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 $PM_{2.5}$."

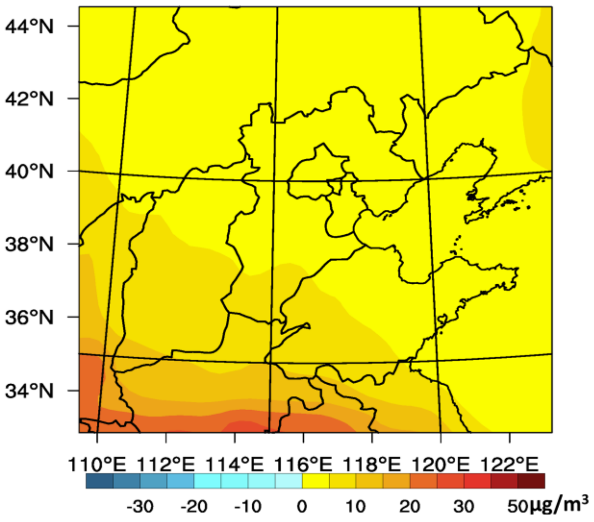

Figure 4 Changes of $PM_{2.5}$ due to boundary conditions from outer domain

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.

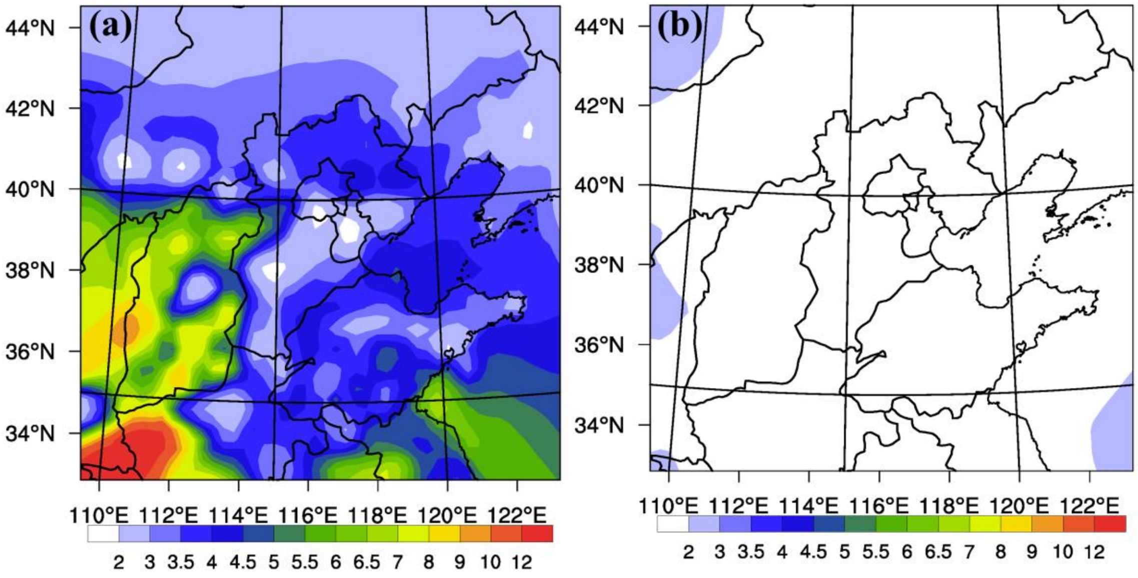

Figure 4 Averaged VOC:NOx ratios in 1960(a) and 2010(b)

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.

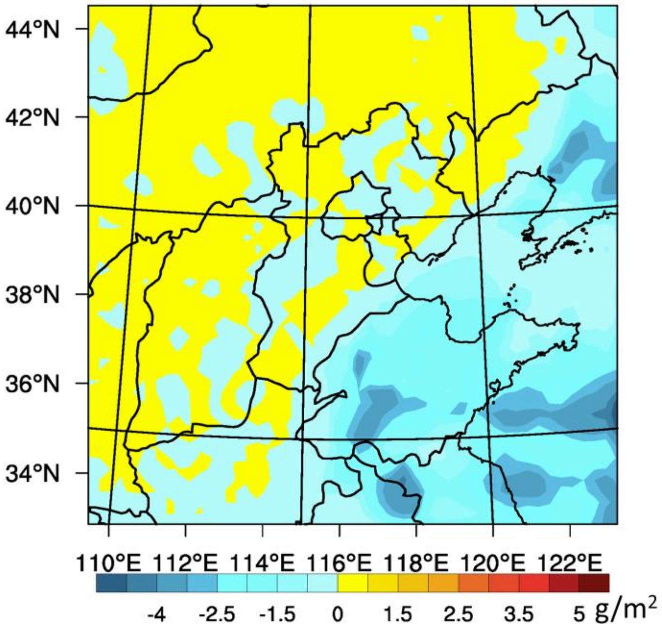

Figure Changes in LWP due to temperature perturbations

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.