Responses to Reviewer #3

This study examined source apportionment of aerosols in Europe over 1980-2018 using the Community Atmosphere Model version 5 with an Explicit Aerosol Source Tagging technique (CAM5-EAST). They found that the near-surface total mass concentration of sulfate, black carbon and primary organic carbon had a 62% decrease and aerosols from foreign sources became increasingly important to air quality in Europe. They also estimated that contributions to the sulfate radiative forcing over Europe from both European local emissions and non-European emissions would decrease at a comparable rate in the next three decades. The CAM5-EAST model showed its advantage in simulating the aerosol source-receptor relationship within one model simulation. The topic is interesting and the manuscript is well organized. I suggest it published in the journal after addressing my minor comments below.

We thank the reviewer for all the insightful comments. Below, please see our point-by-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in an effort to take into account all the comments raised here.

The authors examined sulfate, black carbon and organic carbon aerosols in this study. Why did the author exclude other aerosols like nitrate in the simulation?
Response:

The representation of nitrate and ammonium aerosols requires many additional gas species and chemical/physical process treatments in models. Different from regional air quality models, including complex chemistry and aerosol thermodynamical equilibrium is less efficient for the long-term simulation in global aerosol-climate models. In the 3-mode version of modal aerosol model in CAM5, sulfate is partially neutralized by ammonium in the form of NH_4HSO_4, so ammonium is effectively prescribed, but this model version cannot predict ammonium and nitrate. In the next version of CAM6, which will be released early next year, an advanced aerosol chemistry and microphysics module (called MOSAIC) will be implemented to treat tropospheric trace gas photochemistry, aerosol thermodynamics, kinetic gas-particle mass transfer and particle-phase chemistry, particularly, for nitrate aerosol. As a next step in our research plan, we will implement the tagging tool EAST to the new model version and analyze the source-receptor relationship of sulfate-nitrate-ammonium in future studies.

There seems a lot difference between the source attribution to near-surface concentration and column loading, as demonstrated in Figure 6. Thus, it would be more clear to directly show the transport pattern and source contributions near surface as well as those at higher altitude.

Response:

Thanks for the suggestion. We have now added the horizontal distribution of sulfate-BC-POA concentrations at the surface and 500 hPa, originating from the major tagged source regions, as shown below.

"The transboundary and intercontinental transport of aerosols occur most frequently in the free troposphere rather than near the surface, as horizonal transport pathways at the surface and 500 hPa are indicated on the spatial distribution map of the relative contributions shown in Figures S2 and S3. This also leads to larger relative contributions from non-European sources to aerosol column burdens than to the near-surface concentrations."

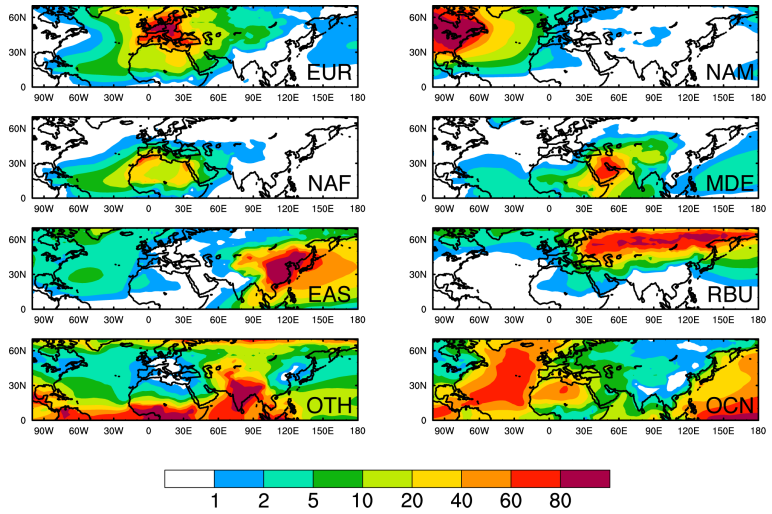

Figure S2. Relative contributions (%) to annual mean near-surface concentrations of sulfate-BC-POA from the major tagged source regions including Europe (EUR), North America (NAM), North Africa (NAF), the Middle East (MDE), East Asia (EAS), Russia-Belarus-Ukraine (RBU), Non-Arctic/Antarctic Ocean (OCN) and other (OTH) regions averaged over 2010–2018.

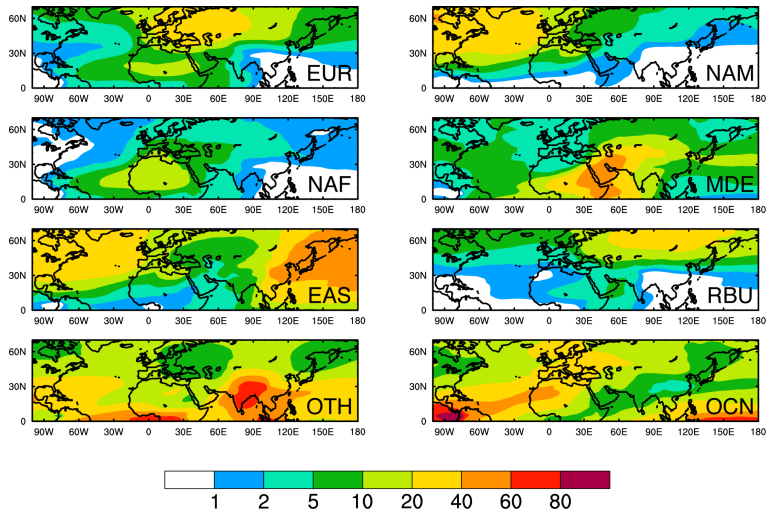

Figure S3. Relative contributions (%) to annual mean concentrations of sulfate-BC-POA at 500 hPa from the major tagged source regions including Europe (EUR), North America (NAM), North Africa (NAF), the Middle East (MDE), East Asia (EAS), Russia-Belarus-Ukraine (RBU), Non-Arctic/Antarctic Ocean (OCN) and other (OTH) regions averaged over 2010–2018.

In Figure 11, the areas represent minimum-to-maximum ranges. Is there a possibility that one SSP scenario produces a minimum decrease in EUR contribution and a maximum decrease in Non-EUR contribution?

Response:

We have now plotted the figure for each SSP scenario individually in Figure S4. All SSPs show that non-European contributions change in a magnitude similar to that of European local emissions.

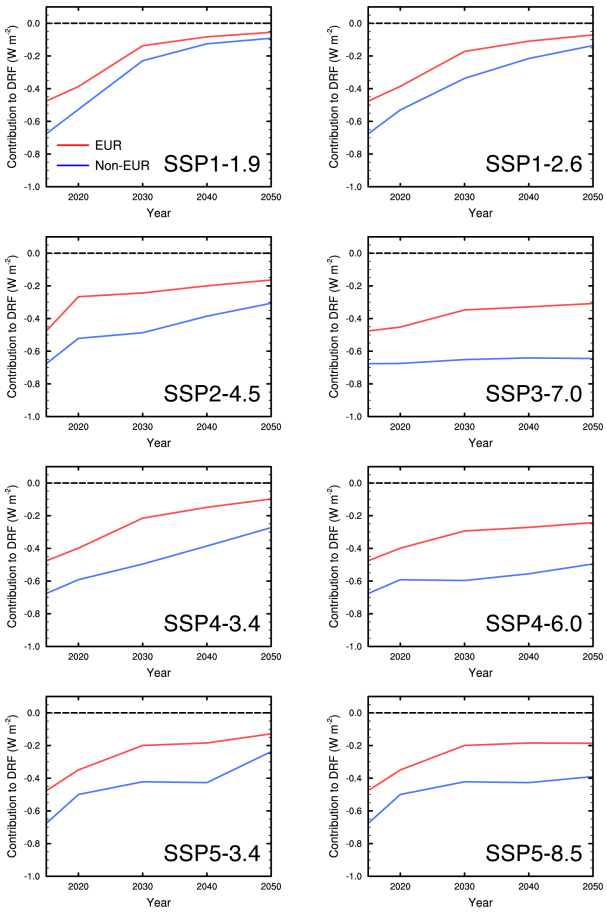

Figure S4. Time series (2015–2050) of estimated annual mean sulfate DRF over Europe contributed by European and non-European emissions from eight SSP scenarios, including SSP1-1.9, SSP1-2.6, SSP2-4.5, SSP3-7.0, SSP4-3.4, SSP4-6.0, SSP5-3.4, and SSP5-8.5. Future DRF of sulfate aerosol over Europe is estimated by scaling historical mean (1980–2018) sulfate DRF using the ratio of SSPs future SO_2 emissions to historical emissions assuming a linear response of DRF to regional emissions.

What is the advantage of using CAM5-EAST rather than CAMx or CMAQ mentioned in the introduction section?

Response:

Influences of remote sources simulated in regional air quality models such as CAMx and CAMQ largely depend on the boundary conditions. They cannot tag and track the emissions outside the regional domain. As we discuss in the text, “However, due to the limitation in domain size of regional air quality models, contributions of intercontinental transport from sources outside the domain are difficult to be accounted.” CAM5-EAST is a global model with aerosol tagging that has been used to examine the transboundary and transcontinental transport of aerosols in previous studies (Yang et al., 2018a,b).

The author analyzed annual averaged source contributions in this study. How is the source-receptor relationship in different seasons? Are they the same as the annual mean results?

Response:

Thanks for the suggestion. We have now added the analysis of seasonal source-receptor relationship of aerosols in Europe and the role of meteorological factors based on an emission normalization method. Please see below:

“Source contributions to aerosols in Europe vary with season due to the seasonality of emissions and meteorology. In general, local sources have the largest contributions to both near-surface concentration and column burden of European aerosols in winter and smallest contributions in summer averaged over 2010–2018 (outer rings in Figure 7). With the contributions normalized by the ratio of seasonal anthropogenic emission to annual mean for each source, the impact of emission seasonal variation on the source contributions can be removed (inner rings in Figure 7) (Yang et al., 2019). Without the influence of emission seasonality, local source contributions decrease in winter and increase in summer, indicating that it was the higher local anthropogenic emissions that result in the larger local source contributions to wintertime aerosols in Europe relative to other seasons. Sulfur sources over oceans account for one fourth to one third of European sulfate concentration and burden in spring likely due to the strong westerlies in this season that transport aerosols from the North Atlantic Ocean to the Europe. Source contributions from Russia-Belarus-Ukraine and North America to BC and POA in Europe show strong seasonal variabilities, which can be explained by the changes in biomass burning emissions considering its large seasonal variability.”

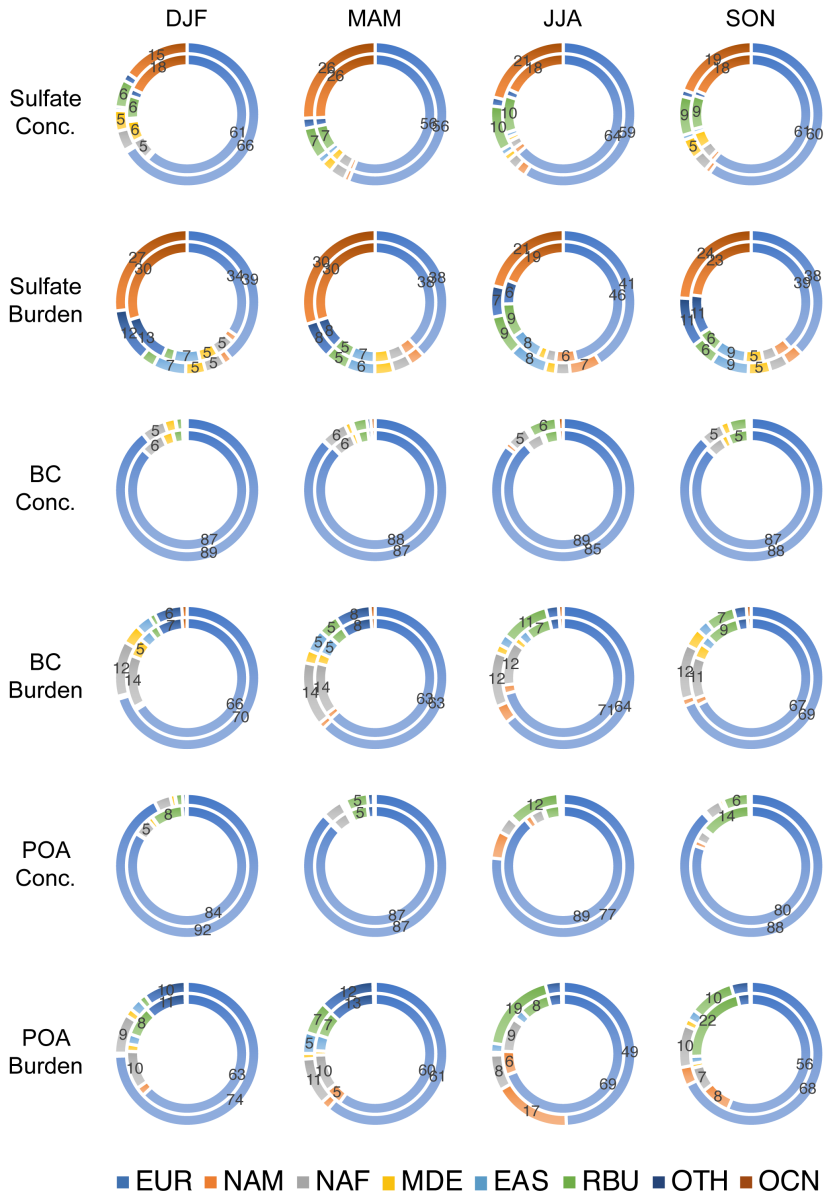

Figure 6. Relative contributions (%) by emissions from major tagged source regions to near-surface concentrations (Conc.) and column burdens of December-January-February (DJF), March-April-May (MAM), June-July-August (JJA) and September-October-November (SON) mean sulfate, BC and POA over the Europe averaged over 2010–2018. Outer rings represent the modeled values and the relative contributions in inner rings is calculated based on absolute values normalized by the ratio of seasonal emission to annual mean. Values larger than 5% are marked.

Page 11: What is temporal resolution of the observational data?

Response:

We have now added a description that “EMEP (European Monitoring and Evaluation Programme, http://www.emep.int) networks provide daily near-surface aerosol concentrations in Europe. The annual mean of daily

observations is used to evaluate the model performance in this study."

Fig.5: specify the abbreviations in the figure
Response:
Revised.

Reference:

Yang, Y., Wang, H., Smith, S. J., Zhang, R., Lou, S., Yu, H., Li, C., and Rasch, P. J.: Source apportionments of aerosols and their direct radiative forcing and long-term trends over continental United States, Earth's Future, 6, 793–808, https://doi.org/10.1029/2018EF000859, 2018a.

Yang, Y., Wang, H., Smith, S. J., Zhang, R., Lou, S., Qian, Y., Ma, P.-L., and Rasch, P. J.: Recent intensification of winter haze in China linked to foreign emissions and meteorology, Sci. Rep., 8, 2107, https://doi.org/10.1038/s41598-018-20437-7, 2018b.

Yang, Y., Smith, S. J., Wang, H., Lou, S., and Rasch, P. J.: Impact of anthropogenic emission injection height uncertainty on global sulfur dioxide and aerosol distribution, J. Geophys. Res.-Atmos., 124, 4812–4826. https://doi.org/10.1029/2018JD030001, 2019.