Response to Referee #1

We sincerely appreciate the Referee #1's comments, which indeed help use to improve the manuscript. We accept all the suggestions proposed by the Referee and revise the manuscript accordingly. Please see the point-point responses to the comments in the following.

Referee: "One global model is used to study the impact of changes in anthropogenic aerosol emissions in China on regional radiation budget. The authors found that, compared to 2008, reductions in black carbon and sulfur dioxide emissions in 2016, separately, had an opposite effect on radiative forcing, resulting in a net radiative forcing of -0.04 W/m2 by the changes in black carbon and sulfur dioxide emissions. The main scientific conclusion of this study is by no means novel. A number of studies in the literature have shown similar conclusions over China, East Asia and/or other regions around the world. In addition, I have several major specific concerns regarding the methodology and model biases as described below"

Response: Accepted. We improve the description of the novelty and the scientific implications of this study in the new text. The novelty mainly includes:

1) The clean-air measures in China led to the heaviest-ever reductions in anthropogenic emissions over the past decade (please see Table 1 in our manuscript), which provides a unique opportunity to investigate the aerosol radiative forcings due to such substantial variations in emissions. We quantified the aerosol-induced radiative forcings at TOA due to both aerosol-radiation interactions and aerosol-cloud interactions over China and the outflow regions (North Pacific) using a global climate model and the latest China's anthropogenic emission inventory (replacing CMIP6 emissions for China). To our knowledge, no modeling studies have shown such results before. Please see Line 60-77 in the revised manuscript (Unless noted otherwise, line numbers are shown for the revised manuscript).

Our results will implicate for predicting climate impacts of emission variations under different SSP (shared socio-economic pathways) scenarios in the future (e.g., Samset et al., 2019). Please see Section 3.3 (Line 320-337).

2) Recent studies mainly emphasized the radiative forcings of SO_2 emission variations over China in the past decade (Fadnavis et al., 2019), but didn't uncover their

interactions with BC and other aerosol species (e.g., nitrate) in determining the net aerosol radiative forcings. Our results suggest that the reductions in SO_2 emissions and the associated sulfate concentrations would facilitate the formation of ammonium nitrate and also diminish the radiative forcing of BC-containing particles through the absorption enhancement effect. These complex effects should be considered in the assessment of aerosol radiative forcings. Please see Line 257-264.

3) This study indicates that the reductions in sulfate and BC led to the increase in downward solar radiation flux onto the Earth's surface. The results provide the first modeling evidence for the cause of surface brightening in China over the past decade, as shown by the long-term observations (Schwarz et al., 2020). Please see Line 301-319.

Major comments

Referee: "1. "the recent decade" in the title and abstract (and a few other places in the main text) should be changed to the exact time period "2008-2016". Why are these two specific years chosen? Many published studies on aerosol radiative forcing use 2014 as "present day". On the other hand, the simulations were only performed for two years with one year discarded and the results were mostly presented as the difference between the two time slices. I don't think the use "trend" in the description of discussion of results is appropriate."

Response: Accepted. We use "2008–2016" in the revised title.

We chose these two years for analysis because the differences of emissions between these two years (2008 and 2016) can reflect the decadal changes in China's anthropogenic emissions caused by a series of emission control policies from 2008 to the present. The SO_2 emissions in China peaked around 2007 and have declined since then (Li et al., 2017). And the toughest-ever clean-air policies were conducted between 2013–2017 and led to further reductions in primary aerosol emissions (Zhang et al., 2019). We added more description on the study period in the revised manuscript. Please see Line 79-83.

We revise the statement with the use "trend" in the main text. Some trend results from previous studies and long-term observation data have been shown to support our simulation results. Please see Line 162-183 and Fig. S6.

Referee: "Given the CAM experiment design, comparison of model results in 2016, which is based on meteorological conditions in 2008, to observations is not apples-to-apples. Meteorology is probably quite important for interannual variability in aerosol concentrations (e.g., Yang et al., 2018). Please discuss how this setup could affect your conclusions."

Response: Accepted. We performed another simulation using both the emissions and meteorology fields for 2016 and discussed how meteorology conditions affect aerosol concentrations and the associated radiative effects. Please see Line 338-346 and Table S4 in the revised manuscript and SI.

Both this study and previous studies demonstrate that the substantial reductions in anthropogenic emissions dominated the mitigation of aerosol pollutions in China in the past decade (Liu et al., 2020; Zhang Q. et al., 2019; Zhang X., et al., 2019). Variation in meteorological conditions only has a minor effect on the decadal variability in aerosol concentrations and their radiative effects due to aerosol-radiation interactions over China (Table S2).

Note that the aerosol-induced cloud radiative forcing cannot be separated from the changes of meteorological fields. This study mainly focuses on the impacts of inter-annual variations in anthropogenic emissions on aerosol radiative effects and meteorological factors are not the key topic of this study.

Referee: "2. I also have a concern about the current experimental design and forcing estimates, which cannot be used to support one of the main conclusion (i.e., the net BC and sulfate radiative forcing of -0.04 W m-2). Forcing induced by BC emissions reduction and forcing induced by SO2 reduction are diagnosed in separate simulations with one of the individual aerosol changes included, assuming that the forcings induced by the individual aerosol changes are additive. Results in the current study and some previous studies (e.g., Chen et al., 2017) have shown nonlinearities between emission and forcing changes even for the same species."

Response: In this study, the forcings induced by BC and SO_2 emissions were calculated online in the same simulations (i.e., differences between Exp16 and Exp08). The separate simulations (mentioned by the referee) with one of the emission change included were only used for comparison instead of drawing the main conclusion. Therefore, the nonlinearities between emissions and forcings have been considered in our study. We add more description for the calculation of aerosol forcings. Please see Line 103-115.

Referee: "On the other hand, the CRF in this study is calculated by scaling results of different simulations with the best estimate of effective radiative forcing in IPCC AR5, which needs to be described more clearly. I am not sure how exactly this was done and why this number is comparable to the DRF calculated in this study. As far as I know, the AR5 effective radiative forcing is calculated differently from the method of Ghan (2013). The latter attributes radiation changes induced by above-cloud light absorbing aerosols to DRF."

Response: Accepted. In this study, we scaled the global-mean CRF (aerosol effects on cloud forcings) calculated in our model to the best estimate of the effective radiative forcing due to aerosol-cloud interactions (-0.5 W m^{-2}) given in Chapter 7 of IPCC AR5 report (Boucher et al., 2013), which makes our CRF results consistent with the current best estimate of CRF. We do not scale the DRF estimate in this study. The DRF is online diagnosed in the model using the method of Ghan (2013) that attributes the above-cloud light absorbing effects by aerosols to DRF. We reword the description for the calculation of CRF in this study. Please see Line 116-126.

Referee: "3. It looks to me that the CAM AOD is so much off comparing to MISR (Figure S2), especially over the continental outflow regions and the high latitudes. A difference plot would be able to show the biases more clearly. There are various studies in the literature that have shown improved aerosol simulations in CAM5 (e.g., Ghan et al. 2016 and references therein). Why aren't those improvements included in this version of the CAM model? Model biases in AOD as well as the vertical distribution of aerosols and how they can impact the forcing estimates in the two regions of interest need to be quantitatively evaluated and more objectively discussed."

Response: Accepted. We add more discussions on the model bias in AOD, compare the vertical distribution of modeled BC concentrations with aircraft measurements, and evaluate the uncertainties in the forcing estimates. Here, we point out that:

1) A difference plot between modeled and observed AOD from MISR and MODIS have been given in the Supplementary Materials (Figure S5). The model biases in AOD over some continental outflow regions and high latitudes (mainly in Southeastern Asia and East of Africa) are commonly found in CAM models (Sockol and Griswold, 2018; He et al., 2015), even if improvements for aerosol simulations have been included. It's quite difficult to quantitatively evaluate the model biases of radiative forcing estimates, but a qualitatively description of the potential bias related to AOD has been added in the revised manuscript. Please see Line 225-236 and Line 274-279.

2) The aerosol simulations have been markedly improved by our group based on standard CAM models (Matsui 2017; Matsui and Mahowald, 2017). Our model uses an advanced two-sectional bin representation of size and mixing states of BC particles, which calculates detailed aerosol processes and their interactions with radiation and clouds. We also include a new pathway of secondary sulfate formation as well as an improved volatility basis-set approach for secondary organic aerosols. Our simulations can generally reproduce the spatiotemporal features of major aerosol compositions over China and outflow areas (Figure 2, Figure S3, and Table S2). In our understanding, the paper mentioned by the referee, i.e., Ghan et al. (2016), does not discus improvements of aerosol simulations.

3) Because the observations of vertical profiles of sulfate and BC for a long period (> 1 month) are quite limited in China mainland, we evaluate the model performances of BC profiles in the continental outflow region (i.e., North Pacific) using the aircraft measurements from the HIPPO campaign. Please see Figure S1 and Line 183-186 in the revised manuscript.

Minor comments and technical edits:

Referee: "Line 57-59: I believe there are quite a few other studies in the literature that are relevant here."

Response: Accepted. We add relevant literatures in the revised manuscript. To our knowledge, very few studies have focused on the aerosol radiative forcings in response to the substantial emission variations in China over the past decade. Please see Line 61-72.

Referee: "Line 71: CAM5 should be spelled out as "Community Atmosphere Model version 5"

Response: Accepted. We reword this sentence. Please see Line 88.

Line 80: Change "simulation" to "simulating"

Response: Accepted. Please see Line 90.

Line 108: How about global emissions for 2016? No changes in emissions of other regions? The CEDS emissions only went up to 2014.

Response: Accepted. The global emissions other than China used in all simulations are from the CEDS for 2008, because we focus on impacts of the China's emission variations on aerosol radiative effects. We add more description for emissions. Please see Section 2.2.

Line 127-128: Why MISR AOD is used here rather than other remote sensing or hybrid products that are often used by the aerosol modeling community to evaluate models.

Response: Accepted. Because the inter-annual variations in observed AOD over China in the recent decade are consistent between MISR and other remote sensing products, such as MODIS (shown in J. Li, 2020), only MISR AOD was used in the original manuscript. We add the spatial maps of MODIS AOD for 2008 and 2016. The detailed values of observations and the comparison with simulations have been given in the main text and supplementary figures. Please see Figure S4 and Figure S5 and Line 211-221.

Line 164-165: this sentence needs clarification. Do you mean less ammonium sulfate leads to more ammonium nitrate over East China in 2016? Assuming that all else being equal?

Response: Accepted. In the case with only SO_2 emission variation included (Exp16SO2), nitrate concentrations increase by almost the same level with those in the 2016 case (Exp16, with changes in all emission variables). Therefore, the reductions in sulfate have led to elevated particulate nitrate concentrations during 2008–2016, given that NH_3 emissions are unchanged. This is caused by the aerosol thermodynamic equilibrium in the sulfate-nitrate-ammonium-water system (Chapter 10 in Seinfeld and Pandis, 2016). We add more sentences to explain the increase of nitrate concentrations resulted from the reductions in sulfate. Please see Line 191-199.

Figure 4: It's very difficult to see the AERONET observations in the figure. Is there only one site that can represent East Asia? I don't see much value added here.

Response: Accepted. We modify Figure 4 to show the location of the AERONET station more clearly. There are some AERONET stations located in China, but only Beijing and Xianghe stations have long-term data available from 2008 to 2016 (also used by Li, 2020). Because these two sites correspond to the same model horizontal grid, we use the averaged value from them to show the long-term variation of AOD for the region. Please see Line 239-243 and Figure 4.

Figure 6: How was the total RF calculated? How does this compared to the TOA forcing difference between Exp08 and Exp16?

Response: Accepted. The total RF induced by the aerosols is calculated by combining the aerosol DRF and CRF at TOA derived from the Exp08 and Exp16 cases. The CRF has been scaled based on the IPCC AR5 report. Please see Line 289-292 and the Figure 7 caption. We understand that the phrase "TOA forcing difference" mentioned by the referee here means the regional-mean change in the net radiation flux at TOA between the two cases. It is estimated to be +0.46 W m^{-2}, quite close to the sum of aerosol DRF and CRF

(+0.50 W m^{-2}). The CRF mentioned here is the original estimate by the model without a scaling.

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