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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Interannual Variability and Trends of Combustion Aerosol and Dust in Major Continental Outflows Revealed by MODIS Retrievals and CAM5 Simulations During 2003–2017" *by* Hongbin Yu et al.

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Referee #1 In this study, the authors examined the 15-year trends and interannual variabilities of dust and combustion aerosols using MODIS retrievals from 2003 to 2017, with the aid of CAM5 simulation. The goal of this paper is clearly stated in the text, and scientifically important. The tables and figures are well prepared. The approaches used is well established. I recommend publication of this paper with several minor modifications.

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Response: We appreciate the reviewer's effort of helping us improve the manuscript. Below is our point-by-point response to the comments. And we will revise the paper accordingly and submit the revised paper for further comments.

General comments: 1. The term of "combustion aerosol" may not be suitable for SOA (shown as green bars in Figures 8_11), since some SOA, like biogenic SOA, is not formed through combustion processes. The authors can either use term "pollution" as in Yu et al., (2009), or explicitly state what aerosol species in CAM5 are considered as "combustion aerosols".

You are definitely right that the "combustion" aerosol is not always suitable for SOA because of contributions by biogenic SOA in some regions. So is the "pollution" aerosol. We changed the early use of "pollution" aerosol to "combustion" aerosol, considering that mineral dust is often referred to as "pollution". The word "combustion" can at least exclude the dust.

We agree that we should state explicitly what aerosol species in CAM5 simulations are considered as "combustion" aerosol. This is done in line 236-239, page 8. The combustion AOD is "a sum of AOD of SO4 (excluding those generated from DMS chemistry), BC, POM, and SOA".

2. Why dust emissions from different regions are not tagged? From Figure 4, it seems that some regions are very likely affected by dust emitted from different regions. It may also help to diagnose the discrepancies between MODIS and CAM5 simulations as shown in Figure 12.

The natural dust and sea salt emissions are calculated online in the model. They are different from anthropogenic aerosol emissions that are provided by offline emission files. We agree with the referee that it would be interesting to diagnose dust source attribution as well, but the dust tagging, which requires additional coding, is not available in the current CAM5 model. We do plan to add the dust source tagging in the future model development.

3. In Figure 8_Figure 11, what types of simulated sulfate are considered? I am assuming sulfate formed from DMS and sulfate formed in coarse mode are excluded in the plots. Is this correct?

Sulfate in Figures 8-11 includes those from anthropogenic sources, such as industry, power plants, agricultural, residential, international shipping, surface transportation, and waste treatment emissions, open biomass burning emissions and volcanic eruptions. Sulfate formed from DMS is excluded here. Sulfate formed in coarse mode is included if it is not formed from DMS.

4. Strictly speaking, the presence of clouds affects MODIS retrievals of aerosols, but not the CAM5 simulations. Is cloud screening performed for CAM 5 analysis also?

Yes, MODIS retrievals are made in cloud-free conditions. For the CAM5 simulations, we did not perform cloud screening, because the coarse resolution of CAM5 makes it difficult to identify meaningful number of cloud-free grids. In addition, CAM5 clouds could be different from MODIS clouds, which may still complicate the comparison.

Specific comments: Line 135: please mention the exact version of CAM5 used in the study.

CAM5.3 was used in this study. It is now mentioned in the manuscript.

Line 142: I think it is called CEDS emission dataset in Hoesly et al. (2018), and the dataset is only available till 2014? Is this correct?

Correct. CEDS historical anthropogenic emission dataset from 1979Âň–2014 (version 20160726) and open biomass burning emissions from 1979–2015 (version 20161213) are used in this study. For anthropogenic emissions in year 2015, emission data are interpolated from SSP (Shared Socioeconomic Pathways) 2-4.5 forcing scenario (Riahi et al., 2017).

Riahi, K., et al.: The Shared Socioeconomic Path ways and their energy, land use, and greenhouse gas emissions implications: An overview, Global Environ. Chang., 42,

153–168, https://doi.org/10.1016/j.gloenvcha.2016.05.009, 2017.

Line 149: By tagging SO2, the source regions of sulfate aerosols can also be tracked. Is this correct? If so, please mention it in the text.

Yes. The sentence has been revised to “Aerosols and their precursor emissions, including SO2, sulfate, BC and POM, are tagged with respect to 14 source regions.”

Line 196 and Table 1: Are fc and fd derived from Figure 3 used for all 13 outflow regions? If I remember correctly, in Yu et al. (2009), different sets of fc and fd for different regions and seasons are derived. This is important since different aerosol characteristics in different regions and seasons.

We used Figure 3 derived fc and fd for all regions without accounting for their spatial and seasonal variations. This is similar to Yu et al. (2009). We don’t have adequate data to refine this assumption. But for marine aerosol fine-mode fraction fm, we do consider seasonal and spatial variations following the method described in Yu et al. (2009).

Line 336 “simulated relative contributions by sulfate, POM, BC, and SOA to the total AOD”. Should it be total AOD or ïAËŻt’c only, because the authors are trying to compare combustion AOD here. We know that, in MAM3/CAM5, SO2 can condense on accumulation mode and coarse mode and form sulfate at the same time. Are sulfate aerosols in coarse mode considered as one contributor of Tau c or Tau d?

The sentence has been revised to “simulated relative contributions by sulfate, POM, BC, and SOA to the total combustion AOD”. Sulfate aerosols in coarse mode of CAM5 are also considered as one contributor of combustion AOD. Note that sulfate converted from DMS is excluded here.

Line 416: The spherical dust assumption may explain the large difference in spring since it is dust storm season in China. However, it can not explain the large difference in June and July, since the occurrence frequency of dust storm in these two months

are not high.

We agree and the cloud contamination may be a major reason for the difference in June and July, We have rephrased the sentences: “It is thus possible that the large MODIS-CAM5 difference during the dust season (March-May) could at least be partially attributed to the spherical dust assumption in the MODIS algorithm. The higher MODIS ïĄťc in June and July is likely a result of cloud contamination and limited sampling.”

Figure 10: It is well known that anthropogenic aerosol concentrations peaks in winter season in EAS region (or China), like in Zhang et al. (2012, https://doi.org/10.5194/acp-12-779-2012). And it is well known that CAM5 fails to reproduce observed seasonality of sulfate aerosols in China. Therefore, it surprises me to see that combustion AOD in NWP does not peak in winter. What are the reasons?

We agree with the reviewer that anthropogenic aerosol concentrations near the surface in eastern China peak in winter. However, what we are looking at in this study is the column combustion AOD in East Asia outflow region. Thus at least two factors could contribute to the difference in seasonality. First, columnal AOD and surface concentration can differ in seasonality, because of the seasonal variation of the mixing layer or the atmospheric boundary layer (ABL). Aerosol is mixed within much shallower ABL in winter than in other seasons, leading to maximum surface PM concentration in China. Second, the seasonality of aerosol outflow over ocean can be largely affected by the aerosol transport and removal processes. For example, aerosol outflow from East Asia has been found to peak in spring when strong ascending airstreams lift pollutants into the free troposphere and then the westerlies carry them across the Pacific (Yang et al., 2015).

With regard to model simulations of sulfate, the default CAM5 using IPCC AR5 emissions failed to reproduce observed seasonality of sulfate aerosols in China, likely in part because IPCC AR5 emissions do not have seasonal variability in anthropogenic

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emissions. In CEDS anthropogenic emissions, seasonal feature of emissions is included, which could have improved the simulated seasonality of sulfate. However, it is possible that the model still has uncertainty in simulating two prerequisites, namely relative humidity and ozone, for sulfate formation as identified in Fang et al. (2019). Detailed analysis of CAM5 sulfate simulation is beyond the scope of this study.

References: Yang, Y., H. Liao, and S. Lou, Decadal trend and interannual variation of outflow of aerosols from East Asia: Roles of variations in meteorological parameters and emissions, Atmos. Environ., 100, 141–153, doi:10.1016/j.atmosenv.2014.11.004, 2015.

Fang, Y., C. Ye, J. Wang, Y. Wu, M. Hu, W. Liu, F. Xu, and T. Zhu, Relative humidity and O3 concentrations as two prerequisites for sulfate formations, Atmos. Chem. Phys., 19, 12295-12307, 2019.

Figure 12. and Line 483: As shown in the figure, it appears to me that the interannual variability in CAM5 simulation is much smaller compared to observations. What is the reason? How comes the nudged simulation can not reproduce observed interannual variability?

Yes, the dust interannual variability in CAM5 simulation is generally much smaller compared to the MODIS observation, except in the Mediterranean region (MED, Figure 12c).

There are many factors that can affect the interannual variability of modeled and observed dust AOD, including biases in model parameterizations of dust emissions, transport, and deposition processes, in reanalysis data used for nudging, and in satellite retrievals. Meteorological nudging used in the CAM5 simulation can only ensure realistic capture of large-scale circulations. Biases in the other aspects of the model could lead to the difference in the mean and interannual variability between model and satellite data.

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