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 $(NH_4)_2SO4$, so it may not be sufficient given the large amounts in SO_2 emissions. In addition, $NH_{3\ emission}$ 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.