

Response to referee comments on “Modeling study of the 2010 regional haze event in the North China Plain”

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

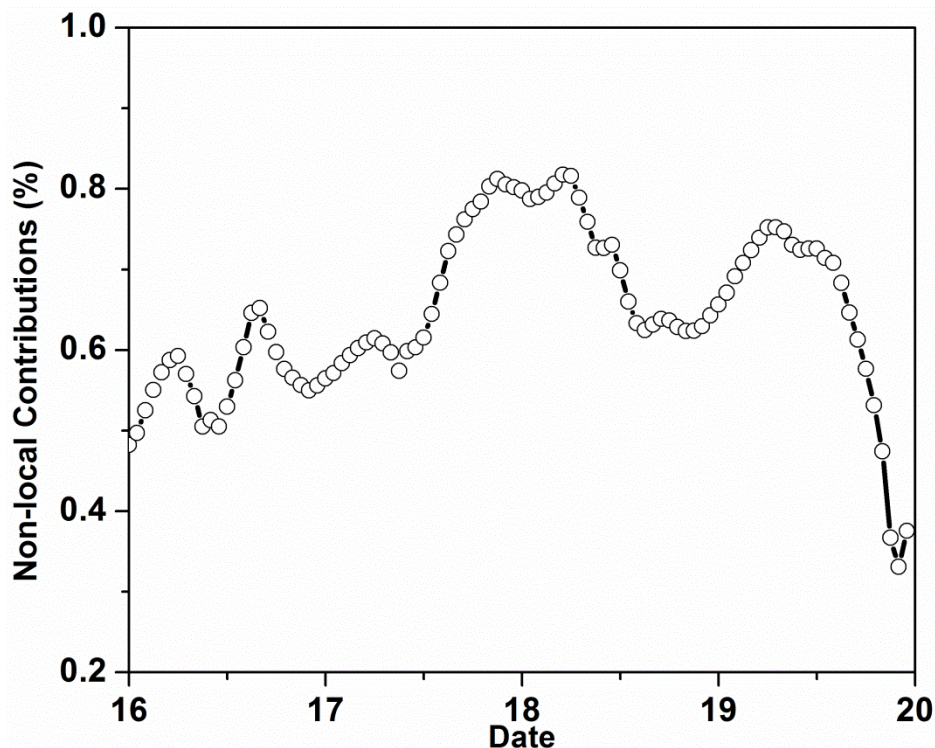
### **To Referee #1**

This manuscript presents a fairly detailed modeling analysis of a haze event in North China in winter. It discusses the chemical composition, transport, and formation mechanism of the haze episode. Using the coupled meteorology-aerosol simulation with WRF-Chem, the paper also estimates the effect of aerosol feedback on meteorology. It concludes that BC contributes to 50% of the overall effect of aerosol feedback on PBL and surface PM<sub>2.5</sub> concentrations. This paper confirms many findings from previous analysis of wintertime hazes in North China, including the importance of secondary inorganic aerosols, regional transport, and meteorological conditions. My main concerns are with the validity of using CO as a proxy of regional PM<sub>2.5</sub> transport and with the uncertainty in BC simulation. Minor issues are with the presentation clarity and some technical details. I recommend publication after these issues are properly addressed.

Major comments:

1). In Section 4.3, the authors use CO to indicate the source regions of PM<sub>2.5</sub> in Beijing. They did a sensitivity analysis by turning off CO emissions in Beijing and used the relative change in CO concentrations to denote the impact of surrounding regions on PM<sub>2.5</sub> pollution in Beijing. Although they showed that CO and PM<sub>2.5</sub> were highly correlated, CO has a much longer lifetime (~ 3 months) than PM<sub>2.5</sub> in winter and also undergoes different loss mechanisms (e.g. CO is not water soluble or lost through deposition). As such the sensitivity simulation using CO may not be appropriate for the sensitivity of PM<sub>2.5</sub>. Why didn’t the authors choose to conduct the sensitivity simulation using PM<sub>2.5</sub> directly? They have a model at their disposal. They can turn off primary sources of PM<sub>2.5</sub> as well as the emissions of its gaseous precursors over Beijing, the same approach as they did for the CO sensitivity simulation, to evaluate the impact of surrounding areas on Beijing.

Response: Following the referee’s suggestion, we have conducted the sensitivity simulation using PM<sub>2.5</sub> directly. The temporal variations of non-local contributions are shown below. The average contribution is about 64.5% from January 16 to January 19. The text has been modified and the CO based results have been replaced with these PM<sub>2.5</sub> based results.



2). A major conclusion of this paper is that BC is responsible for 50% of the aerosol feedback on meteorology which in turn influence surface concentrations of PM<sub>2.5</sub>. That conclusion is obviously dependent on the ability of the WRF-Chem model to simulate BC concentrations correctly as well as its relative contribution to the overall PM<sub>2.5</sub> composition. Figure 7 clearly shows that the model overestimates BC concentrations at the surface, for example by about a factor of two during the severe haze days (i.e. 18-19 January). Because the model underestimates OC and sulfate at the same time, this means the model has a significant overestimation of the fractional contribution of BC in total PM<sub>2.5</sub>. These two factors in combination suggest that the simulated absorbing effect of BC on meteorology in this winter episode should be significantly overestimated because of (1) overestimate in BC absolute concentration and (2) underestimate the role of scattering aerosols. This is an important issue that needs to be acknowledged at a minimum, given the emphasis of this manuscript on the simulated role of BC. But the authors did not discuss or even mention any uncertainty of this point. This is a major shortcoming of this paper and should be addressed before acceptance by ACP.

Response: Thanks for this important suggestion. We have added one paragraph to discuss the uncertainty of the BC. We also added some sentences in summary section to mention this point.

The contribution of BC absorption in aerosol feedbacks depends on the model performance in simulating BC and scattering aerosols (sulfate, OC). As shown in Figure 7, BC was overestimated, and sulfate and OC were underestimated in Beijing. The overestimation could be as large as a factor by 2 in some days. As a result, the contribution of BC absorption in aerosol feedbacks may have been overestimated in this study. To explore the uncertainties of the BC

absorption contribution, we conducted simulations by reducing BC emissions by 50%. After this BC emission perturbation, the changes of PBLH and  $PM_{2.5}$  concentrations at 2p.m. due to aerosol feedbacks and BC absorption are shown in Figure 14. The domain maximum increases of  $PM_{2.5}$  concentrations because of aerosol feedbacks and BC absorption are  $19.1\mu\text{g}/\text{m}^3$  and  $10.21\mu\text{g}/\text{m}^3$ , respectively. The domain maximum decreases of PBLH due to aerosol feedbacks and BC absorption are 235.7m and 114.2m, respectively. These numbers are smaller than before because BC emissions were reduced by 50%. Under these conditions of reduced BC concentrations, the contribution of absorption to the feedbacks was still large (50%) This number can be additionally reduced if OC and sulfate concentrations are simulated well. The underestimations of OC and sulfate were because some secondary formation pathways are missing in the current model. In the future, more accurate contribution of BC absorption in aerosol feedbacks can be estimated after the performances of the WRF-Chem model in simulating BC, OC and sulfate are improved.

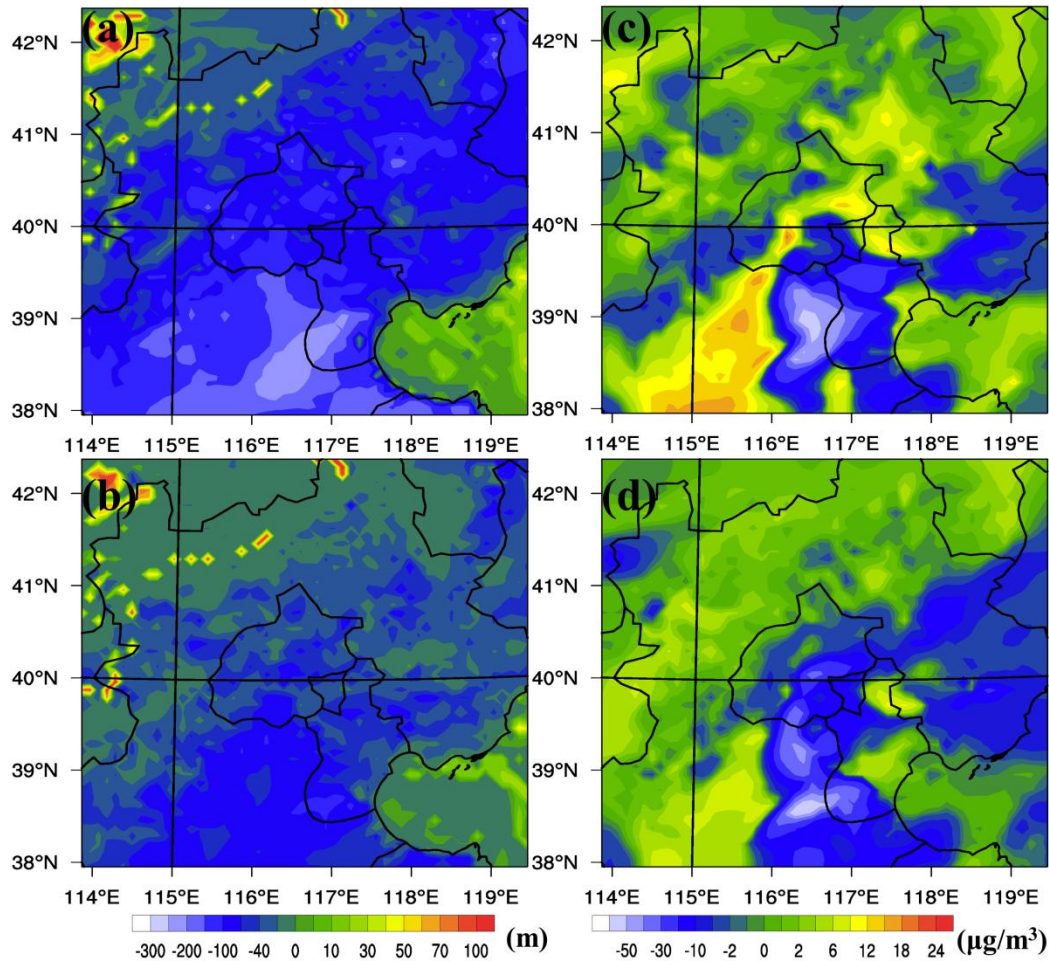


Figure 14. Differences of PBLH (unit: m) and  $PM_{2.5}$  concentration (unit:  $\mu\text{g}/\text{m}^3$ ) at 2p.m. between WF and NF scenarios (a, c) when BC emissions were reduced by half; differences of PBLH (unit: m) and  $PM_{2.5}$  concentration (unit:  $\mu\text{g}/\text{m}^3$ ) at 2p.m. between WF and NF scenarios (b, d) when BC emissions were reduced by half

## Minor Comments

1) pg 22784, 13: add “the time period” before “from 2001 to 2001”.

Response: We have added it.

2) Section 4.1: are the analysis presented in this section based on model simulations or observations? It’s not clear to me.

Response: The analysis in section 4.1 is based on model simulations. “Simulated” can be found in those figure titles (figure 5, figure 6).

3) Pg 22791, 17: upwards should be northward, since the discussion is on a 2-D surface pattern.

Response: We have corrected it.

4) Pg 22791, 18-110: the discussion of the high pressure is confusing. First, there is no clear indication of existence of a high pressure on Figure 4. Figure 4 shows only winds, not pressure fields. Second, the authors suggested high pressure would act to disperse pollution and low pressure leads to pollution accumulation. This is contradictory to the common understanding that high pressure is not conducive for pollution dispersion because of the subsidence and stability, and low pressure (e.g. cyclones) usually acts to reduce pollution.

Response: Thanks for this suggestion. Firstly, we plotted the pressure system but did not put it in the paper. Zhao et al. (2013) discussed the pressure system during this haze episode, so we have added the reference in this section. Hope it is not confusing now. The pressure system can be found in Figure 2 of Zhao et al. (2013).

Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W. W., Dong, F., He, D., and Shi, Q. F.: Analysis of a winter regional haze event and its formation mechanism in the North China Plain, *Atmos. Chem. Phys.*, 13, 5685-5696, doi:10.5194/acp-13-5685-2013, 2013.

Secondly, thank you for pointing out this misleading sentence. We agree that high pressure is not conducive for pollution dispersion because of the subsidence and stability, and low pressure usually acts to reduce pollution, but this relationship depends on the relative location to NCP and magnitudes of the pressure system. Actually, this haze event was terminated by the Mongolia anticyclone, which is a high pressure system. We were trying to say that the low pressure during this episode is unfavorable for the dispersion of air pollutants because air flows converged at the surface. To avoid misunderstanding, we have revised this sentence to “The weak high pressure system was replaced by a low pressure system that lasted until January 20, and this weather condition was not conducive for dispersion of air pollutants (Zhao et al., 2013)”. Hope it is better now.

5) Pg 22793, 115-17: Remove the sentence “However, few modeling studies have ...”. There have been quite some modeling studies on secondary aerosols during winter haze in China.

Response: We have removed this sentence.

6) Pg 22793, line 24-26: Are these factors from model or observations? If from models, how do they compare with observed factors?

Response: Thanks for this question. I made a mistake in figure 7. The plotted PM2.5 components were hourly data, it should be daily mean. We have corrected the figure. Those factors were from model. We calculated the concentrations and factors again using daily mean. We also compared them with observed factors.

We added one sentence in the manuscript: “The increasing factors for observed primary aerosols and SIA are 2.9 and 6.9, which are close to those factors from simulations.”

Table 2. Primary Aerosol, SIA and SOA ( $\mu\text{g}/\text{m}^3$ ) during Haze Days and Non-haze Days in Beijing

	Primary	SIA	SOA
Haze days	56.4	81.9	1.1
Non-haze days	14.2	10.8	0.3
Ratio	4.0	7.6	3.7

7) Pg 22797, line 23: add “and” before as a result

Response: We have added “and”.

8) Pg 22798, line 4: increase of temperature inversion should be changed to decrease of temperature gradient from surface to aloft, because Figure 10e shows only the difference in temperature between the two runs, not temperature profile.

Response: We have corrected.

9) Pg 22798, line 15-17: below Beijing should be changed to south of Beijing.

Response: We have corrected.

## To Referee #2

### General comments:

This manuscript, using the online coupled Weather Research and Forecasting-Chemistry (WRF-Chem) model, to investigate a haze event in NCP, the contributions of Secondary inorganic aerosols and transportation, particle composition, aerosols' feedback on the local meteorology and PM<sub>2.5</sub> itself, and feedbacks associated to Black Carbon. The aim the study is meaningful. The model simulations are in certain agreement with observations. Each point of the paper discussed (cause of haze event, composition, transport, radiative feedback) is important and worth doing. Unfortunately the paper involves too many aspects, but could not concentrate on the one or two targets to study and discuss them in detail. I recommend its resubmission basically in a revision in accordance with the following comments:

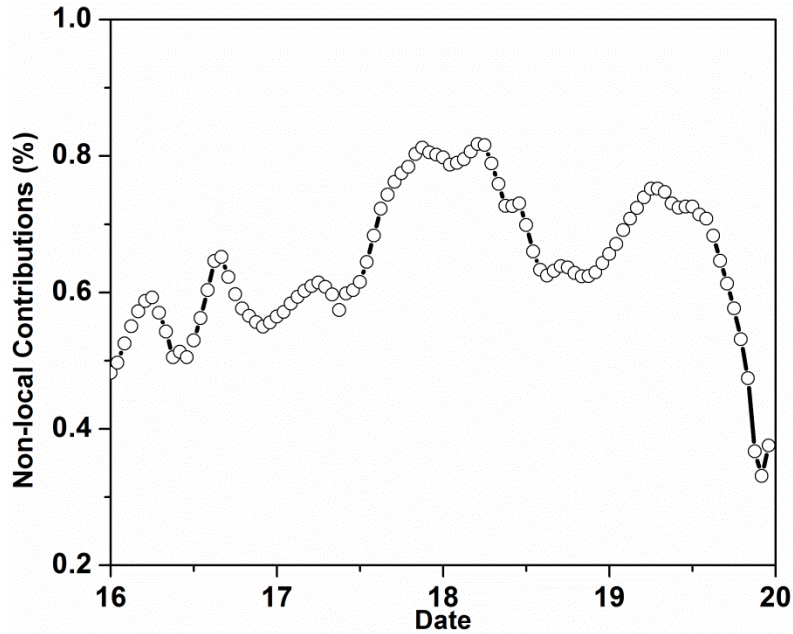
### Major comments:

1. Table S1-1 and Figure S2-10 are all supplement files. The authors spend too much space to discuss them in the manuscript which could not be seen by readers in fact and this may lead to misunderstanding. I suggest if they are necessary, please put them into the formal figures and tables in the paper, otherwise reject them and the use texts in the manuscript.

Response: Thanks for this suggestion. We have moved some important figures, like figure S4, figure S7 and S11 from supplement files to manuscript. Although the tables and figures in supplement files are not as important as those in the manuscript, they are still helpful to understand this haze event, so we did not discard them and put them in the supplement files. If the readers are interested in those materials, they can download the supplement file.

2. Though there is a high correlation between CO and PM<sub>2.5</sub> concentrations, it is groundless to use the CO transportation contribution as that of PM<sub>2.5</sub> directly. It is not difficult to calculate the transportation amount outside Beijing by model output parameters directly.

Response: Following the referee's suggestion, we have conducted the sensitivity simulation using PM<sub>2.5</sub> directly. The temporal variations of non-local contributions are shown below. The average contribution is about 64.5% from January 16 to January 19. The text has been modified and the CO based results have been replaced with these PM<sub>2.5</sub> based results.



3. AOD is a basic parameter to calculate aerosols direct radiative feedback. The AOD difference between model results and observation (CALIPSO) is obvious (Figure 3) and the model AOD is not good enough to support the aerosols radiative feedback calculation. Further AOD evaluations are needed (MODIS, etc.) for modeling the aerosols radiative feedback reasonably.

Response: Thanks for this suggestion. The difficulty is that during these heavy haze events there is little data available. We analyzed the CALIPSO data as well as AOD from surface sites (Figure S5). The surface sites did not report data during the heavy haze period, but the model well predicted the values before and after. Actually, we did download MODIS data, but most are missing during the study period due to extremely high aerosol loadings. If anything we have probably underestimated the feedback effects due to our underprediction of PM<sub>2.5</sub> and AOD based on CALIPSO.

#### Minor comments

1. Please examine the quota format in the manuscript carefully (different quota format appears in the paper).

Response: We have checked the format and they look fine. The publishing assistant also checked it before publication in ACPD, and I corrected following her instructions.

2. Figure1, 3, 4, 11,12 are not clear and need to be redrawn.

Response: We redrew figure 1,3,4,11 and 12. Hope they are clear now.

3. Figure 4 and its related content (page 22791), wind vectors in the figure needs legend explanation. Pressure system is explained on line 1-10, but it is not drawn in the figure. Please examine the similar questions in other figures (Figure 4, 5, 7, etc.)

Response: Thanks for this suggestion. Firstly, we plotted the pressure system but did not put it in the paper. Zhao et al. (2013) discussed the pressure system during this haze episode, so we have added the reference in this section. Hope it is not confusing now. The pressure system can be found in Figure 2 of Zhao et al. (2013).

Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W. W., Dong, F., He, D., and Shi, Q. F.: Analysis of a winter regional haze event and its formation mechanism in the North China Plain, *Atmos. Chem. Phys.*, 13, 5685-5696, doi:10.5194/acp-13-5685-2013, 2013.

4. Please examine the figure captions under the figure and the explanation in the manuscript. They are not same for some figure.

Response: Thanks for this comment. I have checked all figure titles and made corrections. Although some expressions are different, they denote the same meaning.