BC aerosols were often observed in the atmosphere. In this work, we found that the effects of BC morphology are spatially-dependent. Compared to the spherical BC model, the fractal BC model generally presents a larger clear-sky ARI, which may lead to the underestimations of BC ARI in the climate models. The relative differences in the time-averaged clear-sky ARI are 12.1% – 20.6% and 10.5% – 14.9% in typical polluted urban cities and fre sites, respectively. Furthermore, the regional-mean clear-sky ARI is also signifcantly affected by the BC morphology, and relative differences of 17.1% and 38.7% between the fractal model were observed in eastern China and in the northwest US, respectively, while the existence of cloud would weaken the BC morphological effects. The results imply that current climate modeling may signifcantly underestimate the BC ARI uncertainties as the morphological effects on BC ARI are ignored in most climate models. However, this work is by no means exhaustive. This work assumed that BC aerosols are externally mixed with other chemical components, while BC aerosols are often internally mixed with other components, such as organic aerosols, sulfate, etc (China et al., 2013a; Adachi et al., 2010; Wang et al., 2021b). BC absorption can be signifcantly enhanced by the "lensing Effect" even if BC aerosols are internally mixed with non-absorbing materials, which may lead to larger BC ARI (Chung et al., 2012; Liu et al., 2017a). Previous studies have shown that the morphologies of internally mixed BC would signifcantly affect its absorption enhancement (Luo et al., 2019; Wang et al., 2021a; Luo et al., 2021c), so lead to larger uncertainties in the estimation of BC ARI. Thus, the sensitivities of BC morphologies on the ARI estimated in this work may be smaller than those in real cases.

Futhermore, we found that the spherical assumption generally unerestimates the clear-sky ARI for externally mixed BC, hile oppsite phenomenon may be found for internally mixed BC. A core-shell spherical morphology was widely used to epresent the internally mixed BC. However, many partially coated BC aerosols exist in the atmosphere, while the coreshell spherical BC model commonly assumes the BC is fully embedded in a coating shell. The core-shell morphology may verestimate the absorption of partially coated BC (Wang et al., 2021a; Zhang et al., 2018), so overestimate the ARI. Thus, the RI of internally mixed BC with complex morphologies should be further investigated in the future.

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Comments: I was wondering in the current generation of GCMs or ESMs such as CMIP5/6, is the BC model sphere or fractal-like? Or maybe some of them are sphere/fractal. I suggest authors provide some more information on this in the *introduction* section. If most of the current models use simplified sphere BC models, then the contributions of this study would be more significant and the authors should add some discussions in the *discussion* section.

Response: Thanks very much for your comments. In the current models, the spherical BC model is used, and we have added some descriptions in the introduction:

An important cause of the discrepancy is BC’s complex morphology. BC aerosols are assumed as spheres, and the optical properties are calculated using the Mie theory in most climate and atmospheric chemical transport models, such as Community Earth System Model (CESM) (Danabasoglu et al., 2020), the Model for Interdisciplinary Research on Climate (MIROC) (MIROC-SPRINTARS) (Takemura et al., 2005, 2009), Weather Research and Forecasting coupled to Chemistry (WRF-Chem) (Grell et al., 2005; Fast et al., 2006), and GEOS-Chem.

Besides, some discussion was added in the revised manuscript, as shown above.

Comments: In line 254, the authors provided an explanation why different structure may lead to different AAOD. However, such explanation is missing in some other analyses. I suggest the authors add similar explanations like line 254 in the descriptions of other results (e.g., why fractal structure produces higher ARI, maybe more solar radiation is reflected by sphere-structure?).

Response: Thanks very much for your comments. We have added some explanations like line 254. The reason is that the fractal structure can absorb more light, but the total extinction is not significantly modified. We have added some descriptions in the revised manuscript:

"As explained above, the fractal BC can absorb more light than spherical BC, while the total scattering is not significantly modified. Thus, the fractal BC leads to larger positive clear-sky ARI."

Comments: The authors cited several BC forcing values at the beginning of *section 4.4*. Is there any value could be used to compare with the simulation from this study? There are four ARI values for each location in this study, is there any value that is more realistic?

Response: Thanks very much for your comments. The ARI in previous studies were also simulation results, and the ARI is difficult to be directly measured. Thus, it is difficult to say which value is more realistic. In this work, we compare the simulation with previously reported values just to show that our simulations generally agree with previous studies, but not to show which value is more realistic. Our main aim is to investigate the sensitivities of ARI to the BC morphology, and which model is more realistic for reproducing the real ARI should be investigated in the future.

Comment: It is confusing to see the "relative variations" of 10.4%-15.3% in the *abstract*. What is the relative variation? Day-to-day variation? Please define it. In *section 4.4*, there is "relative uncertainty", are they the same? In the *conclusions*, it is switched to "relative variations" again.

Response: Thanks for pointing it out. In the revised manuscript, we have re-written these sentence in the revised manuscript.

Minor comments:

Comments: The writing needs some polishing (e.g., Line 48, contribute to…)

Response: Thanks for your comments. We checked the English in the revised manuscript, and all the modifications are marked in the revised manuscript.

Comments: Line 95, with a larger D*f*?

Response: Thanks for pointing it out. It is indeed "with a larger D_f", we have corrected it in the revised manuscript.

Comments: Figure 3, lower panel, the four lines are overlapped. You may try to use thicker lines underneath and use thin lines above to make them clearer. It is the same for Figure 6 and 8.

Response: Thanks for your comments. We have replotted the figures in the revised manuscript.