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Responses to Reviewer #1

The study by Yang et al., attempted to quantify the contribution of major source regions in the world towards aerosol loading in Europe. The study has certain flaws which needs to be addressed before it can be accepted for publication at ACP.

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.

Line 163: Any specific reason on why future DRF due to aerosols other than sulphate was not estimated in this study for future. If not, I would suggest doing the same.

Response:

Here in this study we focus both historical and future sulfate DRF rather than other aerosol species. Aerosol DRF is defined in this study as the difference in clear-sky radiative fluxes at the top of the atmosphere between two diagnostic calculations in the radiative transfer scheme with and without specific aerosol species accounted, respectively. Therefore, the DRF estimate requires additional calculations of radiative fluxes. In default CAM5, less than 9 additional radiation calculations are allowed. Since that the simulation was designed to output the DRF related to emissions from Europe as well as few key source regions of the world that were used in our previous studies, it was not feasible to separate all aerosol species for the radiation diagnostic calculations. Considering that sulfate AOD accounts for the largest portion (91%) of the decrease in total combustion AOD in Europe, the sulfate DRF is calculated to roughly represent the DRF due to the total combustion AOD change.

We have now added an explanation in the revised manuscript: “Rather than sulfate, DRF of other aerosol species is not calculated in this study due to the computational limitation considering multiple source regions. However, because sulfate dominates the decrease in total combustion AOD in Europe shown below, the sulfate DRF is calculated to roughly represent the DRF caused by the total combustion AOD change.”

Line 175: Why was nitrate and ammonium aerosols were not considered in this study? I would suggest including nitrate at least.

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.

Line 218: I would strongly suggest to not compare the sum of BC, OC and sulphate with PM2.5 from observations until aeolian dust, sea salt, nitrate and ammonium are presented/simulated. Additionally, I feel it is meaningless to compare the model AOD (without components like nitrate, ammonium) with AERONET.

Response:

Thanks for the suggestion. We have removed the $PM_{2.5}$ comparison in the manuscript and revised corresponding descriptions. For the comparison of model and observed AOD, although the sum of sulfate (or NH_4HSO_4 specifically), BC, POA, SOA, dust and sea salt cannot represent total aerosols in the real world, the comparison is still meaningful. The purpose here is to show the decreasing trend of AOD in Europe. Including nitrate aerosol in the simulation is unlikely to reverse the trend. So, we decide to keep the AERONET lines in the figure.

Line 220: Any specific reasons on why the model does not have the capability to simulate ammonium and nitrate aerosols.

Response:

Please see the responses above.

I strongly suggest the authors to include a section on seasonal source-receptor relationship for Europe supported by meteorological factors (like wind directions). I understand it is computationally expensive to carry out this for all the years considered in the study. However, performing seasonal analysis for a single representative year would suffice.

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.”

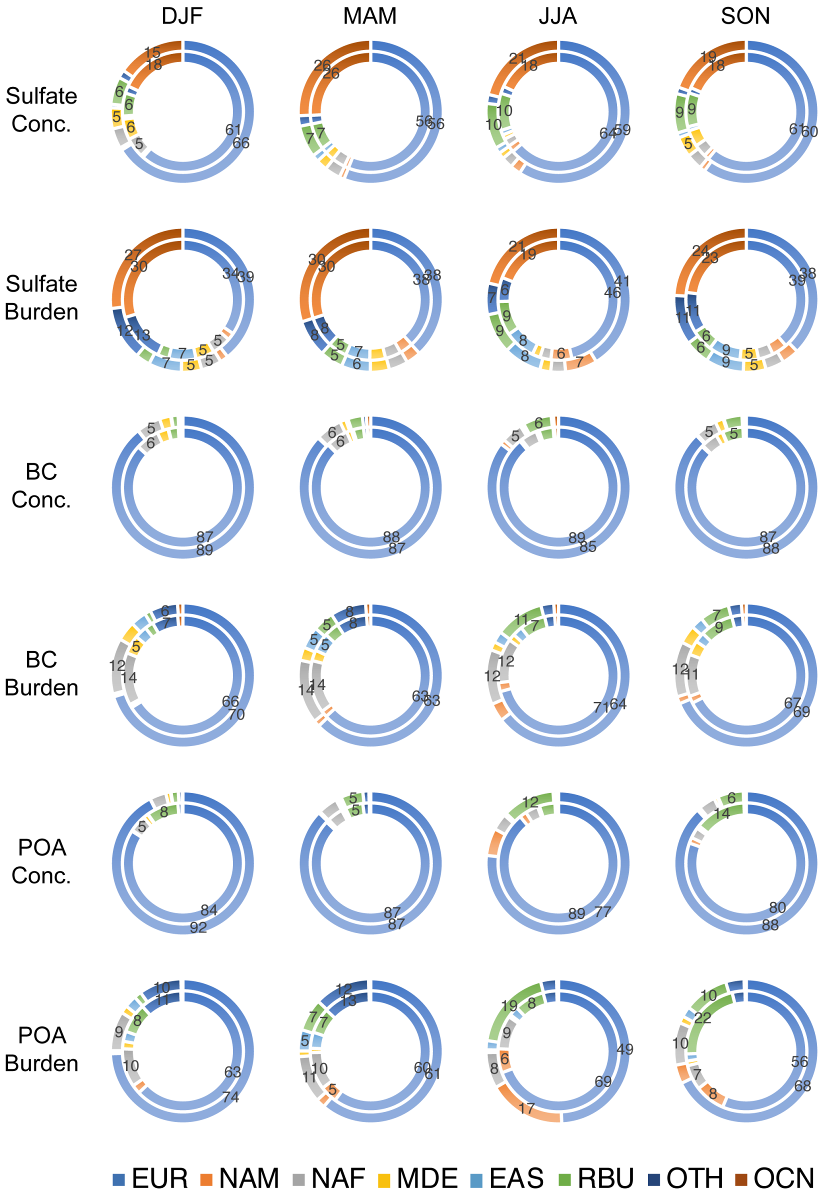

Figure 6. Relative contributions (%) by emissions from major tagged source regions to near-surface concentrations (Conc.) and column burdens of December-January-February (DJF), March-April-May (MAM), June-July-August (JJA) and September-October-November (SON) mean sulfate, BC and POA over the Europe 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.

Reference:

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.