Response to Comments of Reviewer #1

Manuscript number: acp-2021-611
Title: Simulated impacts of vertical distributions of black carbon aerosol on meteorology and $PM_{2.5}$ concentrations in Beijing during severe haze events

General comments:
This study presents the impacts of vertical distributions of black carbon aerosol on local $PM_{2.5}$ concentration and meteorology using the Weather Research and Forecasting with Chemistry model (WRF-Chem) and airborne measurement of black carbon vertical profiles. The manuscript is well written and easy to follow, and the simulated impacts are well documented and reported quantitatively. However, my main concern is that the scientific wisdom gained from this research, contributing/adding to the current knowledge of the community, is not clearly conveyed in the current form of the manuscript. In other words, I found multiple places where the authors can discuss more on the implications of the reported impacts/results, as well as the physical reasonings behind them, instead of only reporting the changes from one simulation to another. Therefore, I suggest major revision.
Overall, I think this is a nicely designed and conducted modelling study, which can make valuable contribution to the field. Here I provided some specific comments and suggested changes regarding my main concern of the manuscript.

Thanks to the referee for the helpful comments and suggestions. We have revised the manuscript carefully and the point to point responses are listed below.

Major concerns/questions:

1. Abstract:
   A general comment, the current form reads like a report summary, could the authors reconstruct the abstract in a way that scientific questions/goals of the study are clearly posed in the beginning, followed by a concise summary of the key findings (not only reporting the quantitative statistics, but also the logical flows behind these changes), and ended with implications of the study.

Response:

We have revised the abstract following the Reviewer's suggestion in revised manuscript.

2. Numerical experiments:
   When VerBC_obs and VerBC_hs1-6 (RT) are compared with noBCrad (which is ran with the default BC profiles, except the optical properties are set to zero), it seems you're attributing the simulated differences between them solely to radiative effects, while assuming difference in vertical BC profiles between these simulations and noBCrad has no non-radiative effects (e.g. microphysical or chemical effects). Could you please justify this?

Response:

The experimental design in our study serves two purposes: (1) to compare the direct radiative effects (DRE) of BC with original and modified vertical profiles in two severe haze events and (2) to investigate the roles of parameterized BC vertical profiles in influencing meteorological conditions and $PM_{2.5}$. All the numerical experiments are summarized in Table 2 (see below).

For the first purpose, the differences in model results between CTRL (VerBC_obs) and NoBCrad experiments (CTRL (VerBC_obs) minus NoBCrad) represents the DRE of BC with original (modified) profiles on meteorology and $PM_{2.5}$ concentrations.

For the second purpose, six BC profiles parameterized as exponential functions (VerBC_hs1-6) and one profile of transport-dominated feature (VerBC_RT) were considered, and the differences between VerBC_hs1-6 and NoBCrad (VerBC_hs1-6 minus NoBCrad) as well as the difference between VerBC_RT and NoBCrad (VerBC_RT minus NoBCrad) were quantified.

We have explained in the last paragraph of Section 2.4 that 'In VerBC_obs, VerBC_hs1-6, and VerBC_RT experments, the modifications of BC vertical profiles were performed only when the direct radiative effect of BC was calculated. All other physical and chemical processes in these experiments still used the original BC vertical profiles simulated by the model.'.

We have also explained in the second paragraph of Section 2.4 that 'In the case of NoBCrad, the BC DRE was turned off by setting the BC mass concentration equal to zero when calculating the optical properties of BC, following the studies of Qiu et al. (2017) and Chen et al. (2021).' Therefore the differences in BC profiles between VerBC_obs (hs1-6, RT) and NoBCrad had no non-radiative effects in this study.

We have discussed the limitation of this study in the last paragraph of Section 6: 'There are channels for further improvement in near-future research. We distribute BC mass vertically according to the observed fractions of BC in individual model layers for each day without considering the hourly variations of BC vertical profiles due to the lack of data. Such assumed distribution of BC based on observation may not be consistent with the dynamical (winds, temperature, etc.) and chemical processes of the atmosphere. Further efforts are needed to examine the roles of BC vertical profiles in coupled chemistry-weather models.'.

Table 2. Numerical experiments. Y indicates "on", and N indicates "off".

Simulations	BC direct radiative effect (DRE)		
	DRE	BC vertical profiles for calculation of DRE	
	Turn on/off	Types description	Modified dates
CTRL	Y	Simulated by model	No modification
NoBCrad	N	Simulated by model	No modification
VerBC_obs	Y	Modified according to intraday observations	11-12 and 16-19 December

VerBC_hs1-6	Y	Modified according to $C(h)=C_0 \times e^{-h/hs}$ function[a]	12 and 16-19 December
VerBC_RT	Y	Modified according to observations on 11 December 2016	12 and 16-19 December

[a] The values of *hs* in VerBC_hs1, VerBC_hs2, VerBC_hs3, VerBC_hs4, VerBC_hs5 and VerBC_hs6 are 0.35, 0.48, 0.53, 0.79, 0.82 and 0.96, respectively.

3. Model evaluation:
   L309-311: Indeed the model overestimate $PM_{2.5}$ concentration if one compares the averages over the 9 day period, however, there is a lot more one can say about this model/obs comparison. For example, it seems the overall overestimation in the simulation is mostly coming from the clean days (DEC 13-15), whereas during the 2 haze events, the model seems to do fairly good job, comparing to obs, quantitatively, but there seems to be a timing difference, which could be due to discrepancies in advection between obs & model. Only reporting the mean biases doesn't help the reader understand the difference between model and obs that much.

Response:

Following the Reviewer's suggestion, we have divided the studied period (11-19 December 2016) into: (1) the first pollution event (11-12 December), (2) the clean days (13-15 December), (3) the second pollution event (16-19 December) and have added the statistical metrics for $PM_{2.5}$, SO_2, NO_2, CO and O_3 for clean days and the two haze events as Table S2 (see below) of the Supplementary Material.

We have also added the following sentences to describe Table S2 in the second paragraph of Section 3.1: 'It should be noted that the model performance in simulating $PM_{2.5}$, SO_2, CO and O_3 is better during the two haze events than on clean days. For hourly $PM_{2.5}$, for example, the MBs (NMBs) are 29.1 μg m^{-3} (82.5%) on clean days and 6.3 μg m^{-3} (3.5%) during the two haze events. The possible reasons for the overall overestimation of $PM_{2.5}$ are as follows: (1) the model biases in underestimating WS10 and daytime PBLH; (2) the uncertainties in anthropogenic emission data (e.g. the overestimation in the BC emissions) (Qiu et al., 2017; Chen et al., 2021). Overall, the model can capture fairly good the two severe pollution events in Beijing during 11-19 December 2016.'.

Table S2. Statistical metrics for $PM_{2.5}$, SO_2, NO_2, CO and O_3 on clean days and in two haze events.

Periods	Variables	SIM	OBS	R	MB	NMB	MFB
Clean days	$PM_{2.5}$ (μg m^{-3})	64.4	35.3	0.15	29.1	82.5%	84.1%
	SO_2 (ppbv)	5.5	3.6	-0.02	1.9	53.4%	18.8%
	NO_2 (ppbv)	28.8	20.8	0.55	7.9	38.0%	38.5%
	CO (ppmv)	11.0	14.2	0.64	-3.1	-22.0%	-50.7%

	O_3 (ppbv)	0.9	0.7	0.18	0.2	30.0%	37.6%
	$PM_{2.5}$ (μg m^{-3})	186.1	179.8	0.64	6.3	3.5%	8.0%
Two haze events	SO_2 (ppbv)	9.1	9.9	0.29	-0.7	-7.4%	-13.5%
	NO_2 (ppbv)	57.2	48.2	0.70	8.9	18.5%	12.5%
	CO (ppmv)	4.6	3.2	0.88	1.4	43.0%	-39.4%
	O_3 (ppbv)	2.2	2.4	0.30	-0.2	-9.3%	-8.4%

Moreover, what is the meteorological conditions during the clean period, is there any precipitation event? Cloud formation? These can also help the reader understand these events better. I wonder if a meteorological overview of this 9-day period can be added to the beginning of Section 3 or section 3.2?

Response:

As suggested, we have added a meteorological overview of this 9-day period at the beginning of Section 3.2. The observed hourly precipitation (mm) and 3-hourly total cloud cover (%) in Beijing are added as Figures 5g-5h (see below).

'The first haze event started on December 11 when southeasterlies transported polluted air from southern BTH to Beijing (Fig. 2a). Although the southeasterlies turned into northeasterlies in Beijing on December 12, $PM_{2.5}$ concentrations were still high because of the high relative humidity (63.2%) that was conductive to the formation of secondary aerosols. With the relatively high wind speed of 3.6 m s^{-1} and low relative humidity of 37.2% in Beijing during 13-15 December, the haze pollution gradually disappeared (Fig. 2c-2e). From 16 to 19 December, $PM_{2.5}$ began to accumulate again with unfavorable diffusion conditions (WS10 of 1.4 m s^{-1}) and enhanced formation of secondary aerosols under high relative humidity of 67.1% (Li et al., 2019; Dai et al., 2021). Throughout the simulated period of 11-19 December 2016, Beijing had no precipitation and was partly cloudy (Fig. 5g-5h).'.

We have also added the following sentences in the first paragraph of Section 3.2 to describe the Fig. 5f-5h: 'The simulated SWDOWN in CTRL experiment agrees well with the observations with R and MB of 0.76 and -14.9 W m^{-2}. Due to the limitation of the model outputs, the model provides only information of whether there is cloud in the grid or not. The model can reproduce well the presence of cloud during 11-19 December 2016. Both observations and model results show no precipitation in the studied time period.'

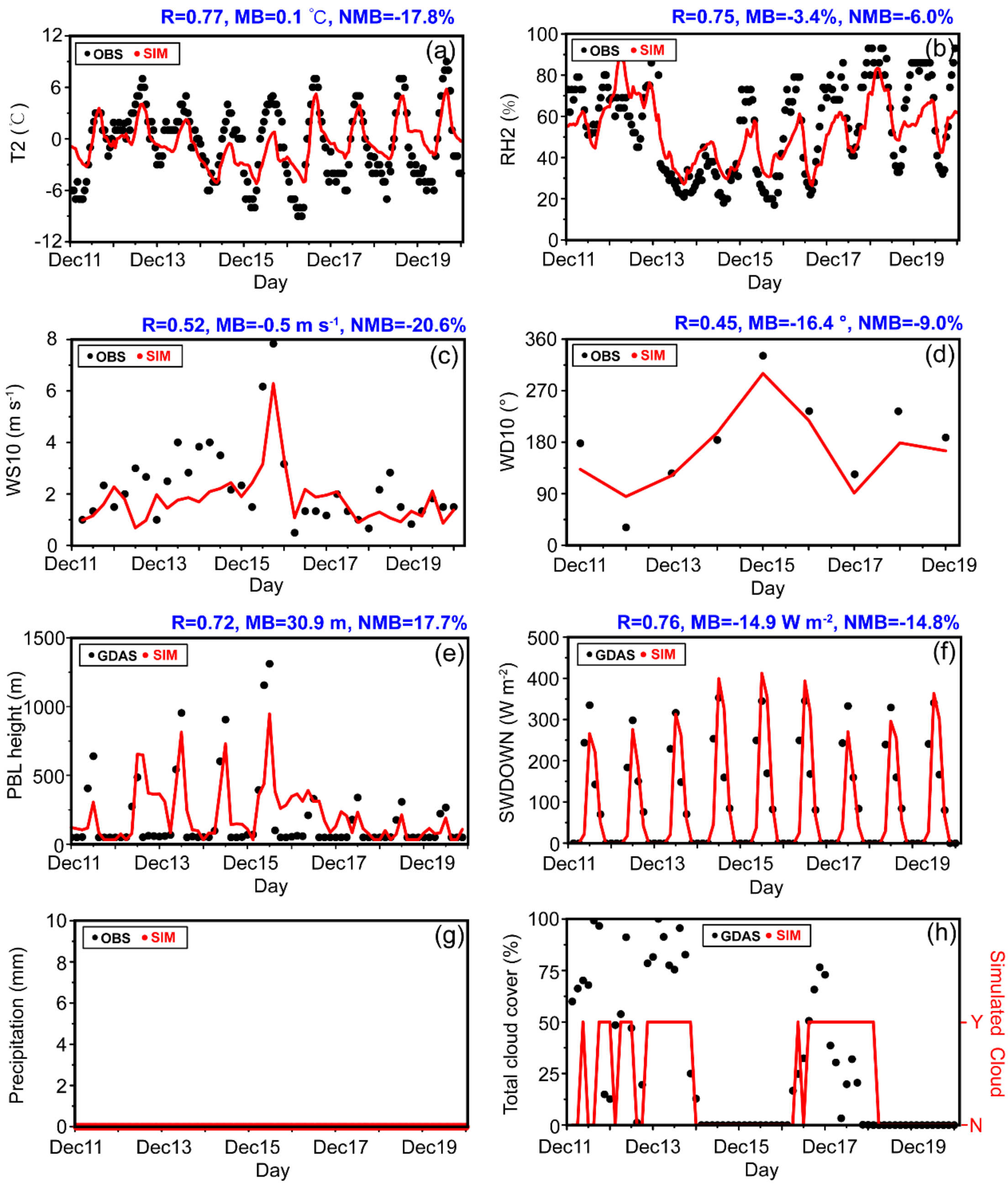

Figure 5. Comparisons of simulated meteorological parameters from CTRL simulation with measurements. The black dots and red lines are the observed (reanalysis) and simulated hourly data of T2 (°C), RH2 (%), precipitation (mm), and 3-hourly data of PBL height (m), SWDOWN (W m^{-2}), total cloud cover (%), 6-hourly data of WS10 (m s^{-1}), and daily data of WD10 (°) in Beijing from 11 December 2016 to 19 December 2016. PBLH, SWDOWN, and total cloud cover are taken from GDAS. The WRF-Chem model output shows only a grid has cloud (Y) or no cloud (N).

L350-353: Again, more details are needed here. Even though model overestimate PBLH in the mean, daily maximum PBLH values from obs exceed that from the model, and the overestimation is mainly due to the fact that obs has '~0m' PBLH during most of the day, is this an artifact or obs mis-characterize PBL? More details here would be helpful.

Response:

We have added the following sentences to discuss more details about PBLH in the first paragraph of Section 3.2:'The model overestimates PBLH by 30.9 m (17.7%) in Beijing as averaged over 11-19 December 2016. The overestimation is mainly in hours of 0:00-8:00 LT and 17:00-23:00 LT. It is noted that the observed PBLH values provided by GDAS of NOAA were mostly 50-60 m at 0:00-8:00 LT and 17:00-23:00 LT in Beijing, far below the simulated mean value of 154.5 m in these hours. There might be biases in the observed PBLH from GDAS. Several previous studies showed that the values of observed PBLH from Lidar measurements were about 200 m at night during haze events (Wang et al., 2012; Luan et al., 2018; Chu et al., 2019).'.

4. Section 4:

   Is there attempt to compare the simulations with obs-corrected/modified BC vertical distribution to observation? I wonder if getting the vertical structure of BC close to obs help improve the overall simulation relative to observations? And this could be an important result of this study, such that getting observationally constrained BC vertical distribution help (or does not help) improve the simulated local meteorology and $PM_{2.5}$ concentration.

Response:

It is a good idea to compare the performance of CTRL (with original BC vertical profiles) and VerBC_obs (with modified BC vertical profiles) simulations in simulating meteorology and $PM_{2.5}$ during the two haze events. We have added Tables S3 and S4 as well as Figure S4 in the Supplementary Material (see below). We have also added a new Section 4.3 to describe Tables S3, S4 and Figure S4:

'4.3 Model performance in simulating meteorology and $PM_{2.5}$ with the original and modified BC vertical profiles

It is of interest to compare the performance of CTRL (with original BC vertical profiles) with that of VerBC_obs (with modified BC vertical profiles) in simulating meteorological parameters and $PM_{2.5}$ during the two haze events. Figure S4 shows the comparisons between observed T2, RH2, WS10, WD10, PBLH and the simulated values from the CTRL and VerBC_obs simulations in Beijing in the two haze events (11-12 December and 16-19 December 2016). Relative to CTRL simulation with original BC vertical profiles, VerBC_obs simulation with modified BC vertical profiles has better performance in simulating T2, WS10 and PBLH except for WD10 and RH2 in the first pollution event. While the MBs of T2, RH2, WS10, WD10 and PBLH are 0.2°C, 0.0%, -0.4 m s^{-1}, 3.8°, and 45.0 m in CTRL, they are 0.0°C, 2.2%, -0.1 m s^{-1}, 10.9°, 29.0 m in VerBC_obs, respectively (Table S3). In the second pollution event, the positive bias in PBLH (MB=43.7 m, NMB=42.9%) in CTRL is reduced to 33.9 m and 33.3% in VerBC_obs.

Table S4 shows the statistical comparison between observed hourly surface-layer

$PM_{2.5}$ and the model results from CTRL and VerBC_obs in Beijing for each day during the two haze events. The model with modified BC vertical profiles can enhance the capability in simulating the temporal variation of $PM_{2.5}$ for each day; the correlation coefficient between simulated hourly concentrations and hourly observations in each day of the studied period increased from 0.04-0.84 in CTRL to 0.24-0.93 in VerBC_obs.

Table S3. Statistical analyses of the performance of CTRL (with original BC vertical profiles) and that of VerBC_obs (with modified BC vertical profiles) in simulating meteorological parameters. The values in RED indicate better performance in VerBC_obs than in CTRL.

		Obs/Sim			MB		NMB	
		Obs	CTRL	VerBC_obs	CTRL	VerBC_obs	CTRL	VerBC_obs
The first pollution event	T2 (°C)	0.2	0.4	0.2	0.2	0.0	N.A.	N.A.
	RH2 (%)	65.5	65.6	67.7	0.0	2.2	0.0%	3.3%
	WS10 (m s^{-1})	1.8	1.4	1.8	-0.4	-0.1	-22.9%	-2.6%
	WD10 (°)	105.2	109.0	116.1	3.8	10.9	3.6%	10.4%
	PBLH (m)	152.2	197.2	181.2	45.0	29.0	29.6%	19.0%
The second pollution event	T2 (°C)	-1.1	0.0	0.2	1.2	1.3	N.A.	N.A.
	RH2 (%)	65.3	55.2	57.3	-10.1	-8.0	-15.5%	-12.3%
	WS10 (m $^{s-1}$)	1.4	1.4	1.1	0.0	-0.3	-0.1%	-19.7%
	WD10 (°)	196.2	165.7	173.0	-30.5	-23.2	-15.5%	-11.8%
	PBLH (m)	101.9	145.6	135.8	43.7	33.9	42.9%	33.3%

Table S4. Statistical analyses of the performance of CTRL (with original BC vertical profiles) and that of VerBC_obs (with modified BC vertical profiles) in simulating $PM_{2.5}$ concentrations. The values in RED indicate better performance in VerBC_obs than in CTRL.

	Obs/Sim			R		MB		NMB	
	Obs	CTRL	VerBC_obs	CTRL	VerBC_obs	CTRL	VerBC_obs	CTRL	VerBC_obs
Dec 11	159.7	214.1	235.9	0.81	0.93	54.4	76.2	34.1%	47.7%
Dec 12	212.3	185.9	189.6	0.04	0.24	-26.4	-22.7	-12.4%	-10.7%
Dec 16	100.7	117.7	115.3	0.56	0.65	17.0	14.6	16.9%	14.5%
Dec 17	184.7	190.8	192.9	0.63	0.82	6.0	8.2	3.3%	4.4%
Dec 18	219.5	190.4	199.8	0.38	0.38	-29.1	-19.6	-13.2%	-9.0%
Dec 19	208.4	217.8	220.5	0.84	0.89	9.4	12.1	4.5%	5.8%

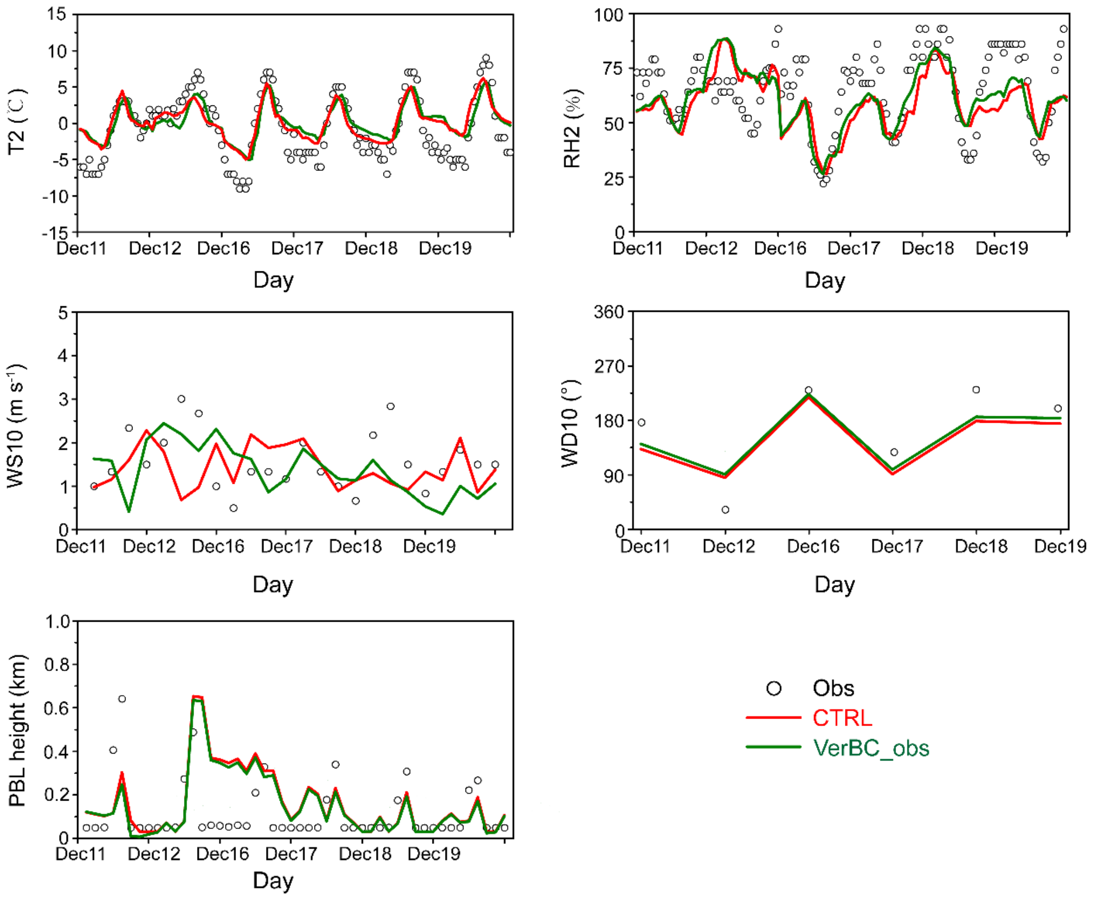

Figure S4. Comparisons of simulated hourly T2 (°C), hourly RH2 (%), 3-hourly PBL height (m), 6-hourly WS10 (m s^{-1}) and daily WD10 (°) from CTRL (original BC vertical profiles; red lines) and VerBC_obs (modified BC vertical profiles; green lines) experiments with observations (black circles) in Beijing during two pollution events (11-12 December and 16-19 December 2016).

L405-411: I feel like Figure S4 is an important figure which can be moved to the main text, as it shows the role of BCrad on regional circulation/wind pattern, which further leads to changes in local $PM_{2.5}$ concentration. However, how does BC DRE enhance the northerlies north of NCP and weakened the wind speed in central and southern Beijing is still not clear to me. I also think investigating the physical mechanisms behind this is critical to the whole study. I suggest more detailed discussion and further analyses here.

Response:

We have moved the original Figure S4 to be Figure 8 in the main text of the revised manuscript. We have also added in Figure 8 the changes in T2 and the sea-level pressure (SLP) caused by BC DRE with either original or modified vertical profiles (see below) to investigate the physical mechanisms of the changes in regional circulation.

We have added the following sentences in the second paragraph of Section 4.1 to describe Figure 8: ‘Figure 8 shows the spatial distributions of changes in T2, sea-level pressure (SLP) and wind at 10 m caused by BC DRE with original and modified vertical profiles. BC DRE with both original and modified vertical profiles produced anomalous northeasterlies in eastern BTH during the two haze events. The mechanism of such changes is that BC DRE induced a strong warming over the Bohai Sea in the east of BTH with a maximum warming of about 1.8 ℃, resulting in an anomalous low-pressure here and consequently anomalous northeasterlies in eastern BTH (Fig. 8c-8d). The similar changes in winds caused by the heating effect of BC were also reported in previous studies (Gao et al., 2016; Qiu et al., 2017; Chen et al., 2021).'.

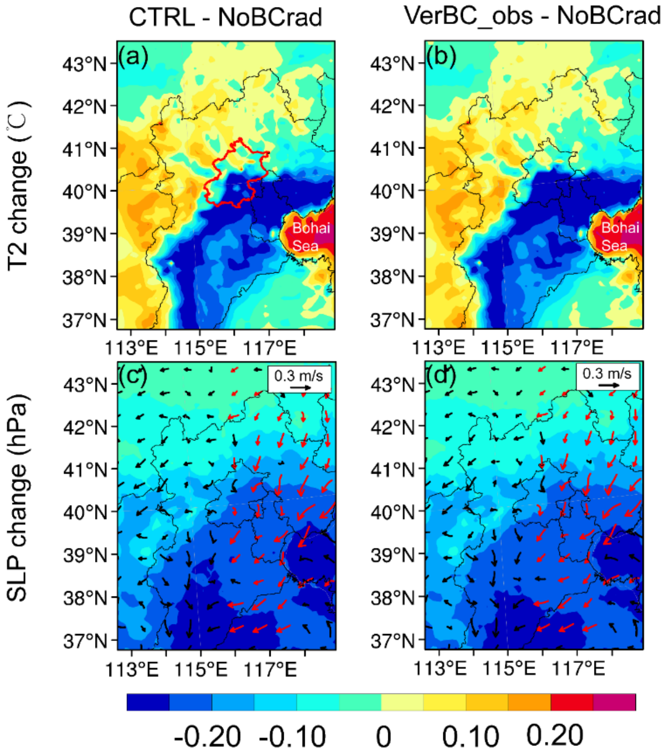

Figure 8. The changes in T2, SLP and wind at 10 m induced by BC DRE with original (a and c; CTRL minus NoBCrad) and modified vertical profiles (b and d; VerBC_obs minus NoBCrad) averaged over the period of 12:00 – 18:00 LT of the two haze events, respectively. (c-d) The northeasterlies in the east of BTH are denoted in red.

Related to Fig. 9: What are the implications of these results from the IPR analyses?

Response:

Figure 9 is now Fig. 10 in the revised manuscript. BC DRE changes $PM_{2.5}$ concentrations mainly through processes of VMIX, CHEM and TRA. The results show that BC DRE increases $PM_{2.5}$ concentrations below 256 m because of positive contribution of VMIX and CHEM. However, in the upper layers (from 256 to 1555 m), the contribution of VMIX and CHEM becomes negative which led to decreases in $PM_{2.5}$ concentrations, indicating that the decrease in PBLH caused by BC DRE inhibits the transport of low-layer pollutants to the upper layer.

5. Section 5:

   L500-509: Could you discuss more on how does larger delta_T_BC result in larger reduction in PBLH?

Response:

We have added the following sentences in the second paragraph of Section 5.1:

'BC aerosol leads to cooling at the surface and warming in the upper PBL, both of which weaken the convection in the boundary layer and consequently reduce the PBLH (Ding et al., 2016; Wang et al., 2018; Chen et al., 2021). In our study, with *hs* values increasing from 0.35 to 0.96, ΔT_{BC} increased from 0.17 to 0.42°C, and the ΔT_{BC} value was 0.51°C in VerBC_RT case (Fig. 13a). The larger ΔT_{BC} indicates stronger cooling at the surface, such temperature inversion at 12:00-18:00 LT resulted in more stable stratification and further inhibited the development of PBL. The cooling at the surface also reduced sensible heat flux from the surface (Fig. S5), suppressing vertical turbulence and hence reducing PBLH (Wilcox et al., 2016).'

Related to Fig. 13: Again, what are the implications of these results? Are these results case/event dependent? Or they can be generalized, e.g. to make arguments on the role of BC vertical distribution on local accumulation and advection of pollutant?

Response:

Figure 13 is now Fig. 14 in the revised manuscript. We get a general conclusion here: "A larger *hs* means less BC at the surface and more BC in the upper atmosphere, which results in a stronger temperature inversion and hence larger reductions in PBLH (larger BC-induced increases in surface-layer $PM_{2.5}$)."

6. Conclusions:
   Again, the current conclusion section reads like a summary of the simulation results, which feels redundant and repetitive. I suggest reconstruction of this section, in a way that the repetitive results summary can be minimized or summarized in higher level languages, and the authors are encouraged to discuss more on the implications of the results and the study in general, e.g. whether these results are event specific or they can be generalized. Languages on the role of BC vertical distribution in affecting local meteorology and transport/accumulation of pollutant should be added.

Response:

As suggested, we have reconstructed the conclusions in Section 6 in revised manuscript.

L580-582: You mentioned results from this study highlights the importance of accurate representation of BC vertical profiles in models. I don't think this point has been made clear, as you haven't shown how simulations with obs-modified BC profiles compared to observations, is there improvement at all?

Response:

We have added new Section 4.3 to investigate the model performance in simulating meteorology and $PM_{2.5}$ with the original and modified BC vertical profiles. The results show that, relative to the CTRL simulation with original BC vertical profiles, VerBC_obs simulation with modified BC vertical profiles has better performance in simulating T2, WS10, WD10 and PBLH except for RH2 in the first pollution event.

Meanwhile, the model with modified BC vertical profiles can enhance the capability in simulating the temporal variation of $PM_{2.5}$ for each day. See also our responses to your Comment 4 (Major concerns/questions).

Minor comments:

1. L44, L477: is "sharper" the right word? To me, the sharpest decline is associated with hs=0.35 where BC drops from 13 to 4 in the first 500m, whereas when hs=0.96, BC drops from 6 to 4 (much slower decline).

Response:

We have deleted the word of 'sharper' and rewritten the sentence as: 'A larger *h*s means less BC at the surface and more BC in the upper atmosphere.'.

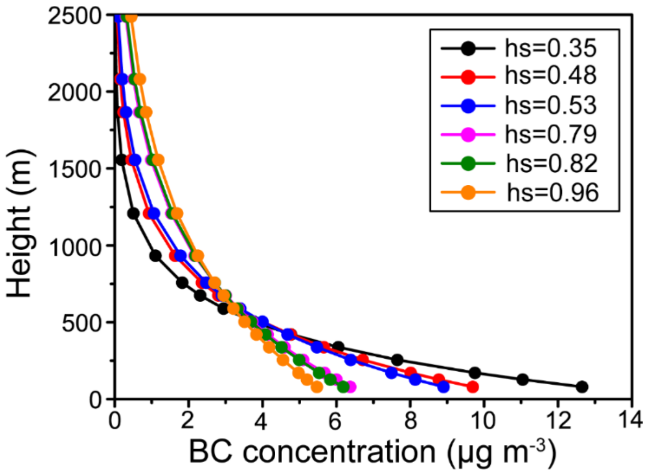

Figure 11. Vertical profiles of BC concentrations parameterized as six exponential functions for 12 and 16-19 December 2016.

2. Section 2.1 Model configuration:
   How does the current model configuration deal with properties at domain top, are the top of the domain forced by free-tropospheric properties/motions from NCEP? Is there nudging in the simulation?

Response:

In our model, the top layer was set to 50 hPa. The global tropospheric analyses from NCEP have 57 vertical layers from the surface to 30 hPa altitude. So, the top of the domain in the WRF-Chem model was forced by free-tropospheric properties from NCEP. Analysis nudging was on in outer domain (d01) and off in inter domain (d02) in our simulation.

3. Section 2.3 Observational data:
   Regarding MODIS dataset, did you use Aqua or Terra, at what resolution (i.e. what level of the dataset). More details here could be useful.

Response:

We have clarified this in the first paragraph of Section 2.3:

‘The MYD03 (Level-1A) product with 1 km spatial resolution from Aqua platform and the MOD03 (Level-1A) product with 1 km spatial resolution from Terra platform were used in this study.’.

4. L275-280: A bit hard to follow here, on L275, you meant ‘VerBC_hs1~6’ instead of ‘VerBC_hs1’, right? When I first read this part, it confused me, but when I saw figure 11, it started to make sense. I suggest some clarifications here.

Response:

Yes. We have changed ‘VerBC_hs1’ to ‘VerBC_hs1~6’.

5. L367: I don’t think “well simulate” is the right word to describe Fig. S3, even the horizontal distribution seems off between MODIS and simulation.

Response:

We have changed ‘well simulate’ to ‘generally reproduce’.

6. L414-418: repeated in the figure caption (Fig. 8), suggest reconstruction.

Response:

Figure 8 is now Fig. 9 in the revised manuscript. We have rewritten this sentence as: ‘Figure 9 illustrates the impacts of BC DRE with original and modified vertical profiles on surface-layer $PM_{2.5}$ as well as the differences in simulated surface-layer $PM_{2.5}$ between VerBC_obs and CTRL (VerBC_obs minus CTRL) in Beijing during the two haze events.’.

7. L498-499: not following this sentence, suggest rewording.

Response:

We have reworded the sentence as: ‘In current regional air quality models, the uncertainties in BC profiles could influence the capability of a model to simulate a cooling effect of BC on surface-air temperature (Wang et al., 2019).’

8. Figure 2: is the $PM_{2.5}$ concentration shown here for near-surface or 850 hPa? Blue and red squares are hard to see on a printed copy. 2(i), could you also show BTH time series as well?

Response:

It is the surface-layer $PM_{2.5}$ (covers from 0 m to 79.5 m). We have added ‘surface-layer’ in figure caption (Fig. 2). Squares are also added in Figs. 2e and 2i. The daily surface-layer $PM_{2.5}$ concentrations averaged over BTH from 11 to 19 December 2016 are added in Fig. 2j (see below).

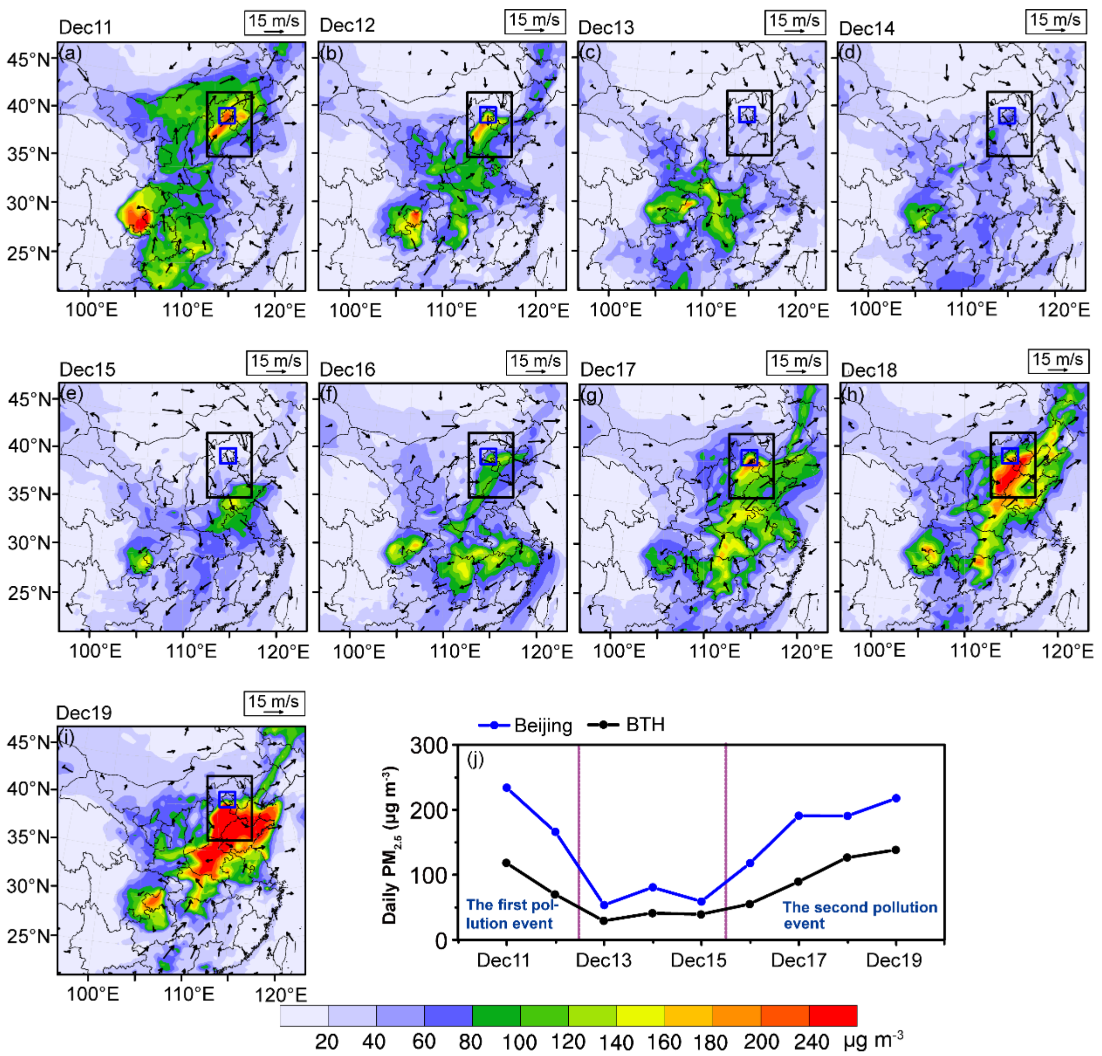

Figure 2. (a-i) Simulated spatial distributions of surface-layer $PM_{2.5}$ concentrations (μg m^{-3}) and winds (m s^{-1}) at 850 hPa at 2 pm local time from 11 to 19 December 2016. Black and blue squares in each panel denote the regions of Beijing-Tianjin-Hebei and Beijing, respectively. (j) Time series of simulated mean daily $PM_{2.5}$ concentration from 11 to 19 December 2016 averaged over the Beijing (blue square) and BTH (black square).

9. Figure 3: SO_2 panel, is the NMB really 0.0% or a precision/rounding issue?

Response:

Corrected. The NMB is 1.4% in SO_2 panel.

10. Figure 5: Wind speed and direction are critical to understanding the difference between obs and model, I wonder if daily or 6-hr (or even smaller intervals) averages of wind speed and direction can be shown to get a cleaner comparison between obs and model. It's pretty hard to compare the two in the current form of Fig. 5, especially WD10.

Response:

We now show the time series of observed and simulated 6-hourly WS10 and daily WD10 in Fig. 5c and 5d.

11. Figure 6: suggest indicating the 1:1 line with a different color (other than red, as it thought it was a fitting line for the red dots at first).

Response:

We have changed the red line to green line (see below).

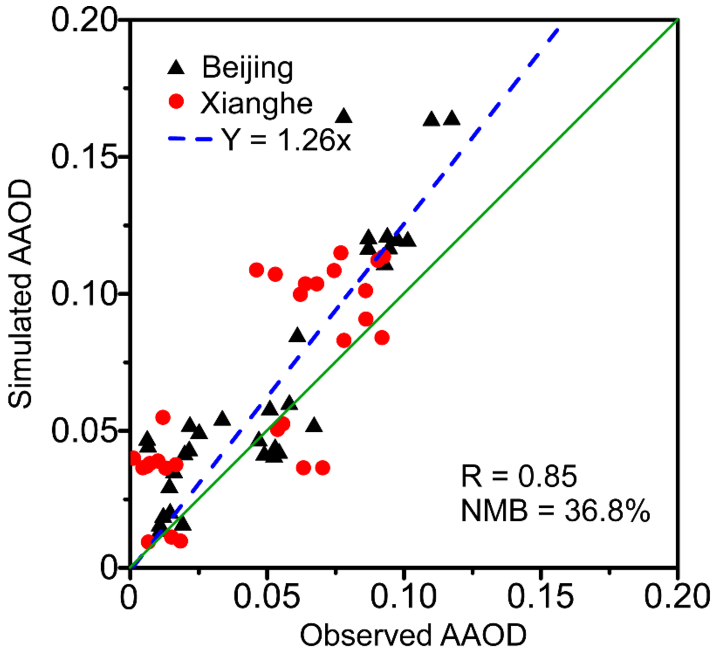

Figure 6. Comparison of simulated absorption aerosol optical depth (AAOD) at 550 nm with observations in Beijing (116.38°E, 39.98°N) and Xianghe (116.96°E, 39.75°N) station from 11 to 19 December 2016.

12. Figures 3, 5, 7, 8, 11: needs x-label titles, e.g. day, time …

Response:

Added.

References:

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Response to Comments of Reviewer #2

Manuscript number: acp-2021-611

Title: Simulated impacts of vertical distributions of black carbon aerosol on meteorology and $PM_{2.5}$ concentrations in Beijing during severe haze events

General comments:

In the current work, the authors used WRF-Chem with IPR analysis and sensitivity tests to assess the effect of vertical distribution of BC on meteorology and surface $PM_{2.5}$ concentrations during Beijing haze episodes. The research topic is very well suited to the scope of the current journal and the results presented are very interesting to their readers. However, there are some uncertainties, especially in the methodology, that require some modification before it is accepted. Without a concrete explanation of the methodology, the reviewer is uncertain whether the model settings chosen by the author will actually answer the authors' research questions. See the general and specific comments listed below.

Thanks to the referee for the helpful comments and suggestions. We have revised the manuscript carefully and the point to point responses are listed below.

Major concerns/questions:

1. There are no presentations on clouds and precipitation throughout the manuscript. In the presence of clouds and precipitation, the radiation effect of aerosols is quite different from that of sunny days. Please show and compare measured and simulated clouds, precipitations, or solar radiation.

Response:

The comparison between simulated and observed hourly precipitation (mm), 3-hourly total cloud cover (%) and 3-hourly shortwave downward radiation flux (SWDOWN, W m^{-2}) in Beijing are added in Figures 5f-5h (see below). The corresponding statistical metrics are added in Table 3 (see below). We have also added the following sentences in the first paragraph of Section 3.2 to describe the comparison.

'The simulated SWDOWN in CTRL experiment agrees well with the observations with R of 0.76 and MB of -14.9 W m^{-2}. Due to the limitation of the model outputs, the model provides only information of whether there is cloud in the grid or not. The model can reproduce well the presence of cloud during 11-19 December 2016. Both observations and model results show no precipitation in the studied time period.'.

Table 3. Statistical metrics for temperature at 2 m (T2; °C), relative humidity at 2 m (RH2; %), wind speed at 10 m (WS10; m s^{-1}), wind direction at 10 m (WD10, °), PBLH (m), SWDOWN (W m^{-2}), $PM_{2.5}$ (μg m^{-3}), SO_2 (ppbv), NO_2 (ppbv), CO (ppmv) and O_3 (ppbv).

Variables	SIM[a]	OBS[b]	R[c]	MB[d]	NMB[e]	MFB[f]
T2 (°C)	-0.5	-0.6	0.77	0.1	-17.8%	-13.1%
RH2 (%)	52.5	55.8	0.75	-3.4	-6.0%	-0.3%

WS10 (m s^{-1})	1.8	2.3	0.52	-0.5	-20.6%	-11.5%
WD10 (°)	165.6	182.0	0.45	-16.4	-9.0%	0.7%
PBLH (m)	205.8	174.9	0.72	30.9	17.7%	72.9%
SWDOWN (W m^{-2})	86.0	100.8	0.76	-14.9	-14.8%	-17.4%
$PM_{2.5}$ (μg m^{-3})	145.6	132.3	0.77	13.2	10.0%	15.7%
SO_2 (ppbv)	7.9	7.8	0.38	0.1	1.4%	-2.9%
NO_2 (ppbv)	47.7	39.2	0.78	8.5	21.6%	20.2%
CO (ppmv)	1.8	1.9	0.73	-0.1	-4.9%	6.4%
O_3 (ppbv)	6.7	6.8	0.66	-0.1	-1.2%	-36.0%

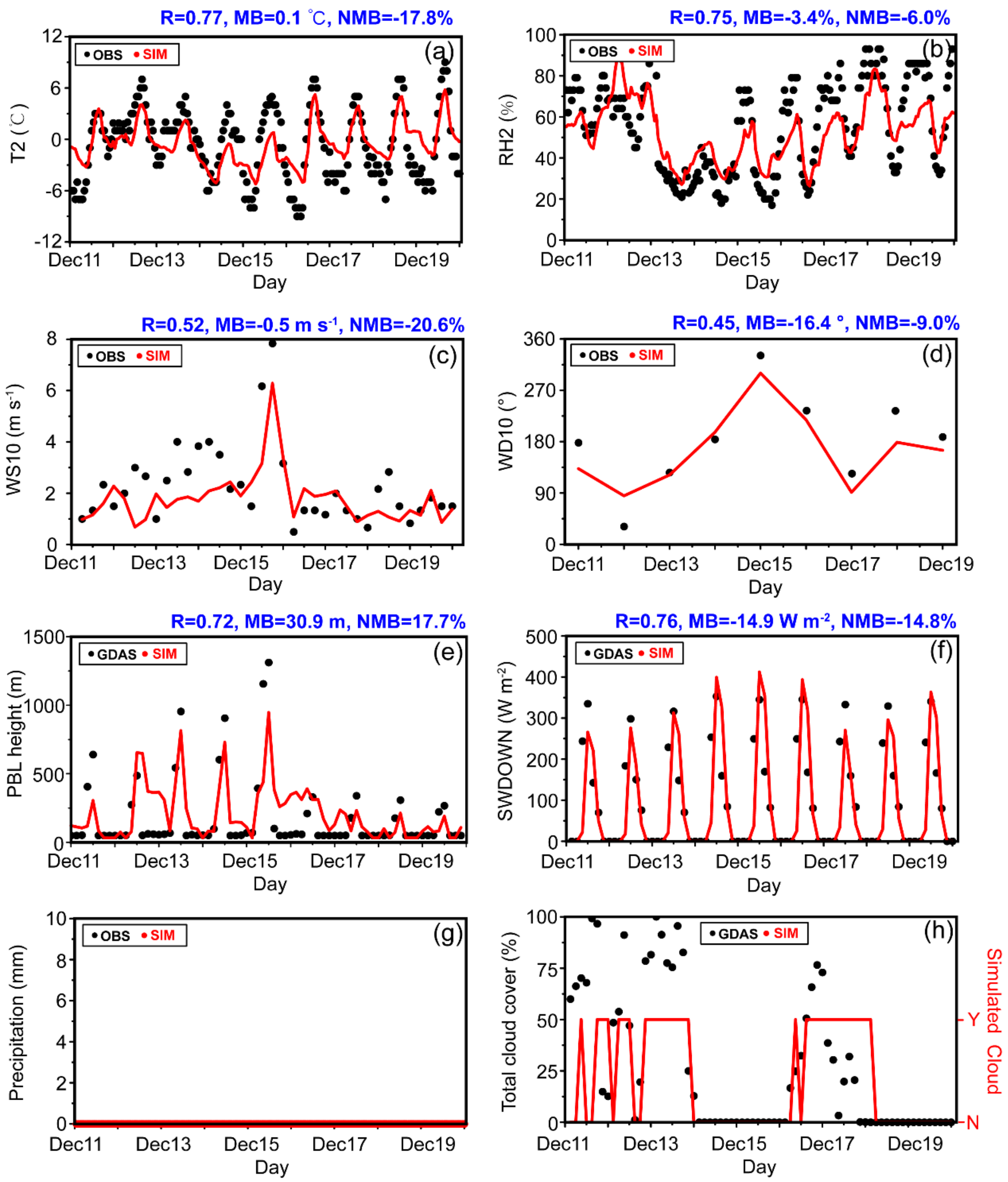

Figure 5. Comparisons of simulated meteorological parameters from CTRL simulation with measurements. The black dots and red lines are the observed (reanalysis) and

simulated hourly data of T2 (°C), RH2 (%), precipitation (mm), and 3-hourly data of PBL height (m), SWDOWN (W m^{-2}), total cloud cover (%), 6-hourly data of WS10 (m s^{-1}), and daily data of WD10 (°) in Beijing from 11 December 2016 to 19 December 2016. PBLH, SWDOWN, and total cloud cover are taken from GDAS. The WRF-Chem model output shows only a grid has cloud (Y) or no cloud (N).

Specific comments:

1. Abstract:
   The authors simply repeated "$PM_{2.5}$ concentration", but more specifically, "$PM_{2.5}$ surface air concentrations" ($PM_{2.5}$ can be aloft). Please define "$PM_{2.5}$ concentration" as "surface air concentrations of $PM_{2.5}$" when it is first appeared.

Response:

We have clarified in the abstract that we mean the surface-layer $PM_{2.5}$ (covers from 0 m to 79.5 m).

2. P. 8, Ln. 154, "NCEP": Please be more specific. For example, specify the datasets number.

Response:

We have added the following details of NCEP in the first paragraph of Section 2.1:

'Meteorological initial and boundary conditions in this model were derived from National Center for Environmental Prediction (NCEP) Final (FNL) Operational Model Global Tropospheric Analyses (ds083.2) with a spatial resolution of 1°×1°.'.

3. P. 9, Ln. 168, "FINN": Please provide horizontal and temporal resolution.

Response:

We have added the following details of FINN in the second paragraph of Section 2.1:

'Biomass burning emissions were taken from the Fire INventory from NCAR (FINNv1.5) which provides daily emissions at a horizontal resolution of ~1 km^2 (Wiedinmyer et al., 2011).'.

4. P. 10, Ln. 189, "updrafts": The convection scheme also includes downdrafts and precipitation. Which parameterization did you use for convection? Please list it in Table 1. The subgrid-scale wet deposition is calculated in the convection model so they can be counted as CONV here, but they need to be count as WET (wet deposition).

Response:

We have revised this sentence as 'CONV refers to the transport within the sub-grid wet convective updrafts, downdrafts and precipitation (Chen et al., 2019).'. We used Grell 3-D scheme (an improved version of the GD scheme) for convection. We have also added it in Table. 1 (see below).

Table1. Physical and chemical options for WRF/Chem.

WRF/Chem Model Configuration	Description

Microphysics scheme	Lin microphysics scheme (Wiedinmyer et al., 2011)
Longwave radiation scheme	RRTMG scheme (Zhao et al., 2011)
Shortwave radiation scheme	RRTMG scheme (Zhao et al., 2011)
Gas phase chemistry scheme	CBMZ (Zaveri and Peters, 1999)
Aerosol module	MOSAIC (Zaveri et al., 2008)
Photolysis scheme	Fast-J (Wild et al., 2000)
Boundary layer scheme	Yonsei University Scheme (YSU) (Hong et al., 2006)
Pavement parameterization scheme	Noah Land Surface Model scheme
Cumulus option	Grell 3-D ensemble scheme

5. P. 10, Ln. 192, "cloud": How do you separate cloud chemical formation from in-cloud scavenging? The formation of $PM_{2.5}$ due to cloud chemistry occurs only when the cloud and rain droplets are completely evaporated. On the other hand, $PM_{2.5}$ in the cloud and rain droplets are not counted as $PM_{2.5}$ and are removed from the air (although they are not completely removed unless droplets reach the ground).

Response:

In the WRC-Chem model, the cloud chemistry is an independent module. We quantify the changes in $PM_{2.5}$ before and after calling this module as the impact of cloud chemistry on $PM_{2.5}$. In-cloud scavenging is included in wet scavenging (WET). WET considers the wet removal of aerosols by in-cloud scavenging and below-cloud washout.

6. P. 10, Ln. 194, "WET represents the wet removal processes of aerosols": In-cloud and below-cloud? Again, how do you separate "cloud" in CHEM from in-cloud scavenging in WET here?

Response:

See our response to your previous question.

7. P. 10, Ln. 195, "OTHER": What are they? Dry deposition should be the one but any other processes?

Response:

OTHER is calculated by subtracting the six physical/chemical processes of $PM_{2.5}$ (CONV, VMIX, CHEM, TRA, WET and EMI) from the total $PM_{2.5}$.

8. P. 10, Ln. 202: "Beijing station", where is it and what does it belong to? Does the station belong to NOAA? Maybe not, but the data was obtained from NOAA's website.

Response:

'Beijing station' is 'Beijing-Capital International Airport station (station number: 54511099999)' in NOAA. We have changed 'Beijing station' to 'Beijing-Capital International Airport station (40.08°N, 116.58°E)' in the revised manuscript. The station (54511099999) belongs to NOAA and it was obtained from the Integrated Surface Dataset (Global).

9. P. 10, Ln. 205, PBL of GDAS: what is the horizontal resolution of GDAS? Even

though the GDAS is the analysis (or one can call it observation), is their PBL also assimilated with observed PBL? If not, PBL of GDAS cannot be regarded as observation as you show in Fig. 5. If it is assimilated with observed PBL, please specify which observation data was assimilated.

Response:

The Global Data Assimilation System (GDAS) has a horizontal resolution of 1°×1°. GDAS continuously collects observational data from the Global Telecommunications System (GTS) and other sources. The PBL is calculated by the meteorological data in GDAS. We have added extra notes in the first paragraph of Section 2.3 : 'Due to the limited observations of planetary boundary layer heights (PBLH), shortwave downward radiation flux (SWDOWN) and total cloud cover in Beijing, the reanalysis data of 3-hourly PBLH, SWDOWN and total cloud cover for Beijing from the Global Data Assimilation System (GDAS) with a spatial resolution of 1°×1° (http://ready.arl.noaa.gov/READYamet.php) were used for model evaluation. More details about the GDAS dataset can be found in Rolph (2013) and Kong et al. (2015).'. We have also changed 'OBS' to 'GDAS' in three panel of PBLH, SWDOWN and total cloud cover in Fig. 5 (see below).

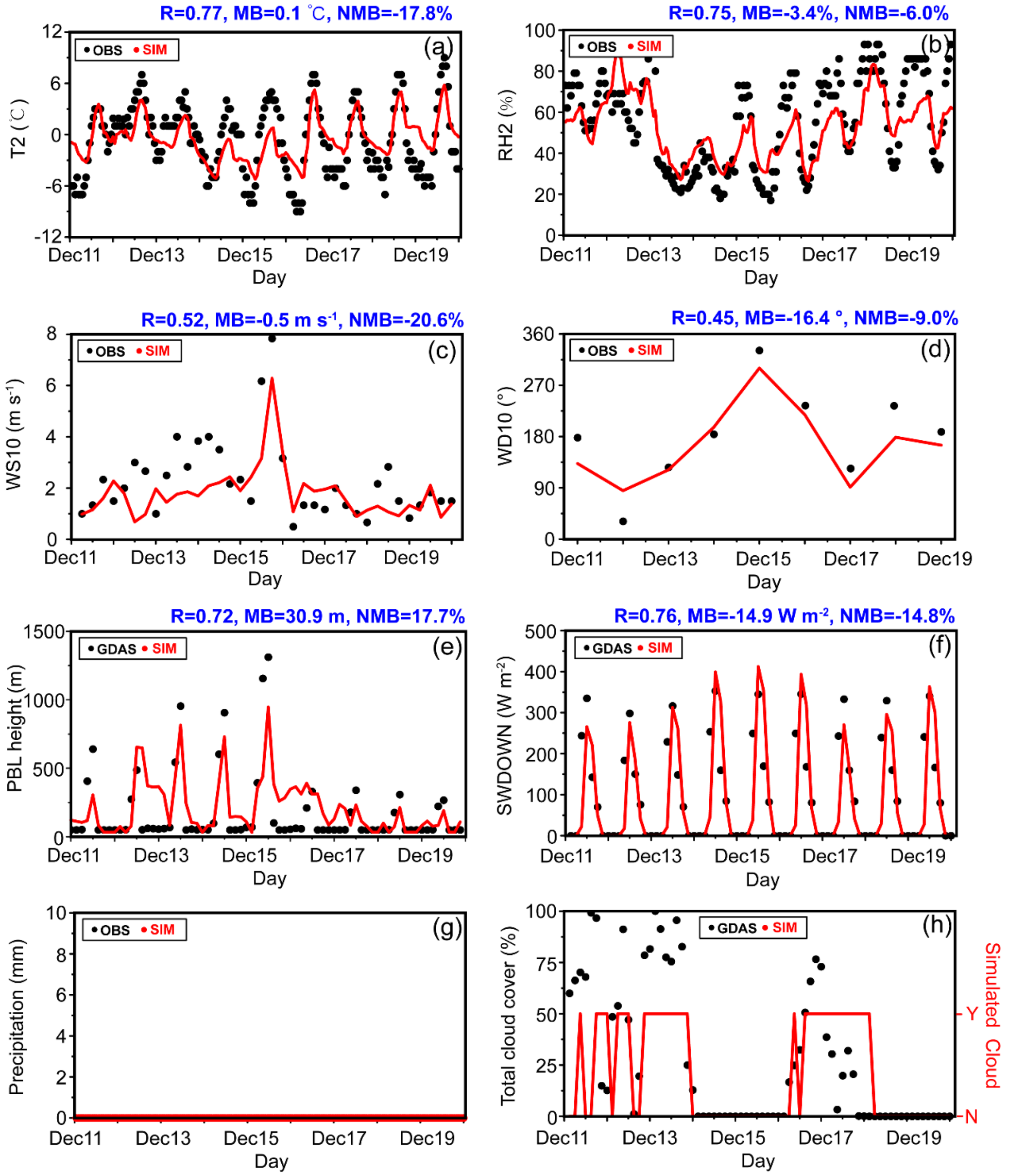

Figure 5. Comparisons of simulated meteorological parameters from CTRL simulation with measurements. The black dots and red lines are the observed (reanalysis) and simulated hourly data of T2 (°C), RH2 (%), precipitation (mm), and 3-hourly data of PBL height (m), SWDOWN (W m^{-2}), total cloud cover (%), 6-hourly data of WS10 (m s^{-1}), and daily data of WD10 (°) in Beijing from 11 December 2016 to 19 December 2016. PBLH, SWDOWN, and total cloud cover are taken from GDAS. The WRF-Chem model output shows only a grid has cloud (Y) or no cloud (N).

10. P. 12, "indirect radiative effects": how? The authors used the Lin's scheme for cloud microphysics, which is a single moment scheme and thus cloud albedo and cloud lifetime effects are not considered. Is it intended simulation settings?

Response:

To run with indirect radiative effects, we turn on the aerosol direct effect and select Lin's microphysics scheme. Then we turn on the prognostic number density option to allow the Lin's scheme to be double moment and be able to communicate the desire to run indirect effect. Such method is based on WRF-Chem User's Guide (https://ruc.noaa.gov/wrf/wrf-chem/user-support.htm).

11. P. 15-16, discrepancy of vertical profiles on December 11: Are the observation and simulation average times same? The simulated profile appears to be at night or very stable during the day, but the observed profile looks only be during the day.

Response:

We have clarified here that 'The observed BC vertical profile is at 16:20 LT and the simulated BC vertical profile is an average of 16:00 and 17:00 LT on December 11.'

12. It is necessary to discuss the reason for the difference in vertical profile between the simulation and observation on December 11th. Judging from the profile, the simulated surface air concentration is four times the observed value, but the overestimation of the simulated surface $PM_{2.5}$ concentration is not so high (Fig. 3). The simulated night T2 of the day has a significant overestimation (+6 deg C). It is 0 deg C in the simulation and -6 deg C in the observation. Is it due to overprediction of simulated clouds to prevent radiative cooling at night?

Response:

We have added the following sentences in the third paragraph of Section 3.1 to discuss the reason for the difference in BC vertical profile between observation and simulation on December 11:

'Possible reasons for model's failing to represent BC vertical profile on December 11 are as follows: (1) the model cannot capture the wind at high altitudes and does not reproduce the high-altitude BC concentrations in the surrounding areas of Beijing; (2) the model underestimates the daily maximum PBLH in Beijing which inhibits the upward transport of surface-layer BC.'

Fig. 5h shows that there was no cloud in the model at night (0:00-8:00 LT) on December 11, so the overestimation of T2 might be caused by the overestimation of surface-layer BC concentrations in the model. The surface-layer BC can absorb solar radiation during daytime and warm the air temperature at night (Ding et al., 2019).

13. Caption of Fig. 1, "The BC vertical profiles were modified for the red box which covers …" should be written in the main text.

Response:

We have added the following sentence here: 'The BC vertical profiles were only modified in the blue square shown in Fig. 1a.' in the last paragraph of Section 2.4.

14. Fig. 2, "blue and red squares" are hardly legible.

Response:

We have changed blue and red squares to black and blue squares (see below).

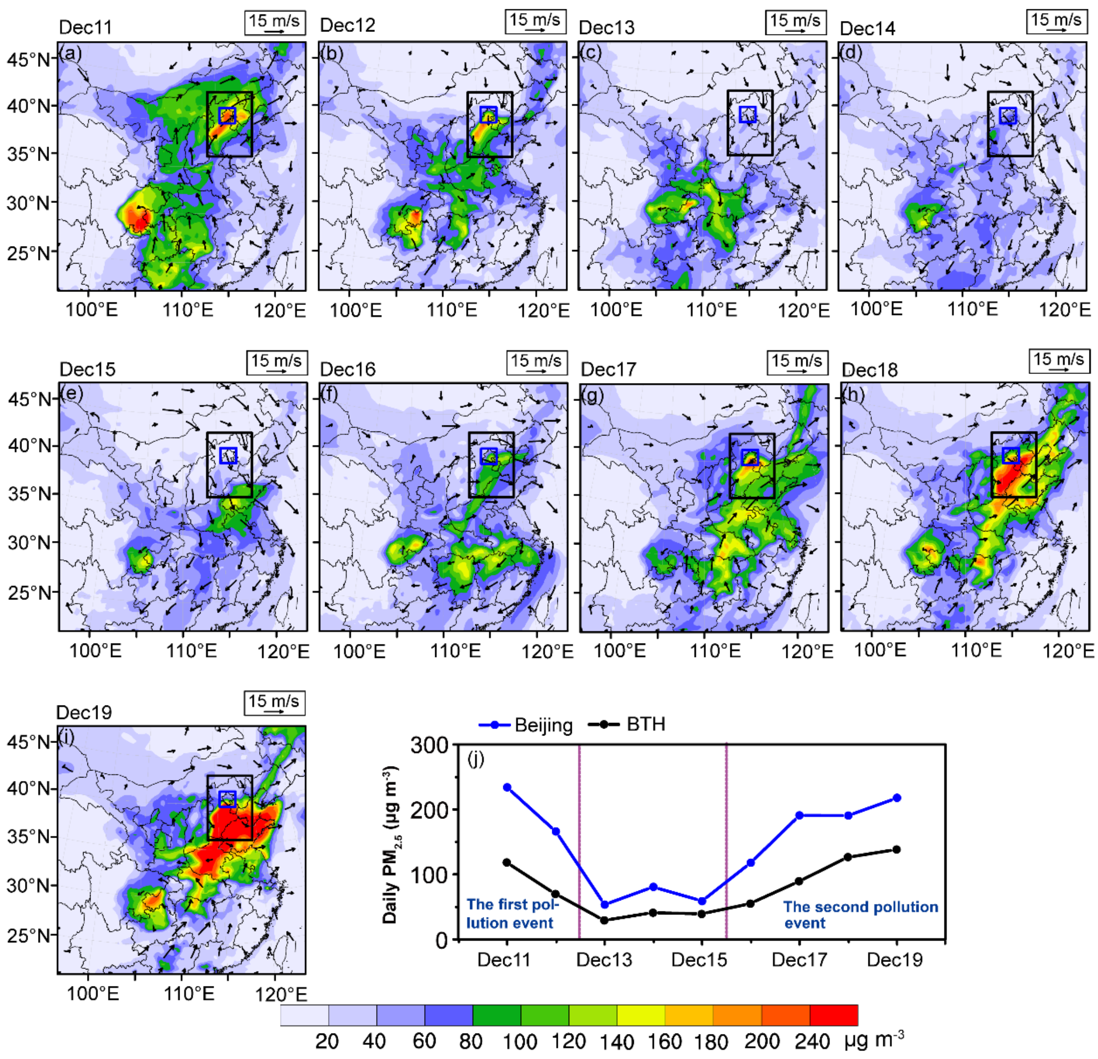

Figure 2. (a-i) Simulated spatial distributions of surface-layer $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$) and winds ($m\ s^{-1}$) at 850 hPa at 2 pm local time from 11 to 19 December 2016. Black and blue squares in each panel denote the regions of Beijing-Tianjin-Hebei and Beijing, respectively. (j) Time series of simulated mean daily $PM_{2.5}$ concentration from 11 to 19 December 2016 averaged over the Beijing (blue square) and BTH (black square).

15. Fig. 2(j), Does "Beijing" mean spatial average of the blue square region? Or one grid of the center of Beijing region? Please specify. Throughout the manuscript, it is hard to get whether the authors indicate values of only one grid point, one observation site, or those of spatial average.

Response:

We have clarified this in the figure caption (Fig. 2). 'Time series of simulated daily mean $PM_{2.5}$ concentration from 11 to 19 December 2016 averaged over Beijing (blue square) and BTH (black square) region.'.

Meanwhile, we have added the definition of Beijing domain in the manuscript when we mentioned the model results for Beijing for the first time in Section 3.1 that 'The model results for Beijing in this paper are the averages over the region of blue square shown in Fig. 1a unless stated otherwise.'.

16. Fig. 3, "Beijing". Again, Beijing point or Beijing area? Both for simulation and observation.

Response:

We have added the following sentence in the figure caption (Fig. 3): 'The observations and simulations in Beijing were averaged over 12 observational sites and corresponding grid points, respectively.'.

17. Fig. 3: Even though the model did not consider SOA, the simulated $PM_{2.5}$ was in perfect agreement with what was observed. Is the SOA negligibly small compared to the POA during the observation period, or do the OM/OC ratio(s) assumed for OC emission in the simulation well represent those of SOA and POA in the BTH region? Specify the number of OM/OC ratio(s) used in the simulation and how the author determined the value(s).

Response:

Although the model did not consider SOA, the surface-layer $PM_{2.5}$ was overestimated by the model. The model performance in simulating $PM_{2.5}$ was described in the second paragraph of Section 3.1 that 'The model can reasonably reproduce the temporal variations of $PM_{2.5}$ and the correlation coefficients between simulated and observed hourly concentrations are 0.77. For hourly $PM_{2.5}$, for example, the MBs (NMBs) are 29.1 μg m^{-3} (82.5%) on clean days and 6.3 μg m^{-3} (3.5%) during the two haze events. The possible reasons for the overall overestimation of $PM_{2.5}$ are as follows: (1) the model biases in underestimating WS10 and daytime PBLH; (2) the uncertainties in anthropogenic emission data (e.g. the overestimation in the BC emissions) (Qiu et al., 2017; Chen et al., 2021).'.

Gao et al. (2016) used the WRF-Chem model and showed that although the total $PM_{2.5}$ was overestimated by 43.3 μg m^{-3} (36.5%) during haze event, OC was underestimated by 44.5% due to the large uncertainty of OC emission inventory and missing secondary organic aerosol formation in the selected CBMZ-MOSAIC coupled mechanism. We also summarized the measured OC concentrations during haze days in Table R1. In this study, the mean simulated surface-layer OC concentrations was 33.0 μg m^{-3} in Beijing averaged over two haze events (mean $PM_{2.5}$ was 186.1 μg m^{-3}) shown in Fig. R1. Compared to measured OC during haze days from previous studies, the simulated OC concentrations were lower in our study.

Table R1. A summary of the measured OC concentration (μg m^{-3}) from previous studies.

Location	$PM_{2.5}$ (μg m^{-3})	OC (μg m^{-3})	Reference
Beijing, China	215.3-372.4	66.0-129.8	Gao et al., 2016
Beijing, China	150.0-250.0	22.5-37.5	Ji et al., 2017
Beijing, China	209.6	54.1	Qiu et al., 2017

Beijing, China	110.0	33.2	Chen et al., 2019
Shijiazhuang, China	216.1	79.2	Chen et al., 2019

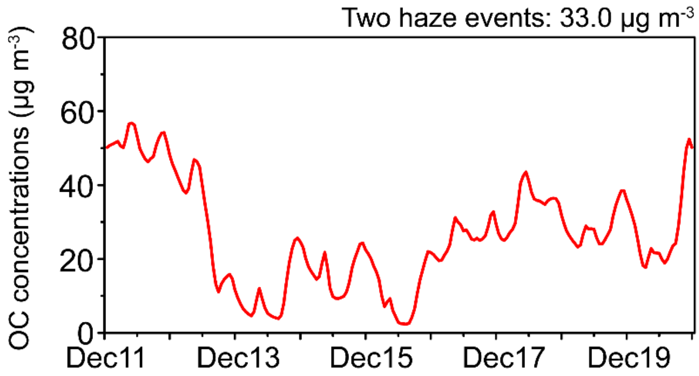

Figure R1. Time series of simulated hourly OC concentrations from 11 to 19 December 2016 averaged over Beijing. The mean value of OC averaged over the two haze events was indicated above the panel.

18. Caption of Fig. 4: What time? Both for observation and simulation.

Response:

The specific observation time was summarized in Table S1. We have also added the time of observation in Fig. 4 (on top of each panel, see below). The model results are the two-hour averages around the observation time.

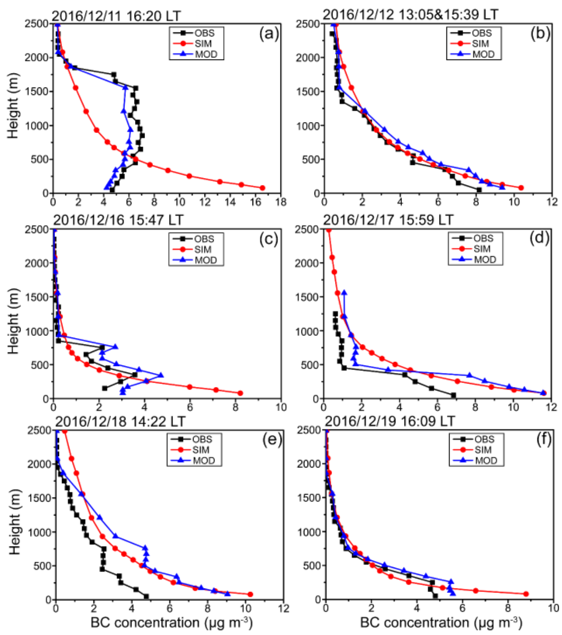

Figure 4. Observed (black line), simulated (red line) and modified (blue line) BC vertical profiles in Beijing on 11-12 and 16-19 December 2016. The time of observation is indicated on top of each panel. The model results are two-hour averages around the observation time.

19. Fig. 5: Can you compare downward solar radiation at ground surface here? It could also effectively evaluate the model performance of aerosols, and even clouds.

Response:

The comparisons between simulated and observed 3-hourly total cloud cover (%) and 3-hourly shortwave downward radiation flux (SWDOWN, W m^{-2}) in Beijing are added in Figure 5f and 5h. The corresponding statistical metrics are added in Table 3. See also our responses to your Comment #1 (Major concerns/questions).

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