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Responses to Reviewer #1

This study quantifies global source-receptor relationships of concentration, direct and indirect radiative forcing of sulfate aerosols utilizing an online chemistry-climate model but nudging it with reanalysis winds. They found that sulfate concentrations are mainly local origin in polluted regions, and their concentration efficiencies in terms of unit precursor emissions are high over arid regions with weak export. In addition, they found the indirect radiative forcing of sulfate aerosols is much larger than the direct radiative forcing. I found topic of this paper is interesting and is suitable for publication in this journal. However, substantial improvements are needed before publication. Following are the major and specific issues:

We thank the referee for all the insightful comments to the manuscript and helpful suggestions for improving the presentation quality. Below, we explain how the comments and suggestions are addressed (our point-by-point responses in blue) and make note of the changes that have been made to the manuscript, attempting to take into account all the comments raised here.

Major issues:

1. The authors should articulate the novelty or advance in science or methodology of this study when comparing to previous works. In the introduction, the authors listed a number of similar studies. However, the authors did not describe clearly their motivations to repeat this kind of work as well as the uniqueness of their findings.

Response:

In previous studies about source attribution of sulfate, only a limited number of anthropogenic source regions over the Northern Hemisphere were considered and examined (Park et al., 2004; Heald et al., 2006; Chin et al., 2007; Hadley et al., 2007; Yu et al., 2013; Bellouin et al., 2016; Stjern et al., 2016). Continents and subcontinents over the tropics and Southern Hemisphere are also important source and receptor regions for the sulfate radiative forcing, especially indirect forcing due to stronger aerosol-cloud interactions in clean environments (Koren et al., 2014). Although Liu and Mauzerall (2007) and Liu et al. (2008, 2009) included ten anthropogenic source regions, they only focused on source attribution of sulfate mass concentration without examining sulfate radiative forcing. In addition, few studies have quantified the global source-receptor relationships of sulfate indirect radiative forcing that can be attributed to local/non-local source regions and anthropogenic/natural source sectors.

Certainly, this study is not a repeat of previous work on source-receptor relationships. This is the first study that examines source attribution of sulfate radiative forcing with tagged anthropogenic and natural sources covering the whole globe. In this study, we quantify sixteen source region/sector contributions (fourteen major source regions and two natural source sectors) to regional and global sulfate mass concentrations, and direct and indirect radiative forcing of sulfate. Another novel aspect of this study is that we are using the new emissions datasets generated for the CMIP6 activities. Thus our model configuration and results could potentially be more comparable to future modeling results coming out of the CMIP6 activities than most of the previous studies.

We have revised the introduction section to show these novelties and discussed the differences between our study and previous studies. Please see responses to the more specific comments below.

2. It is unnecessary to discuss the source-receptor relationships in detail since previous works have already reported similar results. These discussions are lengthy and should be shortened substantially (i.e., abstract, sections 4 and 5). Some figures and discussions could be put into the supporting information.

Response:

We have significantly shortened the details of source-receptor relationships in the abstract as suggested. We would like to stress that our systematical analysis is from a variety of angles to describe source-receptor relationships of sulfate concentrations and radiative forcing, including both the near surface concentration and column burden, both direct and indirect radiative forcing, both oceanic and continental regions, both Northern and Southern Hemisphere, both anthropogenic and natural sources, and both absolute and relative contributions. For all these aspects, we don't see overall similar results to previous studies, especially, in a quantitative way. However, we do value the comment on our lengthy discussions, and have tried our best to shorten the source-receptor descriptions in sections 4 and 5.

3. For the method section, the authors may divide it into several subsections (e.g., model description, tracer tagging, model configurations, . . .). In addition, the parameterizations of calculating the DRF and IRF of sulfate need to be described in detail. The method used to calculate the DRF of sulfate from the tagged regions/sectors is also unclear.

Response:

We have now divided it into the suggested subsections: model description, sulfur source-tagging, emissions, and model configurations. We also clarified on the configurations of the list of simulations performed in this study.

We have also revised the description of parameterizations and the approach of calculating DRF and IRF to include more details, as the following: "Sulfate is internally mixed with other species in the same aerosol mode and

then externally mixed between modes. Sulfate refractive indices at visible wavelengths is $1.43+0.00i$. Activation of cloud droplets uses the scheme from Abdul-Razzak and Ghan (2000). The model simulates aerosol-cloud interactions in stratiform clouds using a physically based two-moment parameterization (Morrison and Gettelman, 2008). In addition to the standard radiative fluxes calculated in the model by taking into account all aerosols, the CESM has the capability of diagnosing radiative fluxes in parallel for a subset of aerosol species. The difference between the standard and the diagnosed radiative fluxes can then be attributed to the difference in aerosols considered in the radiation calculations. For example, the difference in shortwave radiation fluxes at the top of the atmosphere (TOA) represents the DRF of the excluded aerosol components in the diagnostic calculation (Ghan, 2013). Using this same method, the DRF of sulfate from any of the sixteen individual tagged regions/sectors can be derived from a pair of diagnostic radiation calculations with and without the particular tagged sulfate considered. To estimate IRF of sulfate from different sources, we define in this study an incremental IRF, calculated as $\Delta(F_{\text{clean}} - F_{\text{clear, clean}})$, where F is the radiative flux at TOA, F_{clean} is the flux calculated neglecting scattering and absorption by aerosols, $F_{\text{clear, clean}}$ is the flux calculated neglecting scattering and absorption by both clouds and aerosols, and Δ refers to the differences between the base and emission perturbed simulations.”

4. An incremental IRF is defined in this study to quantify the indirect radiative forcing of sulfate. However, there is no validation about this calculation. As the authors mentioned, anthropogenic sources contributed substantially to the incremental IRF over oceans, but few measurements over remote oceans were used to validate their sulfate calculation. The authors may use some aircraft measurements to verify their results over those remote regions.

Response:

The sulfate indirect effect has been fully validated in McCoy et al. (2017) with the same model (CAM5.1-MAM3-PNNL in their study). McCoy et al. (2017) reported that the CAM5.1-MAM3-PNNL model did quite well at producing a reasonable sensitivity of cloud to sulfate mass concentration compared to MODIS satellite data. In addition, in another multi-model intercomparison study including the base simulation results from this work, Fanourgakis et al. (in preparation) evaluates aerosol, CCN and cloud sensitivity in global models against several observational datasets. The incremental IRF in this study is also derived based on the sensitivity of cloud forcing to sulfate (20% of sulfate precursor emission). We have now cited these studies in the revised manuscript instead of duplicating the work.

5. In the introduction, the authors have mentioned that numerous previous studies have examined the sulfate radiative forcing from different sources and regions. However, in the discussion section, they did not carefully compare

their results to previous works. I would suggest the authors pay more attention to the difference between this study and previous works.

Response:

Because we are using different emission datasets and source regions from those in previous studies, a quantitative comparison of source attributions is not so meaningful. However, it is more interesting to compare the radiative forcing efficiency with previous studies. We have now added the Table S9 (see below) to show the comparisons and discussed it at the last section of the manuscript, as the following:

“Table S9 compares the annual sulfate radiative forcing efficiencies simulated in this study to those in previous multi-model studies (Yu et al., 2013; Bellouin et al., 2016; Stjern et al., 2016). As in the previous studies, the DRF efficiency is calculated as the response of global DRF to a 20% reduction in local emissions divided by the 20% of sulfur emissions based on two separate simulations rather than 100% of local emissions in a single simulation (Table S6). The efficiencies based on the 20% emission reduction are very similar to those of the 100% emission reduction, indicating a nearly linear relationship between sulfate DRF and emissions. Compared to Yu et al. (2013) and Stjern et al. (2016), the DRF efficiencies in this study are around the lower bound for all source regions. Another multi-model intercomparison study also reported a lower sulfate DRF simulated in CAM5 compared to other models (Myhre et al., 2013). The difference in DRF efficiencies likely arises from differences in the estimates of aerosol optical properties. With aerosol-cloud interactions included, the total radiative forcing efficiencies in this study are similar to the best estimates provided by Bellouin et al. (2016). The global IRF in CAM5 was also found to be larger than other models in a nine-model intercomparison study, which was attributed to an strong aerosol induced cloud scattering (Zelinka et al., 2014).”

Table S9. Comparison of annual sulfate radiative forcing efficiency ($\text{mW m}^{-2} (\text{Tg S yr}^{-1})^{-1}$) in this study and previous studies. The sulfate DRF efficiencies are calculated as the response of global DRF to a 20% reduction in local emissions divided by the 20% of sulfur emissions.

Direct radiative forcing (DRF) efficiency						
	EUR	EAS	NAM	SAS	RBU	MDE
Yu et al. (2013)	-9.8~-5.0	-7.6~-3.2	-10.0~-5.0	-10.8~-5.0		
Stjern et al. (2016)	-15.7~-5.6	-12.1~-4.6	-15.5~-4.1	-28.0~-6.3	-8.9~-4.3	-32.4~-10.9
This study	-5.4	-3.8	-4.8	-7.2	-3.9	-9.4
Total (direct + indirect) radiative forcing efficiency						
	EUR	EAS				
Bellouin et al. (2016)	-13.0 (-22.7~-4.4)	-9.5(-13.6~-2.6)				
This study	-12.0	-11.6				

Specific comments:

1.L139-145: the description about parameterizations and approach that were used to calculate the DRF and IRF is not very clear. Please provide more details.

Response:

We have revised the description. Please see the response to comment #3 of major issues.

2.L157: black carbon only occurs in the accumulation mode in MAM3, so the comparison is meaningless.

Response:

We have deleted this sentence.

3.L162: Please show some details about this validation.

Response:

We have added Fig. S1 to compare the sulfate concentration and surface air temperature between the no-tagging and tagging simulations to validate the sulfur tagging technique used in this study.

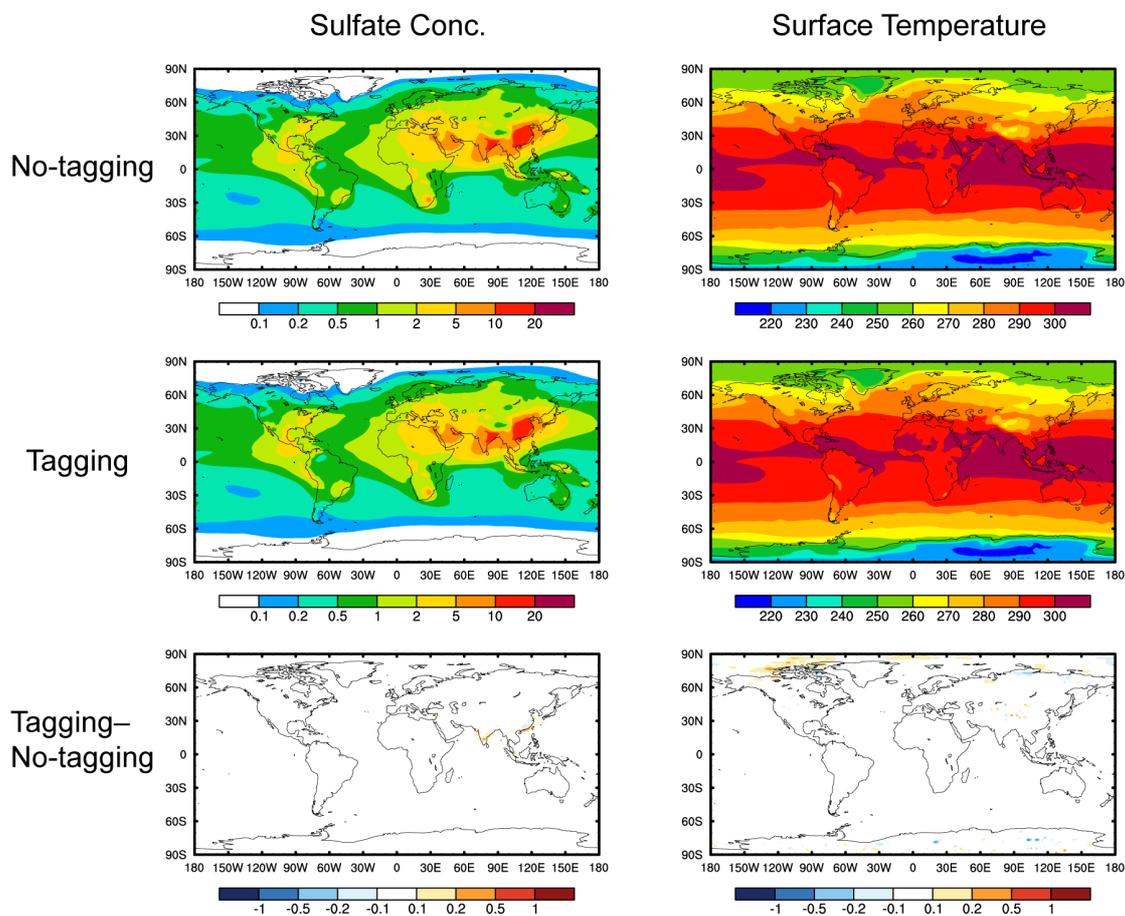


Figure S1. Spatial distribution of annual mean near-surface sulfate concentrations (left, $\mu\text{g m}^{-3}$) and surface air temperature (right, K) from no-tagging (top), tagging (middle) simulations and their differences (bottom).

4.L198: It is not necessary to show the spatial distributions of SO₂ emissions from each tagged region individually. May put Figure 2 into supporting information.

Response:

We have moved this figure to the supporting information as suggested.

5.L203-210: Need some explanations about these seasonal variations.

Response:

We have added some explanations, as “East Asia, RBU and Europe have seasonal peak emissions in boreal winter due to high residential emissions from heating in this season together with higher SO₂ emission from the energy sector. Southern Africa shows larger emission in boreal summer from biomass burning in this season, while emissions from North America are comparable in winter and summer. DMS is emitted over oceans with a boreal winter peak due to phytoplankton blooms over the Southern Ocean.”

6.L219: Only North America is used to validate the decomposition of global incremental IRF. Since different regions may have distinct chemical composition and meteorology, and the sensitivity to regional sulfur emissions could vary significantly by region. I think the authors should validate more regions, especially those with large SO₂ emissions, e.g., East Asia, Europe and South Asia.

Response:

We agree with the reviewer that using only North America to validate the decomposition of global incremental IRF may not be sufficient. However, it is computationally infeasible to test many of the source regions. Given the large emissions from East Asia, we also performed an additional sensitivity simulation with a 20% reduction in regional sulfur emissions over East Asia and have added the IRF comparison in Fig. S10, with results also now included in the text.

We have also revised the description of the comparison as “The 20% emission from North America results in negative IRF over Eastern U.S. and downwind ocean regions. The 20% emission in East Asia emissions produces negative IRF over the northwestern Pacific. Globally, DMS, North America and East Asia contribute to $-0.230 (\pm 0.012)$, $-0.014 (\pm 0.002)$, and $-0.028 (\pm 0.003)$ W m^{-2} , respectively, of sulfate incremental IRF from the method with sulfur tagging technique, similar to $-0.248 (\pm 0.020)$, $-0.018 (\pm 0.019)$, and $-0.028 (\pm 0.018)$ W m^{-2} , from the individual emission-perturbation simulations.”

We have also added a discussion of the noisy spatial distribution of IRF in the emission perturbation method shown in comment #12.

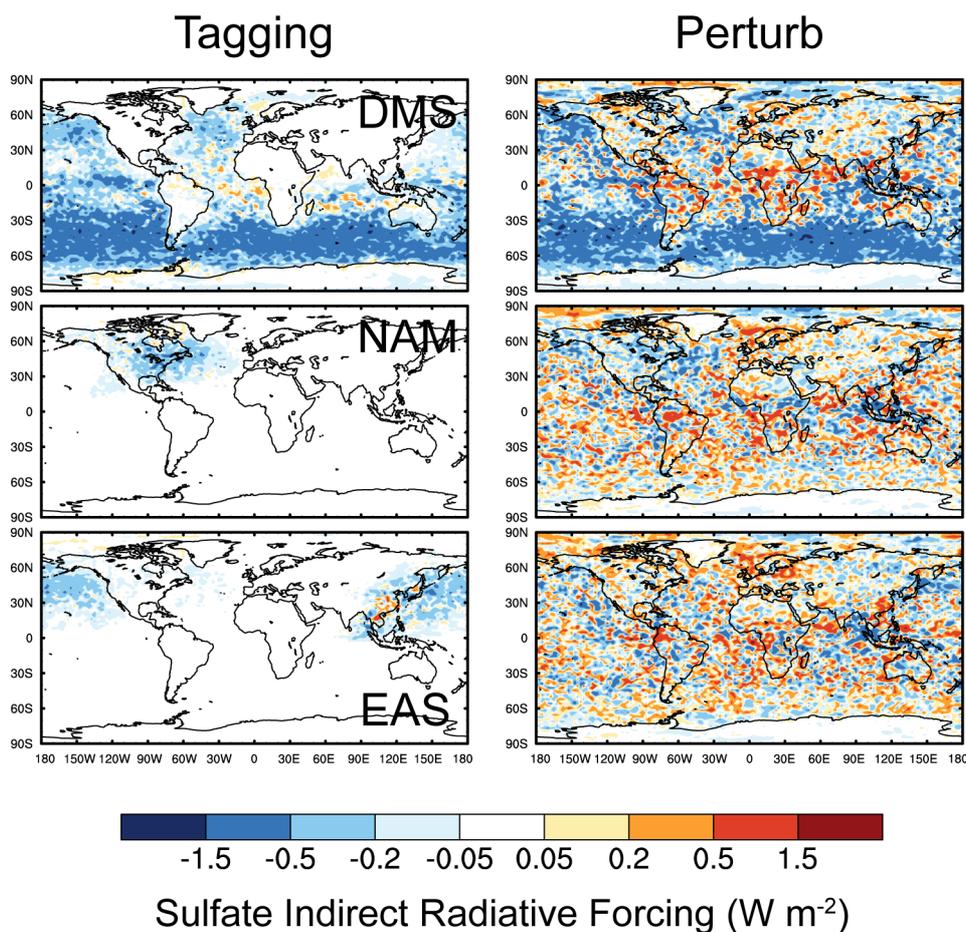


Figure S10. Spatial distribution of annual mean IRF of sulfate (W m^{-2}) induced by a 20% reduction in sulfur emissions from the decomposition using the sulfur tagging method (left panels) and a simple 20% regional/sector emission perturbation (right) for source from DMS (top panels), North America (middle panels), and East Asia (bottom panels).

7.L267: Sulfate has a longer lifetime than black carbon? Need a reference.

Response:

We were thinking about the additional time for the gas-to-particle conversion. It seems to cause some confusion, so we have deleted this sentence.

8.L282-288: If this bias came from the retrieval algorithm, why this overestimation happened more significantly in China than other regions?

Response:

Not only in China, the simulated SO_2 burden is 3 times larger than OMI data over North America, 7 times over Europe, and 5 times over Southeast Asia. We have added these in the manuscript.

9.L294: Here the model results indicated that the export of SO₂ from China is under-estimated. However, on Line 291, the authors stated that the inconsistency between simulated results and satellite observations may suggest an overestimation of SO₂ at higher altitude. In general, the transport is more efficient in the free troposphere, therefore this indicates a potential overestimation of exporting SO₂ from China. Moreover, I would suggest the authors validate total sulfur (SO₂+SO₄) concentrations and total precipitations over China and downwind region.

Response:

Observational data of SO₂ and sulfate are from different sites and have different time coverage. It is difficult to validate total sulfur over China. Nonetheless, considering that both SO₂ and sulfate are underestimated in the model compared to site observations, the total sulfur is likely to be underestimated. We have added Fig. S5 to validate total precipitation. Over China, CAM5 overestimates precipitation over northern China, which leads to a strong aerosol scavenging and low sulfate concentration over this region. We have added these in the manuscript.

We have also revised the discussion of model-observation comparison for clarification, as “The simulated near-surface SO₂ concentrations, however, are also underestimated by 25% compared to observations over thirteen sites in China (Gong et al., 2014) shown in Fig. S4a, also suggesting a large bias in satellite retrievals or too much SO₂ simulated in higher altitude. In general, the transport is more efficient in the free troposphere. If too much SO₂ is simulated in higher altitude, the near-surface SO₂ concentration is likely to be overestimated over downwind regions. However, the modeled SO₂ concentrations over downwind regions of China are underestimated by 45% compared to observations from EANET sites (Fig. S4b). This indicates that bias in the satellite retrievals may be a significant cause of the inconsistency between modeled and satellite-estimated SO₂ burden.”

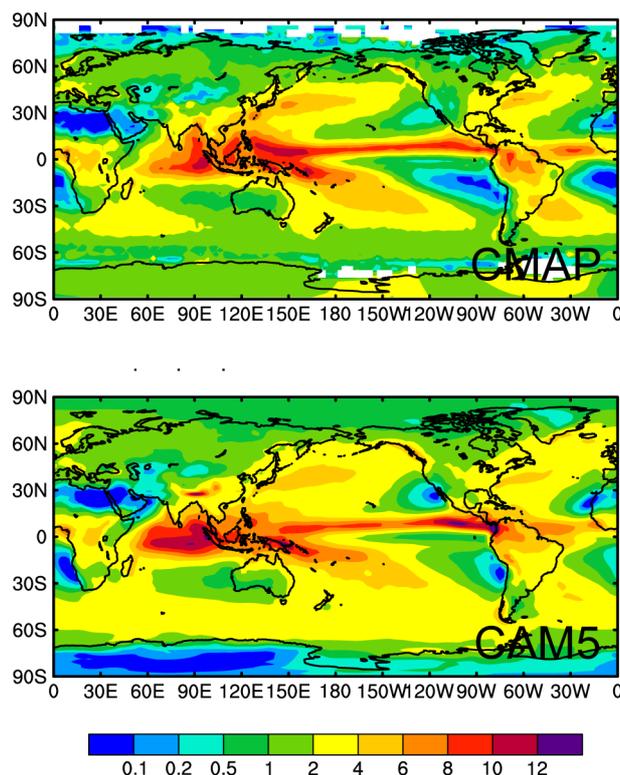


Figure S5. Spatial distribution of annual mean precipitation (mm day^{-1}) from CMAP (Climate Prediction Center’s Merged Analysis of Precipitation, top) and simulated in this study (bottom) averaged over 2010–2014.

10. Sections 4 and 5 are too long and need to be shortened. The authors should pay more attention to the major advance (or unique findings) of this study and explain the difference between their results and previous works.

Response:

We have tried our best to shorten the source-receptor description in sections 4 and 5. As we explained in the response to the major comment above, we don’t necessarily expect similar results to previous studies. As shown in the newly added Table S9, previous studies only examined influence from limited source regions (2–6). In this study, we have 16 tagged source regions partly owing to the computationally efficient sulfur tagging technique, which extends the source-receptor relationship to the whole globe. For the comparison to limited source regions examined in previous studies, we have discussed the differences and possible biases of the model in response to comment #5 of major issues.

11.L423: In Table S3, why is the concentration efficiency of sulfate over MDE in SON greater than 1?

Response:

The efficiencies over the Middle East show high values in almost all seasons due to dry atmospheric conditions favoring long aerosol lifetime,

especially in DJF and SON. We have emphasized it in the manuscript. The concentration efficiency is calculated as local contribution to the near-surface sulfate concentration divided by local sulfur emission (seasonal emissions multiplied by 4). In SON, The MDE local contribution to concentration is $3.40 \mu\text{g m}^{-3}$ and its local SO_2 emission is $0.835 \text{ Tg S yr}^{-1}$. Efficiency= $3.40/(0.835*4)=1.02 \mu\text{g m}^{-3} (\text{Tg S yr}^{-1})^{-1}$. Since the efficiency is not normalized, it does not have to be less than 1.

12.L522-525: The sensitivity test with a 20% reduction in regional sulfur emissions over North America indicated a large uncertainty associated with this method. Therefore, I would suggest the authors to discuss more on the uncertainties of this calculation.

Response:

Thanks for the suggestion. We have added a discussion of this large uncertainty, as “The latter method has larger noise, seen in both the spatial distributions and large uncertainties (standard deviation) of the incremental IRF. The three emission-perturbed simulations produced similar system noise, with a magnitude of $\sim 0.02 \text{ W m}^{-2}$. The incremental IRF signal is larger than the noise around the source regions whereas noise masks the signal in other regions, leading to large uncertainties. However, in the simulation with all source emissions reduced by 20%, the IRF signal overwhelms noise almost everywhere. With the sulfur tagging technique and decomposition method, the noise is also decomposed into smaller pieces which are, in turn, much smaller than the decomposed incremental IRF signal.”

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