Response to Referee 1

Referee 1: The authors have addressed most of my previous comments. Some concerns remain:

1) Regarding my comment on the CRF estimates, there is no detailed description on how the scaling to AR5 forcing is done and, more importantly, what the justification is. I don't think the CRF estimated from the simulations in this study can be scaled to the AR5 forcing for the following reasons:

• The AR5 effective radiative forcing is estimated for the global present-day condition, compared to the preindustrial condition, which is different from the contrast between 2008 and 2016 with emission changes only in East Asia.

• I also had a comment about the difference in methodology for cloud forcing estimate between this study (using the Ghan method) and the AR5. The cloud radiative forcings are incomparable even just for East Asia.

• On the other hand, if the emission reductions in China during 2013-2017 had a significant impact on the estimated CRF over East Asia, which was NOT included in the AR5 forcing estimates, how can the AR5 forcing be relevant to the East Asia CRF in this study?

Response: We sincerely appreciate the referee's further comments, as they are helpful for us to improve the manuscript, especially for the description on the calculation of cloud radiative forcings. To address the concerns from the referee, we'd like to show the followings:

1) First, we explain why and how we scaled the aerosol CRF to match the IPCC AR5 estimate.

To compared with the AR5 cloud radiative forcing, we have performed the global simulation using the preindustrial condition (emissions for 1750) and obtained a global and annual mean aerosol CRF (SW+LW) of -2.7 W m⁻² between 1750 and 2008, which is much more negative than the best estimate (-0.5 W m⁻²) as well as the median value of the multiple model results (-1.4 W m⁻²) in the IPCC AR5. Our result is close to but still more negative than the estimate (-1.7 W m⁻²) by another CAM5 model in Ghan et al. (2012).

Such gaps should have been mainly originated from the differences between climate models in processes controlling cloud amounts and lifetime and how aerosols affect cloud albedo (Zelinka et al, 2014). We therefore infer the possibly higher CRF by aerosol reductions over East Asia between 2008 and 2016 in our simulations. A constant scaling factor is derived from the ratio of the global mean CRF in our simulations between preindustrial and present-day conditions to the corresponding AR5 estimate. The scaling can yield a reasonable range of CRF estimates in the context with previous CRF estimates.

(2) As the referee commented, the methodology for cloud forcing estimate is different with the IPCC AR5 because this study followed the methodology in Ghan, 2013, which attributed radiation changes (+0.42 W m⁻² globally) induced by abovecloud light absorbing aerosols to DRF rather than CRF. Here, if we add this term to our global mean industrial-era CRF estimate, it is changed to -2.3 W m⁻² and still more negative than the best estimate in the IPCC AR5.

The best estimate of ERFaci in IPCC AR5 was given by the expert judgement based on existed studies using climate models and observations. The estimation methods and model structures related to clouds and aerosols differed between each other. One of the model estimates was given by Ghan et al. (2012), in which the methodology to calculate aerosol forcings was used in our study. Thus, it is reasonable that we compared the global mean aerosol CRF in this study with the IPCC AR5 estimates.

(3) The global and annual mean CRF for 1750–2008 was used for comparison with the IPCC AR5 result for 1750–2011 to derive the scaling factor. This factor was then used to adjust the CRF estimates for other periods (2008–2016). In another word, the emission changes between 2008 and 2016 cannot influence the scaling factor.

To sum up, we clarify the methodology for CRF estimates (please see Section 2.2) and show the CRF values both with and without the scaling (following the suggestions by another referee) in the revised manuscript. Please see Line 297-311 in Section 3.3.

Referee: 2) The aerosol radiative forcing is largely determined by column AOD and the vertical distributions. Fig. S5 shows that the CAM5 AOD in East Asia and over the North Pacific is lower than both MISR and MODIS observations. The model also has a large bias in simulating the BC profiles (Fig. S1). Please clarify how this would affect the estimated BC direct forcing. How about the sulfate profiles?

Response: Accepted. We add more discussions on the potential causes of model underestimation of AOD temporal changes, and suggest that the model treatments of dust aerosols and biomass burning emissions could be partially responsible for the gap between simulations and observations.

The BC profiles in Fig. S1 reflect the model capability in simulating BC emissions and their transport from the East Asia continental sources to the remote atmosphere. In line with previous studies for the same region (see figures in Schwarz et al., 2013), the differences between the simulated and observed BC profiles in the troposphere are generally within one order of magnitude. We point out how the bias in the profiles may affect the BC DRF in the northern Pacific in the manuscript. Please see Line 187-190 in the revised manuscript.

To our knowledge, observations of sulfate profiles for a long period (>1 month) are not available in China and continental outflows. But we show that the simulated

aerosol extinction profiles from the surface to the upper troposphere are in general agreement with corresponding CALIPSO data in China. Please see Fig. S7 and Line 249-263 in the revised manuscript.

Referee: 3) Please include the information of simulations in the caption of Fig. 1. It appears that the surface sulfate and BC concentrations are unchanged between 2008 and 2016 over regions other than East Asia. This can be very misleading. The other regions probably should be masked out if their emissions are fixed at 2008. Emissions in the surrounding regions, especially in South Asia, has been increasing from 2008 to 2016. As also shown in the MISR and MODIS observations (Fig. S4), AOD in South Asia had a significant increase from 2008 to 2016. The non-local emissions can have an impact on aerosols in East Asia. This raises a concern about the estimated forcing in East Asia.

Response: Accepted. We supplement the information of simulations used in Figure 1. We also point out the probable changes in aerosol concentrations in South Asia between 2008 and 2016 that were not reproduced in these simulations. Please see revised Figure 1 caption.

This study focuses on how the aerosol forcings respond to changes in anthropogenic emissions from China, and the contribution of foreign emissions on the aerosol concentration changes is suggested to be negligible during the recent decade (Yang et al., 2018). We have described the use of emissions. Please see Section 2.3.

Referee: 4) Line 306-319: As brought up previously, I don't think it's ok to provide the forcing trends based on results from two individual years. The emission reductions started from 2013. There is no physical basis to suggest the linear forcing trends. The estimated trends should be removed or changed to forcing difference instead.

Response: Accepted. We change them to forcing differences and reword the related sentences. Please see Line 312-328 in the revised manuscript.

References:

Ghan, S. J., 2013: Technical Note: Estimating aerosol effects on cloud radiative forcing. Atmos. Chem. Phys., 13, 9971-9974.

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

Schwarz, J. P., and Coauthors, 2013: Global-scale seasonally resolved black carbon vertical profiles over the Pacific. Geophys Res Lett, 40, 5542-5547.

Yang, Y., and Coauthors, 2018: Recent intensification of winter haze in China linked to foreign emissions and meteorology. Sci. Rep., 8, 2107.

Response to Referee 3

Referee 3: In this work, the authors simulate the radiative effect of the reduction in Chinese aerosol emissions over the 2008-2016 period. They show that the reduction in aerosol emissions produced a significant reduction in the aerosol burden and radiative forcing. The combination of the SO2 and black carbon emissions reductions produces a larger effect that the effects of the individual components due to interactions between them.

The paper demonstrates some interesting effects, particularly the interaction between different aerosol components in an internally mixed aerosol scheme. Unfortunately, some of the method for calculating radiative forcing values (particularly the cloud component) is unclear, which makes interpretation of some of the results difficult.

(I have been asked to comment primarily on the radiative forcing aspects).

<u>Response</u>: Thanks very much for the referee's helpful comments. We accepted all of them and revised our manuscript carefully. Please see the point-to-point responses below.

Major points

Referee 3: The scaling to the AR5 mean value is unclear - I assume it means that the ERFaci forcing pattern in the model (for the 2008 simulation) is scaled such that the global mean is -0.5 Wm-2. This means that the same scaling factor is applied everywhere? This should be explained clearly as it is important for the interpretation of the results.

Response: Accepted. We detail why and how we scaled the cloud radiative forcings (CRFs).

We have performed a global simulation using the preindustrial condition (emissions for 1750) and obtained a global and annual mean aerosol CRF of -2.7 W m⁻² between 1750 and 2008. A scaling factor is derived from the ratio of this industrialera CRF simulation result to the corresponding best estimate of ERFaci in IPCC AR5 (-0.5 W m⁻²). We then used this globally constant factor (5.4) to scale down the online-calculated CRF in China and northern Pacific regions for 2008–2016.

Details for the scaling method and the results with and without the scaling have been added in the revised text. Please see Section 2.2 and Line 297-311 in the revised manuscript.

Referee 3: I am not convinced that this scaling to the AR5 value for the ERFaci is appropriate. The spatial pattern of the ERF and particularly the cloud component varies significantly between models (e.g. Zelinka et al, JGR, 2014) and between the RFaci and adjustments (e.g. Gryspeerdt et al, ACP, 2020). This means that the ratio of the forcing in the study region to the global mean varies between models and processes. The AR5 estimate is based primarily on observation-based estimates, which themselves have been suggested to underestimate the ERFaci in recent reviews (e.g. Bellouin et al, Rev. Geophys, 2020).

<u>Response</u>: Accepted. The reasons for the scaling are given in the revised text and mainly include:

1) We find that the global and annual mean industrial-era aerosol CRF (-2.7 W m^{-2}) calculated online in our model is more negative than the estimates given in the IPCC AR5 and many other reports, like Zelinka et al. (2014) and Bellouin et al. (2020). The magnitude of this estimate is approximately at the upper bound of the intermodel spread in ERFaci. Not only for the global-mean value, the regional estimate over China ($-20 \text{ to} -10 \text{ W m}^{-2}$) is also more negative than previous studies, like Ghan et al. (2012) and Zelinka et al. (2014). So it is plausible that the online-calculated CRF over China for the 2008–2016 period is too high.

2) The large gap is probably caused by the systematic differences between climate models in physical processes controlling cloud amounts and aerosol effects on cloud albedo (Zelinka et al., 2014). The globally constant scaling factor of 5.4 used in this study is based on IPCC AR5 and may vary if we used other model results as reference. Since the IPCC AR5 estimate has been suggested to be underestimated (mentioned by the referee and Gryspeerdt et al, 2020), the CRF calculations for China during 2008–2016 with and without the scaling can be regarded as the lower and upper bounds.

Therefore, we follow the referee's suggestion to show the original global estimate first and then compare it with other reports. Both our results with and without the scaling for East Asia and outflows are shown in the main text. The scaling cannot influence the major conclusion, but importantly gives a reasonable range of CRF estimates. All the references mentioned here are cited in the revised text. Please see Section 2.2 and Line 297-311.

Referee 3: Given it is not clear why the global mean ERFaci is over/underestimated in this work (and it doesn't appear to be stated anywhere), it is not clear that a scaling derived at a global level is applicable to forcings calculated only over China and the north Pacific. Are both 2008 and 2016 simulations scaled to the AR5 value? As mentioned by another reviewer, if the effect of these emission reductions is significant, this creates a further pattern effect which impacts the scaling performed.

Response: Accepted. The scaling factor is derived from the comparison of our 1750 and 2008 simulations with the corresponding estimate from the IPCC AR5 (1750–2011). We used this factor to scale down the ERFaci for the period 2008–2016 globally including over China and northern Pacific Ocean. Therefore, the emission reduction between 2008 and 2016 does not influence the scaling factor. Please also refer to the reason for the scaling of ERFaci estimate shown in the last response.

Referee 3: It might be more appropriate to quote the model values (unscaled) and note that the model over/underestimates the global ERFaci. Where the forcing values need to be compared to the AR5 forcings/a measured value, this difference/scaling factor could be noted.

Response: Accepted. We point out the differences of global CRF forcing estimates between our model and the IPCC AR5 (please see Section 2.2), and show the estimates of aerosol forcings in China and outflow regions with and without the scaling. Please see Line 297-311.

Specific points

L112 - The contribution on meteorology vs aerosol changes is a potentially important one. Given the simulated change in AOD is smaller than observed when using the nudged simulation, is the observed AOD change reproduced when comparing the new 2016 simulations the authors have done?

Response: Accepted. The comparison with the observed AOD change is actually not improved in the new 2016 simulation (that is, the contribution of meteorology is not important). To better interpret the bias of AOD changes, we made the following efforts in the revised manuscript.

1) The modeling of sulfate concentrations was improved by comparing to corresponding observations (Fig. 1a). We increased the SO_2 uptake coefficients for the heterogeneous sulfate formation in each simulation. The revised coefficients lie in the range of previous studies (e.g., Huang et al., 2014; Wang et al., 2015). The greater reductions of sulfate concentrations in our simulations improved the comparison with the observed AOD change.

2) We add the comparison of modeled aerosol extinction profiles with the corresponding CALIPSO data in China. The results suggest that the inadequate treatments of biomass burning emissions and dust aerosols in our simulations could be partially responsible for the AOD underestimation.

Please see Figs. S7-S8 and Line 226-266 in the revised manuscript.

L148 - HIAPER Pole-to-pole

Response: Accepted. Please see Line 150.

L171 - 'The variation' - in this and other locations, I think 'variation' is used to mean 'change'. Is that the case?

Response: Accepted. We replace 'variation' to 'change' throughout the manuscript.

L230 - This would be a great opportunity to show if the 2016-meteorology simulations match the observations better than the 2008-meteorology simulations

<u>Response</u>: Accepted. We find that the 2016-meteorology simulation did not improve the comparison of AOD changes with the observations. The potential factors resulting in the model biases in AOD are discussed in Section 3.2.

L273 - contribute less

Response: Accepted. We reword it. Please see Line 293.

L306 - annual-mean

Response: Accepted. We reword it. Please see Line 316.

References:

Bellouin, N., and Coauthors, 2020: Bounding Global Aerosol Radiative Forcing of Climate Change. Rev. Geophys., 58, e2019RG000660.

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

Gryspeerdt, E., and Coauthors, 2020: Surprising similarities in model and observational aerosol radiative forcing estimates. Atmos. Chem. Phys., 20, 613-623.

Zelinka, M. D., T. Andrews, P. M. Forster, and K. E. Taylor, 2014: Quantifying components of aerosol-cloud-radiation interactions in climate models. J. Geophys. Res.-Atmos, 119, 7599-7615.