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Responses to Reviewer #1

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 concentrations 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.

Response:

We have addressed the major issues mentioned in the general comments: (1) We now clarify the significance of our findings in the abstract and in the

Introduction section. We have added one sentence at the end of the abstract: “Considering that the IAVs in meteorological fields are mainly associated with natural variability in the climate system, the IAVs in aerosol concentrations driven by meteorological parameters have important implications for the effectiveness of short-term air quality control strategies in China”. We have also added the following sentences at the end of the first paragraph of the manuscript: “Understanding interannual variations in aerosols driven by variations in meteorological parameters is especially important for air pollution control. For example, the Action Plan for Air Pollution Prevention and Control released by the State Council of China in year 2013 aims to reduce the annual mean $PM_{2.5}$ concentrations in the regions of Beijing-Tianjin-Hebei, Yangtze Delta, and Pearl River Delta by 25%, 20%, and 15% respectively, as the concentrations in year 2017 are compared with those in 2012. The role of interannual variations in meteorological parameters needs to be separated from the impact of the reductions in emissions in these targeted reductions.”

(2) We have performed an new sensitivity study ANNemis to examine the impact of variations in anthropogenic emissions on interannual variations of aerosols. Anthropogenic and biomass burning emissions are allowed to vary from 2004 to 2012. Meteorological parameters and hence natural emissions are kept at the year 2006 values. Note that biomass burning emissions are partly anthropogenic and partly natural. We allow biomass burning emissions to vary over 2004-2012 in all simulations, so that comparison of ANNmet and ANNemis tells us the relative importance of variations in meteorological parameters and anthropogenic emissions in IAVs of aerosols.

(3) We have shortened our descriptions of numbers of MAD and APDM in Sections 3.2, 4.1, and 4.2, and tried to give more physical meanings of these values.

Our point-to-point responses to the reviewer’s comments are listed below.

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].
Response:

We do consider monthly variations in SO_2, NO_x, and NH_3 in our simulations. We have clarified in Section 2.2 that “Monthly variations in SO_2,

NO_x, and NH_3 follow those in Wang et al. (2013)".

We have cited "Murray et al. (2012)" in the description of lightning NO_x.

Zhang et al. (2009) has been added as the reference for David Streets' emission inventory.

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.

Response:

We have revised the sentence as "Meteorological fields, natural emissions, and biomass burning emissions were allowed to vary from 2004 to 2012, while anthropogenic emissions were kept at the year 2006 values."

3. P11184, L9: I appreciate that the authors added interannual 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.

Response:

The IAVs of anthropogenic emissions were the same at all the grid cells in China, since we implemented the same annual scaling factors for all grid cells. Based on the annual scaling factors taken from Zhang et al. (2012) and Lu et al. (2011), the annual total emissions of NO_x, SO_2, OC, and BC in China had APDM values of 7%, 5%，3%, and 5%, respectively, over years 2004–2012. This is now described in Section 5 of the revised manuscript where we describe the model results of the new ANNemis simulation.

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?

Response:

The major reason for the differences in seasonal variations presented here and those in Zhang et al. (2010) is that the aerosol concentrations showed in Zhang et al. (2010) were averaged over eastern China (110°–120°E, 20°–45°N). If we average our aerosol concentrations over the same region (110°–120°E, 20°–45°N), we also obtain minimum aerosol concentrations in JJA.

We have added in the text the comparisons with other studies: "Our simulated seasonal variations in sulfate concentrations agree well with those in

Wang et al. (2013).” Sulfate exhibited a maximum in JJA and a minimum in MAM in NC, whereas minimum concentrations in JJA and maximum values in DJF in SC and SCB.

5. P11185, L19: “The simulated distributions : : :: : :.to those of the emissions”, what about the features of their distributions of emissions?
Response:
We have rewritten the sentence as “The simulated distributions of OC and BC were similar to those of their emissions, with the highest values in NC”.

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.
Response:
We do consider monthly variations in SO_2, NO_x, and NH_3 in our simulations. However, anthropogenic emissions of OC and BC have no monthly variations in the GEOS-Chem model. As a result, the seasonal variation in precipitation is the major reason for simulated seasonal variations of OC and BC. Our assumption of no monthly variations in BC and OC emissions should not influence the simulated interannual variations in these two aerosols reported in our study.

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

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.
Response:
These results were from simulation ANNmet, driven by variations in meteorological parameters alone.

9. P11186, L13-14. In what season and what domain, and why it has large IAVs? The descriptions should be clarified and interpreted clearly.
Response:
This sentence is an expansion of the description of the averaged IAVs. Based on the definitions of MAD and APDM, the MAD and APDM values

presented are averages over the studied years. We just want to note that year by year variations can be larger than the averages reported here. Here we do not intend to present specifically the year by year variations in aerosols in different seasons and regions.

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.
Response:

Thanks for pointing this out. We have revised the description of ammonium as "The distribution and magnitude of the APDM values of ammonium generally followed those of nitrate over polluted eastern China."

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?
Response:

We have revised the sentence as: "Because BC is a chemically inert tracer, the IAVs in BC obtained in ANNmet were caused by the variations in transport and deposition."

About the consideration of SOA, we have the following sentences in Section 2.1 where we describe the GEOS-Chem model: "Considering the large uncertainties in chemistry schemes of SOA, SOA in our simulations was assumed to be the 10% carbon yield of OC from biogenic terpenes (Park et al., 2003) and 2% carbon yield of OC from biogenic isoprene (van Donkelaar et al., 2007)."

As described in Section 2.3, interannual variations in biomass burning emissions have been considered in all of our simulations; the impact of long-range transport is included in our results.

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 contributions of nitrate to PM2.5 here, please list the ratio of the ranges.
Response:

That sentence was indeed confusing. We were talking about APDM instead of MAD values. We have deleted the sentence to avoid confusion.

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.

Response:

We had the same thoughts as the reviewer at the beginning of our study; we tried to show a map similar to Fig. 3 and Fig. 4 using MODIS AOD. However, most of the grid cells had missing MODIS AOD data during 2004-2012. Also, as shown in our Fig.5, MODIS AOD cannot represent well the IAVs of aerosol concentrations in SC and SCB. If we present such a map of the IAVs of MODIS AOD with those of aerosols, the results would be misleading to the readers. With respect to the AERONET AOD Level 2.0 Quality Assured Data, AERONET has only two stations in China covering the years 2004-2012, in which only Beijing station is within the studied regions (NC, SC, and SCB).

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.

Response:

It would be difficult and confusing to show MAD and APDM maps of U and V wind, since winds have directions (both positive and negative values). For example, the averages of winds are needed for the calculation of MAD and APDM values. Either assuming winds with directions or using absolute values of wind speed to calculate the averages of winds would be scientifically confusing. It is better to show MAD and APDM values of transport fluxes, as

we do in Figs. 9–14.

The variations of boundary layer height (BLH) are indeed important by influencing the transport of aerosols. Since we present the variations in transport fluxes at the boundaries of the defined domains when we perform process analyses in Figs. 9-14, the impacts of variations of BLH are already accounted for.

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?

Response:

The values of seasonal transport fluxes in the model can be positive or negative according to directions. Since $\%PC_i$ of aerosols are determined by MAD_i (equations (5) and (3) in the manuscript), absolute values of transport fluxes are used in process analyses.

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.

Response:

Here wind includes the horizontal circulations as well as the vertical convections.

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.

Response:

Following the reviewer's suggestion, we have performed an new sensitivity study ANNemis in the revised manuscript to examine the impact of variations in emissions alone on interannual variations of aerosols. All anthropogenic and biomass burning emissions are allowed to vary from 2004 to 2012, and meteorological parameters and hence natural emissions are kept at the year 2006 values.

Model results from ANNemis show that, in NC and SC over 2004–2012, the variations in emissions alone have smaller impacts on interannual variations of aerosols than the variations of meteorological parameters alone. We note at the end of the Conclusion section that the changes in anthropogenic emissions on longer time scales may lead to linear trends in simulated aerosol concentrations (Yang et al., 2014). For studies on longer time scales, the MAD and APDM values need to be calculated after detrending the time series, following the approach used in previous studies that examined interannual variations in ozone concentrations (Camp et al., 2003), sea surface temperature, partial pressure of CO_2 (Gruber et al., 2002), sea level pressure (Thompson et al., 1998), and North Atlantic Oscillation index (Jung et al., 2003).

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 emissions. That is completely different descriptions and conclusions.

Response:

We agree with the reviewer that the differences between ANNmet_ATM and ANNmet represent the differences in IAVs with and without natural emissions. This is now clarified in the text.

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

Response:

Changed as suggested.

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.

Response:

Thanks for the suggestion. We have added the following sentence at the end of abstract: “Considering that the IAVs in meteorological fields are mainly associated with natural variability in the climate system, the IAVs in aerosol concentrations driven by meteorological parameters have important implications for the effectiveness of short-term 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?

Response:

This is indeed the first IPR analysis in the GEOS-Chem model and it is easy to implement, since the GEOS-Chem model has outputs of most of the processes used in IPR analysis.

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?

Response:

We agree with the reviewer that it is better to have the met-sensitive natural emissions held constant at year 2006 values in ANNmet_ATM. However, it is much easier to turn them off without leading to wrong conclusions in ANNmet_ATM. If all natural emissions do not lead to large interannual variations of aerosols, the perturbations in natural emissions as a result of the variations in meteorological parameters would have even smaller impacts. The model adds up natural and anthropogenic emissions as total emissions, so this design does not cause nonlinearity problems.

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.

Response:

We have cited the study of Kharol et al. (2013) and noted that high nitrate in China is linked to both high NO_x emissions and high NH_3 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?

Response:

The dip of MODIS AOD occurred among October-January in some of the

years examined. Considering the special topography (basin with surrounding mountains) and weather conditions in winter (wintertime clouds and fog events at high frequency) in Chengdu, the quality of MODIS AOD dataset in Chengdu was not as reliable as those in Beijing and Changsha. Based on the ground measurements we could find from the literature, observed aerosol concentrations in Chengdu during 2006-2007 did not exhibit obvious low aerosol concentrations in October-January (Zhang et al., 2012).

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Responses to Reviewer #2

This study simulates the surface PM2.5 (and its major compositions) concentrations in China during 2004-2012, with the goal of understanding the processes affecting its interannual variability (IAV). The authors showed, through model sensitivity experiments, that regional wind and precipitation were the principle factors driving the IAV of surface PM2.5 concentrations.

In my opinion, this is an important topic that will help us understand the relationship between meteorology/climate and air quality. The topic is of great interest to the community and suitable for publication in ACP.

However, there are some important flaws in the current manuscript, which would require major revision before it can be published in ACP. The most critical flaw in this manuscript is the lack of discussion of the implications from the results of the sensitivity experiments. This greatly dimishes the value of the paper. Also, many details of the experiments were not clearly described. I would suggest reducing the too-lengthy reporting of numbers and instead focus on the implication of the results.

Response:

We have addressed the major comments in the following aspects:

(1) We now clarify the significance of our findings in the abstract and in the Introduction section. We have added one sentence at the end of the abstract: “Considering that the IAVs in meteorological fields are mainly associated with natural variability in the climate system, the IAVs in aerosol concentrations driven by meteorological parameters have important implications for the effectiveness of short-term air quality control strategies in China”. We have also added the following sentences at the end of the first paragraph of the manuscript: “Understanding interannual variations in aerosols driven by variations in meteorological parameters is especially important for air pollution control. For example, the Action Plan for Air Pollution Prevention and Control released by the State Council of China in year 2013 aims to reduce the annual mean $PM_{2.5}$ concentrations in the regions of Beijing-Tianjin-Hebei, Yangtze Delta, and Pearl River Delta by 25%, 20%, and 15% respectively, as the concentrations in year 2017 are compared with those in 2012. The role of interannual variations in meteorological parameters needs to be separated from the impact of the reductions in emissions in these targeted reductions.”

(2) We have tried our best to reduce the too-lengthy reporting of numbers in Sections 3.2, 4.1, and 4.2 and to focus on the implication of the results.

Our point-to-point responses to the reviewer’s comments are listed below.

Specific comments:

P11183 Line 3: ’... in which anthropogenic emissions ...’: This wording is confusing. Did Park et al. (2004) used the Streets 2006 emission inventory over Asia? Or was the Streets inventory used to overwrite the emission inventory of Park et al. (2004) over Asia in this work? Please clarify.

Response:

We have rewritten the sentences as “Global emissions of aerosol precursors and aerosols in the GEOS-Chem model generally follow Park et al.

(2003) and Park et al. (2004). Anthropogenic emissions of NO_x, SO_2, BC, and OC (including emissions from power, industry, residential, and transportation) in the Asian domain are overwritten by David Streets' 2006 emission inventory (http://mic.greenresource.cn/intex-b2006) (Zhang et al., 2009) in this work."

P11183, Line 3-6: David Streets' 2006 emission inventory should be cited as Zhang et al. (2009)
Response:
Cited as Zhang et al. (2009).
.
P11184, Line 4-7: Were natural emissions turned off completely? Or were the natural emissions kept at levels without interannual variability (this seems like the better approach for the authors' purpose)? Please clarify.
Response:
We agree with the reviewer that it is better to have the met-sensitive natural emissions held constant at year 2006 values in ANNmet_ATM. However, it is much easier to turn them off without leading to wrong conclusions in ANNmet_ATM. If all natural emissions do not lead to large interannual variations of aerosols, the perturbations in natural emissions as a result of the variations in meteorological parameters would have even smaller impacts.

P11184, Line 14-21: This paragraph is confusing. Please consider revising to make the significance of the experiments clearer.
Response:
We have clarified the purpose of experiment ANNmet_ATM by saying that "We performed one sensitivity simulation ANNmet_ATM with natural emissions turned off. The differences between ANNmet and ANNmet_ATM represent the differences in IAVs with and without natural emissions."

Section 3.2: The report of MAD and APDM values in this section is somewhat tedius and confusing. I would suggest that, instead of reporting values, it would be more useful to simply refer to Table 2+3 and Fig 3+4 and then point out the implication of these values. Also, please avoid non-meaningful sentences. E.g., (line 22) ".. MAD or APDM of OC was similar to that of BC..." and (lines 25-26) "The APDM values of BC were about the same as those of OC".
Response:
These are very good suggestions. We have removed description about MAD values and are now focused on presenting APDM values. We have changed as suggested to describe the APDM values of BC as "The APDM values of BC were about the same as those of OC."

P11188, Lines 10-13: The high correlation between observed and simulated AOD over Beijing is many driven by the model's ability in reproducing the

annual cycle. Looking at Fig 5, the high correlation does not indicate the model's ability at reproducing the observed IAV in NC, as the authors postulated.

Response:

We also have comparisons of the IAVs at the end of this paragraph: "The IAVs of observed AODs agreed fairly well with the IAVs of surface-layer aerosol concentrations. For example, the seasonal-mean APDM values of observed AODs were 18% (DJF), 15% (MAM), 24% (JJA), and 16% (SON) for Beijing, close to the seasonal-mean APDM values of surface-layer PM2.5 shown in Fig. 4."

Section 3.3: Why is Changsha chosen to represent Southern China?

Response:

There are two reasons: (1) Changsha is a metropolis with high emissions and aerosol concentrations in our simulations, and (2) Changsha is an inland city with relatively more accurate satellite measurements than coastal cities such as Shanghai and Guangzhou.

Section 3.3: The authors show that AOD, column PM2.5 burden, and surface PM2.5 concentration in NC (Beijing) are all higher in summer and lower in winter. This is somewhat surprising, as it seems to contradict surface PM2.5 measurements (e.g., Zhang X. Y. et al. (2012)). What is the cause of this discrepancy? Did the authors plot the simulated wet or dry PM2.5 concentrations in Fig. 5?

Response:

According to the measurements of Zhang et al. (2012), concentrations of all aerosol species in northern China were the highest in winter and lowest in summer. Our simulated $PM_{2.5}$ concentrations were the highest in summer because of our simulated high nitrate aerosol concentrations in summer. We have discussed about this in Section 3.1: "Wang et al. (2013) reported that high nitrate in the GEOS-Chem model is likely caused by the overestimate of NH_3 emissions, and Kharol et al. (2013) demonstrated that the persistent nitrate in GEOS-Chem in China is, overall, as much linked to high NO_x emissions as it is to high NH_3 emissions."

Wet and dry deposition should not be the reasons for the discrepancies, since we could reproduce well the seasonal variations of BC and OC aerosols.

Both simulated and MODIS AOD values were the highest in summer, because of the large water uptake by aerosols in this season. Qi et al. (2013) also showed the highest aerosol AOD in summer over North China during 2006–2009 using satellite measurements.

Section 3.3: What is the cause of the bi-modal feature in the annual AOD cycle over SC (Changsha)? This same feature is seen in the surface PM2.5 many SC sites in Zhang X. Y. et al. (2012). The model was unable to reproduce this feature, which the authors attributed to topographical complexity and cloud

contamination of satellite AOD retrieval near Chengdu. However, the fact that several SC sites show the same surface PM2.5 annual cycle is indicative of a more regional mechanism at work.

Response:

Zhang et al. (2012) showed the bi-modal feature in PM_{10} aerosol concentrations in 2006-2007 in southern China. Their speciated measurements indicated that the bi-modal feature could be explained by seasonal variations in mineral dust and sulfate, the two aerosol species with the largest contributions to local PM_{10} concentrations. Observed mineral dust concentration was the maximum in spring and had the second peak in autumn. For sulfate aerosol, strong photochemistry facilitated sulfate formation in spring, summer, and fall, but prevalent precipitation in southern China in summer led to enhanced wet removal of sulfate in that region.

We simulated $PM_{2.5}$ concentrations over 2004-2012 in our study and the model was unable to reproduce the bi-modal feature, indicating that future studies are called for to improve aerosol simulations in southern China. Issues that need to be examined include the seasonal variations in emissions, simulation of mineral dust, and the uncertainties in assimilated precipitation fields.

Section 4.1: Again, instead of reporting values, I would suggest that the authors discuss the implications of the IAV of various meteorological parameters. Also, I do not think it make any sense to compare with Piao et al. (2003) or Qian and Lin (2005). The IAV of meteorological variables for a different periods do not necessarily have to be the same.

Response:

We have removed description on MAD values and added the following sentences to discuss the implications of the IAVs of various meteorological parameters: “The variations in temperature and specific humidity can influence chemical reactions of sulfate, nitrate and ammonium, while those in precipitation are important for wet deposition of all aerosol species. The relatively large APDM values of these meteorological parameters in DJF suggested large IAVs of aerosols in this season.”

Climatologically, IAVs in meteorological parameters are mainly associated with natural variability in the climate system. The magnitude of IAV of a meteorological parameter should be about the same even if different time periods are considered.

Section 4.2 and Figs 9-14: This section is not well explained. How were these budgets constructed? Are the budgets presented in Figs 9-14 based on the standard simulation ANNmet? If so, then this should be clearly indicated in the text and in the caption. Or did the authors conducted multiple sensitivity tests to isolate the contribution of each of the processes shown in Figs 9-14? If the latter, then the authors should be better describe the sensitivity experiments

conducted in the Methods section.

Response:

We have added the following sentences in Section 4.2 to describe how the budgets were constructed: “For an aerosol species, the budget (mass flux from each process) was constructed for the selected region considering the mass balance of this aerosol. Chemical production and removal, transport, as well as wet and dry deposition of the aerosol were diagnosed at every time step and summed over each season in simulation ANNmet.”

Figs 9-14 are based on simulation ANNmet and we have indicated this in the text and in figure captions.

Throughout: What is the value of showing both the MAD and APDM in all analyses? If there is value in showing both, the authors should better explain the implication of the differences between MAD and APDM.

Response:

We have described the differences between MAD and APDM in the second paragraph of the introduction section. If we are examining aerosol concentration (or flux), MAD represents the absolute IAV in aerosol concentration (or flux), and APDM represents the IAV relative to the average concentration (or flux) over the studied years.

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Responses to Reviewer #3

This manuscript presents modeling analysis to quantify the interannual variations (IAVs) of aerosol species over China during the period 2004 to 2012 driven by meteorological parameters. Three sets of simulations were conducted, all with varying meteorology but differ in treatment of emissions: fixed anthropogenic emissions at 2006 levels (ANNmet), turning off natural emissions that are coupled with meteorology, varying anthropogenic emissions. Through comparing the IAVs of the three sets of simulations, the authors conclude that the IAVs of aerosols in China are mainly caused by meteorology, rather than by natural emissions or by anthropogenic emissions. IAVs of different processes are diagnosed from the model simulations (with fixed anthropogenic emissions) and the processes with largest IAVs are selected as the key processes that drive the IAVs of different aerosols. Gas-phase formation of sulfate is found to drive the IAVs of sulfate over NC, while the gas-to-aerosol partitioning of nitrate is the major factor leading to large IAVs of nitrate in China.

Overall the paper presents interesting new analysis of the driving factors of aerosols variations in China. The paper is well organized and thoroughly written. It is suitable for publication in ACP once several revisions have been made, as described below.

Major comments:

1. Section 3.3 comparison of simulations with MODIS AOD: this section evaluates the simulated IAVs of aerosols with AODs retrieved by MODIS. The evaluation was based on the ANNmet simulation (i.e., simulations with fixed

anthropogenic emissions), but the MODIS AOD should reflect both changes in meteorology and emissions. Therefore, the ANNall simulation (i.e., simulations with changing anthropogenic emissions) should be better suited for comparison with MODIS AOD.

Response:

As shown by the title of the manuscript and the title of Section 3, we are mainly focused on examining the impacts of variations of meteorological parameters on interannual variations (IAVs) of aerosols. Our results from Section 5 also show that variations in emissions have smaller impacts on the IAVs than variations in meteorological parameters in NC and SC. To address the reviewer's concern, we now also show in Figure 5 the simulated AOD from simulation ANNall.

2. Although this manuscript is focused on IAVs of aerosol in China, some of the conclusions can be strengthened by expanding the analysis to the changes in absolution concentrations of aerosols by different factors/processes, or at least discussing this aspect in Section 4 or 5. The impact of the large increases of anthropogenic emissions in China on aerosols is completely neglected in the manuscript, although the authors have conducted simulations with increasing anthropogenic emissions. I understand that the authors based their analysis on the IAVs, since the IAVs of increasing anthropogenic emissions are comparable to the IAVs of fixed anthropogenic emissions. Still, to discuss the different role of meteorology factors and anthropogenic emissions on aerosols in China will strength the analysis of the paper and have a bigger impact.

Response:

The changes in absolute concentration of an aerosol by different factors/processes are represented by the mass budget of the aerosol (Figs. 9, 11, 13). We have added the following sentences in Section 4.2 to describe how the budgets were constructed: "For an aerosol species, the budget (mass flux from each process) was constructed for the selected region considering the mass balance of this aerosol. Chemical production and removal, transport, as well as wet and dry deposition of the aerosol were diagnosed at every time step and summed over each season in simulation ANNmet."

As suggested by the reviewers, we have performed an new sensitivity study ANNemis to examine the impact of variations in emissions alone on interannual variations of aerosols. The improved discussions on the different roles of meteorological factors and anthropogenic emissions on aerosols in China are presented in Section 5.

Minor comments:

1. pg 11180, line 5-7: check the spelling of MODIS and MISR.

Response:

We have checked the spellings.

2. pg 11181, line 21: should be regional-scale GEOS-Chem model since the manuscript uses the nested-grid capability of GEOS-Chem.

Response:

The nested-grid GEOS-Chem is a global model with East Asia zooming in.

3. pg 11187, line 1: what's the faction of nitrate in PM2.5? How does this compare with observations?

Response:

That sentence was confusing. We were talking about APDM instead of MAD values. We have deleted the sentence to avoid confusion.

4. pg 11182, line 17-20: not sure how dusts and sea salts are treated in the simulations. Are they allowed to vary with meteorology in all the three simulations? Are their natural emissions turned off in the ANNmet_ATM simulation? Do the PM2.5 results presented in the paper include dust or sea salt? This needs to be clarified.

Response:

We have described in Section 2.1: "We do not examine IAVs of mineral dust and sea salt aerosols in this study, because sea salt aerosol is not a major aerosol component in China based on measurements (Ye et al., 2003; Duan et al., 2006) and mineral dust aerosol simulation has very large uncertainties (Fairlie et al., 2007, 2010)."

We have defined at the beginning of Section 3.1 that $PM_{2.5}$ is the sum of sulfate, nitrate, ammonium, OC, and BC.

References:

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