Response to referee comments on "Response of winter fine particulate matter concentrations to emission and meteorology changes in North China"

We thank the reviewers for valuable comments. This document is organized as follows: the referees' comments are in black and our responses are in blue.

To Referee #1

This paper investigated the responses of $PM_{2.5}$ concentrations to changes in emissions (incl. SO_2, NO_x, NH_3, BC and OC) and meteorology (incl. T, RH and wind speed) for a severe haze season in North China. Multiple sensitivity cases along with the baseline scenario in January 2010 were conducted with an online coupled meteorological and chemical transport model (i.e., WRF-Chem). The result suggests that dramatic changes in emissions are main cause of the increasing haze event in north China, and the winter $PM_{2.5}$ is largely contributed by SO_2 and NH_3 emissions, as well as wind speed and aerosol feedbacks. The study is an important and very worthwhile exercise. Publication of the manuscript is recommended with minor revisions as suggested.

General Comments:

Detailed WRF configuration and emission processing is necessary. For example, there is little information regarding the WRF parameterization, nudging, how the feedback was set up. It is also necessary to provide basic details of model domain, particularly for the vertical profile. How the emissions were located vertically across the PBL? How the emissions and meteorological variables were adjusted in the sensitivity runs at different vertical heights?

Responses: Thanks for these great suggestions. We used same WRF parameterization as Gao et al., ACP, 2016 except the innermost 9km domain was not included here; The vertical profile used is the WRF default 27 vertical pressure profile; Analysis nudging was used for the outer domain; The feedback information was included in the method session: "In this study, we used a configuration that includes direct and indirect feedbacks."; The emissions were assigned to 6 layers from surface based on sources, for example emissions from large point sources (like chimneys) were assigned to higher layers; In the sensitivity runs, emissions and meteorological variables were uniformly adjusted at vertical layers.

We have added the following descriptions in the revised manuscript:

"Two nested domains with 81km and 27km horizontal grid resolutions from outer to innermost and 27 vertical grids were used (Figure S1 in supplementary material). Analysis nudging of meteorology variables was used for the outer domain. The model physics configurations also follow the settings in Gao et al. (2016)."

“We assigned emissions to the first 6 layers from surface based on sectors. For example, emissions from large point sources (such as chimneys) were assigned to higher layers.”

“At different vertical heights, emission and meteorological variables were uniformly perturbed.”

Besides, discussion about the limitation of this study is necessary, such as lack of proper SOA simulation, no consideration of VOC and primary PM emission other than BC/OC.

Responses: Thanks for these valuable suggestions.

About the lack of proper SOA simulation, we used the MADE/SORGRAM SOA scheme to investigate SOA during the same period in Gao et al, ACP, 2016 and found that it is not significant during winter haze, which might not be correct. However, current understanding of SOA is still limited and current SOA schemes largely underestimated SOA, especially during winter. Thus, we did not include it in this study.

Actually, VOC is considered in the model (“The anthropogenic emission inventory used is the MACCity (MACC/CityZEN EU projects) emissions dataset, which provides monthly CO, NO_x, SO_2, VOC, BC, OC, and NH_3 emissions from different sectors for years between 1960 and 2020).

We added the following paragraph to address limitations:

In our previous modeling study of the same period (January 2010), we found that SOA contribution was small, so we did not include SOA in this study. But this indication might be problematic due to current poorly parameterized SOA scheme. In the future, how changes in emissions and meteorology variables affect productions of SOA during winter should be further studied using more advanced SOA schemes. In addition, we did not consider primary PM except BC and OC in the model because there is no information in the MACCity emission inventory, which is another direction for improvements in future studies.

The simulation period was just one month in winter, results might be different in other seasons. The 13 simulation cases need to be discussed in the text, although they’ve been well summarized in Table 1. I would suggest to elaborate them in the method section.

Responses: We agree that the simulation in other seasons might be different, but it is beyond the scope of this paper. This paper focuses on winter haze pollution. We have elaborated the following descriptions of 13 simulation cases in method section.

“CTL case uses emissions for year 1960 and EMI2010 case uses emissions for year 2010. SO_2, NH_3, and NO_x emissions were perturbed separately from 1960 to 2010 (i.e., SO_2-2010 NH_3-2010 NO_x-2010 cases). In the CTL_NF and EMI2010_NF cases, aerosol-radiation interactions are excluded based on emissions for year 1960 and 2010.”

“reflect conditions of early decades (CTL_T2, CTL_RH10, CTL_WS20, EMI2010_T2, EMI2010_RH10, and EMI2010_WS20 cases).”

Specific Comments:

Page 5 Line 9-10, some of the previous studies also used online coupled model, e.g., Wang et al ERL 2014, please clarify it.

Responses: Thanks for this point. The focus of Wang et al ERL 2014 is aerosol feedbacks, while we were trying to say that previous studies about responses of $PM_{2.5}$ to changes in emissions and meteorology use offline models. Our expression (The models used in previous studies referenced above) might be confusing. So we changed the sentence to “The models used in previous studies of emission and meteorology perturbation referenced above”. Hope it is clear now.

Page 7 Line 16, did those changes apply to all vertical layers or just the ground? Is there any evidence to support those numbers?

Responses: Thanks for this good question. These changes are uniformly applied to all vertical layers. The evidence are documented in those papers listed in line 11-14, Page 7. (i.e., It was pointed out surface air temperature in North China increased at the rate of 0.36 ℃ per decade (Guo et al., 2013), the linear trends coefficient of relative humidity anomaly in North China is about -0.60% per decade (Wang et al., 2004), and national mean wind speed decreased 16% in the recent 50 years (Wang et al., 2004)).

Page 9 Line 12, elevated sources such as power plants and industry boilers are even more important than near-ground sources in China.

Responses: We agree with this suggestion. We have plotted total column emissions and updated Figure 1 and the changing factors. We changed the sentence to “In general, the domain averaged NO_x emissions in North China increased by ~990% from 1960 to 2010.”

Page 10 Line 5, there is no data about natural dust provided in Table 2.

Responses: In Table 2, the sum of sulfate, nitrate, ammonium, OC and BC are smaller than $PM_{2.5}$. The differences mostly come from natural dust. Thanks for pointing this out. We have added this description in Page 10 Line 5 (natural dust (the difference between PM2.5 and the sum of sulfate, nitrate, ammonium, BC, OC) to make it clear.

Page 10 Line 16-17, does that mean SOA is not important in China? It seems problematic.

Responses: The simulated using MADE-SORGRAM shows that it is not important in winter in North China, which agrees with the previous SOA simulations in China (Jiang et al., 2012: Regional modeling of secondary organic aerosol over China using WRF/Chem). Currently, SOA is not well represented in the model due to incomplete understanding of SOA, so it might be problematic. We have added one paragraph in the summary session to mention this limitation.

Page 12 Line 10-11, please elaborate the reason.

Responses: We have added the reason: "due to trivial NO_x emissions".

Page 12 Line 11-12, NH_3NO_3 should be NH_4NO_3. The sentence is confusing. Does that mean north China exhibits NH3 poor condition in winter?

Responses: Thanks for this correction. We have changed it to NH_4NO_3. That means NH_3 is relatively poor compared to NO_x. NH_3 also reacts with sulfuric acid to form $(NH_4)_2$SO4, so it may not be sufficient given the large amounts in SO_2 emissions. In addition, $NH_{3\ \mathrm{emission}}$ is lower than in other seasons because NH_3 is mainly from agriculture and agriculture activity is reduced in winter.

Page 15 Line 20-21, please elaborate the reason, e.g., changes in T vertical profile?

Responses: Thanks for this great suggestion. We have added the reasons. "The monthly domain average daytime PBLHs decrease about 2.3% due to changes in temperature vertical profiles."

Page 16 Line 8-9, a little confusing...but PM shows clear increases in Beijing.

Responses: The amounts in changes are relatively small compared to PM concentrations in Beijing. We used daily mean threshold 35 and 75μg/m3 to define haze days. Due to T perturbation, the numbers of haze days do not show significant changes.

Page 19 Line 20, the sensitivity represents the response per unit change, I suppose here it means the response to total changes. Please clarity it.

Responses: Thanks for pointing this out. We have changed the expressions to "$PM_{2.5}$ shows more notable increases in response to changes in SO_2 and NH_3 as compared to increases in response to changes in NO_x emissions". We also changed expressions of sensitive in other places.

Page 20 Line 10, how was the prediction conducted?

Responses: The predictions were conducted by perturbating emissions by those amounts. We have added the description ((by perturbating SO_2, NO_x and NH_3 emissions by -26%, 19% and 14%)) to make it clear.

Figure 1, I suggest to present total emissions in column rather than at surface only.

Responses: Thanks for this suggestion. We have changed the Figure to total emissions in column.

Figure 2, is it based on the meteorology in Jan 2010?

Responses: Yes, it is based on the meteorology in Jan 2010.

Figure 4, negative scale is too large to show any difference, please consider to modify it.

Responses: Thanks for this good advice. We have modified the scale.

Response to referee comments on "Response of winter fine particulate matter concentrations to emission and meteorology changes in North China"

We thank the reviewers for valuable comments. This document is organized as follows: the referees' comments are in black and our responses are in blue.

To Referee #2

Overall I'd rate this manuscript as minor revisions - some rewriting is required, but I have no major concerns, aside from the missing SOA in the regional model. This is a significant omission - not so big that I'd require the authors to repeat their simulations with this fixed, but they definitely need to explain its absence and how this is justified.

My largest concern: Page 10, lines 14-15. I find the omission of a secondary organic aerosol formation mechanism in the model a concern, given the large amount of work in the literature showing the importance of this process towards net aerosol production. The authors reference Gao et al 2016 mentioning its impact is small – this reasoning should be repeated here. It's potentially a significant omission, given the increases in the region's VOC emissions – what are the unique local conditions that allow the authors to justify its omission? Given the availability of several different parameterizations for SOA formation in the literature, why did the authors not ("just") include one in their model?

Responses: Thanks for this important point. The simulated SOA using MADE-SORGRAM in my previous study showed that it is not important in winter in North China, which agrees with the previous SOA simulations in China (Jiang et al., 2012: Regional modeling of secondary organic aerosol over China using WRF/Chem). In Jiang et al. 2012, it was pointed out that high SOA in summer is due to high biogenic VOCs' emissions and intensive photochemical reactions (higher temperature and radiation in summer), and SOA is larger in South China. However, photochemical reaction is very weak in this study period due to high aerosol loadings (stagnant weather conditions in winter) and weak biogenic VOCs in winter. SOA is very complicated and not well understood, so the results from this SOA scheme might be problematic. Given current SOA modules is not good and the CBMZ-MOSAIC in WRF-Chem does not consider SOA, we did not include it in this study. We have added one paragraph in the summary session to mention this limitation.

"In our previous modeling study of the same period (January 2010), we found that SOA contribution was small, so we did not include SOA in this study. But this indication might be problematic due to current poorly parameterized SOA scheme. In the future, how changes in emissions and meteorology variables affect productions of SOA during winter should be further studied using more advanced SOA schemes. In addition, we did not consider primary PM except

BC and OC in the model because there is no information in the MACCity emission inventory, which is another direction for improvements in future studies.”

Not a "concern" per se, but something worth pointing out to the readers and perhaps investigating a bit further. Three points, all related:

(1) Page 7, lines 14 through 17: Later in the discussion the authors mention the manner in which this was done, by perturbing the initial and boundary conditions of the individual meteorological components to create these changes. This should be mentioned here as well, in a single sentence. This methodology later seems to result in a response from the fully coupled model which counteracts the meteorological perturbations. Some discussion of the mechanisms by which these counteracting effects takes place would be warranted.

Responses: Thanks for this great point. We have added a single sentence description here. “These were conducted by perturbing the initial and boundary conditions of these individual meteorological variables”.

For temperature and RH, the responses are generally consistent with perturbations (domain mean 2K increase for temperature and 9.3% decrease for RH). For wind speeds, we can see from Figure 9(a-b) that the monthly mean wind changes are in the opposite direction of the wind fields in base case, which is consistent with the decreasing perturbations. The responses of PM to decreases in wind speeds are also consistent, with increasing PM in highly polluted region and decreasing PM in northwest (when wind speeds are low, natural dust is low).

(2) Sections 3.3, 3.4, 3.5. The impact of these changes in the meteorology initial and boundary conditions may themselves be partially due to the response of the aerosols through feedbacks; affecting radiative transfer, etc. When the IC and BC temperature increases, the domain temperature decreases (section 3.3). When the IC and BC RH increases, the domain RH decreases (section 3.4). When the IC and BC wind speeds increase, the domain wind speeds decrease (question: section 3.5: I assume that the first sentence should read “carried out when initial and boundary condition wind speeds were increased”? Please explain in more detail how the winds were perturbed, and whether this was surface or 3D winds.

Responses: Thanks for this great question. It seems that the reviewer misunderstands our presentation here. We perturbed temperature, RH and wind speeds to represent them in 1960s.

In section 3.3, “we decrease temperature by 2 ℃ in the initial and boundary conditions to reflect conditions more like 1960. As a result, the monthly domain mean surface temperature increases 2.0 ℃”. So, from 1960 to 2010, when IC and BC temperature increase, the domain temperature increases 2.0 ℃. We increases RH in IC and BC to represent 1960. So, from 1960 to 2010, IC and BC RH decreases, the domain RH decreases. For wind speeds, the case is similar. IC and BC wind speeds decrease from 1960 to 2010, and domain wind speeds decrease.

We agree that these changes are partially due to aerosol feedbacks, but the changing directions are generally consistent with the perturbations.

We have added "in initial and boundary conditions" to the first sentence of sect. 3.5. We added one sentence in sect. 2.2 to explain the perturbation in detail: "At different vertical heights, emission and meteorological variables were uniformly perturbed." The wind speeds were perturbed at different heights, not just surface.

(3) Each of these meteorological perturbations to the initial and boundary conditions resulted in a model response which acted counter to that change. Would the authors concur that the feedback meteorological system is acting to damp or counteract meteorological perturbations? This is something worth mentioning in the paper, along with how the feedbacks act this way (e.g. temperature increases leading to increases in the type of aerosols which reduce surface temperatures, etc.).

Responses: As I responded in point (1) and (2), the model responses are consistent with meteorological perturbations. We agree that feedbacks would partially affect meteorology, but it might not counteract meteorological perturbations. We have added sentences to mention this point in the revised manuscript.

Relatively minor issues:

Page 2, lines 19-20. The statement regarding long-term trends in atmospheric circulation potentially being important due to $PM_{2.5}$'s sensitivity to wind speed and aerosol feedbacks is unclear; it's not clear how the latter imply the former. I'm not sure, having scanned through the paper, that the authors have made a good case for that connection.

Responses: Since wind direction and speeds are mostly driven by position and intensity of large scale systems, such as Siberian High. Aerosol feedbacks change PM through suppressed PBL, while PBL height is also caused by the dominant large scale system (such as abnormal high at higher layers). This is just an indication for future studies.

There are frequent references to Gao et al 2016 (perhaps intended as a companion paper) – for the benefit of those who do not have this paper, things like "domain 2 in Gao et al, 2016) (page 7 line 19) should be given more explicitly in this work (e.g. by showing the region on one of the figures). Similarly (Page 9, line 22), readers unfamiliar with the geography of the region might benefit from some symbols with the locations of the cities and regions mentioned appearing on the maps (maps are too small for names appearing on the maps themselves)

Responses: This is really a good suggestion. We added map and description in the revised supplementary material for readers.

Page 3, line 22: would be better as "increases sulfate concentration due to the temperature dependence of SO2 oxidation and resulting higher SO2 oxidation rates".

Responses: Thanks for this suggestion. We have changed the sentence to this better form.

Page 6 line 16: Please describe how the VOC emissions are speciated into CBM-Z VOCs in this description. That is, a speciation profiles must have been used – are these specific for different emissions sources, more generic, etc.? Or are the emissions data used already pre-speciated into the individual VOCs required for the model's chemical mechanism?

Responses: The VOC emissions were speciated into CBMZ VOCs by referring the mapping information in (Meng Li et al., ACP, 2014, Mapping Asian anthropogenic emissions of non-methane volatile organic compounds to multiple chemical mechanisms) and the definitions of each species in the inventory and CBM-Z scheme. We use a generic distribution for different sources since detailed information is limited in the inventory.

Page 8 line 19. There is recent work by McLinden et al in Nature Geoscience (May 2016), which uses satellite-based estimates of SO2 emissions to show regions and particular large sources which have been underreported in emissions inventories. Do the regions this reference shows have underestimates in SO2 production spatially correspond to the regions the authors of the current work have shown have underestimates in sulphate? If so, this would be worth mentioning.

Responses: Thanks for mentioning this interesting new paper. In fact, we used surface SO_2 measurements to evaluate model and found SO_2 emissions might have been overestimated in the study region. In addition, we doubled SO_2 emissions and found it does not significantly increase sulphate. Thus, we don't think errors in SO_2 emission is the main cause.

Figure 3 discussion on page 10: to what extent do boundary conditions account for these changes? How much do the boundary conditions change between the two simulations? This should be discussed in the manuscript.

Responses: Results in Figure 3 are based on data in the innermost domain, which takes boundary conditions from the outer domain. To quantify the impact of changes in the outer domain, we simulated another case with the innermost domain emission fixed in 1960 and outer domain emission changed to 2010. The impacts of boundary conditions mostly occur around the south boundary, and show nearly no impact on PM2.5 in Beijing. We added one figure and discussion on Page 10.

"To quantify how much of the changes in Figure 3 are from the impacts of boundary conditions, we simulated another case with the innermost domain emissions fixed in 1960 and the outer domain emissions changed from 1960 to 2010. The impacts of boundary conditions mostly occur around the south boundary and show nearly no impact on $PM_{2.5}$ in Beijing (shown in Figure 4), which are consistent with the continuous weak southerly winds during the study period (Gao et al., 2016). On domain average, the impacts of boundary conditions result in 5.0ug/m^3 increase in the study domain, accounting for about 33.9% of the total changes in $PM_{2.5}$."

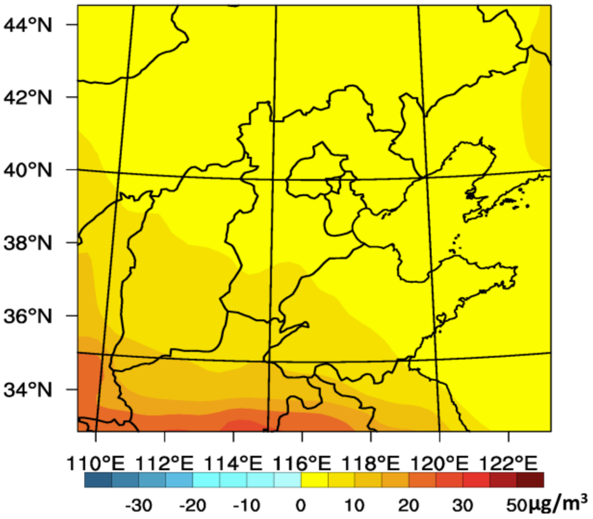

Figure 4 Changes of $PM_{2.5}$ due to boundary conditions from outer domain

Page 12, line 18 to Page 13, line 11: Please include a plot of VOC:NOx ratios at the start and the end of the period to show how the ratio has changed in response to the emissions changes.

Responses: Thanks for this great suggestion. We have added the plots of VOC:NOx ratios at the start and the end of the period. Before emission changes, the domain maximum VOCs: NOx ratio is 16.2 and domain mean ratio is 4.2. After emission changes, domain maximum ratio is 2.8 and domain averaged ratio is 1.2.

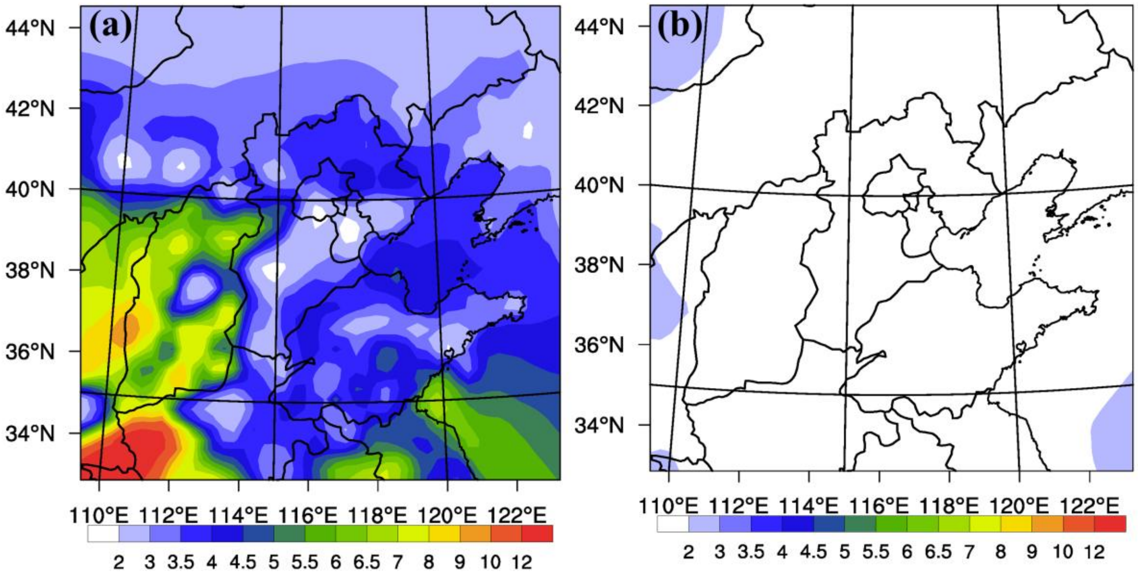

Figure 4 Averaged VOC:NOx ratios in 1960(a) and 2010(b)

Page 15, lines 8-14: The OH increase has been attributed to the temperature perturbation (which makes sense in that this is the boundary condition which has been changed), but this does not necessarily mean that the temperature-dependent reaction rates are the main pathway by which temperature has increased the OH concentration. Another possible path might be through decreases in cloudiness leading to increases in photolysis, leading to increases in OH. Were there any changes in cloudiness in response to the temperature perturbation (or is this meteorological perturbation not fully interactive in which case, yes, temperature alone could be responsible for the OH change)?

Responses: This is a good point that I did not consider. We have plotted the difference in liquid water path (which is the integration of cloud water) and added this factor into the revised manuscript.

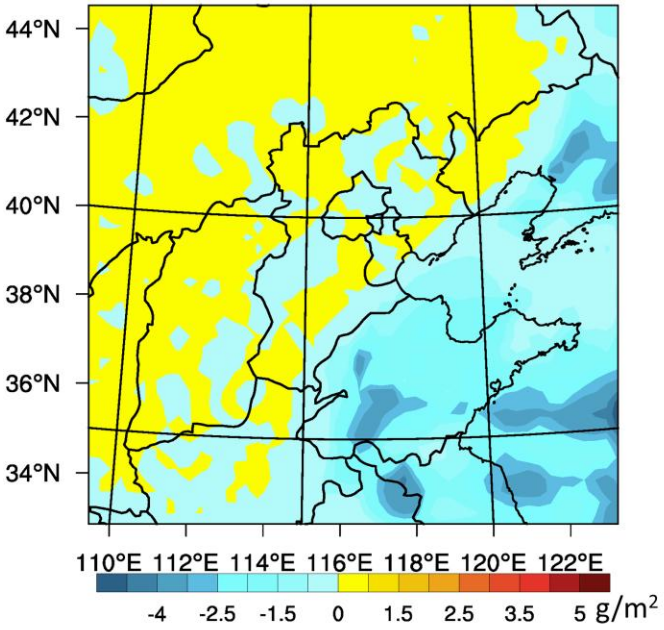

Figure Changes in LWP due to temperature perturbations

Page 19, lines 21 to 23 versus Page 20, lines 10 through 12. In the first set of lines, the authors recommend reductions in SO2 and NH3 as a means of reduction particulate matter; in the second set of lines they show how increases in NH3 and NOx result in particle nitrate formation increasing in the future, despite SO2 decreases, in the winter. The authors need to clarify why / how NH3 is more important for future reductions of PM2.5 than NOx. Is the region relatively ammonia-poor, hence particle nitrate formation will be controlled by NH3 rather than NOx levels? The first set of statements needs to be justified, given the second set of statements, which could be due to either or both of the changes in NH3 and NOx.

Responses: Thanks for this advice for improvements. The reduction of NH3 is more important than NOx because (1) As shown in Figure 4, the increases in NH3 emissions lead to much more

PM than increases in NOx; In winter, this region is VOC limited, so the reduction in NOx might not be effective; (2) This region is relatively ammonia-poor in winter, which is consistent with previous findings in Europe (Megaritis et al., 2013) that reducing NH3 emissions seems to be the most effective control strategy to reduce $PM_{2.5}$. We have added justifications in the revised manuscript.

Response to short comments on “Response of winter fine particulate matter concentrations to emission and meteorology changes in North China”

We thank Dr. Frank Dentener for valuable comments. This document is organized as follows: the comments are in black and our responses are in blue.

To Short Comments

The authors perform emission and meteorology perturbation studies over East Asia (1960-2010), and I would like to point to the similarity of this paper to another paper in this special issue (Kasoar et al.).

I suggest that the magnitude of the emission perturbations Kasoar study (zero-out of anthropogenic) emissions, versus this study (1960-2010 emission changes) is sufficiently similar to warrant some comparison- e.g. of column amounts or AOD. Could something be said about ‘local’ temperature responses as calculated by the model? One other analysis aspect that I find somewhat missing is not only the role of meteorological boundary conditions, but also the chemical ones. With some extra simulations (combinations of BC and emission perturbations) these aspects could also be evaluated, making the publication even more valuable for the HTAP special issue.

Responses: Thanks for mentioning this great work (Kasoar et al.) published in the same issue. In Kasoar’s study, the results are presented due to removal of SO2 emissions, but our results are due to all anthropogenic emission changes from 1960-2010. We plot column AOD and temperature responses and compared with the results in Kasoar’s study. As shown below, due to reductions in 2010 from 2010 to 1960, AOD at 600nm decreases by -0.1~-0.4 in highly polluted areas, and temperature increases by about 0.1~0.55 degree. The results are closet to the results from HadGEM3-GA4 model in Kasoar study.

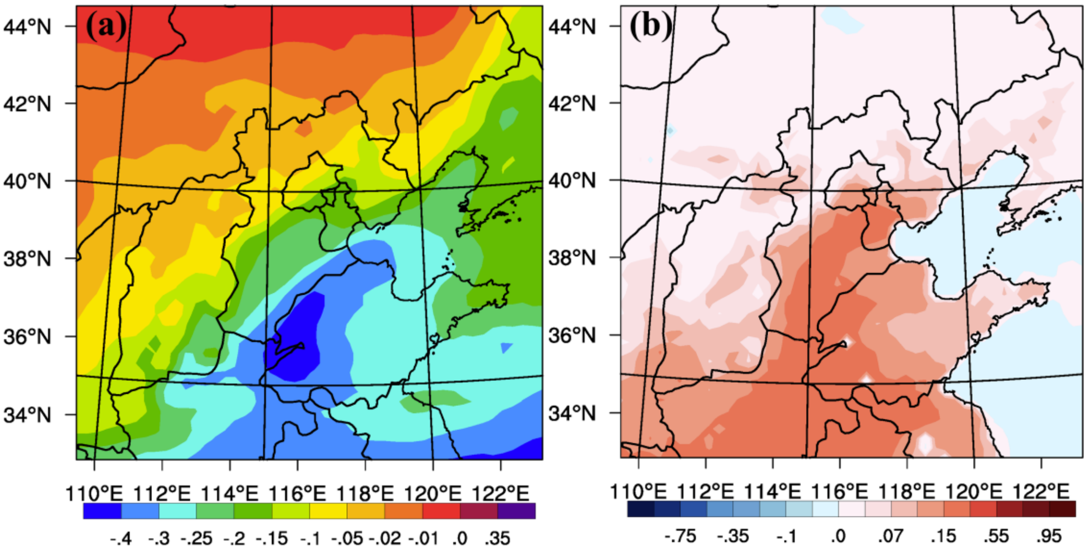

Figure Change in column AOD at 600nm (a) and near surface temperature (b)

Below I offer some comments of technical nature that would warrant additional analysis.

1) Table 1: Assuming that PM2.5 in this study is defined as SO4+NO3+NH4+BC+OC, it seems that PM2.5 value in the first row (1960) is not correct.

Responses: Thanks for this question. Actually, PM2.5 in this study is not just SO4+NO3+NH4+BC+OC. It also includes na,cl, and dust. Na and cl concentrations are very low, so we just ignore it in this study. In table 1, PM2.5 is larger than SO4+NO3+NH4+BC+OC because it includes dust. To clarify this, I added "natural dust (the difference between PM2.5 and the sum of sulfate, nitrate, ammonium, BC, OC)" in the revised manuscript. Hope it is not confusing now.

2) Table 2: (and text). I find the difference between the single perturbation studies and the combined one surprisingly high. I am wondering if in the combined Sox-NOx-NH3 perturbation study also VOC and CO were perturbed- which perhaps could explain the large difference?

Anyway the authors should comment on this, because of cause the response of photochemistry to NOx perturbations can be quite different depending on VOC emissions. One diagnostic analysis is budget analysis: emissions, budget, transport (in-out), lifetime would be very valuable to show.

Responses: Thanks for this great question and suggestion. Yes, you are right, in the combined Sox-NOx-NH3 perturbation study, VOC and CO were also perturbed, which might be the cause of the large difference. We have added this comment on the cause of this difference in the revised manuscript. We also added the plots of VOCs: NOx ratio in 1960 and 2010, which will help readers to understand the photochemical background in 1960 and 2010.

3) page 6, clarify whether the Mozart simulations also used 1960 (2010) emissions. The use of January 2010 warrants some discussion on how representative or typical this month was for a longer climatic period. While even for aerosol with lifetimes of a few days a spin-up of 5 days is rather short, it is certainly not capturing the lifetime of ozone and other components that feedback through oxidants on chemistry. As the authors seem to find large non-linear effects, I think they should consider trying to do longer simulations, if possible.

Responses: Thanks for pointing out this issue. The Mozart simulations used 2010 emissions but we only used boundary and initial conditions for the outer domain, which used emissions for 1960. We downloaded MOZART simulations from NCAR website, which does not cover early years (1960). The results presented used the innermost domain, which takes boundary conditions from outer domain. We have clarified this point in the revised manuscript. This study focuses on a month based simulation, but we are working on running long simulations from a climate change perspective, which will be another interesting story.

4) a table with domain emissions in experiments would be useful. It is not clear how much SO2 was changing (p.9). Clarify what was done with VOC, CO. I assume that the BC/IC were not changing along with the sensitivity studies, but it should be clarified.

Responses: Thanks for this great question. We have added the changing factor for SO_2 (increased by +220%) in the revised manuscript. Changes in other species were also added and modified based on column values. CO and VOCs were also projected from 1960 to 2010. We have added this clarification. Yes, IC/BCs were not changing. We have also added this point in the revised manuscript.

5) section 3.4 (and 3.3) I am a bit wondering about the consistency of changing RH and T separately- while obvious the parameters are closely connected. I think this warrants more discussion. Would changing absolute humidity make more sense? Would dynamics change when changing RH?

Responses: Thanks for this great question. Yes, T and RH are closely connected. The increases in T will lead to changes in RH, and the changed RH will affect some chemical reactions. But these changes are also due to changing T. Under global warming background, chemical reactions might change via changed RH. Some previous studies perturb absolute humidity using offline models. But it is not easy to implement it in the fully online coupled model. We found that perturbing initial and boundary conditions is one of the solutions, but in WRF, RH is provided in boundary and initial conditions, not absolute humidity. Changing absolute humidity might be more interesting and deserve future investigation. When changing RH, wind fields slightly changed, not significant.

Response of winter fine particulate matter concentrations to emission and meteorology changes in North China

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Abstract

The winter haze is a growing problem in North China, but the causes have not been well understood. The chemistry version of the Weather Research and Forecasting model (WRF-Chem) was applied in North China to examine how the $PM_{2.5}$ concentrations change in response to changes in emissions (sulfur dioxide (SO_2), black carbon (BC), organic carbon (OC), ammonia (NH_3), and nitrogen oxides (NO_x)), as well as meteorology (temperature, relative humidity (RH), and wind speeds) changes in winter. From 1960 to 2010, the dramatic changes in emissions lead to +260% increases in sulfate, +320% increases in nitrate, +300% increases in ammonium, +160% increases in BC and 50% increases in OC. The responses of $PM_{2.5}$ to individual emission specie indicate that the simultaneous increases in SO_2, NH_3 and NO_x emissions dominated the increases in $PM_{2.5}$ concentrations. $PM_{2.5}$ shows more notable increases in response to changes in SO_2 and NH_3 as compared to increases in response to changes in NO_x emissions. In addition, OC also accounts for a large fraction in $PM_{2.5}$ changes. These results provide some implications for haze pollution control. The responses of $PM_{2.5}$ concentrations to temperature increases are dominated by changes in wind fields and mixing heights. $PM_{2.5}$ shows relatively smaller changes in response to temperature increases and RH decreases, compared to changes in response to changes in wind speed and aerosol feedbacks. From 1960 to 2010, aerosol feedbacks have been significantly enhanced, due to higher aerosol loadings. The discussions in this study indicate that dramatic changes in emissions are the main cause of increasing haze events in North China, and long-term trends in atmospheric circulations maybe another important cause since $PM_{2.5}$ is shown to be substantially affected by wind speed and aerosol feedbacks. More studies are necessary to get a better understanding of the aerosol-circulation interactions.

1 Introduction

$PM_{2.5}$ (particulate matter with diameter equal to or less than 2.5μm) is a main air pollution concern due to its adverse effects on public health (Gao et al., 2015; Pope et al., 2009). Pope et al. (2009) estimated that a decrease of 10μg $PM_{2.5}$ is related to about 0.6 year mean life expectancy increase. $PM_{2.5}$ is also associated with visibility reduction and regional climate (Cheung et al., 2005). Many cities in North China are experiencing severe haze pollution with exceedingly high $PM_{2.5}$ concentrations. In January 2010, a regional haze occurred in North China and maximum hourly $PM_{2.5}$ concentration in Tianjin was over $400\mu g/m^3$ (Zhao et al., 2013). In January 2013, another unprecedented haze event happened, and the daily $PM_{2.5}$ concentrations in some areas of Beijing and Shijiazhuang reached over $500\mu g/m^3$ (L. T. Wang et al., 2014), and instantaneous $PM_{2.5}$ concentration at some urban measurement sites were over $1000\mu g/m^3$ (Zheng et al., 2015).

It is well known that particulate matter levels are strongly influenced by emissions and meteorological conditions (Steiner et al., 2006). The PM in the atmosphere can be directly emitted from sources like wildfires, combustion, wind-blown dust, and sea-salt, or formed from emitted gases through secondary aerosol formation mechanisms. Meteorology affects PM levels via changing emissions, chemical reactions, transport and deposition processes (Mu and Liao, 2014). For example, increasing wildfire emission in North America is mainly caused by warmer temperatures and precipitation changes (Dawson et al., 2014), and increased temperature leads to higher biogenic emissions, which are important precursors of secondary organic aerosols (Dawson et al., 2014; Heald et al., 2008; Jacob and Winner, 2009). Increasing temperature also increases sulfate concentration due to the temperature dependence of SO_2 oxidation and resulting higher SO_2 oxidation rates (Aw and Kleeman, 2003; Dawson et al., 2007) and semi-volatile aerosols may decrease due to evaporation under higher temperature (Sheehan and Bowman, 2001; Dawson et al., 2007; Tsigaridis and Kanakidou, 2007). Higher relative humidity (RH) favors the formation of nitrate and increasing precipitation decreases all PM species via wet scavenging (Dawson et al., 2007; Tai et al., 2010). Furthermore, increasing clouds promote in-cloud sulfate production (Tai et al., 2010) and changes in wind speed and mixing height determines the dilution of primary and secondary PM (Jimenez-Guerrero et al., 2012; Megaritis et al., 2014; Pay et al., 2012).

With rapid economic and industrial developments, emissions in China have grown during the past years. It is estimated that NO_x emissions in China increased by 70% from 1995 to 2004 (Zhang et al., 2007), Black Carbon (BC) by ~50% from 2000 to 2010 (Lu et al., 2011), Organic Carbon (OC) by ~30% from 2000 to 2010 (Lu et al., 2011), and SO_2 by ~60% from 2000 to 2006 (Lu et al., 2011). Apart from emission changes, it was observed that the winter is warming up in China, especially in the northern part (Guo et al., 2013; Hu et al., 2003; Ren et al., 2012). In addition, wind speed in North China has lowered (Shi et al., 2015; Wang et al., 2004) and RH has decreased in China (Song et al., 2012; Wang et al., 2004).

Many studies have investigated the impacts of emission changes on aerosol formation (Aksoyoglu et al., 2011; Andreani-Aksoyoglu et al., 2008; Megaritis et al., 2013; Tsimpidi et al., 2012a; Tsimpidi et al., 2012b) and the effects of climate/meteorology changes on $PM_{2.5}$ concentrations (Dawson et al., 2007; Megaritis et al., 2013; Megaritis et al., 2014; Tagaris et al., 2007; Tai et al., 2012a; Tai et al., 2012b) in Europe and in the United States. The haze pollution is growing in China, especially in North China, but the causes of the growth are not well understood. For haze pollution in China, it has been reported that aerosol feedbacks that change radiation and temperature can worsen pollution (Gao et al., 2016; Petäjä et al., 2016; Xing et al., 2015c; Zhang et al., 2015). In addition, the connections between haze and meteorological conditions have been established in many former studies (Fu et al., 2014; Jia et al., 2015; Leng et al., 2015; C. Li et al., 2015; Wang and Chen, 2016; Yang et al., 2016; X. Y. Zhang et al., 2015; Zhang et al., 2016). However, the roles of the large emission changes during the last 4 to 5 decades and the observed meteorology changes in North China are not known.

The main objective of this study is to investigate the responses of $PM_{2.5}$ and its major species to changes in emissions, including SO_2, BC, OC, NO_x and NH_3, and to temperature, RH and wind speed changes in North China region. Winter haze in North China has a large contribution from secondary inorganic aerosols and secondary inorganic aerosols are influenced by emissions, temperature and RH. The models used in previous studies of emissions and meteorology perturbations referenced above are all offline models, which are not capable of considering the feedbacks of changing meteorology on other meteorological variables, and the impacts of aerosols on meteorology. However, as pointed by Gao et al. (2016) and J. Wang et al. (2014) aerosol feedbacks should not be neglected when modeling aerosol in China. In this study, we consider aerosol feedbacks using the fully online coupled WRF-Chem model.

This paper is organized as follows. First, the WRF-Chem model, model settings and domain settings are briefly described and then in the next section, emission changes from 1960 to 2010 and accordingly $PM_{2.5}$ changes are discussed. After that, the responses of $PM_{2.5}$ to changes in each emission species are analyzed. At last, the impacts of temperature, RH and wind speed changes on $PM_{2.5}$ are analyzed and discussed.

2 Methodology

2.1 WRF-Chem model

The WRF-Chem model is the chemistry version of the Weather Research and Forecasting model, which is fully online coupled that allows gases and aerosols simulations at the same time as meteorology simulations. In this study, we used a configuration that includes direct and indirect feedbacks. The gas phase mechanism used in this study is the Carbon Bond Mechanism version Z (CBM-Z), which includes 67 species and 164 reactions (Zaveri and Peters, 1999; Zaveri et al., 2008). The gas-particle partitioning module used is the MOSAIC module, which considers all important aerosol components, such as sulfate, nitrate, ammonium, BC, and OC (Zaveri et al., 2008). Eight size bins version of MOSAIC was used and the aerosol sizes ranged from 0.039µm to 10µm. Wind-blown dust was modeled online using the AFWA scheme. Two nested domains with 81km and 27km horizontal grid resolutions from outer to innermost and 27 vertical grids were used (Figure S1 in supplementary material). Analysis nudging of meteorology variables was used for the outer domain. The model physics configurations also follow the settings in Gao et al. (2016). Inputs into the model include meteorological boundary and initial conditions (BCs and ICs) from NCEP FNL 1 °×1 °data and chemical boundary and initial conditions from MOZART model simulations (Emmons et al., 2010). Chemical BCs and ICs are not changing along with the sensitivity simulations, but the studied domain (innermost domain) takes boundary conditions from outer main, where emissions are perturbed. The anthropogenic emission inventory used is the MACCity (MACC/CityZEN EU projects) emissions dataset, which provides monthly CO, NO_x, SO_2, VOC, BC, OC, and NH_3 emissions from different sectors for years between 1960 and 2020 (Granier et al., 2011). We compared the MACCity emission inventory for 2010 (Granier et al., 2011) with MIX emission inventory for 2010 (M. Li et al., 2015) in the China region, and the magnitudes of emissions in China from these two datasets are very close. For example, the SO_2 emissions in China in 2010 were estimated to be 28663 Gg in the MIX emission inventory, and were 26876.3 Gg in the MACCity emission inventory. Simulations for evaluating roles of emission changes were conducted using emissions for year 1960 and year 2010. We assigned emissions to the first 6 layers from surface based on sectors. For example, emissions from large point sources (such as chimneys) were assigned to higher layers. Biogenic emissions were estimated online using the MEGAN model (Guenther et al., 2006). The simulation period was January 2010 and five days in previous month were modeled as spin-up to overcome the influences of initial conditions.

2.2 Sensitivity experiments

We explored the sensitivities of the winter time haze event in 2010 to changes in emissions and meteorology features through a series of simulations using 1960 and 2010 emission baselines. Specifically, the influences of emission changes of SO_2, BC, OC, NH_3, and NO_x, and meteorology (temperature, RH and wind speeds) changes on $PM_{2.5}$ and its major species were evaluated using a series of simulations. They are listed and explained in Table 1. All base simulations use meteorology of January 2010. CTL case uses emissions for year 1960 and EMI2010 case uses emissions for year 2010. SO_2, NH_3, and NO_x emissions were perturbed separately from 1960 to 2010 (i.e., SO_2-2010 NH_3-2010 NO_x-2010 cases). In the CTL_NF and EMI2010_NF cases, aerosol-radiation interactions are excluded based on emissions for year 1960 and 2010. It was pointed out surface air temperature in North China increased at the rate of 0.36 ℃ per decade (Guo et al., 2013), the linear trends coefficient of relative humidity anomaly in North China is about -0.60% per decade (Wang et al., 2004), and national mean wind speed decreased 16% in the recent 50 years (Wang et al., 2004). To estimate the impacts of changes in temperature, RH and wind speed that happened in the past several decades, we decreased temperature by 2 degrees, increased RH by 10%, and increased wind speeds by 20%, to reflect conditions of early decades (CTL_T2, CTL_RH10, CTL_WS20, EMI2010_T2, EMI2010_RH10, and EMI2010_WS20 cases). These were conducted by perturbing the initial and boundary conditions of these individual meteorological variables.

At different vertical heights, emission and meteorological variables were uniformly perturbed. The changes of $PM_{2.5}$ and its major components due to perturbations in emissions and meteorology are analyzed for the North China region. The North China region is defined using domain 2 in Gao et al. (2016) and the statistics of changes are calculated within domain 2 for the January 2010 month.

2.3 Model Verification

The WRF-Chem model performance has been evaluated using multiple observations, including surface meteorological, chemical and optical data, and satellite data in Gao et al. (2016). The model was shown to capture the variations of surface temperature, RH, while wind speed was slightly overestimated (Gao et al., 2016), which has been reported as a common problem of current WRF-Chem model under low wind speed conditions. The Root Mean Square Error

(RMSE) of temperature were all less than 3.2K and RMSEs of RH varied from 6.4 to 11.1%. The RMSE of wind speeds were below the proposed criteria (2m/s) (Emery et al., 2001) at the Beijing, Tianjin and Baoding stations, but larger than that criteria at the Chengde station. The time series of simulated surface $PM_{2.5}$, NO_2, and SO_2 showed good agreement with observations as did simulated aerosol optical depth (AOD) (Gao et al., 2016). Mean Fractional Bias (MFB) ranged from -21.8% to 0.4% and Mean Fractional Error (MFE) ranged from 26.3% to 50.7% when comparing against $PM_{2.5}$ observations (Gao et al., 2016). In addition, the comparison between model results and satellite found that the vertical distribution of aerosol and horizontal distribution were captured well by the model (Gao et al., 2016). Compared with observed $PM_{2.5}$ composition, sulfate and OC were underestimated and nitrate was overestimated by the model (Gao et al., 2016). The underestimation of sulfate may be due to underestimation of SO_2 gas phase oxidation, errors in aqueous-phase chemistry, and/or missing heterogeneous sulfate formation (Gao et al., 2016).

3 Results and Discussion

3.1 $PM_{2.5}$ sensitivity to emission changes from 1960 to 2010

The emission changes of SO_2, NO_x, NH_3, BC and OC and resulting impacts on $PM_{2.5}$ from 1960 to 2010 were examined based on the MACCity dataset for years 1960 and 2010. Figure 1(a-e) displays SO_2, NO_x, NH_3, BC and OC emissions for 1960 and Figure 1(f-j) shows the changes from 1960 to 2010. Populated regions of North China, such as urban Beijing, urban Tianjin, and urban Shijiazhuang, exhibit large emissions of SO_2, NO_x, NH_3, BC and OC in 1960. However, NH_3 emissions exhibit different spatial distribution patterns from SO_2, NO_x, BC and OC emissions, because NH_3 is mainly associated with agriculture while SO_2, NO_x, BC and OC are mainly related with industrial and residential activities. From 1960 to 2010, SO_2, NO_x, NH_3, BC and OC increased over the entire North China domain and markedly increased in the Jing-Jin-Ji city cluster. In general, the domain averaged SO_2 emissions and NO_x emissions in North China increased by ~220% and ~990% from 1960 to 2010, respectively. The domain averaged NH_3 emissions in North China increased by ~390% from 1960 to 2010, but the most significant increases occurred not in the Jing-Jin-Ji city cluster, but in Inner Mongolia. Unlike NH_3 emissions, BC emissions increased the most in urban Beijing from 1960 to 2010. This is because residential sources are the biggest contributor to BC in winter (Li et al., 2016) and the population in urban Beijing sharply increased with rapid urbanization. From 1960 to 2010, the mean BC emissions in North China increased by ~154%. Similar to BC emissions, OC emissions increased substantially in the center of Beijing, and the domain averaged increasing ratio is about 54% from 1960 to 2010. The enhancements of SO_2, NO_x, NH_3, BC and OC emissions in North China are expected to result in substantial increase in regional $PM_{2.5}$ concentrations.

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Figure 2 shows the simulated monthly mean concentrations of $PM_{2.5}$ and its major components (sulfate, nitrate, ammonium, BC and OC) based on emissions for year 1960. As listed in Table 2, the domain averaged concentrations of sulfate, nitrate, ammonium, BC, OC, and $PM_{2.5}$ are 1.9, 0.8, 0.8, 1.5, 4.6, and 19.2μg/m^3, respectively. For year 1960, $PM_{2.5}$ concentrations are mainly dominated by sulfate, OC and natural dust (the difference between $PM_{2.5}$ and the sum of sulfate, nitrate, ammonium, BC, OC). Figure 3 displays the changes of sulfate, nitrate, ammonium, BC, OC, and $PM_{2.5}$ due to changes in SO_2, NO_x, BC and OC emissions from 1960 to 2010. The predicted monthly mean concentrations of $PM_{2.5}$ components and $PM_{2.5}$ increase everywhere over the entire domain due to emission changes resulting from the rapid urbanization and industrialization from 1960 to 2010 (Figure 3(a-f)). As listed in Table 2, the predicted monthly domain mean sulfate increases the largest (5.0 μg/m^3), followed by nitrate (2.6 μg/m^3) and OC (2.5 μg/m^3).

From 1960 to 2010, the predicted BC increased by ~157% and OC increased by ~54% due to 153% increase in BC emissions and 52% increase in OC emissions. The nearly linear response of both BC and OC aerosols to their emissions is due to the omission of a secondary organic aerosol formation in the chosen CBMZ/MOSAIC mechanism. Thus, both of them were treated as primary aerosols in these simulations. Our previous analyses indicate that SOA contribution in this time period was small (Gao et al., 2016). The domain mean $PM_{2.5}$ concentrations increased by 14.7μg/m^3 and the domain maximum increase is about 45μg/m^3 (Figure 3(f) and Table 2).

To quantify how much of the changes in Figure 3 are from the impacts of boundary conditions, we simulated another case with the innermost domain emissions fixed in 1960 and the outer domain emissions changed from 1960 to 2010. The impacts of boundary conditions mostly occur around the south boundary and show nearly no impact on $PM_{2.5}$ in Beijing (shown in Figure S2~~X~~), which are consistent with the continuous weak southerly winds during the study period (Gao et al., 2016). On domain average, the impacts of boundary conditions result in 5.0ug/m^3 increase in the study domain, accounting for about 33.9% of the total changes in $PM_{2.5}$."

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To explore how emission changes can affect haze days, we calculated the number of haze days in urban Beijing for the CTL and EMI_2010 cases, using daily mean thresholds of 35 and 75μg/m^3

(China National Ambient Air Quality Grade I and Grade II Standard, L. T. Wang et al., 2014). In urban Beijing, there are 4 days when daily mean $PM_{2.5}$ concentrations are above 35µg/m^3, and 0 days with daily mean $PM_{2.5}$ concentrations above 75µg/m^3 for the CTL case. For the EMI_2010 case, these two numbers increase to 15 and 8, indicating that the large increases in emissions over the past several decades have significantly affected haze occurrences in Beijing.

3.2 Sensitivity to changes in individual emission species

The results discussed above show that in the winter period, the concentrations of secondary inorganic aerosols (sulfate, nitrate, and ammonium) has increased dramatically. Thus it is important to explore how sensitive secondary inorganic aerosol is to perturbations in precursor emissions. Three sensitivity simulations (change SO_2, NH_3 and NH_3 emissions separately) were conducted to examine how changes in emissions of each species affect aerosol concentrations. The predicted changes of $PM_{2.5}$ and major $PM_{2.5}$ components at the ground-level are shown in Figure 4 and monthly domain mean aerosol changes are summarized in Table 3.

3.2.1 Changes in SO_2 emissions

Due to changes in SO_2 emissions from 1960 to 2010, domain averaged sulfate increase by 3.4µg/m^3 (178.3%), nitrate decreases by -0.3µg/m^3 (-32.3%), and ammonium increases by 0.2µg/m^3 (29.4%). NH_3 reacts preferentially with SO_2 (Seinfeld and Pandis, 2006) and free NH_3 reacts with enhanced H_2SO_4 due to increasing SO_2. As a result, ammonium increases and less

HNO_3 gas is transferred to the aerosol phase, which is consistent with the responses to increasing SO_2 emissions in Kharol et al. (2013).

3.2.2 Changes in NH_3 emissions

As shown in Figure 4 and Table 3, changes in NH_3 emissions from 1960 to 2010 result in significant increases in nitrate (1.5μg/m^3, +76.0%) and ammonium (0.6μg/m^3, +84.0%). The domain mean changes of sulfate due to increase in NH_3 is close to zero (about 0.1μg/m^3), because sulfate formation is only indirectly associated with NH_3 availability (Tsimpidi et al., 2007). The significant changes in nitrate and ammonium occurred in south Hebei, Shandong, and Henan province, where anthropogenic NO_x emissions are very high (Figure 1). Although NH_3 emissions substantially increased in Inner Mongolia (Figure 1), responses of nitrate and ammonium are not significant there due to trivial NO_x emissions. The substantial increases of nitrate after NH_3 emission increase indicate that NH_3 limits the NH_4NO_3 formation in North China region in this period.

3.2.3 Changes in NO_x emissions

After changing NO_x emissions from 1960 to 2010 levels, domain mean surface $PM_{2.5}$ decreases by about 0.2μg/m^3, but the changes of individual $PM_{2.5}$ inorganic components vary. The increase of NO_x emissions cause 0.7μg/m^3 (-39.1%) decrease in monthly domain mean sulfate and the domain peak sulfate reduction is about 2.9μg/m^3. The OH radical is critical in the sulfate formation in the regions where SO_2 concentrations are high and there is a competition between NO_x and VOCs to react with OH (Tsimpidi et al. 2012b). When the VOCs/NO_x concentration ratio is close to 5.5:1, the OH reacts with NO_x and VOCs at an equal rate (Seinfeld and Pandis, 2006). When the concentration ratio is lower than 5.5:1, the OH primarily reacts with NO_x, and the region with this concentration ratio is called VOC-limited region. In VOC-limited regions, an increase of NO_x will cause a decrease of OH and ozone concentration. When the VOCs/NO_x concentration ratio is higher than 5.5:1, the OH will preferentially react with VOCs, and the region with this high ratio is called NO_x-limited region. In the NO_x-limited region, an increase of NO_x will increase OH and ozone concentrations. In the simulated winter month, biogenic emissions are low and NO_x emissions in North China are very high, leading to lower VOCs to NO_x ratios, and it can be considered as VOC-limited region. Fu et al. (2012) pointed out that north East Asia is VOC-limited in January and urban areas of Beijing are VOC-limited in both January and July. The model averaged VOCs/NOx ratio changes from 4.2 to 1.2 due to emission perturbations from 1960 to 2010 (shown in Figure S3). As a result, the large increases in NO_x emissions from 1960 to 2010 result in a 47.9% decrease in daytime surface ozone concentration and 55.6% decrease in daytime surface OH concentration, which are shown in Figure 5. Over the entire domain, ozone and OH decrease due to NO_x emission increases (Figure 5). Consequently, sulfate aerosol decrease over the entire domain, as shown in Figure 4(i). Decreases in sulfate might also be related to changes in thermodynamics of the ammonium-sulfate-nitrate system. Although OH decreases, nitrate still rises (0.6μg/m^3, +76.0%) due to the increase in NO_x emissions. The domain mean ammonium decreases by about 5.1% (-0.04μg/m^3). The net effects of NO_x emission increases bring about 0.2μg/m^3 decrease in monthly domain mean $PM_{2.5}$ concentration and the domain peak decrease is about 1.1μg/m^3 (Table 3).

3.2.4 Coupled changes in SO_2, NH_3 and NO_x emissions

As shown above, increasing SO_2 emissions significantly increases $PM_{2.5}$ concentrations in North China region, increasing NH_3 emissions also increases $PM_{2.5}$ concentrations but to a lesser extent, and increasing NO_x emissions slightly decreases $PM_{2.5}$ concentrations. The effects of coupled changes in SO_2, NH_3 and NO_x emissions are not a simple addition of the effect of changing them separately. As listed in Table 3, the monthly domain mean sulfate, nitrate, ammonium, and $PM_{2.5}$ increases more than the effects of changing emissions separately. Domain mean sulfate increases by 5.0μg/m^3 (+264.0%), nitrate increases by 2.6 μg/m^3 (+322.5%), ammonium increases by 2.3μg/m^3 (295.2%) and $PM_{2.5}$ increases by 9.9μg/m^3 due to coupled changes in SO_2, NH_3 and NO_x emissions from 1960 to 2010. The simultaneous increases in SO_2, NH_3 and NO_x emissions promote dramatic increases of secondary inorganic aerosols in North China, which is partially due to perturbations in VOCs and other species at the same time~~.~~

3.2.5 Changes in BC and OC emissions

Since BC and OC are treated as primary aerosols in the chosen CBMZ/MOSAIC mechanism, changes in their emissions do not show any impact on other aerosol components. As listed in Table 3, monthly domain mean $PM_{2.5}$ increases by 2.3μg/m^3 and 2.5μg/m^3 due to changes in their emissions from 1960 to 2010, respectively.

3.3 Effects of temperature increases

The model used in this study is a fully online-coupled model, which simulates meteorological variables and chemical variables together. Therefore, it is not possible to increase temperature uniformly, as was done in previous studies using offline models (Dawson et al., 2007; Megartis et al., 2013; Megartis et al., 2014). To examine the sensitivity of $PM_{2.5}$ to temperature change (reflecting the winter warming trends), we decrease temperature by 2 ℃ in the initial and boundary conditions to reflect conditions more like 1960. As a result, the monthly domain mean surface temperature increases 2.0 ℃ (CTL-CTL_T2), but in a non-uniform manner. These responses in domain temperature are partially due to aerosol feedbacks. The spatial distributions of monthly mean surface temperature and temperature changes are shown in Figure 6(a). The monthly mean surface temperature increases more along top left domain boundaries and less over the Bohai sea. The influence of increasing temperature on biogenic emissions is included using temperature-sensitive biogenic emission model MEGAN (Guenther et al., 2006).

Due to the perturbation in temperature as mentioned above, sulfate, nitrate, ammonium and $PM_{2.5}$ are predicted to increase in most areas of the domain (Figure 7). Predicted monthly mean sulfate increases by 0.06μg/m^3 (+3.1%), nitrate increases by 0.03μg/m^3 (+4.2%), and ammonium increases by 0.02μg/m^3 (+2.8%). The increases of sulfate, nitrate and ammonium are mostly attributed to the increasing OH radicals, as shown in Figure 6(b). After the temperature perturbation, daytime OH increases by about 3.6% on domain average. It was found that higher temperature increased volatilization of ammonium nitrate and partitioned it to the gas phase (Megaritis et al., 2014), but it is not significant here due to the low temperature in winter. In addition, the increase of sulfate, nitrate, and ammonium could be partially due to accelerated gas-phase reaction rate at higher temperature (Dawson et al., 2007; Megaritis et al., 2014). It may also due to enhanced photolysis caused by decreases in cloudiness after temperature perturbations (as shown in Figure S4: changes in liquid water path).

As shown in Figure 7 (d-e), the concentrations of primary aerosols (BC and OC) also increase after the temperature perturbations. This is due to changes in other physical parameter, such as wind direction, wind speed, and PBLHs, which are key factors in the diffusion of air pollutants. Figure 6(c) shows that monthly PBLHs in most North China areas decrease after the temperature perturbation, and PBLHs over the Bohai sea decrease the most, with monthly mean decrease over 50 meters. The monthly domain average daytime PBLHs decrease about 2.3% due to changes in temperature vertical profiles. Surface horizontal winds also change (Figure 6(d)), which directly affect the distributions and magnitudes of $PM_{2.5}$ concentrations in North China along with PBLH changes.

The responses of $PM_{2.5}$ concentrations to temperature perturbation are different from the responses of sulfate, nitrate, ammonium, BC and OC (Figure 7), with decreases in northwestern regions and increases in most areas of the North China Plain. This is because natural dust is dominant in northwestern regions (as shown in Figure 2(f)), and the concentrations of natural dust decrease under lower horizontal wind speeds (Figure 6(d)). The monthly $PM_{2.5}$ concentration decreases by 0.01μg/m^3 on domain average due to temperature perturbation. Because of temperature increase, the numbers of haze days (defined using the daily mean threshold 35 and 75μg/m^3) in urban Beijing do not change.

The discussions shown above are based on emission levels in 1960. The responses to temperature perturbations were also investigated based on emission levels in 2010, and the results are shown in Figure S2, S3 and Table 3. The spatial distributions of the changes are similar to the results shown above, but with larger magnitudes. The domain mean PBL heights decreases slightly more (-8.6 compared to -8.3 meters). The domain mean $PM_{2.5}$ concentrations and $PM_{2.5}$ components exhibit larger increases in North China, although daytime OH concentrations increases less (2.6×10^{-9} compared to 3.3×10^{-9} ppmv), suggesting that the responses of $PM_{2.5}$ concentrations are mostly due to changes in PBL heights and wind fields.

3.4 Effects of RH decreases

The RH was enhanced by 10% in model initial and boundary conditions to represent RH for the previous decades. As a result, the simulated monthly mean RH decreases by 9.3% on domain average. Due to RH perturbation, domain mean $PM_{2.5}$ concentration decreases by 0.7 $\mu g/m^3$. As shown Figure 8(a), $PM_{2.5}$ concentrations decrease in the Jing-Jin-Ji region but increase in southern areas of the domain. The ammonium nitrate formation equilibrium depends on RH (Tai et al., 2010), so HNO_3 may be shifted to the gas phase under lower RH. In addition, the changes in RH can also affect the wet deposition rate. The increases in southern areas of the domain are mainly due to suppressed in-cloud scavenging, as the decreases in RH inhibit the formation of clouds. As shown in Figure 8(b), liquid water path (LWP) decreases by 75.0%. As a result, the in-cloud scavenging loss rate decreases. The changes of predicted aerosol optical depth at 600nm are shown in Figure 8(c). In most regions, visibility decreases due to lower RH. Because of RH decreases, the numbers of haze days (defined using the daily mean threshold 35 and 75$\mu g/m^3$) in urban Beijing do not change. The responses to RH perturbations were also investigated based on emission levels in 2010, and the results are shown in Figure S4 and Table 3. The responses are also similar to changes based on emission levels in 1960, but with larger magnitudes.

3.5 Effects of wind speed decreases

Simulations were also carried out when wind speeds in initial and boundary conditions were increased to estimate the wind speeds for the previous decades. The predicted domain averaged monthly mean wind speed decreases by about 0.7 m/s. As shown in Figure 9(a), the monthly mean near surface horizontal winds are pronounced in mountainous areas (northwest areas of the domain) and relatively smaller in other areas. Figure 9(b) shows the changes of wind speeds (CTL-WS20) due to model perturbations. The predicted monthly mean $PM_{2.5}$ concentrations decrease by 2.3μg/m^3 on domain average, but the responses of $PM_{2.5}$ vary within the domain. As shown in Figure 9(c), $PM_{2.5}$ concentrations decrease in the northwestern areas because of lower production of natural dust under lower horizontal wind speeds. However, in most areas of the North China Plain, $PM_{2.5}$ concentrations increase under lower wind speeds (Figure 9(c)). The domain peak increase is about 2.4 μg/m^3, which is based on low predicted $PM_{2.5}$ concentrations using emissions for year 1960. If the concentration in base case is higher, the responses will be enhanced. As shown in Figure 9(d), the domain maximum increases in $PM_{2.5}$ increases from 2.4 to 9.4 μg/m^3. Because of wind speed decreases, number of haze days that daily mean $PM_{2.5}$ concentrations are above 35μg/m^3 increases by 1.

3.6 Effects of changes in aerosol feedbacks

As mentioned in Gao et al. (2016), high concentrations of aerosol enhance stability of boundary layer and increase $PM_{2.5}$ concentrations. Due to dramatic changes in emissions from 1960 to 2010, the strength of aerosol feedbacks may also have changed. To quantify these changes, we simulated four cases (i.e., CTL, CTL_NF, EMI2010, and EMI2010_NF). CTL-CLT_NF and EMI2010-EMI2010_NF are used to represent the contributions of aerosol radiative effects in

1960 and 2010. The changes in monthly mean daytime PBL heights and $PM_{2.5}$ concentrations are shown in Figure 10. In 1960, the domain averaged PBL height decreases by 6.7 meters due to aerosol radiative effects, and the domain maximum decrease is 25.4 meters. Correspondingly, the domain averaged $PM_{2.5}$ increases by 0.1 μg/m^3 and the domain maximum increase is 0.9 μg/m^3. In 2010, the domain averaged PBL height decreases by 13.8 meters and the domain maximum decrease is 55.2 meters (more than two times compared to 1960). Correspondingly, the domain averaged $PM_{2.5}$ increases by 0.7 μg/m^3 and the domain maximum increase is 5.1 μg/m^3. The enhanced strength of aerosol feedbacks is another important cause of degraded aerosol pollution. Thus, controlling emissions will have a co-benefit of reducing strength of aerosol feedbacks.

3.6 Implications for the effects of emission and meteorology changes on $PM_{2.5}$ concentrations

The simulated responses of $PM_{2.5}$ concentrations to emission changes and meteorology changes presented here, along with the previous presented effects of aerosol feedbacks (Gao et al. 2016), provide important implications for the causes of the dramatic increases in winter $PM_{2.5}$ concentrations.

We calculated domain maximum changes in $PM_{2.5}$ concentration averaged over four stagnant days (January 16-19) owing to emission changes from 1960-2010 (EMI2010-CTL), temperature increases (CTL-CTL_T2), RH decreases (CTL-CTL_RH10), wind speed decreases (CTL-CTL_RH20), and aerosol feedbacks (CTL-CTL_NF). The values are 137.7, 2.0, 2.6, 7.5 and 4.0μg/m^3, respectively. When the perturbations are based on emission levels in 2010, domain maximum changes in $PM_{2.5}$ concentration due to temperature increases (EMI2010-EMI2010_T2), RH decreases (EMI2010-EMI2010_RH10), wind speed decreases (EMI2010-

EMI2010_WS20), and aerosol feedbacks (EMI2010-EMI2010_NF) are 4.8, 4.7, 26.4 and 25.5µg/m^3. The effects of emission changes on haze formation are dominant and the effects of aerosol feedbacks are comparable to the effects of wind speed decreases.

The comprehensive comparisons of these factors are also summarized in Table 3. Based on the monthly domain mean responses of $PM_{2.5}$ concentrations to these factors, dramatic emission changes due to urbanization and industrialization are the main causes of degraded air quality and frequent haze occurrences in in North China. $PM_{2.5}$ shows significant responses to changes in SO_2, NH_3, NO_x emissions than BC and OC (about 106.3% higher). In addition, $PM_{2.5}$ shows significant increases in response to changes in SO_2 and NH_3 emissions, as compared to increases in response to changes in NO_x emissions. This region is relatively ammonia-poor in winter, so reducing NH3 emissions might be effective, which is consistent with previous findings in Europe (Megaritis et al., 2013). SO2 is the precursor of sulfate, which accounts for a large fraction of PM in this region. Thus, they should be preferentially controlled in order to reduce $PM_{2.5}$ levels. To control SO_2 emissions, the usage of natural gas or other clean energy should be promoted to reduce the usage of coal. NH_3 emissions in China are mainly from agriculture sources (about 90%), including livestock, fertilizer, and agricultural soil (Huang et al., 2012). Lelieveld et al. (2015) found that agricultural emissions make the largest relative role in $PM_{2.5}$ concentration in eastern USA, Europe, Russia and East Asia. To control NH_3 emissions from agriculture sources, some animal feeding and animal housing strategies should be taken. In addition, controlling emissions will also have a co-benefit of reducing strength of aerosol feedbacks.

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According to the ECLIPSE_GAINS_4a emission dataset, SO_2 emissions in China will decrease by -26%, NO_x emissions in China will increase by 19%, and NH_3 emissions in China will increase by 14% from 2010 to 2030. We predicted (by perturbating SO_2, NO_x and NH_3 emissions by -26%, 19% and 14%) that these changes will lead to large decreases in winter sulfate (-2.3μg/m^3 on domain average). Nitrate will increase by 1.5μg/m^3 and ammonium will slightly decrease (-0.05μg/m^3) on domain average. The net change of domain averaged $PM_{2.5}$ concentration is not significant (-0.8μg/m^3), so more efforts are needed to control these important gaseous precursors.

From the information listed in Table 3, the responses of $PM_{2.5}$ concentrations to temperature and RH perturbations are not as significant as to wind speed perturbations. From Sect. 3.3, we also found that the effects of temperature perturbation on $PM_{2.5}$ concentration are dominant by changes in PBLH and wind fields. Previous studies have pointed out the occurrences of haze events are highly associated with atmospheric circulation anomalies (Chen and Wang, 2015; Zhang et al., 2016). Thus, changes in atmospheric circulations may be another important cause of growing haze pollution, in addition to emission changes. Furthermore, aerosol can also change atmospheric circulation, especially in severely polluted East Asia. Thus, controlling emission may have co-benefits of mitigate aerosol effects on atmospheric circulation.

The effects of changing atmospheric circulations on winter haze pollution in China is beyond the scope of this paper, but should be investigated in future studies.

4 Conclusions

A fully online coupled meteorological and chemical transport model, WRF-Chem was used to study responses of winter $PM_{2.5}$ concentrations to changes in emissions of SO_2, BC, OC, NH_3, and NO_x and to meteorology (temperature, RH, and wind speeds) changes in North China region, where people are suffering from severe winter haze pollution.

The detailed historical emissions dataset MACCity for year 1960 and 2010 were used to evaluate the impacts of changes in emissions of SO_2, BC, and OC. From 1960 to 2010, the dramatic changes in emissions lead to +264.0% increases in sulfate, +322.5% increases in nitrate, +295.2% increases in ammonium, +157.0% increases in BC and 54% increases in OC. The domain mean $PM_{2.5}$ concentrations increase by 14.7μg/m^3 and the domain maximum increase is about 45μg/m^3. The responses of $PM_{2.5}$ to individual emission species indicate that the simultaneous increases in SO_2, NH_3 and NO_x emissions dominated the increases in $PM_{2.5}$ concentrations. $PM_{2.5}$ shows significant increases in response to SO_2 and NH_3 emission changes. The increases in NO_x emissions may decrease surface ozone concentration and surface OH radical concentrations, because North China region is VOC-limited in the winter. In addition, OC accounts for a large fraction in $PM_{2.5}$ changes.

The sensitivities of $PM_{2.5}$ to emission changes of its precursors provide some implications for haze pollution control. SO_2, NH_3 and OC should be preferentially controlled. In China, the residential sector, particularly biofuel usage is the primary sources of OC (Lu et al., 2011). The usage of natural gas or other clean energy should be promoted to reduce the usage of coal and biofuel to reduce SO_2 and OC. To control NH_3 emissions from agriculture sources, some animal feeding and animal housing strategies should be taken.

The effects of changes in winter time meteorology conditions were also studied. Emission changes from 1960 to 2010 substantially increase numbers of haze days, but meteorology perturbations do not show any significant impacts. The perturbations in temperature and RH do change $PM_{2.5}$ concentrations, but the strength is not as significant as the effects of wind speed and emission changes. The effects of temperature perturbation are dominated by the changes in surface wind fields and PBLHs. The effect of aerosol feedbacks is comparable to the effect of decreasing wind speeds and the strength of aerosol feedbacks significantly increased from 1960 to 2010.

The above discussions indicate that aerosol concentrations are mainly controlled by atmospheric circulations, except emission changes. Thus, long-term trends in atmospheric circulations maybe another important cause of winter haze events in North China. More studies are necessary to get a better understanding of the aerosol-circulation interactions.

In our previous modeling study of the same period (January 2010), we found that SOA contribution was small, so we did not include SOA in this study. But this indication might be problematic due to current poorly parameterized SOA scheme. In the future, how changes in emissions and meteorology variables affect productions of SOA during winter should be further studied using more advanced SOA schemes. In addition, we did not consider primary PM except BC and OC in the model because there is no information in the MACCity emission inventory, which is another direction for improvements in future studies.

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Table 1. Simulation cases and descriptions

Cases	Descriptions
CTL	Base case, anthropogenic emissions are from MACCity dataset for year 1960

EMI2010	Anthropogenic emissions are from MACCity dataset for year 2010
SO_2-2010	Same as CTL case except SO_2 emissions are for year 2010
NH_3-2010	Same as CTL case except NH_3 emissions are for year 2010
NO_x-2010	Same as CTL case except NO_x emissions are for year 2010
CTL_T2	Same as CTL case except temperature BCs and ICs are decreased by 2K
CTL_RH10	Same as CTL case except RH BCs and ICs are increased by 10%
CTL_WS20	Same as CTL case except wind speed BCs and ICs are increased by 20%
CTL_NF	Same as CTL case except aerosol-radiation interactions are excluded
EMI2010_T2	Same as EMI2010 case except temperature BCs and ICs are decreased by 2K
EMI2010_RH10	Same as EMI2010 case except RH BCs and ICs are increased by 10%
EMI2010_WS20	Same as EMI2010 case except wind speed BCs and ICs are increased by 20%
EMI2010_NF	Same as EMI2010 case except aerosol-radiation interactions are excluded

Table 2. Monthly domain mean concentrations of $PM_{2.5}$ and its major components for year 1960, and domain maximum and mean concentrations for changes from 1960 to 2010 due to emission changes ($\mu g/m^3$)

Years		SO_4^{2-}	NO_3^-	NH_4^+	BC	OC	$PM_{2.5}$
1960	Domain mean	1.9	0.8	0.8	1.5	4.6	19.2
1960-2010	Domain maximum	18.9	7.8	6.8	9.9	11.1	45.0
	Domain mean	5.0 (264.0%)	2.6 (322.5%)	2.3 (295.2%)	2.3 (156.6%)	2.5 (54.0%)	14.7 (76.4%)

Table 3. Monthly domain mean changes of sulfate, nitrate, ammonium and $PM_{2.5}$ concentrations (μg/m^3) due to emission and meteorology perturbations, and aerosol feedbacks (the two values of $PM_{2.5}$ changes

are for meteorology perturbations and aerosol feedbacks based on 1960 and 2010 emission levels, respectively)

	$SO4^{2-}$	NO^{3-}	NH^{4+}	$PM_{2.5}$
Changes in SO_2 emissions	3.4(178.3%)	-0.3 (-32.3%)	0.2 (29.4%)	3.4
Changes in NH_3 emissions	0.1 (5.3%)	1.5 (189.6%)	0.6 (84.0%)	2.3
Changes in NO_x emissions	-0.7 (-39.1%)	0.6 (76.0%)	-0.04 (-5.1%)	-0.2
Changes in SO_2, NH_3, NO_x emissions	5.0 (264.0%)	2.6 (322.5%)	2.3 (295.2%)	9.9
Changes in BC emissions	-	-	-	2.3
Changes in OC emissions	-	-	-	2.5
Temperature perturbations	-	-	-	-0.01/0.3
RH perturbations	-	-	-	-0.7/-1.1
Wind speed perturbations	-	-	-	-2.3/-0.5
Aerosol feedbacks				0.1/0.7

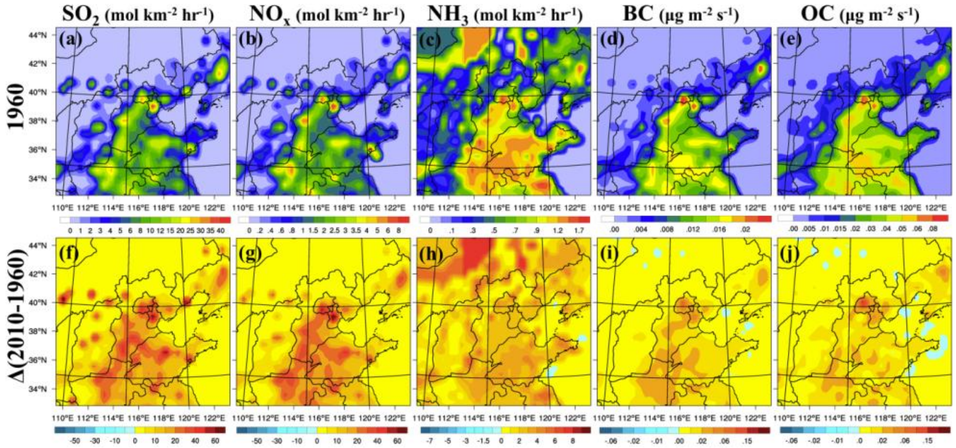

Figure 1. SO$_2$, NO$_x$, NH$_3$, BC and OC emissions for year 1960 (a-e), and the changes of them from 1960 to 2010 (f-j)

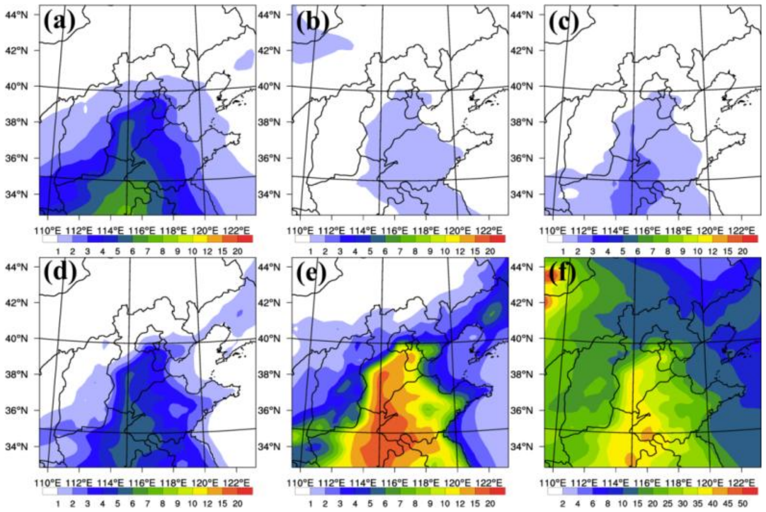

Figure 2. Predicted monthly mean sulfate (a), nitrate (b), ammonium (c), BC (d), OC (e) and PM$_{2.5}$ (f) concentrations based on emissions for year 1960

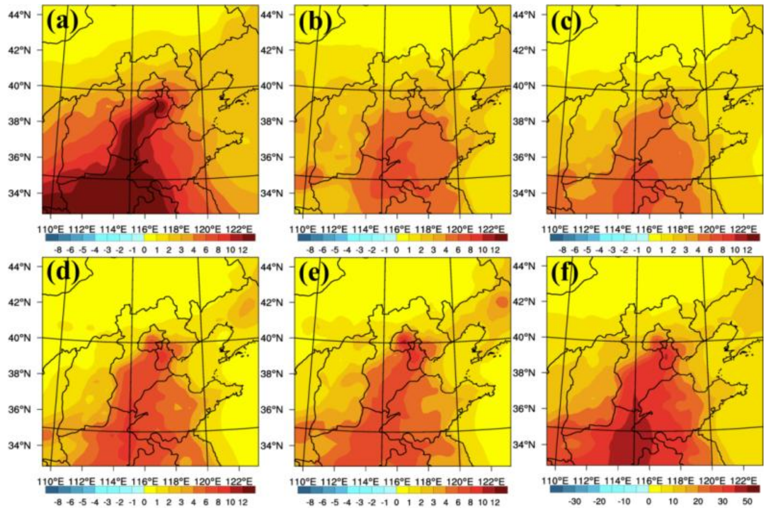

Figure 3. Predicted monthly mean changes of sulfate (a), nitrate (b), ammonium (c), BC (d), OC (e) and $PM_{2.5}$ (f) due to emission changes from 1960 to 2010

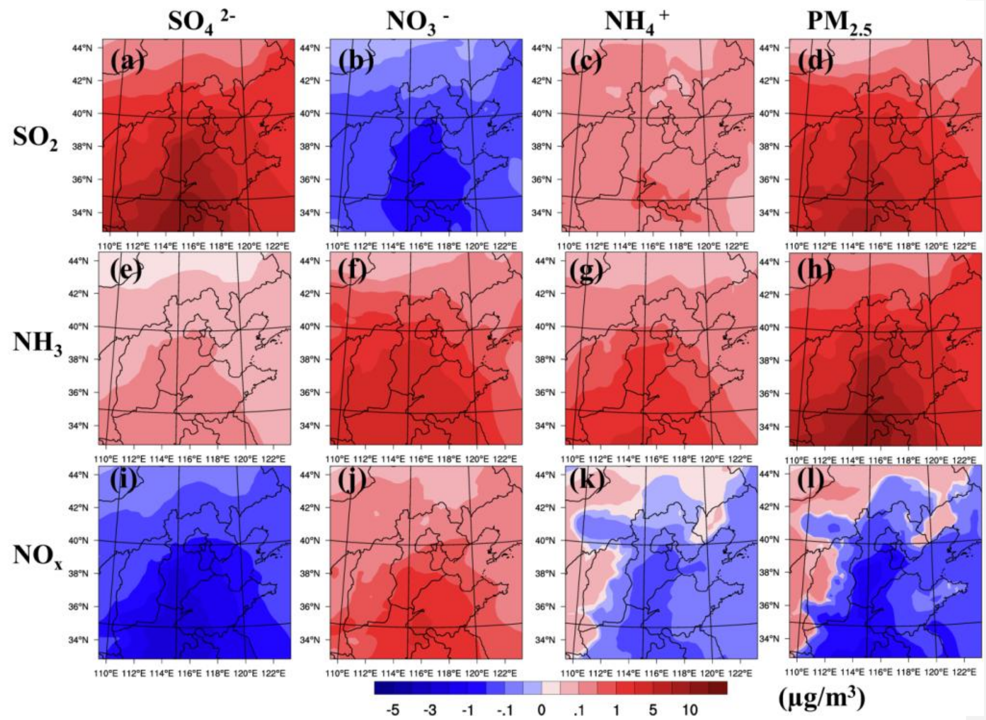

Figure 4. Responses of $PM_{2.5}$ and major $PM_{2.5}$ inorganic species (sulfate, nitrate, and ammonium) to individual changes in SO_2, NH_3 and NO_x emissions from 1960 to 2010

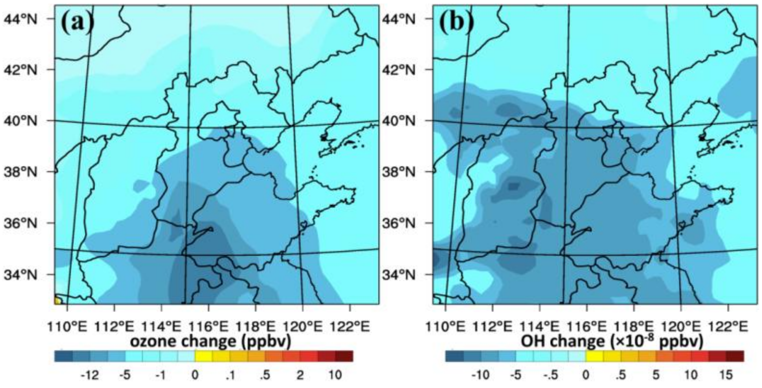

Figure 5. Daytime ozone (a) and daytime OH (b) changes due to NO_x emission increases

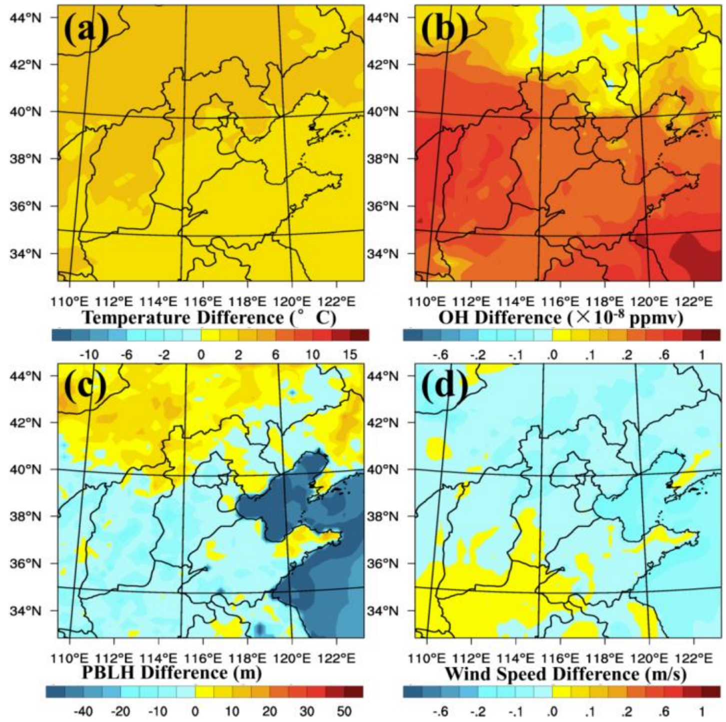

Figure 6. Monthly mean temperature difference due to perturbation in initial and boundary conditions (a), and daily mean OH (b), mean PBLH (c) and mean near surface wind speed changes (d) due to temperature increase

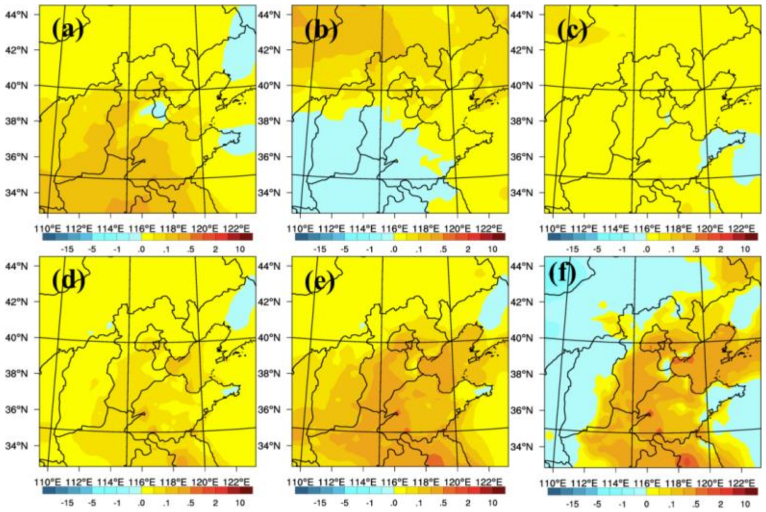

Figure 7. Monthly mean changes of sulfate (a), nitrate (b), ammonium (c), BC (d), OC (e), and PM$_{2.5}$ (f) and due to temperature increase

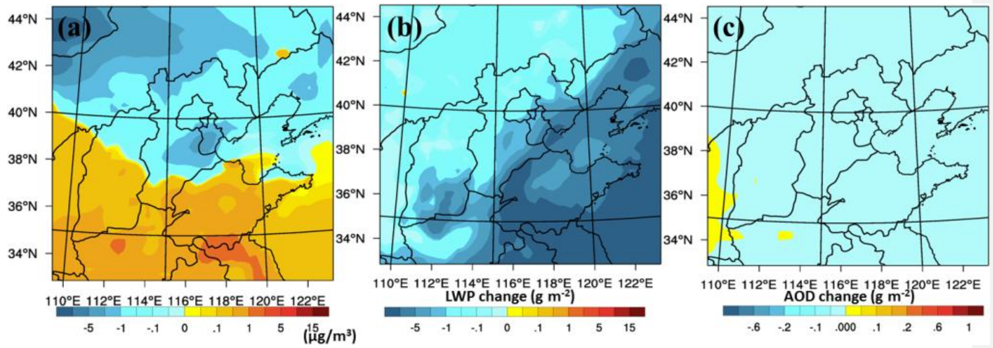

Figure 8. Monthly mean changes of PM$_{2.5}$ (a), LWP (b), and AOD at 600nm (c) due to RH decrease

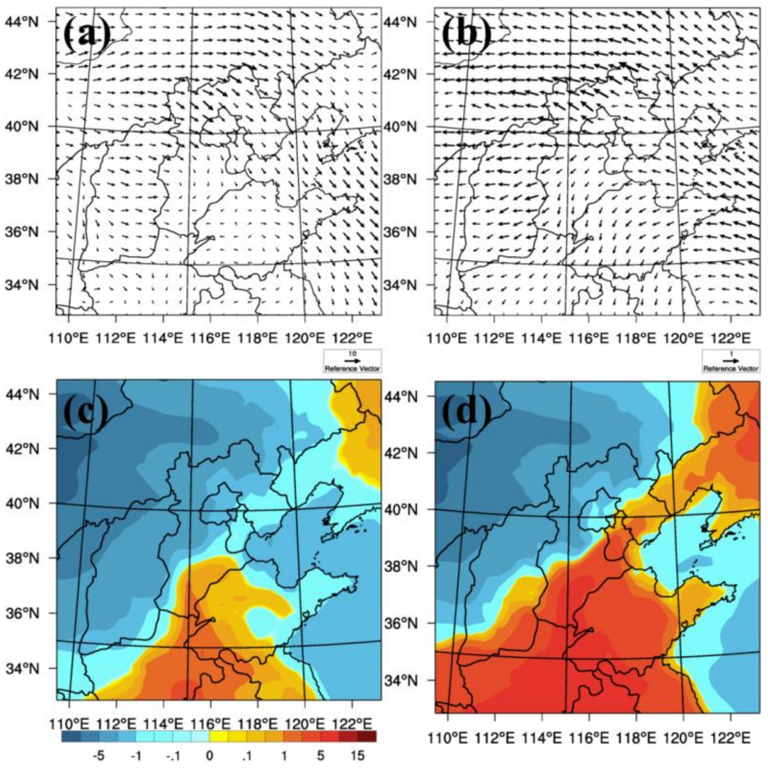

Figure 9. Monthly mean wind fields for WS20 case (a) and changes of wind speeds (CTL-CTL_WS20) (b), and mean changes of $PM_{2.5}$ concentrations based on 1960 emission levels (c) and 2010 emission levels (d)

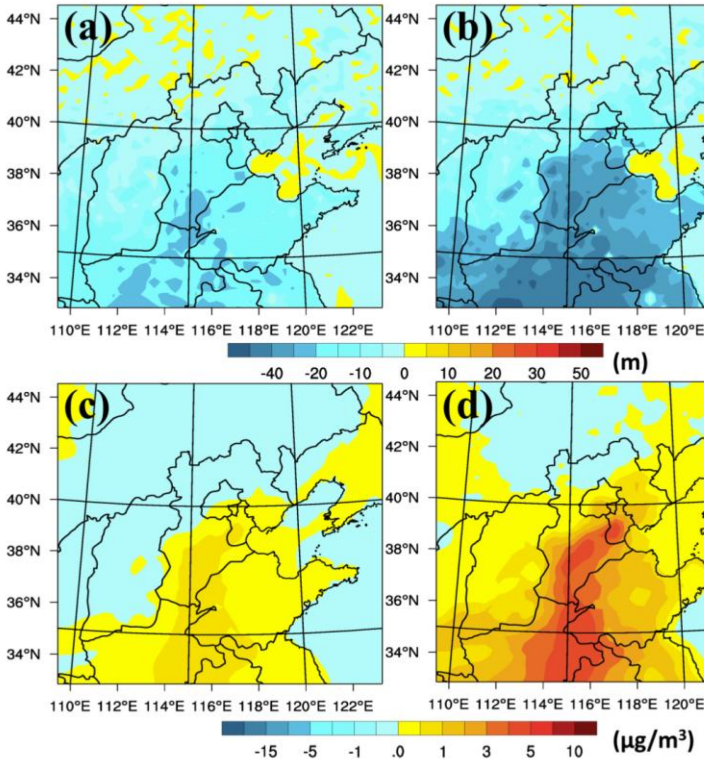

Figure 10. Monthly mean changes of daytime PBL heights for year 1960 (a) and 2010 (b), and of daytime $PM_{2.5}$ concentrations for year 1960 (c) and 2010 (d) due to aerosol-radiation interactions