

## Manuscript # acp-2017-303

### Responses to Reviewer #2

The manuscript by Y. Yang et al. examines the source of sulfate concentration and its direct and indirect radiative effect based on a novel source-receptor analysis technology embedded in CESM. Sources from both anthropogenic and natural emissions are identified for different regions over the globe. The model results comply with the expectation from common knowledge and provide qualitative and comprehensive understanding. This research addresses an important and interesting question of where the sulfate aerosol comes from globally and provides some implication for pollution alleviation. But in terms of scientific significance I would not rank this research in the highest catalog because this method has been used in previous studies (Wang et al., 2013; Yang et al., 2017) with different chemical species and regions. Considering that this research provide large amount of detailed and solid analysis that improves our knowledge on the question, I would like to recommend the publication of this manuscript. Some comments are given below

We thank the referee for all the comments to the manuscript for improving the presentation quality and the recommendation for publication. Regarding the major comment on the relatively low scientific significance, we would argue that the sulfur tagging technique was implemented in the Community Earth System Model (CESM), for the first time, in this study. Our previous studies used a black carbon (BC) tagging method to study the source attributions and impact of BC emitted from different source regions/sectors. The sulfur-tagging and BC-tagging share the same idea, but compared to BC, sulfur has additional gas-phase and aqueous-phase chemical reactions and there are more size-modes to treat sulfate particles. Thus the sulfur-tagging code implementation, testing and validation did take a large amount of additional efforts, and this tool is indeed novel and unique to the present study.

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.

Line 151: Tagging sulfate is “for the first time”, however, not the first time used in CESM. Suggest not emphasizing the novelty.

Response:

Please see our response above. Sulfate tagging is indeed for the first time implemented and used in CESM.

Line 156 to 159: The same reason as above. Suggest just elaborate the method and avoid using phrases such as “In contrast”.

Response:

Thanks for the suggestion. We don't have to contrast it to BC-tagging, so we changed the description and deleted this phrase.

Line 215: why using 20% reduction to evaluate the indirect effect? Although 20% reduction was used in a previous study (Stjern et al., 2016), this increment of emission is arbitrary to me. Moreover, it hinders comparison with the magnitude of DRF, which compares the forcing with and without 100% aerosols.

Response:

We agree that the 20% reduction of emissions is somewhat arbitrary. However, it follows the AeroCom multi-model experiments design in the framework HTAP (Hemispheric Transport of Air Pollution) to examine the significance of emission reduction. There are numerous studies examined air quality and climate responses to a 20% emission reductions (e.g. Fiore et al., 2009; Fry et al., 2012; Yu et al., 2013; Stjern et al., 2016; Bellouin et al., 2016). We use the same amount of reduction for the purpose of having a fair comparison to these studies. We have added an explanation in the model configuration part, as “The 20% is chosen to follow the experiment design in the framework HTAP2.”

We also agree with the reviewer that the DRF calculated with and without 100% sulfate cannot be directly compared with the incremental IRF induced by the 20% change in emissions. Therefore, we have added in Table S8 the incremental DRF/IRF (calculated from the base and 20% emission reduction simulations) and the standard DRF/IRF (based on the present-day and preindustrial emission simulations), as well as their radiative forcing efficiencies. We have also added a discussion of the comparisons, as “For comparison, Table S8 also includes the incremental DRF calculated with the same simulations for the incremental IRF and the standard anthropogenic DRF between present-day and preindustrial conditions, as well as their efficiencies. The forcing efficiencies are also similar between the incremental and the standard anthropogenic DRF. The IRF and its efficiencies are much higher than those of DRF for sources over or around clean oceanic regions (e.g., DMS, volcanic SO<sub>2</sub>, emissions from Australia and South America), but much lower for regions with high emissions (e.g., the Middle East, South Asia).”

**Table S8.** Annual sulfate incremental direct and indirect radiative forcing calculated based on simulations with and without 20% reduction in sulfur emissions globally and standard direct and indirect radiative forcing ( $W m^{-2}$ )

calculated based on simulations using present-day and preindustrial emissions, as well as the forcing efficiencies ( $\text{mW m}^{-2} (\text{Tg S yr}^{-1})^{-1}$ ) for all of the sixteen tagged source regions/sectors.

DRF Forcing								
	NAM	CAM	SAM	EUR	NAF	SAF	MDE	SEA
Incremental DRF	-0.003	-0.002	-0.002	-0.004	-0.001	-0.005	-0.006	-0.002
DRF (PD-PI)	-0.015	-0.010	-0.011	-0.018	-0.005	-0.023	-0.031	-0.008
DRF Efficiency								
	NAM	CAM	SAM	EUR	NAF	SAF	MDE	SEA
Incremental DRF efficiency	-4.8	-6.9	-7.1	-5.4	-8.3	-8.4	-9.4	-5.3
DRF efficiency	-4.9	-7.0	-7.1	-5.4	-8.1	-8.5	-9.1	-5.5
DRF Forcing								
	CAS	SAS	EAS	RBU	PAN	ROW	VOL	DMS
Incremental DRF	-0.001	-0.009	-0.014	-0.002	-0.001	-0.010	-0.007	-0.014
DRF (PD-PI)	-0.006	-0.046	-0.068	-0.011	-0.003	-0.053		
DRF Efficiency								
	CAS	SAS	EAS	RBU	PAN	ROW	VOL	DMS
Incremental DRF efficiency	-5.4	-7.2	-3.8	-3.9	-5.5	-4.6	-2.7	-4.0
DRF efficiency	-5.2	-7.2	-3.8	-3.9	-5.5	-4.8		
IRF Forcing								
	NAM	CAM	SAM	EUR	NAF	SAF	MDE	SEA
Incremental IRF	-0.014	-0.006	-0.016	-0.004	-0.001	-0.016	-0.001	-0.004
IRF (PD-PI)	-0.082	-0.036	-0.072	-0.032	-0.005	-0.061	0.012	-0.017
IRF Efficiency								
	NAM	CAM	SAM	EUR	NAF	SAF	MDE	SEA
Incremental IRF efficiency	-22.8	-19.8	-50.3	-6.6	-6.2	-28.7	-1.7	-14.1
IRF efficiency	-26.3	-25.0	-44.7	-9.5	-7.9	-22.4	3.5	-11.9
IRF Forcing								
	CAS	SAS	EAS	RBU	PAN	ROW	VOL	DMS
Incremental IRF	-0.002	-0.005	-0.028	-0.007	-0.009	-0.042	-0.057	-0.230
IRF (PD-PI)	-0.012	-0.002	-0.117	-0.056	-0.051	-0.202		
IRF Efficiency								
	CAS	SAS	EAS	RBU	PAN	ROW	VOL	DMS
Incremental IRF efficiency	-8.7	-3.7	-7.8	-11.8	-77.3	-18.6	-22.5	-63.2
IRF efficiency	-11.5	-0.3	-6.6	-18.7	-86.6	-18.1		

Line 449: is this 1% difference a coincidence?

Response:

It does not mean '1% difference', but '1750 emission is less than 1% of present-day emission'. We tried to illustrate that DRF of anthropogenic sulfate is calculated here based on present-day and no emission condition ( $\text{DRF}_{\text{PD}} - 0$ ), while the estimate in IPCC AR5 represents the difference between the present-day and 1750 DRF ( $\text{DRF}_{\text{PD}} - \text{DRF}_{1750}$ ). The global anthropogenic  $\text{SO}_2$  emission amount (0.5 Tg/yr) in 1750 is very small, about 0.5% (less than "1%")

of the 2010–2014 level (109.8 Tg/yr) from the CEDS emission dataset. Therefore,  $DRF_{PD} - DRF_{1750} \approx DRF_{PD}$ .

#### References:

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