Response to Referee #2

Thanks very much for the Referee #2's helpful comments. We revise our manuscript according to the annotated document. Please see the point-to-point responses in the following.

Referee: "I recommend a more focused title, rather than a whole sentence."

Response: Accepted. The title has been changed to "Aerosol radiative forcings induced by variations in anthropogenic emissions over China during 2008–2016".

Referee: "result->resulted; latest->the latest"

Response: Accepted. We reword them. Please see Line 6-10 in the revised manuscript.

Referee: "Line 25: This phrase is a non-sequitor with respect to what is given above. Is an aerosol-induced positive radiative forcing truly desirable?"

Response: Accepted. Because aerosol radiative forcings induced by anthropogenic emissions potentially influence surface temperature and precipitation, we imply that targeted regulations of emissions are required to mitigate the risk of climate change. We reword the sentence to highlight implications of this study. Please see Line 26-28.

Referee: "Line 60: the->a; don't->do not"

Response: Accepted. We modify them. Please see Line 67-68.

Referee: "Line 72: A verb is missing from this phrase."

Response: Accepted. We reword the sentence. Please see Line 90.

Referee: "Line 88: ... species for years 2008 and 2016 for...."

Response: Accepted. We reword the sentence. Please see Line 105.

Referee: "Line 91: Also for all the 2016 simulations? what is the reasoning behind that? unless you used the proper 2016 meteorology for the 2016 simulations and the phrase is simply misleading. Line 96: As long as you used the 2008 meteo for the 2008 run and the 2016 meteo for the 2016 run. Otherwise, you cannot separate anything, I think."

Response: Because change in meteorological conditions (clouds, winds, etc.) could also influence the calculation of aerosol radiative effects, we need to separate the contribution of emission variations on aerosol radiative effects by using the fixed meteorological fields from one fixed year. Such kind of separation is commonly adopted in previous studies on evaluating aerosol climate effects due to emission variations for a long-term period (e.g., Leibensperger et al., 2012; Fadnavis et al., 2019).

We add more description for the treatment of meteorological fields in this study and perform a sensitivity simulation to discuss the effects of meteorological conditions on aerosol radiative forcings. Please see Line 113-115, Line 338-346, and Table S4.

Referee: "Table 2: I think you need to define with lat/lon boxes what EC is and also you need to add the reference to the emission inventory in the table legend."

Response: Accepted. We add the definition of EC and the reference to the emission. Please see the Table 2 caption.

Referee: "Line 123: Is this data public? give a website and a reference to this data. Have they been used before in studies? have they been validated? you should make this clear before you use them to validate your model runs."

Response: Accepted. The EANET databases have been widely used for air pollution studies and model evaluation of inorganic aerosol components (e.g., Itahashi et al., 2018). The program has adopted the regular Quality Assurance/Quality Control. Details can be

found at: https://www.eanet.asia/. We add more description for the database. Please see Line 140-144 and Data availability.

Referee: "Line 123: including->include."

Response: Accepted. Please see Line 143.

Referee: "Line 123: From the text above the impression given is that you run your model only for the Chinese domain. Do you mean you also run for Japan? I think a plot showing the model domain with a species of your choice would be a nice addition to your section 2.1. You should also address the issue whether the chosen emission inventory, species concentrations, etc. that apply to China also apply to Japan."

Response: Accepted. The climate model, CAM5/ATRAS, was ran on a global scale in this study. The model configuration is consistent among different regions. The emission inventory used for China is from MEIC and the rest of the world from CMIP6. Please see Line 103-107 and Section 2.2.

Referee: "Line 128: Which version of the data did you use? downloaded from where? did you regrid? you need to provide references for this data and discuss their validation. How do they compare for e.g. against MODIS, which is the more established satellite dataset?"

Response: Accepted. The version and download source of MISR data have been included in the revised manuscript. Further, we add the MODIS AOD data for comparison with the model. The differences between modeled and observed AOD from MISR and MODIS are shown in the Supplementary Materials. The detailed comparison between MISR and MODIS has been provided by Garay et al. (2020) and references therein. Please see Line 155-159 and Figure S4 and S5.

Referee: "Line 132: This agreement is not shown by Figure 1a/b. You need a different representation, either a line plot, since your stations are point locations or a table with numbers.

You also have to be very clear as to how you handdled the ground-based measurements. You are comparing a point observation from the ground with a model with moderately big pixels. How did you deal with representativeness errors? SO2 has a very clear seasonal cycle, how does that apply to your comparisons? do you create monthly mean values from the observations? for which time of the day? There are many choices that can affect your comparisons and those need to be made clear in the text, in Section 2.3."

Response: Accepted. We demonstrate that our model can represent the inter-annual variations in aerosol concentrations on a regional scale for 2008–2016 by comparing with available ground-based and satellite observations as well as previous studies.

1) We add the quantitative comparison between modeled and observed sulfate concentrations (Table S2). Please see Line 162-176, Table S1, and Table S2.

2) More details on the use of ground-based observation data are added in the methods section and supplementary materials. Because monthly observation data for aerosol components are very limited and not publicly available in China (particularly for 2008), we average the data derived from different measurement campaigns and compare them with the annual-mean simulation results extracted from the model horizontal grid closest to the observation sites. Please see Line 140-147, Table S1, and Table S2.

3) Although monthly observations of sulfate are not available, we validate the model performance of SO_2 seasonality using satellite-based SO_2 column measurements by OMI. The model presents a consistent seasonal pattern of SO_2 concentrations with satellite retrievals in China. Please see Figure S2.

4) It's clear that both the observations and simulations have demonstrated notable decreases of sulfate concentrations over East China, as shown in Figure 1 and Table S2. Representation errors of in-situ observations are possible, but they could not change the temporal variations of sulfate concentrations, which are highly dependent on regional emissions in China (Zheng et al., 2015).

Referee: "Line 135: Again, this agreement is not demonstrated and the findings of Liu et al. should be stated here in numbers."

Response: Accepted. We add the detailed result given by Liu et al. (2018). Please see Line 171-172.

Referee: "Line 138: Again, you need to explicitly give the findings of Li et al. that actually agree with your 10microgram m-3 finding."

Response: Accepted. We show the findings of Li et al (2017) to support our results. Please see Line 166-169.

Referee: "Line 141: What percentage of the wind is downwind per annum?"

Response: Accepted. The long-range transport of aerosols in China is dominated by the westerly winds, which are prevailing winds from the west toward the east in the middle latitudes between 30 ℕ and 60 ℕ (Liang et al., 2004). Because our simulations were conducted on a global scale, the quantitative impacts of China's emission variations on the aerosol distributions in different downwind regions can be seen clearly in Fig. 2. We improve the sentence. Please see Line 175-176.

Referee: "Line 142: verity->verify."

Response: Accepted. We modify this word. Please see Line 177.

Referee: "Line 146: You cannot find a trend by two years only. If you had all the years between 2008 and 2016, i.e. 9 years, then you might be able to claim this. Your simulations show a difference between two years, this is not a trend."

Response: Accepted. The results mentioned here are derived from the simulations in 2008 and 2016. Nevertheless, the variations of BC and sulfate in the model are in line with exiting observations, which have demonstrated the decreasing trends over the past decade (Liu et al., 2018; Kanaya et al., 2020). We reword the sentence. Please see Line 177-183.

Referee: "Line 147: You need to mark this island for those of us who do not know where it is and show in a figure this decline. Again, if you are showing results from two years only this is not an "interannual" decline, this is a decline between two years."

Response: Accepted. The decline of BC concentrations in Fukue island between 2008 and 2016 has been noted in Figure 1c, d. The long-term observations in Fukue island show a significantly decreasing trend in BC concentrations (Kanaya et al., 2020) and support our simulation results.

Referee: "Line 150: This fact was not made clear in a satisfactory manner to the audience. You need more precise statistics."

Response: Accepted. As suggested by the referee, we add more detailed statistical data for the comparison between simulations and observations as well as previous studies. We also reword this sentence. Please see Table S3, Line 162-190.

Referee: "Figure 1: You mean, the locations of the EANET stations are shown, not the actual numbers these provide. Replace literatures with literature."

Response: Accepted. We replace the word literature used here. The observed aerosol concentrations (the values) at EANET stations are shown in Figure 1. The inner colors of each circle represent the magnitude of aerosol concentrations, which are overlaid on the map of simulation results. The detailed location information of EANET stations is given in Table S1.

Referee: "Figure 2: This is not a map of China, this is a global map. Does this mean you run your simulations for the entire globe? this should be made clear in Section 2.1."

Response: Accepted. We run the global climate model on a global scale. Please see Line 103-104.

Referee: "Line 162: Not shown clearly. Impossible to tell what colour the dots have."

Response: Accepted. The comparison of simulated nitrate concentrations with observations can be seen in both Table S2 and Figure S3. The observed concentrations are shown in colored dots, which are overlaid on the map of simulations.

Referee: "Line 165: Similar, i.e.? in numbers?"

Response: Accepted. The numbers are added here. Please see Line 196-198 in the revised manuscript.

Referee: "Line 182: Not a trend if only two years are used."

Response: Accepted. We reword the phrase in this section. Please see Line 211.

Referee: "Line 184: This is a very important finding and should be included in the main paper."

Response: Thanks for the suggestion. Because similar findings for the comparison between satellite-observed and modeled AOD have been reported in several previous studies (He et al., 2015; Sockol and Small Griswold, 2017), we only display those results in the Supplementary Materials (Figure S4 and S5).

Referee: "Line 187: This can be due to a million different reasons. You do not give an std, maybe the respective errors compensate for this difference. Also, how exactly did you calculate the annual mean AOD from the two sets of information? did you collocate on a daily basis, then created the monthly and then the annual? another way? these simple choices may affect your numbers greatly."

Response: Accepted. We show the standard deviations (i.e., uncertainties ranges given by the MISR product, please see Garay et al., 2020) and inter-annual trends in AOD values from MISR. The MISR aerosol product at a one-year temporal resolution is used in this study. Please see Figure S6 and Line 155-159 and Line 227-228 in the revised manuscript.

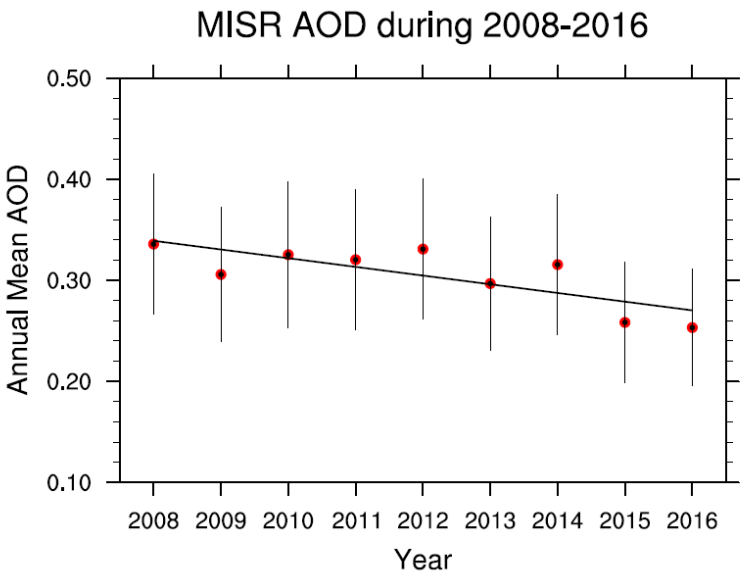

Fig. S6 Inter-annual variation in AOD at 550 nm retrieved by MISR over East China during 2008–2016. The mean values (dots) and standard deviations (vertical lines) are calculated using the gridded AOD and standard deviation from the Level-3 MISR product with a one-year temporal resolution. The black line represents the linear fit of the AOD (T-test: $\alpha=0.013<0.05$).

Referee: "Line 188: How negative? precisely."

Response: Accepted. The numbers are added in the statement. Please see Line 218-220.

Referee: "Figure 4: So, these are the global changes due to the local emission variation? which experiment is this, from Table1? the first?"

Response: Accepted. The changes are derived from the Exp08 and Exp16 simulations. All the experiments are performed on a global scale but only results in China and outflow areas used for analysis. Figure 4 shows the changes in East Asia and continental outflows. Please see Line 103-115.

Referee: "Figure 4: Where are the coloured dots?"

Response: Accepted. We update Figure 4 to show the location of the AERONET station. The detailed values of observations and the comparison with simulations have been given in the main text. Please see Line 237-242.

Referee: "Line 206: That may be the case, but from the material you provide, it cannot be verified."

Response: Accepted. More details on the model evaluation on aerosol optical properties and associated uncertainties are added in the section. We reword this sentence here based on the evidence we provided. Please see Line 222-246.

Referee: "Line 244: You need to show these results."

Response: Accepted. The results have been shown in this sentence. Please see Line 285-287.

Referee: "Figure 5: Why not zoom these maps to show Asia only? more detail would appear and it is clear that the rest of the world is not affected."

Response: Accepted. We zoom out the Figure to underline the variations in Asia and surrounding areas. Please see Figure 5 and Figure 6 in the revised manuscript.

Referee: "Figure 5f: This map is impossibly noisy, neighboring pixels show -1 and 1 Wm-2 forcing, this is not physical."

Response: We would like to point out that the response of cloud radiative forcing to aerosol perturbations is distinguishable from the noise over China and north Pacific (Figure 5f). The aerosol-induced cloud radiative forcing (CRF) is estimated by varying the anthropogenic emissions between 2008 and 2016 in two different simulations. The CRF noise can be caused by the aerosol-meteorology feedback in the global climate model. Such anomalies of aerosol-induced CRF signals are also seen in similar studies (e.g., Shi and Liu, 2019).

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