Responses to Referee #2

Yang et al integrated the Explicit Aerosol Source Tagging (EAST) system into the Energy Exascale Earth System Model version 1 (E3SMv1) to investigate the variations in anthropogenic aerosol concentrations, their sources, and their radiative impacts across four major global emission regions (North America, Europe, East Asia, South Asia) during three key historical periods (1850–1980, 1980–2010, 2010–2017). This research advances our understanding of the historical changes in aerosol pollution, emphasizing the complexity of source-region relationships. The conclusions are primarily derived from simulations performed using this integrated model. However, detailed information about the EAST system within the manuscript is limited. Therefore, I recommend acceptance of the paper after the authors address the following points:

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.

1.The manuscript would benefit from an expanded section on the EAST system. Given the study's reliance on this algorithm for its conclusions, a more in-depth explanation of how and why the EAST system functions is necessary, beyond just referencing previous papers.

Response:

Thank you for the suggestion. In the revised manuscript, we have added the relevant description to clarify the source tagging system introduced in the model as "Source apportionment aims to quantify the contributions to aerosols from specific sources. To examine the source-receptor relationships of aerosols, we implemented the Explicit Aerosol Source Tagging (EAST) in E3SMv1, which play a critical role in attributing aerosol concentrations to their respective emission sources. The EAST follows the BC source-tagging technique introduced in Wang et al. (2014), sulfate source-tagging method used in Yang et al. (2017) and other carbonaceous aerosol-tagging applied in Yang et al. (2018a), which was previously implemented in the CAM5 (CAM5-EAST). This tagging system is different from the traditional emission sensitivity method that zero out or perturb emissions from a given source region or sector in sensitivity simulations along with a baseline simulation, which has to assume a linear response to emission perturbation and requires many additional simulations for estimating the contributions from multiple sources (Wang et al., 2014). EAST independently considers all aerosol physical, chemical, and dynamical processes for each tagged sources through introducing additional aerosol-related variables within one simulation and it does not rely on a linear response to emission perturbations. These capabilities make it physically more accurate and time saving than the sensitivity experiments. This tagging method has previously been adopted in regional models and has now implemented in the global E3SMv1 model to better understand the

intercontinental transport from sources outside the regional domain."

2. While the modeled aerosol concentrations align well with 2017 observations from IMPROVE (USA), EMEP (Europe), and CNEMC (China), the study spans a considerable historical period. Therefore, a more robust validation of the modeled data, particularly for earlier periods, would enhance the study's credibility.

Response:

Thank you for the suggestion. In addition to the year 2017, we have also the evaluated of the performance of E3SMv1 in 2010 (Fig. S3). The spatial correlation coefficient (R) between the E3SMv1 simulated $PM_{2.5}$ concentrations and observations in 2010 is +0.91. The model well reproduces the $PM_{2.5}$ concentrations in the U.S. with the normalized mean biases (NMB) of 0.7%. However, it also largely underestimates the $PM_{2.5}$ concentrations in China and Europe.

In order to evaluate the model performance in reproducing the historical changes in aerosol concentrations during the important periods of emission changes, the variations in near-surface PM2.5 concentrations are compared with observations (Fig. S5) and MERRA-2 reanalysis (Fig. S6). The model well reproduces the decreases in PM2.5 concentrations in the eastern U.S. and Europe and the increases in East Asia and South Asia during 1980–2010, with the spatial R of 0.78 between model results and MERRA-2 data. The model also well simulates the aerosol decline in North America, Europe, and East Asia and a continuous increase in South Asia during 2010–2017, with the R of 0.81 between model results and observational data. We have added these descriptions in the manuscript.

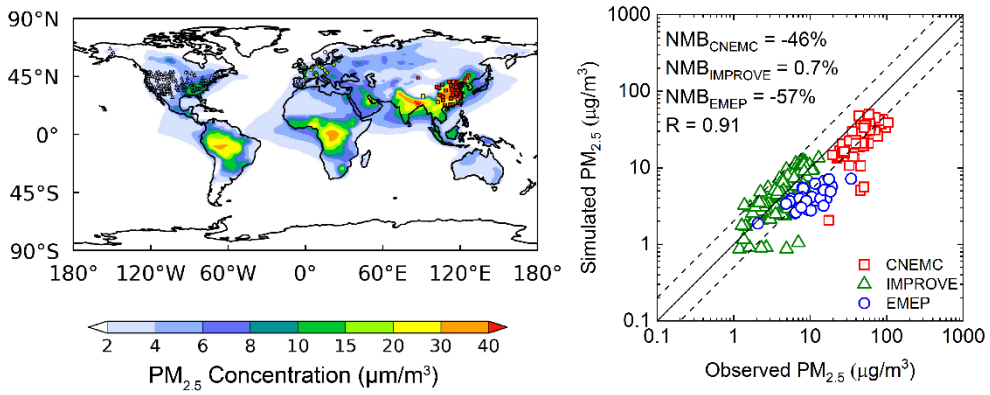

Figure S3. Spatial distribution (left panel) and scatter plot (right) between the simulated and observed annual mean near-surface $PM_{2.5}$ concentrations ($\mu g/m^3$) in 2010. Observational data are from IMPROVE (triangle), EMEP (circle) and CNEMC (square, in 2013). The solid line marks the 1:1 ratio and dashed lines mark the 1:2 and 2:1 ratios. Normalized mean bias (NMB) and correlation coefficient (R) between observation and simulation are shown on the right panel. $NMB = 100\% \times \sum(M_i - O_i)/\sum O_i$, where M_i and O_i are the modeled and observed values at site i, respectively.

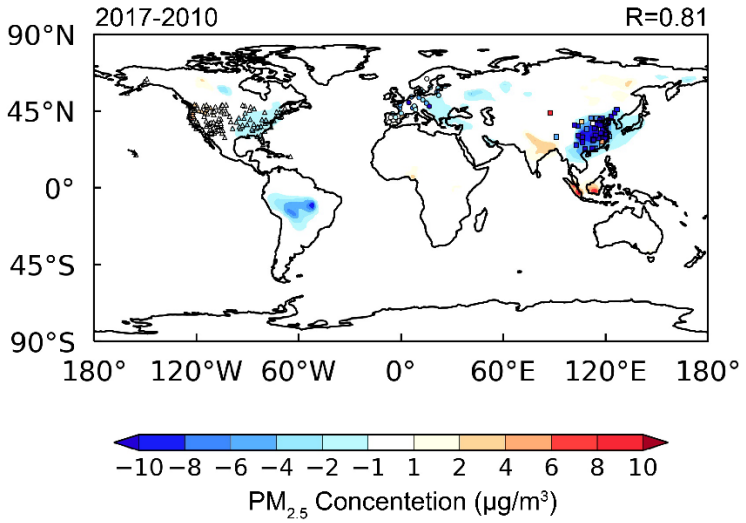

Figure S4. Spatial distribution of the change in simulated and observed annual mean near-surface $PM_{2.5}$ concentrations ($\mu g/m^3$) between 2010 and 2017. Observational data are from IMPROVE (triangle), EMEP (circle) and CNEMC (square, between 2013 and 2017).

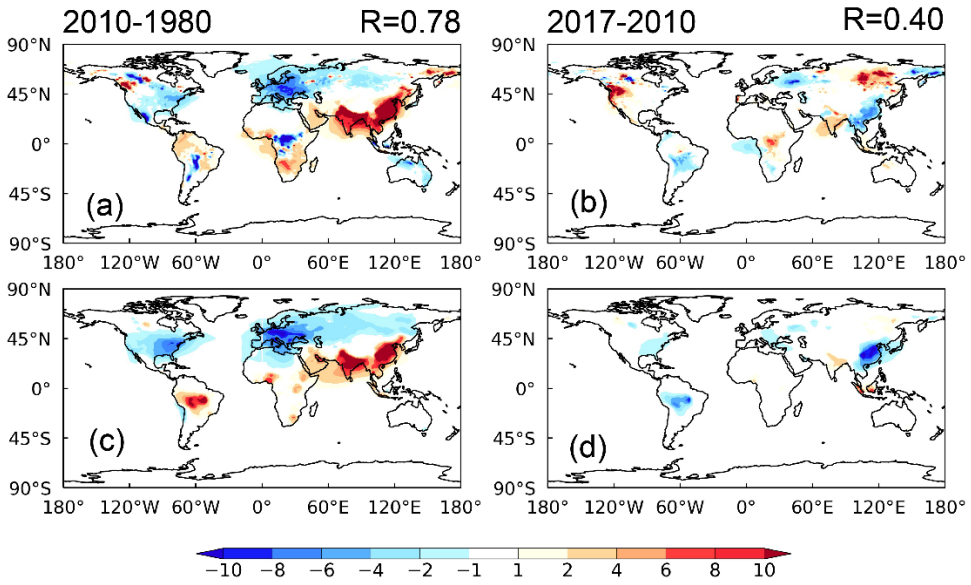

Figure S5. Spatial distribution of the annual mean near-surface $PM_{2.5}$ concentrations ($\mu g/m^3$) from (a, b) MERRA-2 and (c, d) model simulation between 1980 and 2010 (left) and between 2010 and 2017 (right).

3.The use of both column burden and near-surface concentration for discussion is noted. Clarification on the benefits of using column burden in certain contexts would be valuable. Additionally, specifying the defined altitude for 'near surface' in the context of this study would provide clarity.

Response:

Thanks for your suggestion. Aerosol column burden refers to the concentration of

aerosols contained in the air column above a unit area, which can better reflect the aerosol transport within the air column and is more related to the aerosol radiative effect. The near-surface concentration of aerosols represents the concentration of aerosols in the air near the surface (from 1000 to 997 hPa for model layer), which is more related to air quality and human health. We have now added these descriptions in the manuscript.

References:

Wang, H., Rasch, P. J., Easter, R. C., Singh, B., Zhang, R., Ma, P. L., Qian, Y., Ghan, S. J., and Beagley, N.: Using an explicit emission tagging method in global modeling of source-receptor relationships for black carbon in the Arctic: Variations, sources, and transport pathways, J. Geophys. Res., 119, 12888–12909, https://doi.org/10.1002/2014JD022297, 2014.

Yang, Y., Wang, H., Smith, S. J., Easter, R., Ma, P.-L., Qian, Y., Yu, H., Li, C., and Rasch, P. J.: Global source attribution of sulfate concentration and direct and indirect radiative forcing, Atmos. Chem. Phys., 17, 8903–8922, https://doi.org/10.5194/acp-17-8903-2017, 2017.

Yang, Y., Wang, H., Smith, S. J., Zhang, R., Lou, S., Qian, Y., Ma, P., 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, 2018a.