Response to Reviewer #1

This paper discusses the results of simulations for 2000 and 2050, with a focus on China and transboundary transport. It thoroughly discussed the characteristics of the simulated changes between 2000 and 2050, differentiating between emissions and climate impacts.

Major comments:

1. This is a very complete analysis but the reader is little information on how could relate to other models and/or other scenarios. There are no discussions on how the present simulations compare to other simulations; it is clear that specific analyses are made here, but it would still be valuable to put the results of the present study into a bigger context.

Response:

Thanks for the suggestion. We have added the following comparisons of simulated climate-induced changes in aerosol concentrations in the revised manuscript: "Our simulated climate-induced changes in aerosol concentrations are comparable in magnitude with those reported in other studies for other regions. Based on the IPCC A1B scenario, Tagaris et al. (2007) found a 10% decrease in $PM_{2.5}$ throughout the United States and Pye et al. (2009) reported changes in annually averaged sulfate-nitrate-ammonium of up to 0.61 μg m^{-3} in the United States , as a result of 2000-2050 climate change alone. Under the IPCC A2 scenario, climate change over 2000-2050 alone was found to reduce $PM_{2.5}$ concentrations in the United States by −0.9 μg m^{-3} in the study of Avise et al. (2009). Juda-Rezler et al. (2012) reported that from1991-2000 to 2091-2100 PM_{10} over Central-Eastern Europe generally decrease, by up to 1.5 μg m^{-3} in large scale simulations and up to 3.5 μg m^{-3} in fine scale simulations under the IPCC A1B Scenario."

Although a number of observational and modeling studies have examined the present-day long-range transport of aerosols into and from East Asia, no previous studies, to our knowledge, have examined future changes in aerosol transport (as we summarize in the introduction section of manuscript). We have now comparisons of simulated present-day aerosol transport with previous studies in the manuscript (Section 5.1.1).

2. My other comment relates to the use of 3 years and no assessment of statistical significance. For many studies in which climate and chemistry are coupled, a simulation length 3 years is not to ascertain that the shown differences are not simply a reflection of noise. While it is clear that changes in emissions are large, it is still unclear how that translates into changes in concentrations downwind and aloft. I would strongly urge the authors to extend the length of the simulations if at all possible as it strongly undermines the significance of the presented results.

Response:

Following the suggestions of both reviewers, we have extended the length of simulations to 10 years in the revised manuscript. We perform simulations

for four cases: (1) year 2000 climate and emissions, (2) 2050 climate and 2000 anthropogenic emissions of aerosol precursor and aerosols, (3) 2000 climate and 2050 anthropogenic emissions of aerosol precursor and aerosols, and (4) 2050 climate and emissions. Each case is integrated for 10 years (driven by 1996–2005 meteorological fields to represent year 2000 climate or by 2046–2055 meteorology to represent year 2050 climate) following 1 year of model spin-up. All the results presented in this paper are 10-year averages.

With model results from the 10-year simulations, we have performed and presented statistical significance tests for 2000-2050 changes in meteorological fields (Figures 1-4), concentrations of aerosol species (Figures 7-9), and mass fluxes of $PM_{2.5}$ (Figures 11-13) based on the student's two sample t-test.

For the case with changes in emissions alone, since changes in emissions are mostly imposed near the surface, the vertical changes in concentrations of aerosols in the lower troposphere generally follow the sign of changes at the surface-layer, as shown by the altitude-latitude cross-sections of the changes in aerosol concentrations averaged over eastern China (Figure A below). Changes in concentrations above 2 km altitude can be influenced by long-range transport. Because our manuscript is already very long, we do not show such plots of vertical changes in concentrations in the manuscript. Instead, we show future changes in long-range transport owing to changes in emissions alone in Figs. 11-13 of the manuscript. The changes in aerosol concentrations downwind in the US as a result of the changes in emissions alone have been reported in Pye et al. (2009); we are focusing on the changes in aerosol concentrations in China in this study.

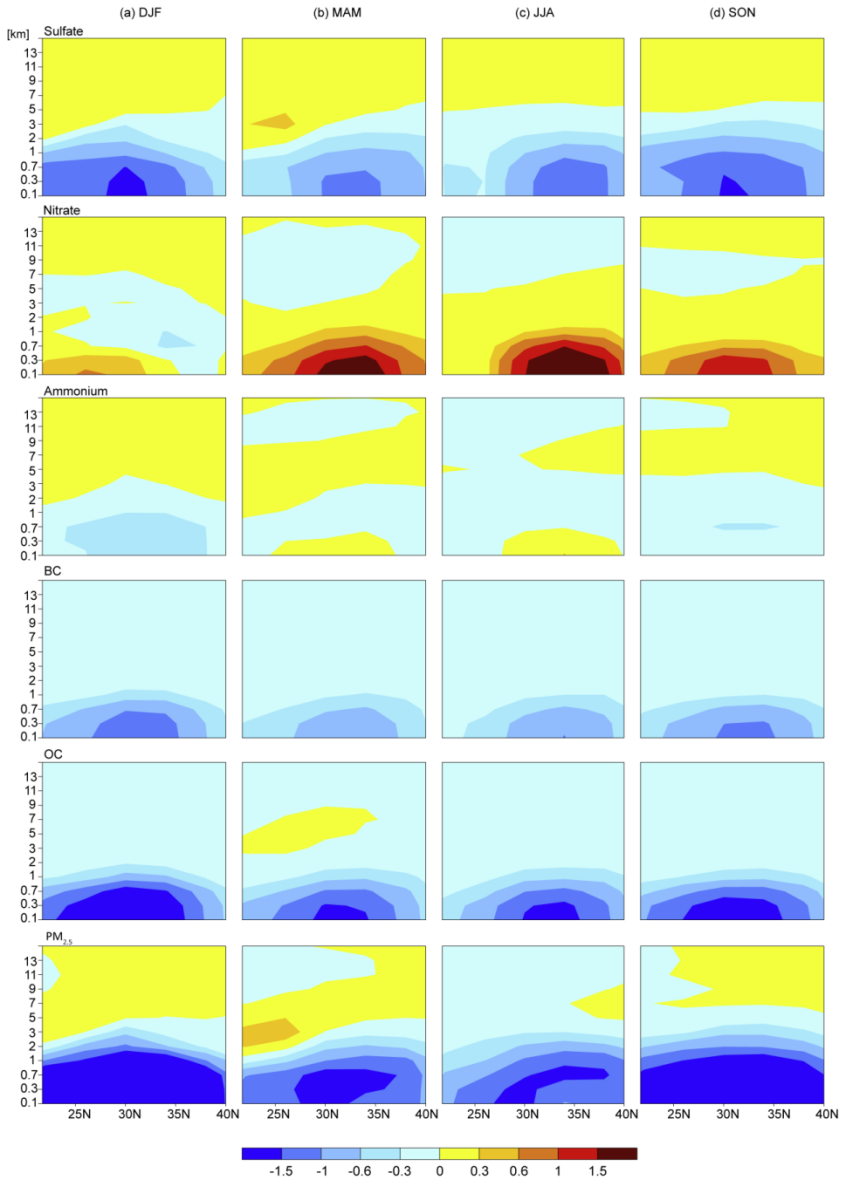

Figure A. The altitude-latitude cross-sections of the changes in aerosol concentrations from the present day (1996-2005) to future (2046-2055) owing to changes in anthropogenic emissions alone. Values are averaged over longitudes from 115°E to 120°E.

Minor comments:

1. Page 6509, line 5: could you be more specific than “fairly well”? A presentation of the actual results would be useful.

Response:

We have added the following sentences to be more specific (see Figures B and C below for our comparisons): “Both the simulated and assimilated surface air temperatures show higher temperatures in southern China than in northern China and also higher temperatures in eastern China than in western China. The maximum temperatures in southeastern China are simulated to be 280–290 K in DJF, 290–300 K in MAM, 300–310 K in JJA, and 290–300 K in

SON, which agree with the assimilated values in all seasons except that the assimilated maximum temperatures in southeastern China are lower than 305 K in JJA. Simulated zonal winds averaged over 100°–120°E longitudes show maximum wind speeds of the jet stream located at 200 hPa altitude of 60, 40, 20, and 30 m s^{-1} in DJF, MAM, JJA, and SON, respectively, which agree closely with the assimilated values."

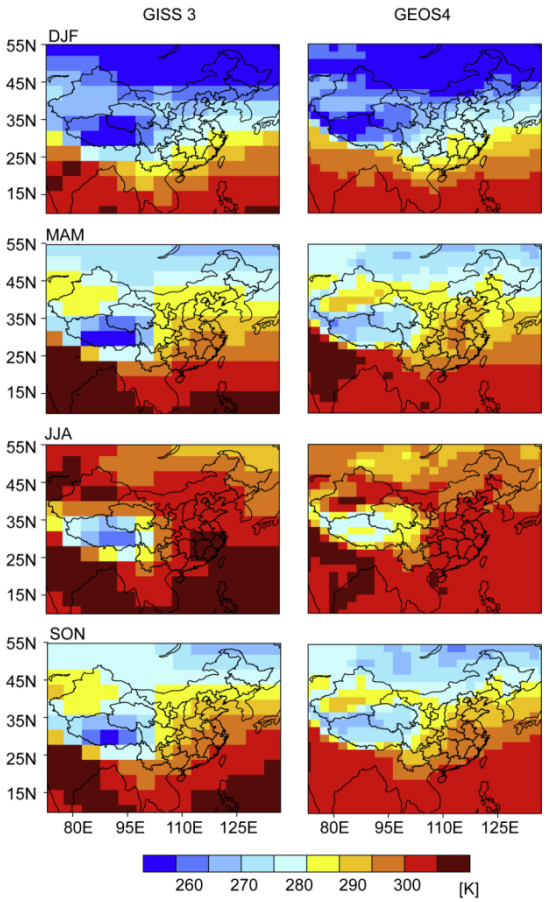

Figure B. Comparisons of simulated (GISS Model 3) and reanalyzed (GEOS-4) surface air temperatures (K) in China for present day (averages over 1996-2005).

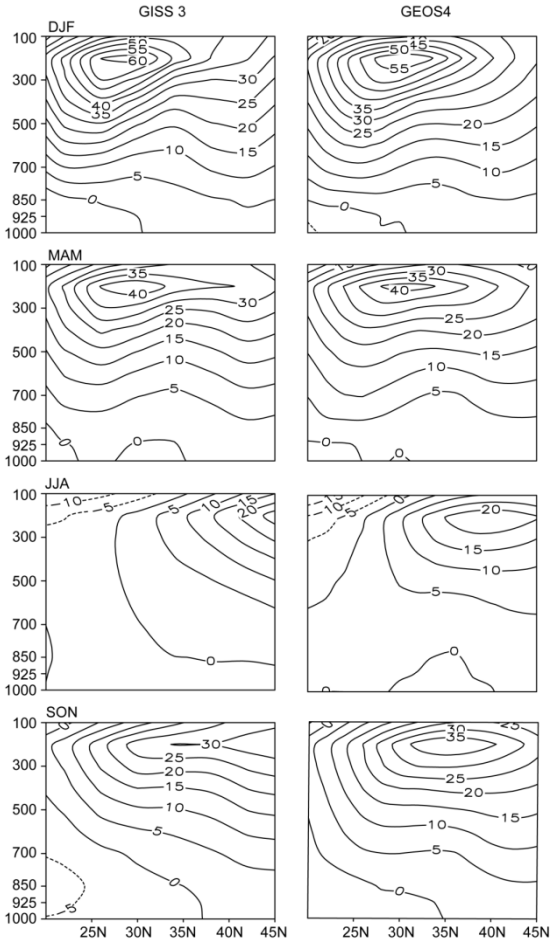

Figure C. Comparisons of simulated (GISS Model 3) and reanalyzed (GEOS-4) zonal winds (m s^{-1}) for present day (averages over 1996-2005). To represent winds over eastern China, winds are averaged over the longitude range of 100°-120°E.

2. Page 6510, line 8-9: why is the PBL depth predicted to decrease. This seems counterintuitive to a warming world.

Response:

The changes in PBL depth result from the simulated changes in atmospheric temperature (or atmospheric stability). As an example, we show in Figure D the 2000-2050 changes in SON air temperatures averaged over 110-120°E and 32-40°N (this area shows large reductions in PBL depth in Figure 4 of the revised manuscript in SON). The simulated increases in temperature in the 500-700 hPa altitude are larger than those in the lower troposphere, leading to a more stable atmosphere and hence a lower PBL depth in 2050. It should be noted that the negative changes in PBL depth were also found for the US in Pye et al. (2009), with the changes in PBL also simulated by the GISS Model 3.

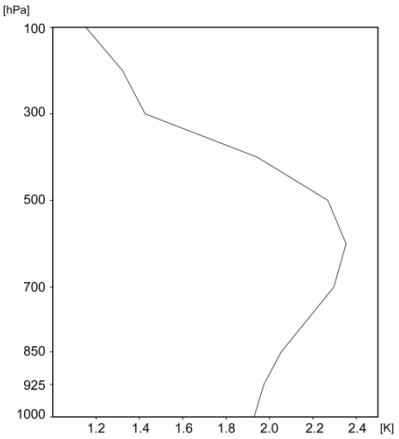

Figure D. The simulated 2000-2050 changes in SON air temperatures averaged over 110-120°E and 32-40°N.

3. Page 6510, line 15: this seems to contradict the above statement on decreasing PBL height. Explain or rephrase.

Response:

There might be a typo in either page number or line number. Line 15 on Page 6510 is “2.4 Simulations” in the ACPD version of the manuscript.

4. Page 6511, line 5: it would be nice to show those results, since it is likely that the model setup is not exactly the same as in the listed publications.

Response:

This comment is about our presentation of simulated O_3. Following the suggestion of the other reviewer, all the presentations on O_3 have been removed in the revised manuscript.

5. Page 6512, line 8: how is the meteorology for the nested domain generated from the climate simulations.

Response:

The nested-grid simulation in Wang et al. (2013) was driven by the assimilated meteorological fields from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). We have now clarified this in the text.

6. Page 6513, line 14: this is clearly a place where the analysis could be place in the context of other climate simulations (as in CMIP3 for example).

Response:

This comment is about our explanation of simulated changes in O_3. Following the suggestion of the other reviewer, all the presentations on O_3 have been removed in the revised manuscript.

7. Page 6514, line 6-7: show comparison of precipitation with observations or at least reanalysis.

Response:

We have compared the present-day precipitation with reanalysis in Section 2.3: "The present-day precipitation simulated by the GISS model shows larger values in MAM and JJA than in DJF and SON, which agree with the assimilated precipitation, but the model overestimates precipitation in the middle and lower reaches of the Yangtze River in MAM whereas underestimates precipitation in that region in JJA."

In this paragraph mentioned by the reviewer, we are describing future changes in aerosol concentrations by future changes in climate alone. Future changes in precipitation can only be compared with simulations from other models. We have mentioned in section 2.3 that "Note that the projected patterns of precipitation changes (the increases in precipitation in northern China in DJF and the increases in precipitation in eastern China in JJA) from the GISS Model 3 generally agree with those from the IPCC AR4 multi-model predictions for China under the A1B scenario (IPCC, 2007)".

8. Page 6514: Figures 7 and 8 should be on the same scale to ensure comparability.

Response:

Figures 7, 8, and 9 show the changes in aerosol concentrations owing to climate change alone, changes in emissions alone, and changes in both climate and emissions, respectively. Since the effects of climate change alone are much smaller than the effects of changes in emissions, we could not see spatial and temporal details of the changes in aerosol concentrations in Figure 7 anymore as we tried to use the same scale in Figure 7 as in Figure 8. We thus decide to retain the scale in Figure 7, and then use the same scale in Figures 8 and 9.

9. Page 6518: from this section on it would be interesting to discuss the potential rates of nonlinearities between the separate and combined impacts of emissions and climate changes.

Response:

Following the reviewer's suggestion, we have added a new column in each of Tables 3-5 to show the rate of nonlinearity defined as *NonL*=(Change in flux by climate change alone + Change in flux by changes in emissions alone)/Change in flux by changes in both climate and emissions. Values of *NonL* of seasonal and annual fluxes of aerosols mostly deviate from 1.0 (perfect linearity).

Response to Reviewer #2

This manuscript estimates the impact on ozone and aerosols over China in 2050 compared to 2000, and tries to separate the impacts of emission changes and climate change. Understanding and presenting the future impact of climate change and emission changes on different regions is important for policy makers, so this is an important area of research. I have not seen the collection of observation data over China in the supplementary section before, so this could be valuable by itself.

My major concern with the work presented is that only 3 years of simulation is used to calculate the effects of climate change, which I expect means that internal variability will be a large component of the climate change signal they use.

Note: because of the limited number of years I did not carefully read the detailed claims in manuscript.

I recommend several other significant changes too, detailed below.

The quality of the written English is excellent.

Overall, this manuscript is potentially a good paper, so I recommend major changes be made before publication.

Major comments:

1. I expect that many of the spatial and seasonal climate change results presented are strongly affected by internal variability. I recommend either a) the simulations are extended for many more years (dependent on the type of ocean model used) or b) the interannual variability in the model is analyzed to prove that this is not important (3 data-points per location and season will give poor estimates of variability, but should be enough to give a rough estimate).

Response:

Following the suggestions of both reviewers, we have extended the length of simulations to 10 years in the revised manuscript. We perform simulations for four cases: (1) year 2000 climate and emissions, (2) 2050 climate and 2000 anthropogenic emissions of aerosol precursor and aerosols, (3) 2000 climate and 2050 anthropogenic emissions of aerosol precursor and aerosols, and (4) 2050 climate and emissions. Each case is integrated for 10 years (driven by 1996–2005 meteorological fields to represent year 2000 climate or by 2046–2055 meteorology to represent year 2050 climate) following 1 year of model spin-up. All the results presented in this paper are 10-year averages.

With model results from the 10-year simulations, we have performed and presented statistical significance tests for 2000-2050 changes in meteorological fields (Figures 1-4), concentrations of aerosol species (Figures 7-9), and mass fluxes of $PM_{2.5}$ (Figures 11-13) based on the student's two sample t-test.

2. I recommend specifying the type of ocean model used in the climate model (specified sea-surface-temperatures, slab ocean model, full dynamical ocean model). This has a large impact on the interannual variability in a climate model, and hence the length of simulation required.

Response:

We have clarified this in Section 2.3 that "The GISS Model 3 was coupled with a ''Q-flux'' ocean as described in Wu et al. (2008)". In the "Q-flux" model, monthly mean ocean heat transport fluxes were calculated iteratively for present-day climate to reproduce observed sea surface temperatures. These ocean heat transport fluxes were then held fixed, while sea surface temperatures and ocean ice were permitted to respond to changes in climate (Wu et al., 2008).

3. The role of ozone in the manuscript is unclear: is the manuscript about aerosols, or aerosols and ozone? It is not mentioned in the title, but it is discussed in the background section, briefly in the validation section, and briefly in the results sections, but not the boundary transport section. As I am sure the authors are aware, ozone is complicated because it depends on the non-linear balance between several different species and cloudiness (that affects photolysis), among other factors. However, the manuscript has only a cursory discussion and presentation about ozone. I suggest the authors consider whether to a) expand their material on ozone, b) delete the ozone sections and focus on aerosols, or c) explain the inclusion of ozone (eg, to help validate the model). I also suggest the authors consider whether to add "ozone" to the title.

Response:

Thanks for pointing this out. Following the reviewer's suggestion, we have deleted all ozone sections and are now focused on aerosols.

4. A lot of the data is effectively presented in tables. I therefore suggest that it is not necessary to repeat information from the tables in the main text, unless it is being used to make a specific scientific point. This will significantly shorten the manuscript.

Response:

Following the suggestion, we have removed some of the descriptions about the tables from the text to shorten the manuscript.

5. The changes in emissions and estimated concentrations are both presented. Since the concentration of many species is strongly driven by its main emissions, extra scientific value would be gained by directly comparing the changes in emissions and estimated concentrations to show the extent to which the change in concentration is non-linearly related to the change in the main emission for each specie.

Response:

We add the discussions on changes in emissions and estimated concentrations in section 4.3: "With reductions in SO_2, BC, and OC in eastern China over 2000-2050 by 28.5%, 58.6%, and 39.8% (Table 1), respectively, annual mean concentrations of sulfate, BC, and OC over eastern China are simulated to be reduced, respectively, by 19.2%, 56.4%, and 35.9% with future

changes in emissions alone whereas by 18.5%, 56.6%, and 36.5% with future changes in both climate and emissions. These results indicate that changes in concentrations of these aerosols are strongly driven by the changes in emissions."

6. I found no mention in the manuscript for how methane was handled in the simulations. This will be important to understanding the simulation results, especially for ozone and secondary aerosols. Since methane is also a greenhouse gas, I recommend that it be described how methane was handled for both a) radiative forcing in the climate simulation and b) the chemistry reactions.

Response:

We have added the following sentences in Section 2.2 to clarify: "For both radiative forcing in climate simulation and chemical reactions, present-day methane levels in the model are based on observations and set to 1750 ppb with a 5% inter-hemispheric gradient (Wu et al., 2008). The future (2046-2055) methane level in GEOS-Chem follows the IPCC A1B scenario and is set to 2400 ppb for simulations in which changes in anthropogenic emissions are considered (Pye et al., 2009)."

7. Section 6 (conclusion and discussion). This section seems to be mostly a summary of the facts and figures presented in the tables and elsewhere. I recommend this section be significantly shortened, and be used to highlight the main scientific understanding gained from this work. The authors might also consider dropping 'discussion' from the section title.

Response:

We have shortened the conclusions to just highlight the main scientific understanding gained from this work and have removed "discussion" from the section title.

8. Figure 6 shows that the model aerosol concentrations are often significantly different to observations. I recommend that the authors comment on how this impacts their conclusions.

Response:

As mentioned in the text, one of the factors that lead to our underestimates of aerosol concentrations is that "the measurements are usually taken in urban areas, whereas the simulated values represent grid cell averages". We do acknowledge in the text the low biases in emissions inventories of BC and OC. Following the reviewer's suggestion, we now comment in the text that "The low biases in BC and OC emissions may lead to underestimates of outflow of carbonaceous aerosols from eastern China in our study."

9. Figure 10. I would normally expect boundary planes to form a box to help close the budget. Hence I would like to see an explanation for why the

boundaries are disjoint. I am particularly interested in why the planes do not cover the India border when section 4.2 indicates it is important for changes over the Himalayan Plateau.

Response:

Since the national border of China is not in a regular shape, the boundary planes are chosen to represent the places with the largest fluxes of aerosols, so that we can capture the major features of transboundary transport of aerosols into and out of China. If the boundary planes form a box, the southern boundary plane will extend to the East China Sea and capture fluxes of aerosols that are not transported into China. Also, if we extend the southern boundary to India, the examining of aerosol fluxes in and out of China would have a problem since a large fraction of aerosol transport may occur within India.

10. The supplementary material with the aerosol observations over China is potentially very valuable. It would enhance the paper to provide some details on how to use it. For example: how far above the ground are they measured? What time of day are the measurements made? Are they hourly averages? daily averages? Could the data be provided with the paper in a spreadsheet file or some other data format?

Response:

We have added a new column of "Notes on measurements" in each of supplementary tables to give the above details the reviewer asked for. We also note at the end of the supplementary tables that "A spreadsheet file of aerosol measurements can be obtained by contacting the corresponding author of the paper".

Minor comments:

1. Consider deleting 'and associated transboundary transport' from the title to shorten it.

Response:

As we summarize in the Introduction section of the manuscript, many previous studies have examined present-day long-range transport of aerosols but few studies have examined future changes in transboundary transport. Future changes in transboundary transport of aerosols are new findings that we want to emphasize in this work, so we think it is better to have this information in the title.

2. Delete 'to' on p6503/line 25.

Response:

Deleted.

3. On p6504/lines 7-8, put the list of aerosol species in brackets rather than

commas.

Response:
Changed as suggested.

4. Often the change in aerosols is only given in ug/m3. I suggest the authors consider adding the percentage changes too.

Response:
We have added the percentage changes in places that we present changes in aerosols.

5. p6506/line 18, I think it should be 2051.

Response:
Changed to the years of simulations (2046-2055) performed in the revised manuscript.

6. p6506, line 20, I suggest adding 'the' before 'present study'.

Response:
Added.

7. p6507, line 11, I suggest changing the '/' symbols to commas.

Response:
Changed.

8. I suggest moving section 2.4 to immediately after section 2.1 (or including it in section 2.1).

Response:
Changed as suggested.

9. p6514, lines 3-7. Is it possible the increase in aerosol could be caused by increased natural emissions?

Response:
As temperatures are higher in 2050, BVOC emissions increase by 10-30% in southeastern China in MAM and JJA, but this has a small impact on OC concentrations, since higher temperatures do not favor SOA formation.

10. Section 5. Over what altitude range was the transboundary flux calculated? If it includes the stratosphere, then it would be good to comment on the effect this has (if any).

Response:

We have clarified in the text that "These fluxes of aerosols are calculated through 3 vertical planes from the surface to 100 hPa altitude."

11. Section 5. Is the transboundary flux calculated within the model every timestep, or is it calculated using time averaged winds and concentrations in the output?

Response:
We have clarified in the text that "The transboundary fluxes are calculated within the model at every time step and the seasonal and annual values are presented here."

12. P6526, lines 1-3. I find this sentence ambiguous. Does it mean 'climate change is important to air quality and long-range transport', or 'climate change and long-range transport are important to air quality'? I suggest rephrasing this sentence.

Response:
We have revised the sentence as "Results from the present study indicate that climate change is important to domestic air quality in China and long-range transport of aerosols."

13. p6534, line 1. I suggest adding 'anthropogenic' before 'emissions'.

Response:
Added.

14. The acronym 'NMVOC' does not appear to be defined in the manuscript.

Response:
We have now defined NMVOCs as non-methane volatile organic compounds in the manuscript.

15. Table 2. The acronym 'HC' does not appear to be defined in the manuscript.

Response:
"HC" in Table 2 has been replaced by "hydrocarbons".

16. Tables 3-5. I assume the units should actually be Tg/year. It would also help to clarify whether the data is for the mass of the whole species, or just one element (as is done in tables 1-2), eg Tg(S)/year, or Tg(SO4)/yr.

Response:
We have clarified in Table captions of Tables 3-5: "The units are Tg season^{-1} for seasonal fluxes and Tg yr^{-1} for annual fluxes of whole species (e.g., Tg(SO4) season^{-1}, Tg(SO4) yr^{-1})."

17. Fig 3a. It would help to clarify whether the cloud fraction is a true fraction (1=total overcast cloud) or a percentage.

Response:

We have clarified in the figure caption that the cloud fraction is a true fraction (1=total overcast cloud).

18. Supplementary tables. I suggest clarifying the aerosol sizes of the data (I think they are all PM2.5).

Response:

We have added a new column of “Notes on measurements” in each of supplementary tables with information about aerosol size.

References

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