Interactive comment on "Relative effects of open biomass and crop straw burning on haze formation over central and eastern China: modelling study driven by constrained emissions" by Khalid Mehmood et al.

Anonymous Referee #1

Received and published: 7 November 2019

General comments:

Open biomass burning (OBB) is one of the major air pollution sources in many regions including central and eastern China (CEC). However, it's challenging to accurately estimate its emission amounts using either bottom-up or top-down methods. The bottom-up method suffers from large uncertainties in estimation components such as surface fuel loading, fuel consumption, and emission factors, while the top-down method has difficulties in detecting small fires like open crop straw burning (OCSB) studied in this work. Here the authors used a two-way coupled chemical transport model (WRF-CMAQ) as well as ground and satellite observations to constrain a presumably biased satellite-based fire emission inventory (FINNv1.5) for the CEC region during a pollution episode in June 2014. They also evaluated regional air quality impacts of biomass burning based on the optimized OBB/OCSB emissions. The topic is within the scope of the journal, and the manuscript is well-organized and -written. However, some concerns regarding the generalization of its method and results exist. Therefore, I suggest its publication in the journal after addressing the following issues.

Response: We thank the reviewer #1 for the constructive comments and address them as below.

Specific comments:

1. The first and the biggest concern is about its scientific significance. This study mainly focused on a short time period less than a month. The major OBB/OCSB burning episode is about 10 days in EP2 from June 5 to 14, 2014. The authors spent their most efforts on scaling fire emissions in three time periods (EP1~3) and different CEC regions (Henan, Anhui, and other provinces over CEC) to reduce the normalized mean bias (NMB) as a metric of modelling performance. Since the studying time period is relatively short, this approach tends to fall into the "overfitting" problem as a common modelling error. It's questionable how robust are these scaling factors as shown in Fig. 7 as they are even distinct in different studying time periods (EP2 vs. EP1/3). It would be more interesting to extend the time scale and produce more generalizable optimization of biomass burning emissions in CEC with increased scientific merit.

Response: Yes, the scaling factors are specific to this case study. In this paper, we focus on a very representative period (i.e., EP2) when the abundant OCSB and other types of OBB outbreak concurrently, and the resultant $PM_{2.5}$ pollution spread extensively. Further, we have achieved a valuable finding that is the degree of the uncertainties associated with these scaling factors for this period. That is to say, this paper has found a very important result in this regard.

2. Regarding the research methodology, the authors essentially used a “trial and error” method to approach an optimal estimate of regional OBB emissions that led to a relatively good agreement between $PM_{2.5}$ simulations and ground observations. Though it is straightforward by tweaking adjustment coefficients of total OBB emissions, this method has several limitations such as computationally expensive and cumbersome, indistinguishable bias sources, and possible over-adjustment. Given large uncertainties in many aspects of $PM_{2.5}$ simulations, it is easy to ascribe the modelling discrepancy to wrong causes and correct the model to get a good-looking result for some wrong reason. It is suggested to do more comprehensive model evaluation in terms of aerosol speciation (to help identify OBB/non-OBB source contributions) and spatial distributions (both horizontal and vertical) before correcting OBB primary emissions.

Response: Given large uncertainties in previous OBB emissions (e.g., FINNv1.5), the “trial and error” method is relatively necessary to optimize those estimates in comparison to the immediate predictions without any constraint. Using this method, we have quantified the critical uncertainties in previous OBB emission estimates to a large extent, which is one of the most important findings in this study. To address the reviewer’s concerns about the model performance on aerosol speciaiton, we add the evaluations of the simulated $PM_{2.5}$ compositions (i.e., K^+, SO_4^{2-}, NO_3^-, OC, and NH_4^+) in the BASE and OPT cases in Sect. 3.4. The observed $PM_{2.5}$ composition was obtained from the CARE-China network (Xin et al., 2015; Liu et al., 2018), which is introduced in Sect. 2.1. Owing to their key features, including the discontiguous samples as well as the 48h temporal resolution, they are not able to support the time series analysis but can be used for the period evaluations. Besides the evaluations at the province scale, the above results at the site level are well complementary for the horizontal evaluations. In addition, considering the lack of directly vertical measurements associated with this study, we supplement the comparisons of the simulated AODs in the OPT case against the observations. This is mainly due to the AODs, as the integration of multiple column properties, generally serve as the vertical proxies to examine the model performance. Correspondingly detailed information has been given an account in the response for the specific comment (2) from Referee #2.

Added/rewritten part in Sect. 2.4: Hourly mass concentrations of surface $PM_{2.5}$ and other chemical species (i.e., CO, NO_2, SO_2, O_3, and PM_{10}) were continuously measured by the Ministry of Ecological Environment of China (http://www.cnemc.cn/, last access: 2 August 2019), including 340 monitoring sites in 65 cities during the study period over CEC. The $PM_{2.5}$ compositions were obtained from the Campaign on Atmospheric Aerosol Research network of China (CARE-China) in 2011, which was mainly supported by the Chinese Academy of Sciences. The CARE-China network, as the first comprehensive measurement platform for atmospheric aerosols across China (Xin et al., 2015; Liu et al., 2018), has embraced 40 ground sites including 20 urban sites, 12 background sites, and 8 rural/suburban sites and measured most of $PM_{2.5}$ compositions (Liu et al., 2018). Table S3 displays detailed information of 5 sites utilized in this study, which were located in Hunan, Anhui, Jiangxi, Shandong, and Jiangsu. Note that, owing to their key features, including the discontinuous samples (i.e., from June 2 to 4, from June 9 to 11, and from June 16 to 18) as well as the 48h temporal resolution, they were not able to support the time series analysis but can be used for the period evaluations.

These monitoring data were used as follows: (1) According to the evolution of surface $PM_{2.5}$ concentrations and their composition over CEC, we characterized changes in spatial and temporal patterns of regional haze induced by OBB. (2) We compared simulated chemical and meteorological fields with surface observations to evaluate model performance. (3) The $PM_{2.5}$ concentrations were used to estimate potential sources by the CWT method. (4) Daily mean values of AOD at 550 nm retrieved from satellite platform were examined during the target period to highlight significant spatial and temporal variabilities of regional haze over CEC. Here, the episode-averaged AOD products from MODIS (MOD08_D3) at 550 nm were utilized (https://giovanni.sci.gsfc.nasa.gov/giovanni/, last access: 5 August 2019).

Added/rewritten part in Sect. 3.2: Besides the analysis of $PM_{2.5}$ concentrations at the provincial levels, the major $PM_{2.5}$ compositions (i.e., K^+, SO_4^{2-}, NO_3^-, OC, and NH_4^+) at the individual sites (Table S5) were also illustrated. We found that there were steady increases in most of the chemical compositions during EP2 at the Changsha and Yucheng sites, which were located in Hunan and adjacent to Anhui, respectively. Such spatiotemporal patterns were highly correlated with those of OBB and $PM_{2.5}$ concentrations. The SO_4^{2-} and OC, as the dominating species, were project to be responsible for such increases during EP2. For example, the former rose by 13.4 μg m^{-3} (65.2 %) and 17.6 μg m^{-3} (134.5 %) in Changsha and Yucheng, respectively. More, we should pay special attention to K^+, which was usually treated as the tracer for OBB (Duan et al., 2004). As expected, with the OBB outbreaks in EP2, the K^+ concentrations dramatically increased at those two sits (25.2 % ~ 153.7 %), indicating the critical role of OBB in Hunan and Anhui. A similar phenomenon also appeared in Qianyanzhou, where the observed K^+ concentrations increased from 0.09 μg/m^3 to 0.56 μg/m^3 (534.2 %). Note that, most of other observed composition in Qianyanzhou and Wuxi remained in the relatively low range.

Added/rewritten part in Sect. 3.4: Besides, we also evaluated the simulated $PM_{2.5}$ composition (K^+, SO_4^{2-}, NO_3^-, OC, and NH_4^+) (Table S5). Compared to the BASE case, the simulated results in the OPT case at the Changsha, Hefei, and Yucheng sites were raised in various degrees and improved in general. Specifically, the NMB values of the simulated SO_4^{2-} and NH_4^+ were reduced from – 47.5 % to -14.1 % and from -30.4 % to -0.9 %, respectively, marking the reliable role of the constraining method to a large extent. The simulated OC concentrations presented similar growth trends but remained inadequate (-48.7 % ~ -6.6 %), indicating the missing mechanisms and insufficient simulations of secondary organic aerosols in the AERO6 scheme. On the contrary, the OPT case was prone to underestimate K^+ (-79.1 % ~ -20.0 %) and fail to capture the spatiotemporal changes in NO_3^- (-69.3 % ~ 391.0 %). This might be associated with the large uncertainties in the chemical speciation in the original OBB emissions. However, it should be noted that the OPT case can effectively stabilize the model performance in reproducing NO_3^- (~ 24.7 %) when their values were larger than 10 μg m^{-3}. In addition, the constrained OBB emissions also improved the model performance at the Qianyanzhou site, while the Wuxi site seemed to be irrelevant to the OBB case. Hence, the constraining method enables the model to optimize the simulated $PM_{2.5}$ composition over CEC to some extent.

Added Table S5:

Table S5. The comparisons of the simulated $PM_{2.5}$ composition (μg m^{-3}) in the BASE and OPT cases with the observations (OBS) as well as their corresponding NMB values (%).

Composition	Time periods		Changsha		Hefei		Yucheng		Qianyanzhou		Wuxi	
			Values	NMB	Values	NMB	Values	NMB	Values	NMB	Values	NMB
K+	From 10 a.m., June 2 to 10 a.m., June 4	OBS	0.59		0.92		0.90		0.09		0.62	
		BASE	0.15	-74.64	0.18	-80.53	0.35	-61.12	0.22	149.12	0.12	-80.73
		OPT	0.21	-64.50	0.28	-69.71	0.45	-50.01	0.25	183.09	0.13	-79.12
	From 10 a.m., June 9 to 10 a.m., June 11	OBS	1.50		1.16		1.49		0.56		0.56	
		BASE	0.49	-67.34	0.55	-52.49	0.78	-47.67	0.41	-26.80	0.38	-32.23
		OPT	0.79	-47.34	0.89	-23.12	0.87	-41.63	0.45	-19.65	0.34	-39.37
	From 10 a.m., June 16 to 10 a.m., June 18	OBS	0.51		0.91		0.51		0.16		0.25	
		BASE	0.34	-33.36	0.54	-40.52	0.71	39.78	0.29	77.65	0.07	-72.05
		OPT	0.38	-25.52	0.78	-14.09	0.74	45.68	0.33	102.15	0.09	-64.07
SO_4^{2-}	From 10 a.m., June 2 to 10 a.m., June 4	OBS	20.48		10.95		13.11		12.56		16.21	
		BASE	11.40	-44.34	5.90	-46.12	8.70	-33.64	1.98	-84.24	9.64	-40.54
		OPT	24.12	17.77	13.54	23.64	15.34	17.01	3.45	-72.53	9.58	-40.91
	From 10 a.m., June 9 to 10 a.m., June 11	OBS	33.83		18.68		30.74		8.97		13.23	
		BASE	20.14	-40.47	14.70	-21.31	15.98	-48.01	2.55	-71.56	8.40	-36.53
		OPT	37.07	9.57	22.30	19.38	31.99	4.07	3.54	-60.52	9.00	-31.99
	From 10 a.m., June 16 to 10 a.m., June 18	OBS	18.94		27.05		21.86		20.95		12.00	
		BASE	13.75	-27.39	16.70	-38.26	8.71	-60.15	4.78	-77.19	6.85	-42.90
		OPT	23.64	24.83	28.90	6.85	18.05	-17.41	5.61	-73.23	7.40	-38.31
NO_3^-	From 10 a.m., June 2 to 10 a.m., June 4	OBS	9.34		8.48		7.49		1.93		6.35	
		BASE	10.97	17.49	5.39	-36.44	3.61	-51.81	1.03	-46.56	1.78	-71.96
		OPT	14.98	60.44	9.81	15.67	6.45	-13.90	2.62	35.94	1.95	-69.28
	From 10 a.m., June 9 to 10 a.m., June 11	OBS	11.57		3.20		12.72		0.60		1.82	
		BASE	9.70	-16.18	7.90	146.66	10.80	-15.13	1.85	207.90	0.98	-46.05
		OPT	12.45	7.58	8.50	165.40	14.45	13.56	2.95	390.98	0.97	-46.60
	From 10 a.m., June 16 to 10 a.m., June 18	OBS	4.85		28.33		15.89		2.22		12.27	
		BASE	7.40	52.45	20.00	-29.39	20.38	28.23	1.88	-15.36	14.95	21.82
		OPT	8.78	80.88	29.80	5.21	21.09	32.69	2.23	0.40	15.78	28.59
NH_4^+	From 10 a.m., June 2 to 10 a.m., June 4	OBS	10.26		9.03		8.45		5.46		9.17	
		BASE	8.45	-17.63	7.54	-16.53	3.78	-55.27	1.03	-81.14	13.50	47.27
		OPT	11.54	12.50	10.92	20.89	7.30	-13.61	1.75	-67.96	13.90	51.64
	From 10 a.m., June 9 to 10 a.m., June 11	OBS	17.68		8.54		15.26		3.71		4.76	
		BASE	11.06	-37.45	4.98	-41.72	13.45	-11.88	1.35	-63.59	2.01	-57.77
		OPT	17.37	-1.76	10.39	21.60	18.74	22.78	1.75	-52.80	3.32	-30.25
	From 10 a.m., June 16 to 10 a.m., June 18	OBS	8.62		21.87		14.95		10.14		9.74	
		BASE	3.84	-55.47	20.06	-8.29	14.20	-4.99	5.97	-41.11	8.70	-10.65
		OPT	10.43	20.94	28.78	31.57	17.64	18.03	6.78	-33.12	8.40	-13.73
OC	From 10 a.m., June 9 to 10 a.m., June 11	OBS	19.80		19.54		18.95		15.70		29.15	
		BASE	10.15	-48.74	10.30	-47.30	10.65	-43.81	6.12	-61.02	15.07	-48.30
		OPT	18.50	-6.57	15.50	-20.70	13.50	-28.77	8.12	-48.28	15.95	-45.28

3. A two-way coupled WRF-CMAQ model was used in this study, which considered aerosol-radiation interactions inherently. However, there is no discussion about this at all in the results and discussion section. The authors list the advantages of this fully coupled model by referring to a series of previous studies in the method section, then this key feature seems to be forgotten in the following sections. Readers would be interested in many questions related with aerosol feedbacks, such as how important those aerosol-radiation interactions are in the regional pollution episode, can they affect local weather systems and pollution severity significantly, etc.

Response: We supplement the additional discussions in Sect. 4 to explain why we have not explored the effects of OBB-induced aerosol-radiation interactions during EP2. This is mainly because that the effects of the aerosol-radiation interactions are projected to be particularly small compared to the large uncertainties in the OBB emissions.

Specifically, the two-way coupled WRF-CMAQ utilized in this study is a standard approach. A lot of previous studies have applied the same or similar model to explore aerosol-radiation interactions. Generally, this process could enhance surface $PM_{2.5}$ concentrations by 8 ~ 12 % over CEC (Wang et al., 2014, 2015; Zhang et al., 2015, 2018; Jung et al., 2019). Compared to the significant underestimations of the OBB emissions (5 ~ 7 times) and the resultant $PM_{2.5}$ pollution (> 50 %) during EP2, the impacts of aerosol-radiation interactions become irrelevant here. More importantly, the associated process analysis has no benefit for addressing the main task of this study.

Added/rewritten part in Sect. 4: To optimize the OBB emissions, previous attempts always focused on the primary emissions but neglected aerosol-radiation interactions (Uranishi et al., 2019; Yang and Zhao, 2019). Theoretically, with the aid of the systematic considerations of aerosol-radiation interactions in the two-way coupled WRF-CMAQ model, this study was projected to achieve more reliable results. A lot of previous studies have applied the same or similar model to explore the impacts of aerosol-radiation interactions. However, it has been found that this process generally enhanced surface $PM_{2.5}$ concentrations by 8 ~ 12 % over CEC (Wang et al., 2014, 2015; Zhang et al., 2015, 2018; Jung et al., 2019). Compared to the significant underestimations of the OBB emissions (5 ~ 7 times) and the resultant $PM_{2.5}$ pollution (> 50 %) during EP2, the impacts of aerosol-radiation interactions become irrelevant here. More importantly, the associated process analysis has no benefit for addressing the main task of this study. Looking forward, it is necessary to advance the scientific understandings of the role of the constrained OBB-induced aerosol-radiation interactions in disturbing the local weather systems as well as surface $PM_{2.5}$ pollution. Considering the distinct topic, we should combine sufficiently specific observations to give impetus to comprehensive process analysis, which will be the topic of a next separate study.

Technical corrections:

1. Please add specific values for these parameters in Eq. (1) and Eq. (2).

Response: We supplement specific values (i.e., Table S1 and Table S2) for B_{size}, B_{hour}, P_{topmax}, and $P_{bottommax}$ based on the look-up tables.

Added/rewritten part in Sect. 2.2: In this study, we determined the heights of the hourly top (P_{top}) and bottom (P_{bottom}) of the OBB plume using a quick plume rise model (Eq. (4) and Eq. (5)). This process was calculated based on the buoyant efficiency (B) available from the corresponding hourly and size class tables (Tables S1 and S2) (Tai et al., 2008; Fu et al., 2012a).

Added Table S1:

Table S1. The fire-related parameters (i.e., B_{size}, P_{topmax}, and $P_{bottommax}$) as a function of the fire size classes.

Fire Class	1	2	3	4	5
Size	0~10	10~100	100~1000	1000~5000	>5000
B_{size}	0.4	0.6	0.75	0.85	0.9
P_{topmax}	160	2400	6400	7200	8000

$P_{bottommax}$	0	900	2200	3000	300

Added Table S2:

Table S2. The activity related parameter (i.e., B_{hour}) as a function of hour of day.

Hour	B_{hour}	Hour	B_{hour}
0	0.03	12	0.7
1	0.03	13	0.8
2	0.03	14	0.9
3	0.03	15	0.95
4	0.03	16	0.99
5	0.03	17	0.8
6	0.03	18	0.7
7	0.03	19	0.4
8	0.06	20	0.06
9	0.1	21	0.03
10	0.2	22	0.03
11	0.4	23	0.03

2. In Fig. 4, please indicate which modelling experiment is the $PM_{2.5}$ simulation based on.

Response: The simulated $PM_{2.5}$ concentrations in Fig. 4 were extracted from the OPT case. We add the corresponding statement in Fig. 4.

The rewritten caption in Fig. 4: Spatial distributions of observed and simulated episode-averaged $PM_{2.5}$ concentrations from the OPT case over CEC during (a) EP1, (b) EP2, and (c) EP3. Colored circles denotes locations of ground measurement sites and corresponding values.

3. In Fig. 12, please add unit in the label bar.

Response: We add the units (μg m^{-3}) in the label bar of Fig. 12**.**

The updated Fig. 12:

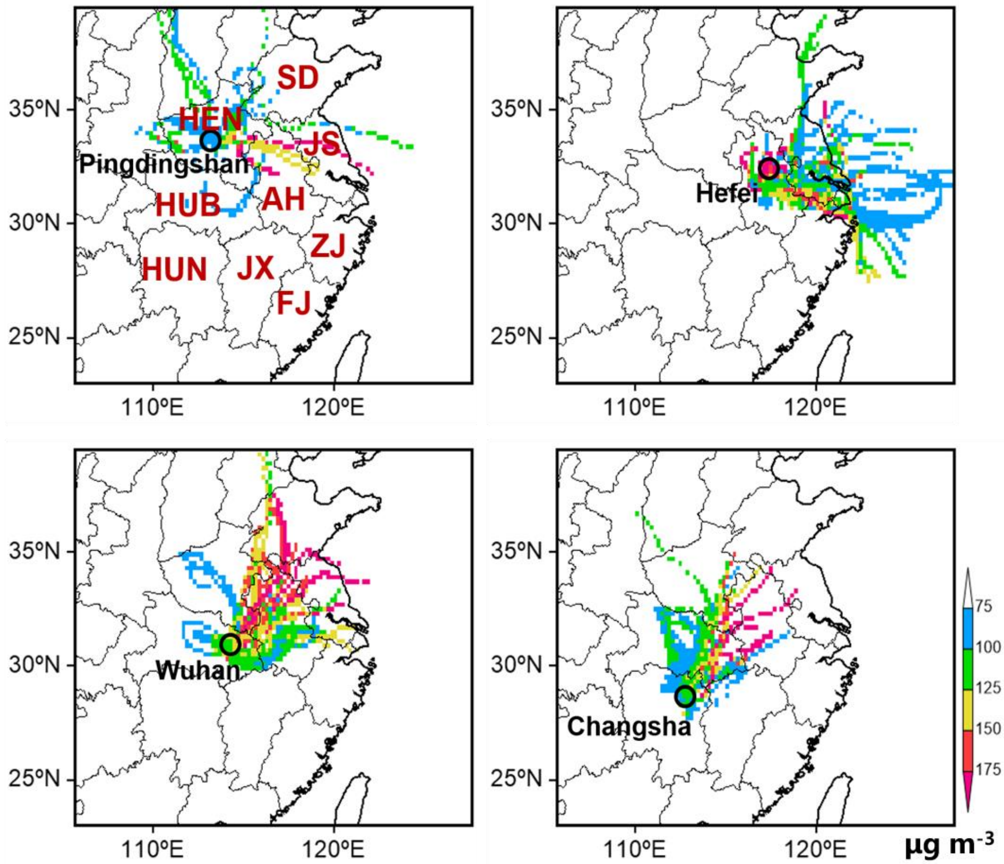

Figure 12. CWT maps during EP2 at four representative cities (Pingdingshan, Hefei, Wuhan and Changsha) that represent Henan, Anhui, Hubei and Hunan, respectively.

4. Table 1 looks fuzzy due to the low resolution. Please improve the presentation quality.

Response: We improve the presentation quality of Table 1.

References:

Duan, F., Liu, X., Yu, T. and Cachier, H.: Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing, Atmos. Environ., 38(9), 1275–1282, 2004.

Fu, J. S., Hsu, N. C., Gao, Y., Huang, K., Li, C., Lin, N.-H. and Tsay, S.-C.: Evaluating the influences of biomass burning during 2006 BASE-ASIA: a regional chemical transport modeling, Atmos. Chem. Phys., 12(9), 3837–3855, 2012.

Jung, J., Souri, A. H., Wong, D. C., Lee, S., Jeon, W., Kim, J. and Choi, Y.: The Impact of the Direct Effect of Aerosols on Meteorology and Air Quality Using Aerosol Optical Depth Assimilation During the KORUS-AQ Campaign, J. Geophys. Res. Atmos., 124(14), 8303–8319, 2019.

Liu, Z., Gao, W., Yu, Y., Hu, B., Xin, J., Sun, Y., Wang, L., Wang, G., Bi, X., Zhang, G. and others: Characteristics of PM 2.5 mass concentrations and chemical species in urban and background areas of China: emerging results from the CARE-China network, Atmos. Chem. Phys., 18(12), 8849–8871, 2018.

Tai, E., Jimenez, M., Nopmongcol, O., Wilson, G., Mansell, G., Koo, B. and Yarwood, G.: Boundary conditions and fire emissions modeling, Prep. Texas Comm. Environ. Qual. Sept., 2008.

Uranishi, K., Ikemori, F., Shimadera, H., Kondo, A. and Sugata, S.: Impact of field biomass burning on local pollution and long-range transport of PM2. 5 in Northeast Asia, Environ. Pollut., 244, 414–422, 2019.

Wang, H., Shi, G. Y., Zhang, X. Y., Gong, S. L., Tan, S. C., Chen, B., Che, H. Z. and Li, T.: Mesoscale modelling study of the interactions between aerosols and PBL meteorology during a haze episode in China Jing--Jin--Ji and its near surrounding region--Part 2: Aerosols' radiative feedback effects, Atmos. Chem. Phys., 15(6), 3277–3287, 2015.

Wang, J., Wang, S., Jiang, J., Ding, A., Zheng, M., Zhao, B., Wong, D. C., Zhou, W., Zheng, G., Wang, L. and others: Impact of aerosol--meteorology interactions on fine particle pollution during China's severe haze episode in January 2013, Environ. Res. Lett., 9(9), 94002, 2014.

Xin, J., Wang, Y., Pan, Y., Ji, D., Liu, Z., Wen, T., Wang, Y., Li, X., Sun, Y., Sun, J. and others: The campaign on atmospheric aerosol research network of China: CARE-China, Bull. Am. Meteorol. Soc., 96(7), 1137–1155, 2015.

Yang, Y. and Zhao, Y.: Quantification and evaluation of atmospheric pollutant emissions from open biomass burning with multiple methods: a case study for the Yangtze River Delta region, China, Atmos. Chem. Phys., 19(1), 327–348, 2019.

Zhang, B., Wang, Y. and Hao, J.: Simulating aerosol--radiation--cloud feedbacks on meteorology and air quality over eastern China under severe haze conditionsin winter, Atmos. Chem. Phys., 15(5), 2387–2404, 2015.

Zhang, X., Zhang, Q., Hong, C., Zheng, Y., Geng, G., Tong, D., Zhang, Y. and Zhang, X.: Enhancement of PM2. 5 Concentrations by Aerosol-Meteorology Interactions Over China, J. Geophys. Res. Atmos., 123(2), 1179–1194, 2018.