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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simulation of the interannual variations of aerosols in China: role of variations in meteorological parameters" *by* Q. Mu and H. Liao

Anonymous Referee #1

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Review Comments on

"Simulation of the interannual variations of aerosols in China: role of variations in meteorological parameters"

General Comments: This study investigated the interannual variations (IAVs) of aerosols over heavily polluted regions in China for years 2004–2012 using the nested grid version of GEOS-Chem. The indexes of IAVs were quantified by the definitions of mean absolute deviation (MAD) and absolute percent departure from the mean (APDM), by using which the authors calculated the IAVs of simulated aerosols con-

centrations and several major meteorological variables during 2004-2012 over three regions in China. The simulated PM2.5 concentrations showed largest IAVs in North China while smallest IAVs in Sichuan Basin. In addition, the manuscript also examined the relative importance of anthropogenic emissions and meteorological parameters in altering the IAVs of aerosols by conducting some sensitivity experiments.

This manuscript presents an interesting idea to examine the IAVs of aerosols and corresponding meteorological parameters. While model and data used are reasonable, it's hard to discern "significance" from this work, as the reader got lost in the many "numbers" of the work that are presented and listed item by item that are not fitted into clear physical meanings and conclusions. My overall suggestion is to greatly reduce the number of details in listing those "numbers" from the tables, instead of and as much as possible combining the results into more discussions and conclusions. It's hard to figure out what to do with a few percentage changes in one region with this type of aerosol, and this many percentage changes in some other regions. I appreciate the authors' efforts in organizing the manuscript into several thematic sections, but it would be helpful to have discussion and statistics closer to the results and not separate them (section 2.3 and section 5).

I have a few reservations regarding the conclusions from the sensitivity experiments in this study. The paper needs to be revised to include additional details and clarifications regarding the interpretations and emissions used in the model. In several places, the presentations and discussions could be improved. All of these issues are listed in the Specific Comments section. Before this paper can be published in ACP, however, additional effort is required to clarify the significance of the findings, and the results need to be cast in a light that is useful to improve our understanding of the recent poor air quality in China. So I recommend this paper for publication in ACP after minor revision if the authors satisfactorily address all the comments and questions.

Specific Comments:

1. P11183, L3 and Table 1: Some of the emission species (e.g. CO, NOx) of Street inventory over China also include the monthly variations, which may have been implemented into GEOS-Chem, the authors should clarify it clearly whether the monthly variations have been included for NOx and SO2 emissions over China from the model code. According to GEOS-Chem version 9.1.2 that the authors used, a lot updates have been added for lightning NOx emissions as described by Murray et al., [2012], that should be clarified, too.

L5: The reference is Zhang et al., [2009].

2. P11184, L1-2: How about other nature emissions, such as lightning, biogenic, soil etc.? Perhaps specify here that these were also allowed to evolve according to the meteorology.

3. P11184, L9: I appreciate that the authors added inter annual variability using scaling factors to anthropogenic emissions from published paper. I recommend that it would be better to include the maps or Table of the IAVs of anthropogenic emissions for each species, similar to that of the aerosol concentrations and meteorological fields, which will show the comparisons of the IAVs among anthropogenic emissions, aerosol concentrations, and meteorological fields.

4. P11185, L4-12: Compared to Zhang et al. (2010), the minimum seasonal-mean surface-layer concentrations of most aerosols and PM2.5 concentrations in current study are not in JJA over eastern China while in MAM, why? Any explanations? In addition to only listing the values of the temporal and special averages, did the authors also compare current results with other previous results?

5. P11185, L19: "The simulated distributions .....to those of the emissions", what about the features of their distributions of emissions?

6. P11185, L20-22: Other than the precipitations seasonal variations, the monthly variations of anthropogenic emissions are also very important to impact on the seasonal or

monthly variations of aerosol concentrations, such as the maximum in winter. Though these monthly variations have not yet been included in some of previous studies over China, actually, the Street emissions have already provided the monthly variations for all the aerosol species, the authors should double checked whether it has been included or added into the model runs, then clarify it clearly.

7. P11185, L26-28: Did the current study use the same NH3 emissions as those of Wang et al., (2013)?

8. P11186, L4-5: Why did these species have larger IAVs over than NC than those over SC? Does it relate to the IAVs of emissions or meteorological fields? Please interpret it with more details.

9. P11186, L13-14. In what season and what domain, and why it has large IAVs? The descriptions should be clarified and interpreted clearly.

10. P11186, L18-19: From Fig. 3-4, it does not look like that the magnitude of IAVs of ammonium generally follows those of nitrate, while the ammonium is much smaller in DJF over NC and SC compared to nitrate. Meanwhile, the seasonal variations of ammonium are also quite different to those of nitrate.

11. P11186, L22-25: "BC is a chemically inert. ...parameters", what are the authors going to emphasize? Does OC include the contributions from SOA and other chemical reactions here? Normally, the OC from biomass burning is much larger than BC, which is a major source from South Asia during spring. Thus, the IAVs of biomass burning, especially over South Asia would also be an important factor that contribute to the long range transport to eastern China during spring. How much of the transport would impact on the IAVs of OC and BC over NC and SC?

12. P11187, L1-2: From Fig. 3-4, the MADs of nitrate and PM2.5 are not quite similar, especially in JJA, that the PM2.5 is almost minimum in JJA, while nitrate is almost the maximum in JJA. Could the authors explain the reasons? How much is the contribu-

tions of nitrate to PM2.5 here, please list the ratio of the ranges.

13. P11187, Section 3.3: One of the weaknesses of this study is the lack of independent measurements to verify the result IAVs, while given the paucity of long-term data available, this is not the authors' fault. It is great that the authors tried to use the MODIS long term AOD observations to validate the IAVs. However, I am surprised that why did the authors just pick up several grid boxes from the level 3 MODIS data to do the comparisons, instead of showing a map similar to Fig. 3 and Fig. 4. It is well known that the average calculations have been applied into the Level 3 data of satellite observations to generate the gridded data, which may includes some bias and errors for site by site comparisons, especially the resolution errors when compare to model results. Also, the resolution of MODIS data and GEOS-Chem are not the same here when the author picked up several grid boxes to represent the sites of the cities. I strongly recommend using the long term measurements of AERONET AOD as the observation if the authors try to validate the model results site by site.

14. P11189, Section 4.1: The authors emphasized that the wind plays an important role in IVAs of OC in section 4.2.3. Why did not also show the MAD and APDM maps of U and V wind in Fig. 7 and 8? In addition, the authors just describe these 3-4 meteorological parameters separately in section 4.1 without make corresponding discussions related to the impacts on IAVs of aerosols concentrations. It is really difficult for the readers to jump here and there to connect it by themselves. Since the study only focused on the surface aerosols, the variations of boundary layer height or boundary mixing may also be important to be considered.

15. P11189, Section 4.2: In this section, the authors listed the transport fluxes at the boundaries of the defined domain. I wonder how did the authors define the directions of the fluxes? Why all of the fluxes values are positive, and does these values include the directions? For instances, all of the vertical fluxes are positive, does it mean that there is always convections with upward movements?

16. P11193, L18: How did the authors define "wind" here? Does it only include the horizontal circulations without consider the vertical convections? But it looks like the vertical fluxes have great contributions from above descriptions.

17. P11193, Section 5: The differences of APDM between ANNmet and ANNall should represent contributions from inter annual variations of anthropogenic emissions. Meanwhile, I have reservation regarding the above differences comparison to the APDM values obtained in ANNmet to get the relative importance of anthropogenic emissions and meteorological parameters in the IAVs of aerosols. Because the system is not completely linear, especially for those chemical reactions which is highly sensitive to the meteorological parameters. Thus, it is not surprised to get such large contributions from meteorological parameters. So it may not be reasonable to conclude the contribution from meteorological parameters using this method. A comparable way would be only keep the same meteorological parameters for a specific year (such as 2006, to be consistent with ANNmet) with other emissions varied inter annually, then getting the differences to ANNall would show the contribution from meteorological parameters.

Even though the authors did not show the IAVs of the anthropogenic emissions, I don't expect that the inter annual variations of the anthropogenic emissions would make large contributions to the IAVs of aerosol concentrations since it may not be as large as the meteorological IAVs. It may only enhance or weaken the IAVs of aerosols. It also should be noted, from the definitions of either MAD or APDM, they can only tell us the magnitudes of inter annul variations, including both positive and negative variabilities. Therefore, it suggests that this method is not the combined effects of to reflect the increasing or decreasing variations. The authors should interpret the associations between the meteorological parameters and aerosol concentrations more clearly since they are not always direct proportions.

18. P11194, L14-16: the differences between ANNmet_ATM and ANNmet do not only represent the IAVs of aerosols caused by variations in meteorology-sensitive natural emissions. Actually, it should represent the differences with or without natural emis-

sions. That is completely different descriptions and conclusions.

19. P11179, L16: these equations would be much easier to read if they were actually typeset as equations rather than inline text.

20. It would be nice if the abstract could end with a sentence regarding the broader impacts and significance of this work, perhaps with regards to the effectiveness of air quality control strategies in China.

21. P11181: Is this the first IPR analysis in GEOS-Chem? Is it computationally easy to implement? If not, could the original implementation and discussion of how this is calculated be cited?

22. It is interesting that the ANNmet_ATM experiment was designed with met-sensitive natural emissions turned off, rather than being just held constant at 2006 values. Was there a reason for this design choice? Were there any concerns regarding nonlinearity of the model response to turning emissions completely off?

23. L11185, L26: Kharol et al. (2013) demonstrated that the persistent nitrate in GEOS-Chem in China is, overall, as much linked to high NOx emissions as it is to high NH3 emissions.

Figure 5: In Chengdu, why is the MODIS AOD have a dip in values nearly every year in months 10 – 1 when the model AOD is high and often peaking?

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