Responses to Referee #1

This manuscript investigates the dynamic changes in anthropogenic aerosol concentrations, their sources, and subsequent radiative impacts across four emission regions during different historical periods. This work provides a valuable contribution to our understanding of historical trends in anthropogenic aerosols, emphasizing the importance of regional variations and their implications for both local and global environmental policies. However, there are notable issues, particularly concerning the validation of the model. Addressing these concerns would significantly improve the manuscript, making it suitable for publication. A revised version will provide a nuanced exploration of aerosol dynamics, contributing to the broader discourse on climate change mitigation and collaborative efforts for sustainable environmental management.

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.

Major Comments:

1. In line no. 105, the authors mention the use of the Energy Exascale Earth System Model version 1 (E3SMv1) for their study. However, there is insufficient detail about the model in the introduction. Is E3SMv1 considered a state-of-the-art model for this type of study? What advantages does this model offer over other available global climate models or regional models? The choice of a climate model is crucial in studies of this nature, and the introduction should elucidate why E3SMv1 was deemed suitable for the investigation. A detailed explanation of the model's capabilities, unique features, and any advancements that make it state-of-the-art in the context of this study would enhance the paper's clarity and transparency.

Response:
  Thank you for the suggestion. In the revised manuscript, we have added the relevant description in the model description as "To study variations in historical anthropogenic aerosols in the major source regions, the E3SMv1 developed by US Department of Energy (DOE) (Golaz et al., 2019) is used in this study. The E3SMv1 model is updated on the basis of Community Atmosphere Model version 5 (CAM5) in order to explore several key emerging issues in the field of environment and climate, and is a branch of the widely-used Community Earth System Model (CESM) (Rasch et al., 2019). E3SMv1 consists of atmosphere, land surface, ocean, sea ice, and river model components. It features numerous upgrades to aerosol, turbulence, chemical, and cloud-related processes, offering multiple spatial resolution options. The model can run simulations for decades or more at higher resolution to help understand past, present, and future changes in Earth's behavior, and to explore how the atmosphere interacts

with other components of the Earth system. The four-mode Modal Aerosols Module (MAM4) in its atmospheric component predicts sulfate, black carbon (BC), primary organic matter (POM), secondary organic aerosol (SOA), marine organic aerosol, mineral dust and sea salt (Wang et al., 2020). The model has been applied to investigate the variations in anthropogenic and natural aerosols related to the air-sea interactions (Yang et al., 2022b; Zeng et al., 2021). Compared to the regional model, the E3SMv1 with an aerosol tagging tool introduced in this study is more suitable for the simulation of transboundary and intercontinental transport of aerosols across the globe."

2. Similar question for the source tagging system (E3SMv1-EAST). Is this the standard system for source tagging? What types of systems are used in other models, and why was this system chosen for this study? Source tagging systems play a critical role in attributing aerosol concentrations to their respective emission sources, influencing the accuracy and reliability of the study's findings. Providing background information on EAST's capabilities, its advantages over alternative source tagging systems, and any specific features that make it particularly suited for integration with E3SMv1 would enhance the clarity of the methodology.

Response:
        Thank you for the suggestion. Yes, this tagging system has been widely applied in regional models. 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."

3. The entire methodology depends on how accurately the emission inventories are incorporated and how precisely the model predicts PM_5 values compared to

observations. However, in the model evaluation section 2.3, lines 151-155, the authors consider observational data from IMPROVE (USA), EMEP (Europe) and CNEMC (China). Observational data from the region of South Asia is not considered, though it is a region where the model study is being run. A major source region of PM$_{2.5}$ in South Asia is India and observational data from this region should also be used in the model evaluation, as this region is a part of the model study. Mishra et al. 2021 have shown that Indian region is a major contributor to PM$_{2.5}$, even 10 to 20% more than China. With observations limited to China, for both East and South Asia, it becomes challenging to comprehensively assess the model's performance. Considering the region's contribution to PM$_{2.5}$, incorporating observational data from South Asia, particularly India, is crucial for a comprehensive assessment of the model's performance.

Response:
We have now added the evaluation of modeled PM$_{2.5}$ concentrations in Indian (Fig. 2). The observational data are collected by the U.S. embassies and consulates in India. We also revised the model evaluation as "The model successfully reproduces the spatial distribution of PM$_{2.5}$ concentrations, with relatively high concentrations in eastern China, India and low concentrations in the U.S. and Europe. The spatial correlation coefficient (R) between the E3SMv1 simulated PM2.5 concentrations and observations is +0.81. The model well reproduces the PM2.5 concentrations in the U.S. with the normalized mean biases (NMB) of –11%. However, it largely underestimates the PM$_{2.5}$ concentrations in China, Europe and India, which has also been revealed in several studies (e.g., Gao et al., 2018; Gao et al., 2023; Navinya et al., 2020; Zeng et al., 2021)."
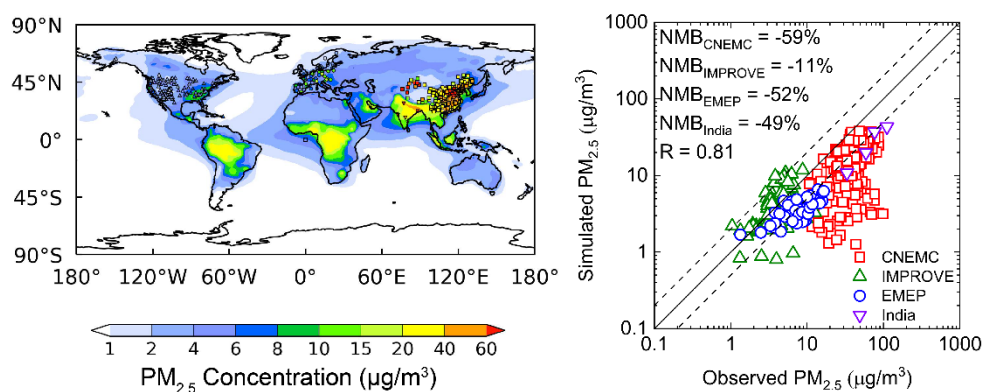
Figure 2. Spatial distribution (left panel) and scatter plot (right) between the simulated and observed annual mean near-surface PM$_{2.5}$ concentrations (μg m^{-3}) in 2017. Observational data are from IMPROVE (triangle), EMEP (circle), India (inverted triangle) and CNEMC (square). The solid line marks 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.

4. The PM$_{2.5}$ data in Figure 4, panel SAS indicates that India's PM$_{2.5}$ mass concentration decreases from 2010-1989 to 2017-2010. However, this contradicts numerous studies (for example: Dey et al 2012, Dey et al 2020, Guttikunda et al 2022, Singh et al 2023) that have found a significant increase in PM$_{2.5}$ levels from 2010 onwards. Additionally, the reported PM$_{2.5}$ mass concentration values in the range of 4 to 5 μg m^{-3} are ten times lower than the values observed by Mishra et al. 2021. This discrepancy raises concerns about the model's performance in the South Asia region, which may cast doubt on the reliability of other results obtained from the model.

Response:

In Figure 4, we plot changes in near-surface mass concentration (μg/m^3) of anthropogenic PM$_{2.5}$ concentrations contributed by the 5 tagged source regions (NAM, EUR, EAS, SAS and ROW from top to bottom) between 1850 and 1980 (left), between 1980 and 2010 (middle), and between 2010 and 2017 (right). It shows the variations between four important historical periods, not the annual average PM$_{2.5}$ value. Therefore, the panel SAS indicates that anthropogenic PM$_{2.5}$ concentration contributed by SAS emissions was on the rise during both 1980-2010 and 2010-2017, which is consistent with the results in Dey et al.(2020), Guttikunda et al.(2022), and Singh et al.(2023). Figure 4 shows that the anthropogenic PM$_{2.5}$ increase by 4-5 μg/m^3 during 2010-2017 in SAS, rather than the average value of PM$_{2.5}$ in 2010-2017. The annual mean PM$_{2.5}$ concentrations in SAS are shown in Fig. 2 and the values are between 20 and 60 μg/m^3, which is similar to the data provided in other studies. By the way, the references for the PM$_{2.5}$ observations have been cited in the manuscript.

5. Line 591: Please write the full caption for Figure 5.
Response:
Revised.

6. Line 600: Please write the full caption for Figure 7.
Response:
Revised.

References:

Golaz, J. C. et al.: The DOE E3SM Coupled Model Version 1: Overview and Evaluation at Standard Resolution, J. Adv. Model. Earth Syst., 11, 2089–2129. https://doi.org/10.1029/2018MS001603, 2019.

Rasch, P. J. et al.: An Overview of the Atmospheric Component of the Energy Exascale Earth System Model, J. Adv. Model. Earth Sy., 11, 23772411, https://doi.org/10.1029/2019MS001629, 2019.

Wang, H. et al.: Aerosols in the E3SM Version 1: New developments and their impacts on radiative forcing, J. Adv. Model. Earth Syst., 12, e2019MS001851, https://doi.org/10.1029/2019MS001851, 2020.

Yang, Y., Zeng, L., Wang, H., Wang, P., and Liao, H.: Dust pollution in China affected by different spatial and temporal types of El Niño, Atmos. Chem. Phys., 22, 14489–14502, https://doi.org/10.5194/acp-22-14489-2022, 2022b.

Zeng, L., Yang, Y., Wang, H., Wang, J., Li, J., Ren, L., Li, H., Zhou, Y., Wang, P., and Liao, H.: Intensified modulation of winter aerosol pollution in China by El Niño with short duration, Atmos. Chem. Phys., 21, 10745–10761, https://doi.org/10.5194/acp-21-10745-2021, 2021.

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. Atmos., 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.

Gao, M., Beig, G., Song, S., Zhang, H., Hu, J., Ying, Q., Liang, F., Liu, Y., Wang, H., Lu, X., Zhu, T., Carmichael, G. R., Nielsen, C. P., McElroy, M. B.: The impact of power generation emissions on ambient PM2.5 pollution and human health in China and India, Environ. Int., 121, 250-259, https://doi.org/10.1016/j.envint.2018.09.015, 2018.

Gao, J., Yang, Y., Wang, H., Wang, P., Li, B., Li, J., Wei, J., Gao, M., and Liao, H.: Climate responses in China to domestic and foreign aerosol changes due to clean air actions during 2013–2019, npj Clim. Atmos. Sci., 6, 160, https://doi.org/10.1038/s41612-023-00488-y, 2023.

Navinya, C. D., Vinoj, V. and Pandey, S. K.: Evaluation of PM2.5 Surface Concentrations Simulated by NASA's MERRA Version 2 Aerosol Reanalysis over India and its relation to the Air Quality Index, Aerosol Air Qual. Res., 20, 1329–1339, https://doi.org/10.4209/aaqr.2019.12.0615, 2020.

Zeng, L., Yang, Y., Wang, H., Wang, J., Li, J., Ren, L., Li, H., Zhou, Y., Wang, P., and Liao, H.: Intensified modulation of winter aerosol pollution in China by El Niño with short duration, Atmos. Chem. Phys., 21, 10745–10761, https://doi.org/10.5194/acp-21-10745-2021, 2021.

Mishra, G., Ghosh, K., Dwivedi, A. K., Kumar, M., Kumar, S., Chintalapati, S., & Tripathi, S. N. (2021). An application of probability density function for the analysis of PM5 concentration during the COVID-19 lockdown period. Science of the Total Environment, 782, 146681.

Dey, S., Purohit, B., Balyan, P., Dixit, K., Bali, K., Kumar, A., Imam F, Chowdhury S, Ganguly D, Gargava P, & Shukla, V. K. (2020). A satellite-based high-resolution (1-km) ambient PM2. 5 database for India over two decades (2000–2019): applications for air quality management. Remote Sensing, 12(23), 3872.

Guttikunda, S., & Nishadh, K. A. (2022). Evolution of India's PM5 pollution between 1998 and 2020 using global reanalysis fields coupled with satellite observations and fuel consumption patterns. Environmental Science: Atmospheres, 2(6), 1502-1515.

Singh, T., et al. (2023). Very high particulate pollution over northwest India captured by a high-density in situ sensor network. Scientific Reports, 13(1), 13201.

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. S4) and MERRA-2 reanalysis (Fig. S5). 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.
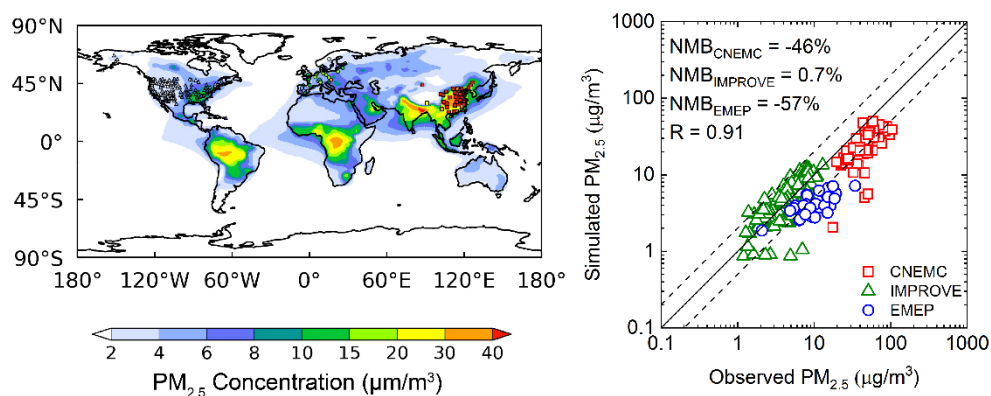
Figure S3. Spatial distribution (left panel) and scatter plot (right) between the simulated and observed annual mean near-surface $PM_{2.5}$ concentrations ($\mu g/m^3$) in 2010. Observational data are from IMPROVE (triangle), EMEP (circle) and CNEMC (square, in 2013). The solid line marks the 1:1 ratio 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.
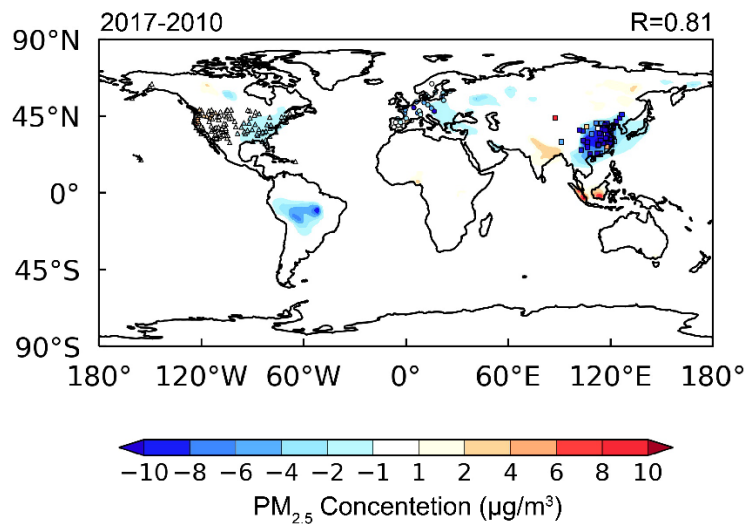
Figure S4. Spatial distribution of the change in simulated and observed annual mean near-surface PM$_{2.5}$ concentrations (µg/m³) between 2010 and 2017. Observational data are from IMPROVE (triangle), EMEP (circle) and CNEMC (square, between 2013 and 2017).
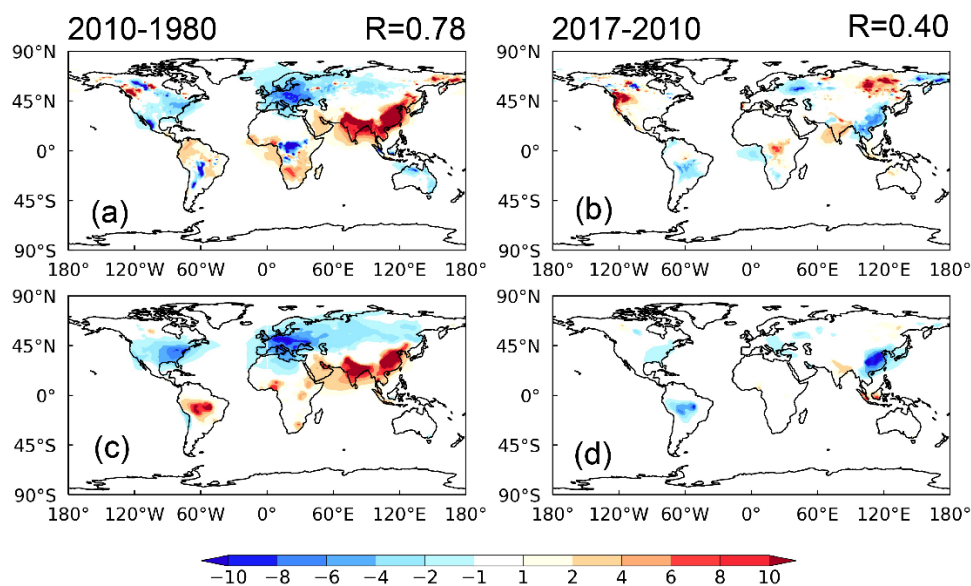
Figure S5. Spatial distribution of the annual mean near-surface PM$_{2.5}$ concentrations (µg/m³) from (a, b) MERRA-2 and (c, d) model simulation between 1980 and 2010 (left) and between 2010 and 2017 (right).

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.