Response to Comments of Reviewer #2

(comments in *italics*)

Manuscript number: acp-2019-245

Title: Assessing the formation and evolution mechanisms of severe haze pollution in Beijing-Tianjin-Hebei region by using process analysis

Review of: "Assessing the formation and evolution mechanisms of severe haze pollution in Beijing–Tianjin–Hebei region by using process analysis" by Chen et al.

The authors quantitatively examined the cause of a severe haze event over Beijing–Tianjin–Hebei (BTH) through decomposing contributions from emissions, physical and chemical processes, using the WRF–Chem model equipped with an improved integrated process rate (IPR) analysis scheme. The IPR scheme also reveals the dominant role of aerosol radiative effects in haze formation is physical rather than chemical. Such an IPR scheme merits application in future relevant studies. Overall, the manuscript is well structured and should be of great interest to ACP readers. I recommend it is publishable after my minor comments/suggestions are addressed.

Response:

Thanks to the reviewer for the valuable comments and suggestions which are very helpful for us to improve our manuscript. We have revised the manuscript carefully, as described in our point-to-point responses to the comments.

General Comments:

1. ***Local vs. regional contributions:*** *From the sensitivity simulations, local emissions and regional transport account for 80% of total $PM_{2.5}$. What does the contribution from "others" (Fig.7) mean? Is it because the non-linear chemical formation for secondary aerosol or contribution from aerosol precursors outside of BTH?*

Response:

According to the experiments listed in Table 2, the contributions of local anthropogenic emission and regional transport to the $PM_{2.5}$ concentrations in BTH (Beijing-Tianjin-Hebei) can be identified by comparing the simulation results between OnlyBTH_Anth and NoAnth (i.e., OnlyBTH_Anth minus NoAnth), and between NoBTH_Anth and NoAnth (i.e., NoBTH_Anth minus NoAnth), respectively.

In addition to the primary source emission (e.g., anthropogenic emission) and regional transport, secondary aerosol formation and their hygroscopic growth are also considered to be a large contributor to severe haze episodes (Huang et al., 2014b; Han et al., 2015; Chen et al., 2019).

As Li et al., (2018) pointed out that a "brute-force" method (e.g., sensitivity analysis used to measure the model outputs response to emission change) is a traditional way to identify source contributions from non-reactive species in a linear process, but it cannot straightforwardly apply to secondary species due to the non-linearity in responses. All these indicate that the actual impact of one factor in a nonlinear process in the presence of others can be separated into (1) pure impact

from the factor, and (2) interactional impacts from other factors.

In this manuscript, we divide the simulated $PM_{2.5}$ concentration into three parts: contributions from local anthropogenic emission, regional transport and others. Besides the impacts of local anthropogenic emissions and regional transport, the rest of the contributions to the $PM_{2.5}$ concentration in BTH may include the impacts from biogenic emission, biomass-burning emission, dust emission, sea salts, aerosols outside the simulation domain, and the non-linear chemical formation for secondary aerosol.

2. ***Advection contribution:*** *A negative value for advection is diagnosed by the IPR scheme. From my understanding, the advection means horizontal transport, which should not be always a negative contribution, and instead it may contribute a lot to $PM_{2.5}$ if taking the value of regional contribution as an equivalent.*

Response:

We totally agree with the reviewer's opinion. In this manuscript, TRAN includes horizontal and vertical advection, which is highly related to wind and aerosol concentration gradients from upwind regions to downwind areas (Gao et al., 2018). When the calculated TRAN in a model grid during a simulation output interval is negative, indicating the process of advection will decrease the aerosol concentrations, and vice versa.

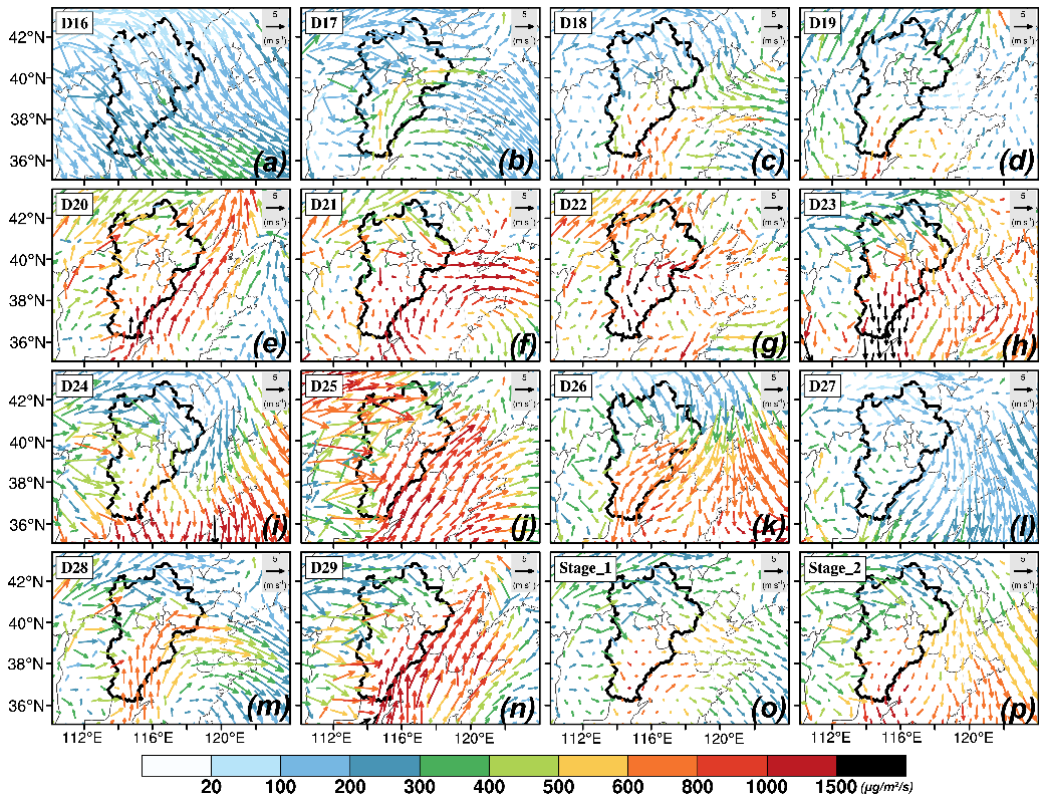

Figure R1. (a-n) Spatial distribution of simulated daily transport fluxes of $PM_{2.5}$ during 16-29 December 2015. The average $PM_{2.5}$ flux in Stage_1 (December 16-22) and Stage_2 (December 23-27) are also shown in (o) and (p).

Figure R1 shows the spatial distributions of simulated transport fluxes of $PM_{2.5}$ during 16-29

December 2015. The average flux in Stage_1 (December 16-22) and Stage_2 (December 23-27) are also shown. During Stage_1 (Fig. R1(o)), the southerly wind over the southern parts of BTH is low, resulting in weak $PM_{2.5}$ fluxes from polluted upstream regions (e.g., Henan and Shandong) to downstream regions (e.g., BTH). However, pollutants in the northern parts of BTH are transported eastwardly to the regions of Huanghai and Bohai Sea. Generally, a negative value of TRAN is diagnosed by the integrated process rate (IPR) scheme in Stage_1. Similar influence of TRAN on $PM_{2.5}$ concentrations in BTH can also be found in Stage_2. This is because prevailing northerly winds in BTH bring aerosols to downstream regions (e.g., Henan and Shandong).

3. ***Aerosol radiative effects:*** *It is considerate to include the aerosol indirect effect, though this process contributes marginally in less-cloud wintertime. But the authors failed to show/discuss how aerosol indirect effect is expressed from IPR result. For example, in Fig. 11 the CLDC (0.5 ug m^{-3}) and WETP (0.2 ug m^{-3}) can be taken as the result of aerosol indirect effect. It needs to clarify.*

Response:

Following the comments from Reviewer#3, simulation results from 16 to 18 December 2015 are also considered in this manuscript to better analyze the formation mechanism of the haze event. According to the time series of simulated daily $PM_{2.5}$ concentrations in BTH (Fig. 6(l)), aerosol accumulation stage (Stage_1) is now considered during December 16-22. All the values in Stage_1 have been re-calculated in the revised manuscript, and now, the CLDC and WETP in Fig. 11 are 0.5 ug m^{-3} and 0.2 ug m^{-3}, respectively.

According to the reviewer's comments, another sensitivity experiment (referred to as NoAIE case) is designed. Same as CTL, but a prescribed vertically uniform cloud droplet number (0.93×10^8 particles per kg of air) is used in NoAIE, which is calculated from the CTL case during the whole simulation period, following Gao et al., (2015) and Zheng et al., (2015). Comparing the simulation results from CTL and NoAIE, the impacts of aerosol indirect effects (AIEs) on haze episode can be analyzed and quantified.

Table R1. Contributions of each process to the change in 24-h $PM_{2.5}$ increase caused by aerosol indirect effects during Stage_1. The 24-h increase in $PM_{2.5}$ concentration (23:00LST minus 00:00LST) in CTL and NoAIE are 43.9 ug m^{-3} and 44.2 ug m^{-3}, respectively.

Process	TRAN	EMIS	VMIX	SGCV	GASC	CLDC	AERC	WETP	$PM_{2.5}$
CTL-NoAIE (ug m^{-3})	0.3	0.0	1.2	0.0	0.0	0.4	-2.3	0.1	-0.3

From Table R1, we can find that when AIE is considered, the 24-h increase of $PM_{2.5}$ concentration during Stage_1 is decreased by 0.3 ug m^{-3}, from 44.2 ug m^{-3} in NoAIE to 43.9 ug m^{-3} in CTL. The reduction induced by AIE can be mainly attributed to the contribution of aerosol chemistry (AERC, -2.3 ug m^{-3}). AERC refers to microphysical nucleation, condensation, and coagulation, as well as the mass transfer between the gas phase and condensed phase. As Zheng et al., (2015) pointed out that when AIE is included, the predicted cloud droplet number is based on the simulated aerosol number different than what is prescribed in the model default setting. This coding strategy allows interstitial air-borne aerosols to become cloud-borne aerosols after activation.

Therefore, the simulated $PM_{2.5}$ concentrations are decreased.

The change induced by AIE in contributions of vertical mixing (VMIX) process during Stage_1 is +1.2 ug m^{-3}, this is because PBLH (planetary boundary layer height) is reduced, and the decreased PBLH can result in the accumulation of aerosol particles in the near-surface layer.

Due to low cloud cover (or small cloud water content) and little precipitation in northern China in winter as shown in Zhao et al. (2015) and Zheng et al., (2015), the contributions of CLDC (cloud chemistry, +0.4 ug m^{-3}) and WETP (wet scavenging, +0.1 ug m^{-3}) induced by AIE to the 24-h $PM_{2.5}$ change are relative small.

Specific comments:

The presentation of this work would be greater if some editorial aspects are improved.

1. *Use of %: P1L27. Here and elsewhere in the text, please round off 250% instead of 250.0%. The decimal doesn't make sense in terms of model bias.*

Response:

According to the reviewer's suggestion, the decimal fraction has been rounded off in the whole revised manuscript.

2. *Please check the appropriate usage of hyphen (-) (e.g., near-surface) and en-dash (–) (e.g., December 20–22).*

Response:

Thanks for your suggestion. The hyphen has been used in the whole revised manuscript.

3. *Fig.1: The purple dot (?) for PBLH is hardly to see. Reduce the size of green triangle or increase its transparency.*

Response:

According to the reviewer's suggestion, Figure 1 has been re-plotted in the revised manuscript. **(Page 31)**

4. *Fig.4: I suggest the use of NMB and correlation coefficient are good enough for model evaluation. Reader gets lost in so many numbers.*

Response:

According to the reviewer's suggestion, several statistics (e.g., mean bias (MB), gross error (GE), and root mean square error (RMSE)) have been removed in the revised manuscript.

5. *Fig.6: I can't see any different for (k) and (i), and there is also no discussion in the text. Remove one of them.*

Response:

Thanks for your suggestion. Only the time series of simulated daily $PM_{2.5}$ concentrations averaged over the BTH region are shown in Fig. 6(l) in the revised manuscript. **(Page 36)**

6. *Fig.8: Move the middle panel (b1 and b2) towards right. Is there any difference in Y-axis of (b1) and (b2). I suppose they are the same.*

Response:

Thanks for your suggestion. The Y coordinates in Figs. 8(b1) and (b2) are the same, and we have re-plotted the figure in the revised manuscript. **(Page 38)**

7. *P1L24: There is any special meaning for "absolute" PM2.5. If not, please "absolute" when it is unnecessary.*

Response:

According to the reviewer's suggestion, the expressions of "absolute $PM_{2.5}$ concentrations" have been changed to "$PM_{2.5}$ concentrations" in the revised manuscript.

8. *P2L6 & P5L8: Remove "and so on".*

Response:

According to the reviewer's suggestion, we have deleted it in the revised manuscript.

9. *P3L7: I don't think severe haze frequently occurs in wintertime over PRD region, and neither of your two references support this.*

Response:

Thanks for the reviewer's suggestion. We have deleted it in the revised manuscript.

10. *P3L10-12: Health threatened by PM2.5 is the most important thing people care about.*

Response:

We totally agree with the reviewer's opinion. Observations show that annual $PM_{2.5}$ concentrations in China are more than 5 times higher than the World Health Organization (WHO) guideline value in some metropolitans (Wang et al., 2014). Sustained exposure to high $PM_{2.5}$ concentrations greatly threatens public health (Hu et al., 2014; Wang et al., 2015; Burnett et al., 2018), including lung cancer (Dockery et al., 1993), cardiopulmonary disease (Pope and Dockery, 2006), bronchitis (Gao et al., 2015) and so on. Source sector contributions of anthropogenic emissions to complex air pollution and their health impacts will be discussed in our upcoming study.

11. *P5L1: Remove "(SPM)". You don't use it in the following text.*

Response:

According to the reviewer's suggestion, we have deleted it in the revised manuscript.

12. *P5L3: Haze is not actually caused by "the interactions between …". It's a synergy effect by these factors.*

Response:

According to the reviewer's suggestion, we have revised the sentence as follows "All these studies discussed above revealed that the formation of haze episode was caused by the synergy impacts of local emissions, regional transport, meteorological conditions, and chemical production". **(Page 5, Line 3-4)**

13. *P5L12-13: Is there any reference saying "substantial efforts since 2009"? Zheng et al. (2018, ACP, Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions) shows emissions dropped substantially only after 2013.*

Response:

Thanks for the reviewer's comments. Since the 9th FYP (Five-Year Plan, 1996-2000), the policies about the emission controls on gases (e.g., CO_2 and SO_2) and energy saving were initiated (Cao et al., 2009). In the 11th FYP (2006-2010), an obligatory target about the emission reductions for each local government was further outlined (Anger et al., 2016), and the policies were maintained and extended in the 12th FYP (Jin et al., 2016). Although the implementations of total control policies were successful and the targets were achieved, the air quality improvement was insignificant (Schreifelds et al., 2012).

A severe haze event happened in January 2013 over many provinces in China. This haze with its unprecedentedly high index of $PM_{2.5}$ concentrations and extremely low visibility was of worldwide concern. Quickly responding to the $PM_{2.5}$ crisis, the Chinese government issued the well-known action plan "China National Action Plan on Air Pollution Prevention and Control" in September 2013, which means the "war" against air pollution was declared. Since then, emissions of multi-pollutants are reduced (Zheng et al., 2018) and industrial structures are optimized (Bao and Yao, 2016).

According to the reviewer's suggestion, we have revised the sentence as follows "Since 2013, substantial efforts have been taken to improve air quality in China, including emission reduction and energy transition". **(Page 5, Line 12-13)**

14. *P7L2: Which year of anthropogenic emission from MIX?*

Response:

The anthropogenic emissions in China are taken from the monthly 2010 Multi-resolution Emission Inventory (MEIC, http://www.meicmodel.org/). According to Chen et al. (2019), the diurnal and weekly variations of anthropogenic emission factors for each sector (power, industry, residential and transportation) are also adopted in this manuscript (Fig. R2).

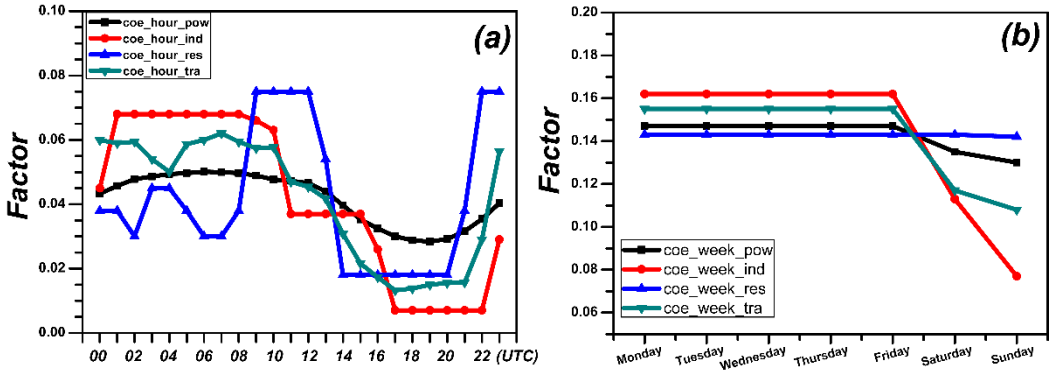

Figure R2. (a) Diurnal and (b) weekly variations of anthropogenic emission factors for power, industry, residential and transportation sectors.

15. *P8L20: Sub-grid convection (SGCV) is found to be zero in the simulation. It can be possibly due to no precipitation in this case. But why gas-phase chemistry is also zero, or what it specifically means? Nitrate formation is dominantly through gas-phase oxidation of NO2+OH.*

Response:

Thanks for the reviewer's suggestion. SGCV (sub-grid convection) refers to the scavenging within the sub-grid wet convective updrafts. The simulated SGCV in the CTL case is zero. This is because the daily accumulated cumulus precipitation averaged in BTH during 16-29 December 2015 is zero.

GASC (gas-phase chemistry) is simulated by CBMZ (Carbon-Bond Mechanism version Z), and this process mainly focuses on the reactions between gases, including the gas phase photooxidation reactions. Detailed parameterizations can be found in Zaveri and Peters (1999). Take gas phase HNO_3 as an example, the principle formation path for HNO_3 (g) in the daytime is $NO_2 + OH \rightarrow HNO_3$. At nighttime, N_2O_5 gas can react with vapor phase of H_2O (gas-phase reaction, the heterogeneous reaction of N_2O_5 ($N_2O_5 + H_2O$ (l) $\rightarrow 2HNO_3$(l)) is treated by the aerosol scheme) to form HNO_3 (g).

In this manuscript, the IPR analysis is applied to diagnose the contributions of physical/chemical processes to the variations in aerosol concentrations (e.g., SO_4^{2-}, NO_3^-, NH_4^+ and $PM_{2.5}$). This is why the calculated contributions of GASC to $PM_{2.5}$ variations in Fig. 8 are zero.

16. *P9L14 & L16: You haven't defined what "two stages" are. Maybe don't need to mention this here.*

Response:

Thanks for your suggestion. We have deleted it, and the section of numerical experiments (Section 2.3) has be revised as follows:

"Table 2 summarizes the experimental designs. To investigate the contributions of regional transport and local emission to the $PM_{2.5}$ concentrations in BTH, four simulations with different anthropogenic emission categories are conducted: (1) CTL: The control simulation with all anthropogenic emissions considered; (2) NoAnth: No anthropogenic emission is considered in the whole domain; (3) NoBTH_Anth: Same as CTL, but anthropogenic emissions in BTH are excluded; (4) OnlyBTH_Anth: Contrary to the NoBTH_Anth case, anthropogenic emissions are only

considered in BTH. All the physical and chemical schemes used in these cases are identical. The contributions of regional transport and local emission to the $PM_{2.5}$ concentration in BTH can be identified by comparing the simulation results of NoBTH_Anth and NoAnth (i.e., NoBTH_Anth minus NoAnth) and OnlyBTH_Anth and NoAnth (i.e., OnlyBTH_Anth minus NoAnth), respectively". **(Page 9, Line 9-16)**

"To quantify the aerosol radiative effects (ARE) on haze pollution, another sensitivity experiment (referred to as NoARE case) is designed by turning off the feedbacks between aerosols and meteorological variables, including eliminating the aerosol direct effect (ADE) and aerosol indirect effect (AIE) in the model. The ADE is turned off by removing the mass of aerosol species from the calculation of aerosol optical properties as did in Qiu et al. (2017). The AIE is turned off by using a prescribed vertically uniform cloud droplet number, which is calculated from the CTL case during the whole simulation period, following Gao et al. (2015) and Zhang et al., (2015a). The differences between CTL and NoARE (i.e., CTL minus NoARE) represent the impacts of aerosol radiative forcing". **(Page 9, Line 17-23)**

"The IPR analysis method is applied to all the designed experiments. Comparing the contributions of each detailed process between pollution accumulation stage and dissipation stage in CTL can quantitatively explain the reason for the variation of the $PM_{2.5}$ concentrations in BTH. Meanwhile, the prominent physical or chemical process responsible for the aerosol radiative impacts on the haze episode can also be investigated by analyzing the IPR analysis method used in CTL and NoARE cases". **(Page 9-10, Line 24-28)**

"All the five simulations are conducted for the period from 13 to 29 December 2015, and the initial three days are discarded as the model spin-up to minimize the impacts of initial conditions. Simulation results from the CTL case during 16 to 29 December 2015 are used to evaluate the model performance". **(Page 10, Line 5-7)**

17. P10L12-19: Please delete unnecessary "marked in" in this paragraph.

Response:

We have revised the paragraph (Section 2.4) according to the reviewer's suggestion.

"Simulated meteorological parameters in CTL case, including 2 m temperature (T_2), 2 m relative humidity (RH_2), 10 m wind speed (WS_{10}) and 10 m wind direction (WD_{10}), are compared with hourly observations at twelve stations, which are collected from NOAA's National Climatic Data Center (https://gis.ncdc.noaa.gov/maps/ncei/cdo/hourly). Due to limited observations of PBL height in BTH, the retrieved PBLH in 3-hour intervals obtained from the GDAS (Global Data Assimilation System) (https://ready.arl.noaa.gov/READYamet.php) in Beijing (39.93 °N, 116.28 °E) is also used to evaluate the model performance. More detailed information about the GDAS meteorological dataset (1 °×1 °) can be found in Rolph et al. (2013), Kong et al. (2015) and https://www.ready.noaa.gov/gdas1.php. Hourly shortwave downward radiation flux (SWDOWN) at the Xianghe station (39.75 °N, 116.96 °E) is taken from WRMC-BSRN (World Radiation Monitoring Center-Baseline Surface Radiation Network, http://bsrn.awi.de) for the energy budget evaluation. The hourly observed surface-layer $PM_{2.5}$ concentrations at the 59 stations are obtained from the CNEMC (China National Environmental Monitoring Center, http://www.cnemc.cn/). The daily measurements of mass concentrations of SO_4^{2-}, NO_3^-, NH_4^+, BC and OC are collected at the sites of (39.97 °N, 116.37 °E) in Beijing and (38.03 °N, 114.53 °E) in Shijiazhuang (Huang et al., 2017;

Liu et al., 2018). Detailed locations of these observations are shown in Fig. 1(b)". **(Page 10, Line 9-21)**

18. *P11L4-6 & P12L10-11: The NMB (IOA) and R are the only metrics you used and they are informative enough. I suggest remove other metrics if possible. Or define them in Table 3.*

Response:

According to the reviewer's suggestion, several statistics (e.g., mean bias (MB), gross error (GE), and root mean square error (RMSE)) have been removed in the revised manuscript. Meanwhile, the detailed calculation methods about the statistics are described in Table 3 in the revised manuscript.

19. *P11L10: Change "options" to "parameterizations"*

Response:

According to the reviewer's suggestion, we have revised it in the manuscript. **(Page 11, Line 12)**

20. *P13L24: Add "air quality" before "threshold value"*

Response:

According to the reviewer's suggestion, the sentence has been revised in the manuscript. **(Page 14, Line 3)**

21. *P14L8: I suggest change "internal" to "dominant" or "leading"*

Response:

Thanks for your comments, and we have changed it in the revised manuscript.

"Previous studies have reported that anthropogenic emission was the dominant cause of haze events in China (Jiang et al., 2013; Sun et al., 2014; Gu and Liao, 2016; Yang et al., 2016b)". **(Page 14, Line 12-13)**

22. *P16L2-4: Please delete "Suspended … during winter haze periods". You have done this in the Introduction section.*

Response:

According to the reviewer's suggestion, we have revised the paragraph as follows "Previous studies have demonstrated that the aerosol radiative forcing could increase the near-surface $PM_{2.5}$ concentrations by about 12%-29% (Gao et al., 2015; Gao et al., 2016; Qiu et al., 2017; Zhou et al., 2018). However, the detailed influence mechanisms (i.e., the prominent physical or chemical process responsible for the aerosol radiative impacts on $PM_{2.5}$ concentrations) are still unclear. In this section, we examine the effects of aerosol radiative forcing on meteorological parameters and $PM_{2.5}$ levels during the haze episode, with a special focus on the detailed influence mechanism by using the IPR analysis". **(Page 16, Line 5-10)**

23. *P16L15-16: Is this process important in your case? If not, saying this here reads misleading.*

Response:

Thanks for your suggestion. The sentence has been deleted in the revised manuscript.

24. *P16L10: I suggest to move Fig. S4 in the main text. It is interesting to show the important role of absorbing aerosol on regional circulation change. This result is consistent with the simulation by Qiu et al. (2017).*

Response:

Thanks for your suggestion. The main purpose of this manuscript is to investigate the formation and evolution mechanisms of a haze event in BTH during 16-29 December 2015, including examining the contributions of local emission and regional transport to the $PM_{2.5}$ concentration in BTH, and the contributions of each detailed physical or chemical process to the variations in the $PM_{2.5}$ concentration. The influence mechanisms of aerosol radiative forcing (including aerosol direct and indirect effects) are also examined by using the process analysis. Figure S4 is only used to explain why aerosol radiative effects had a negative impact on the near-surface $PM_{2.5}$ concentrations in BTH during December 23-24. So we decide to leave the figure in the supplement.

From Fig. S4, we can find that an anomalous northeasterly induced by absorbing aerosols was simulated, leading to a decrease in the near-surface $PM_{2.5}$ concentrations, which is different from previous studies that reported light-absorbing aerosols could worsen air quality (Li et al., 2016; Huang et al., 2018; Gao et al., 2018).

We totally agree with the reviewer's opinion about the important roles of absorbing aerosols on regional circulation changes. More experiments will be designed to examine the changes in atmospheric thermal and atmospheric dynamic caused by absorbing aerosol radiative forcing and their effects on haze episodes in our future studies.

25. *Conclusions and discussions. The "conclusions" part can be shortened and concise, which should make room for more insightful discussion. I came up some ideas. (1) how the IPR scheme can be further improved? (2) the authors could discuss the possible application of IPR scheme in future haze study (both winter and summer), because the work provides a quantitative analysis of how aerosol radiative effects change $PM_{2.5}$ through physical and chemical pathways.*

Response:

According to the reviewer's suggestion, the words in the conclusion part have been cut back by ~20% in the revised manuscript, and more discussions have been added in the last section, including some limitations in the current study and several possible applications of the IPR analysis in future studies. Here are the revised paragraphs:

"There are some limitations in this work. The uncertainty of the MIX anthropogenic emission inventory, the lack of secondary organic aerosols, and the missing mechanisms of some heterogeneous reactions may result in large uncertainties in the final simulation results, especially the predicted aerosol chemical compositions, such as SO_4^{2-}, NO_3^- and NH_4^+. The biases in simulated concentrations of SO_4^{2-}, NO_3^- and NH_4^+ may have impacts on the contributions of AERC and CLDC processes to the air pollution variation. Uncertainties should be quantitatively

analyzed in future studies. Besides, conclusions draw from a case study in BTH cannot represent a full view of the underlying mechanisms of haze formation and elimination. Better understanding will be attained by conducting multiple-case simulations in future. What's more, an anomalous northeasterly induced by absorbing aerosols was observed, leading to a decrease in the near-surface $PM_{2.5}$ concentrations during December 23-24 2015 in BTH, which was different from previous studies that reported light-absorbing aerosols could worsen air quality (Li et al., 2016; Huang et al., 2018; Gao et al., 2018). More experiments should be designed in future to examine the changes in atmospheric thermal and atmospheric dynamic caused by absorbing aerosol radiative forcing and their impacts on haze episodes". **(Page 20, Line 4-15)**

"As Zheng et al. (2018) pointed out that the $PM_{2.5}$ concentration in China has been decreasing in recent years, but the decreased fine particulate matter could stimulate ozone production (Li et al., 2019a; Zhu et al., 2019). Multi-pollutant mixture may be a hot topic in the future, and the IPR analysis can be a useful method to provide a quantitative analysis about the formation mechanism of the complex air pollutions, including figuring out the major physical/chemical process behind these events. Meanwhile, significant differences between model predictions (e.g., O_3 and $PM_{2.5}$) are found among current multi-scale air quality models (Chen et al., 2019b; Li et al., 2019b), even though the same inputs are used. These different performances can be associated with the differences in model formulations, including parameterizations and numerical methods (Carmichael et al., 2008). In order to acquire a quantitative attribution of the cause of differences between simulation results, process analysis method should be developed and implemented in these models, and the IPR analysis will be easier to draw conclusions about the fundamental problems that cause the differences between model predictions". **(Page 20, Line 16-25)**

Reference:

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Gao, M., Guttikunda, S. K., Carmichael, G. R., et al.: Health impacts and economic losses assessment of the 2013 severe haze event in Beijing area, Science of the Total Environment, 511, 553-561, 10.1016/j.scitotenv.2015.01.005, 2015.

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Thank you very much for your comments and suggestions.

Marked-up Manuscript:

Assessing the formation and evolution mechanisms of severe haze pollution in Beijing–Tianjin–Hebei region by using process analysis

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Abstract. Fine particle pollution associated with haze threatens human health, especially in the North China Plain, where extremely high $PM_{2.5}$ concentrations were frequently observed during winter. In this study, the WRF-Chem model coupled with an improved integrated process analysis scheme was used to investigate the formation and evolution mechanisms of a haze event ~~happened~~ over Beijing–Tianjin–Hebei (BTH) in December 2015, including examining the contributions of local emission and ~~outside~~ regional transport to the ~~absolute~~ $PM_{2.5}$ concentration in BTH, and the contributions of each detailed

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physical or chemical process to the variations in the $PM_{2.5}$ concentration. The influence mechanisms of aerosol radiative forcing (including aerosol direct and indirect effects) were also examined by using the process analysis. During the aerosol accumulation stage (December ~~20~~16–~~–~~22, Stage_1), the near-surface $PM_{2.5}$ concentration in BTH was increased from 24.2 μg m^{-3} to 289.8 μg m^{-3}, with the contributions of regional transport increased from 12% to 40%, while the contributions of local emission were decreased from 59% to 38%. ~~the average near-surface $PM_{2.5}$ concentration in BTH was 250.0 μg m^{-3}, which was contributed by local emission of 42.3% and outside transport of 36.6%.~~ During the aerosol dispersion stage (December 23–~~–~~27, Stage_2), the average concentration of $PM_{2.5}$ was 107.9 μg m^{-3}, which was contributed by local emission of 51% and regional transport of 24%.~~. The contribution of local emission increased to 50.9%, while the contribution of outside transport decreased to 24.3%.~~ The 24–~~–~~h change (23:00LST minus 00:00LST) in the near–~~–~~surface $PM_{2.5}$ concentration was +~~50.4~~43.9 μg m^{-3} during Stage_1 and –~~–~~41.5 μg m^{-3} during Stage_2. Contributions of aerosol chemistry, advection ~~process~~ and vertical mixing ~~process~~ to the 24–~~–~~h change were +~~43.8~~29.6 (+17.9) μg m^{-3}, -71.8 (-103.6) μg m^{-3} and ~~–161.6~~177.3 (~~–~~221.6) μg m^{-3} during Stage_1 (Stage_2), respectively. Small differences in contributions of other processes were found between Stage_1 and Stage_2. ~~, such as advection process, cloud chemistry process, and so on.~~ Therefore, the $PM_{2.5}$ increase over BTH during haze formation stage was mainly attributed to the strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes. When aerosol radiative feedback was considered, the 24–~~–~~h $PM_{2.5}$ increase was enhanced by ~~9.6~~4.8 μg m^{-3} during Stage_1, which could be mainly attributed to the contributions of vertical mixing process (+~~39.8~~22.5 μg m^{-3}), advection process (~~–38.6~~-19.6 μg m^{-3}) and aerosol chemistry process (+~~5.1~~1.2 μg m^{-3}). The restrained vertical mixing was the primary reason for the enhancement in near–~~–~~surface $PM_{2.5}$ increase when aerosol radiative forcing was considered.

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1 Introduction

Anthropogenic activities associated with rapidly developed industrialization and urbanization have been leading to a sustained increase in the amounts of atmospheric pollutants, especially in the fast-developing countries (IPCC, 2013). As one of the largest emission sources of aerosols and their precursors, China has been suffering from serious air pollution for years (Lei et al., 2011; Li et al., 2011; Liu et al., 2018a), with severe haze events frequently occurring in winter, especially over large urban agglomerations, such as the North China Plain (NCP) (Han et al., 2014; Gao et al., 2015), the Yangtze River Delta area (YRD) (Ding et al., 2016; Wang et al., 2016a), ~~the Pearl River Delta area (PRD) (Fan et al., 2015; Liu et al., 2018b),~~ and the Sichuan Basin (SCB) (Zhao et al., 2018; Zhang et al., 2019). During severe haze events, the observed maximum hourly surface-layer $PM_{2.5}$ (fine particulate matter with aerodynamic diameter of 2.5 μm or less) concentration exceeded 1000 μg m^{-3} (Wang et al., 2013b; Sun et al., 2016; Li et al., 2017a), which could significantly influence visibility (Li et al., 2014), radiation budget (Steiner et al., 2013), atmospheric circulation (Jiang et al., 2017), cloud properties (Unger et al., 2009), and ~~even~~ human health (Hu et al., 2014; Guo et al., 2017).

Extensive studies have been carried out in recent years to analyze the formation mechanisms of haze episodes in China. Wang et al. (2013a) used a synergy of ground-based observations, satellite, and lidar measurements to study a long-lasting and severe haze episode occurred in eastern China in January 2013, and concluded that stagnant meteorological conditions, which could be generally characterized by weak wind speed, high relative humidity, intense inversion, and low mixing layer height, were tightly associated with severe haze episodes. Based on National Center for Environmental Prediction (NCEP) reanalysis data, Shu et al. (2017) identified five typical synoptic patterns, and pointed out that each synoptic pattern exerted different impacts on particle pollution over YRD. By analyzing the simulation results from a large ensemble climate model (MIROC5), Li et al. (2018a) investigated the contributions of anthropogenic influence to severe haze events happened over eastern China in January 2013 and December 2015, and found that anthropogenic forcing (i.e., increased emissions of greenhouse gases) could modify atmospheric circulation pattern, and these human-induced circulation changes were conducive to the occurrence of severe haze events. Zhang et al. (2015a) used a global 3-D chemical transport model (GEOS-Chem) to quantify the local source contributions to wintertime surface-layer $PM_{2.5}$ concentrations over North China from 2013 to 2015, and reported that emissions from residential and industrial sources and transportation contributed

most to the high concentrations of atmospheric aerosols in Beijing. Many studies reported that regional transport of aerosols also played an important role in haze episodes (Wang et al., 2013b; Jiang et al., 2015; ~~Zheng et al., 2015~~Li et al., 2018b). Wang et al. (2013b) reported that the cross-city clusters transport outside BTH (Beijing, Tianjin, and Hebei) and transport among cities inside BTH contributed 20%–35% and 26%–35% of $PM_{2.5}$ concentrations over BTH, respectively. Secondary aerosol formation and their hygroscopic growth were also confirmed to be a large ~~contribution~~ contributor to severe haze episodes (Huang et al., 2014b; Han et al., 2015; Chen et al., 2019a). The conversion of SO_2 to SO_4^{2-} was strongly associated with high relative humidity, and NO_3^- was found to be produced mainly by photochemical and heterogeneous reactions (Chen et al., 2016; Zhang et al., 2018a).

It is well known that aerosols can scatter and absorb solar radiation to alter the radiative balance of the atmosphere and surface (direct radiative effect), and can serve as cloud condensation nuclei or ice nuclei to affect cloud properties (indirect radiative effect) (Twomey, 1974). These impacts are coupled with atmospheric dynamics to produce a chain of interactions with a large range of meteorological variables that influence both weather and climate (Ramanathan et al., 2001; Huang et al., 2006; Li et al., 2017c; Yang et al., 2017), which will further induce feedbacks on aerosol production, accumulation, and even severe haze pollutions (Petaja et al., 2016; Li et al., 2017d; Zhao et al., 2017; Gao et al., 2018; Lou et al., 2019). Based on multi-year measurements (from 2010 to 2016), Huang et al. (2018) found that aerosol radiative effects led to a significant heating in the upper planetary boundary layer (PBL) and a substantial dimming at the surface over North China. This is because high concentrations of light-absorbing aerosols were observed, and the aerosol-meteorology interactions depressed the development of PBL, and therefore aggravated the haze pollution (Su et al., 2018). The light-absorbing aerosols can also amplify haze in NCP by weakening East Asian winter monsoon wind speeds through ocean and cloud feedbacks (Lou et al., 2019). By using the WRF-Chem model, Gao et al. (2015) analyzed the feedbacks between aerosols and meteorological fields over NCP in January 2013, and found that aerosols caused a significant negative (positive) radiative forcing at the surface (in the atmosphere), resulting in a weaker surface-layer wind speed and lower PBL height (PBLH). The average surface-layer $PM_{2.5}$ concentration was increased by 10–50 μg m^{-3} as a result of the more stable atmosphere. By analyzing the observations from a comprehensive field experiment and simulation results from WRF-Chem model, Liu et al. (2018b~~a~~) concluded that the decreased PBLH associated with increased aerosol concentrations could

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enhance surface-layer relative humidity by weakening the vertical transport of water vapor, and the increased relative humidity at the surface accelerated the formation of secondary particulate matters (SPM) through heterogeneous reactions, leading to the increase of the $PM_{2.5}$ concentration by 63 μg m^{-3} averaged over the NCP during 15-21 December, 2016.

All these studies discussed above revealed that the formation of haze episodes was caused by the interactions betweensynergy impacts of local emissions, regional transport, meteorological conditions, and chemical production. Nevertheless, only the net combined effects on the concentrations of pollutants were provided, without the capabilities of understanding and isolating the atmospheric physical and chemical processes involved. The quantitative assessment of the contributions from each detailed physical/chemical process (e.g., vertical mixing process, advection process, emission source process, aerosol chemistry process, cloud chemistry process, and so on) is necessary for fully understanding of the formation and evolution mechanisms of haze episodes (Goncalves et al., 2009; Xing et al., 2017; Kang et al., 2019). What's more, although many previous studies have identified the positive feedback effects of aerosol radiative forcing on particulate accumulation, the detailed influence mechanisms of the forcing-response relationship at each process chain remain largely elusive (i.e., the prominent physical or chemical processes responsible for the aerosol radiative impacts on haze episodes). Since 20092013, substantial efforts have been taken to improve air quality in China, including emission reduction and energy transition. However, haze events continued to occur frequently all over the country. For example, a severe, long-lasting, and wide-ranging haze episode was observed in December 2015 over the central and eastern China, with the regional average $PM_{2.5}$ concentration exceeding 150 μg m^{-3}. For BTH, a red alert for haze (the most serious level) was issued for the period from 20 to 22 December 2015, with the maximum hourly $PM_{2.5}$ concentration exceeding 1000 μg m^{-3}. The formation and evolution mechanisms, and the aerosol radiative feedbacks of this severe haze episode have not been fully estimated yet.

In this study, we develop an improved online integrated process rate (IPR) analysis scheme (i.e., process analysis) in the fully coupled online Weather Research and Forecasting-Chemistry (WRF-Chem) model, to investigate the formation and evolution mechanisms of the severe haze episode happened over NCP from 20 16 to 29 December 2015. Sensitivity experiments are conducted to examine the contributions of local emission and outside regional transport to the absolute $PM_{2.5}$ concentrations during the haze episode, while the IPR analysis is used to quantify the contributions of each detailed physical/chemical process to the variations in the $PM_{2.5}$ concentrations. The effects of aerosol radiative forcing, including

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direct and indirect effects, on meteorological parameters and $PM_{2.5}$ levels during the haze episode are also quantified, with a special focus on the detailed influence mechanism. We hope that the results concluded in this study may provide better understanding of the formation mechanisms for severe haze events, and help policy makers take targeted measures to improve air quality over North China.

This manuscript is arranged as follows. Model configuration, integrated process rate (IPR) analysis (i.e., process analysis), numerical experiments, and observations are presented in Section 2. Model evaluation is conducted in Section 3. The formation and evolution mechanisms of the haze episode are investigated in Section 4. Section 5 provides the impacts of aerosol radiative forcing. Summaries and discussions are presented in Section 6.

2 Methods

2.1 Model configuration

A fully coupled online Weather Research and Forecasting–Chemistry model (WRF–Chem v3.7) is used to simulate meteorological fields and concentrations of gases and aerosols simultaneously (Skamarock et al., 2008; Grell et al., 2005). The WRF–Chem model is designed with two domains using 219 (west–east) × 159 (south–north) and 150 (west–east) × 111 (south–north) grid points at the horizontal resolutions of 27 and 9 km, respectively (Fig. 1). The outer domain covers nearly the whole East Asia, and the inner domain is located in the NCP. In order to minimize the impacts from IBCs (lateral boundary conditions), we only analyze the simulation results from the inner region of the second domain (i.e., BTH), following Chen et al. (2018) and Wu et al. (2012). The vertical dimension is resolved by 29 full sigma levels, with ~~16~~ 15 layers located in the lowest 2 km for finer resolution in the planetary boundary layer, and the height of the first layer averaged in BTH is about 30 m.

Meteorological initial and lateral boundary conditions used in the WRF–Chem model are taken from the NCEP (National Center for Environmental Prediction) Final Operational Global Analysis data with the spatial resolution of 1° × 1°. Four–dimensional data assimilation (FDDA) with the nudging coefficient of 3.0×10^{-4} for wind (in and above PBL), temperature (above PBL) and water vapor mixing ratio (above PBL) is adopted to improve the accuracy of simulation results

(no analysis nudging is included for the inner domain) (Lo et al., 2008; Otte, 2008; Wang et al., 2016b; Werner et al., 2016). The forecasts from the global chemical transport model MOZART-4 are processed to provide the chemical initial and boundary conditions for the WRF-Chem model (Emmons et al., 2010).

Anthropogenic emission data are obtained from the MIX Asian emission inventory (http://www.meicmodel.org/dataset-mix.html), with a horizontal resolution of 0.25 degree (Li et al., 2017b). It is developed to support the MICS-Asia III (Model Inter-Comparison Study for Asia Phase III) and the TF HTAP (Task Force on Hemispheric Transport of Air Pollution) projects. This inventory includes SO_2 (sulfur dioxide), NO_x (nitrogen oxides), CO (carbon monoxide), CO_2 (carbon dioxide), NMVOC (non-methane volatile organic compounds), NH_3 (ammonia), BC (black carbon), OC (organic carbon), $PM_{2.5}$ and PM_{10}. All these species are from several sectors, such as agriculture, industry, power, transportation and residential, and the emission rate of each species for each hour is based on Gao et al. (2015). The biogenic emissions are calculated online using the MEGANv2.04 (Model of Emission of Gases and Aerosol from Nature v2.04) model (Guenther, 2006). Biomass-burning emissions are obtained from the GFEDv3 (Global Fire Emissions Database v3) (Randerson et al., 2005). Dust emissions and sea salt emissions are calculated online by using algorithms proposed by Shao (2004) and Gong et al. (1997), respectively.

The Carbon-Bond Mechanism version Z (CBMZ) (Zaveri and Peters, 1999) is selected to simulate the gas phase chemistry, and the 8-bin sectional aerosol module, MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) (Zaveri et al., 2008), with some aqueous chemistry, is used to simulate aerosol evolution. All major aerosol species are considered in the MOSAIC scheme, including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), chloride (Cl), sodium (Na), BC, primary organic mass, liquid water, and other inorganic mass (Zaveri et al., 2008). The aerosol size distribution is divided into discrete size bins defined by their lower and upper dry particle diameters (Zhao et al., 2010). In the current CBMZ/MOSAIC scheme, the formation of SOA (secondary organic aerosol) is not included (Zhang et al., 2012; Gao et al., 2016). Aerosol optical properties, including extinction efficiency, single scatter albedo, and asymmetry factor are computed by Mie theory, based on aerosol composition, mixing state, and size distribution (Barnard et al., 2010). The impacts of aerosols on photolysis rates are calculated using the Fast-J photolysis scheme (Wild et al., 2010). Aerosol radiation is simulated by RRTMG (Rapid Radiative Transfer Model for GCMs) for both shortwave (SW) and longwave (LW) radiation

(Zhao et al., 2011). More information about the parameterizations used in this study can be found in Table 1.

2.2 Integrated process rate (IPR) analysis

Most air quality models are configured to output only the pollutant concentrations that reflect the combined effects of all physical and chemical processes. Quantitative information of the impacts of individual process is usually unavailable. Process analysis techniques (i.e., integrated process rate (IPR) analysis) can be used in grid-based Eulerian models (e.g., WRF-Chem) to obtain contributions of each physical/chemical process to variations in pollutant concentrations. Eulerian models utilize the numerical technique of operator splitting to solve continuity equations for each species into several simple ordinary differential equations or partial differential equations that only contain the influence of one or two processes (Gipson, 1999).

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The IPR analysis method has been fully implemented in Community Multi-scale Air Quality (CMAQ) model, and has been widely applied to study regional photochemical ozone (O_3) pollution (Goncalves et al., 2009; Khiem et al., 2010; Xing et al., 2017; Tang et al., 2017). Several WRF-Chem model studies used the IPR analysis to investigate the impacts of physical/chemical process on variations in O_3 concentrations. Gao et al. (2018) investigated the impacts of BC-PBL interactions on O_3 concentrations by analyzing the contributions from photochemistry, vertical mixing, and advection processes. Jiang et al. (2012) calculated the contributions of photochemical reactions and physical processes to O_3 formation by using a simplified IPR analysis scheme.

Applying the IPR analysis to diagnose the contributions of each physical or chemical process to variations in aerosol concentrations in WRF-Chem model is more complex technically, and therefore few studies conducted the IPR analysis for aerosols. In this study, we developed an improved IPR analysis scheme in the WRF-Chem model to isolate the processes impacting variations in aerosol concentrations into nine different processes, namely advection (TRAN), emission source (EMIS), dry deposition (DYRD), turbulent diffusion (DIFF), sub-grid convection (SGCV), gas-phase chemistry (GASC), cloud chemistry (CLDC), aerosol chemistry (AERC), and wet scavenging (WETP). TRAN includes horizontal and vertical advection, which is highly related to wind and aerosol concentration gradients from upwind regions to downwind areas (Gao et al., 2018). DRYD is based on resistance models for trace gases (Wesely, 1989) and aerosol particles (Ackermann et al.,

1998). SGCV ~~includes~~ refers to the scavenging within the sub-grid wet convective updrafts. CLDC refers to aqueous-phase photolytic and radical chemistry reactions in clouds, including the activation processes. ~~the scavenging and aqueous chemistry within the wet convective updrafts.~~ AERC means microphysical nucleation, condensation, and coagulation, as well as the mass transfer between the gas phase and condensed phase. WETP contains in-~~-~~cloud rainout and below-~~-~~cloud washout during grid-~~-~~scale precipitation. The contribution of individual process can be calculated as the difference of aerosol concentrations before and after the corresponding operator.

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Based on the principle of mass balance, IPR can be verified by comparing the variations in aerosol concentrations (the concentration at the current time minus the concentration at the previous time) with the sum of the contributions from the nine processes during each time step. As shown in Fig. S1, the net contributions of all processes match the variations in aerosol concentrations pretty well.

2.3 Numerical experiments

Table 2 summarizes the experimental designs. To investigate the contributions of ~~outside~~ regional transport and local emission to the ~~absolute~~ $PM_{2.5}$ concentrations in BTH, four simulations with different anthropogenic emission categories are conducted: (1) CTL: The control simulation with all anthropogenic emissions considered; (2) NoAnth: No anthropogenic emission is considered in the whole domain; (3) NoBTH_Anth: Same as CTL, but anthropogenic emissions in BTH are excluded; (4) OnlyBTH_Anth: Contrary to the NoBTH_Anth case, anthropogenic emissions are only considered in BTH. All the physical and chemical schemes used in these cases are identical. The contributions of regional transport and local emission to the ~~absolute~~ $PM_{2.5}$ concentration in BTH can be identified by comparing the simulation results of NoBTH_Anth and NoAnth (i.e., NoBTH_Anth minus NoAnth) and OnlyBTH_Anth and NoAnth (i.e., OnlyBTH_Anth minus NoAnth), respectively.

To quantify the aerosol radiative effects (ARE) on ~~the~~ haze ~~episode~~pollution, another sensitivity experiment (referred to as NoARE case) is designed by turning off the feedbacks between aerosols and meteorological variables, including eliminating the aerosol direct effect (ADE) and aerosol indirect effect (AIE) in the model. The ADE is turned off by removing the mass of aerosol species from the calculation of aerosol optical properties as did in Qiu et al. (2017). The AIE is

turned off by using ~~the~~a prescribed vertically uniform ~~constant~~ cloud droplet number ~~concentration (CDNC)~~, which is calculated from the CTL case during the whole simulation period, following Gao et al. (2015) and Zhang et al., (2015a). The differences between CTL and NoARE (i.e., CTL minus NoARE) represent the impacts of aerosol radiative forcing.

The IPR analysis method is applied to all the designed experiments. Comparing the contributions of each detailed process between pollution accumulation stage and dissipation stage in CTL can quantitatively explain the reason for the variation of the $PM_{2.5}$ concentrations in BTH. Meanwhile, the prominent physical or chemical process responsible for the aerosol radiative impacts on the haze episode can also be investigated by analyzing the IPR analysis method used in CTL and NoARE cases.

~~The IPR analysis method is applied to two stages in the CTL case to quantify the contribution of each detailed physical and/or chemical process to the variations in the $PM_{2.5}$ concentration. Comparing the contribution of each process between the two stages can quantitatively explain the reason for $PM_{2.5}$ increase during the stage of haze accumulation and $PM_{2.5}$ decrease during the stage of haze dispersal.~~

~~To quantify the aerosol radiative effects (ARE) on the haze episode, another sensitivity experiment (referred to as NoARE case) is designed by turning off the feedbacks between aerosols and meteorological variables, including eliminating the aerosol direct effect (ADE) and aerosol indirect effect (AIE) in the model. The ADE is turned off by removing the mass of aerosol species from the calculation of aerosol optical properties as did in Qiu et al. (2017). The AIE is turned off by using the constant cloud droplet number concentration (CDNC), which is calculated from the CTL case during the whole simulation period, following Gao et al. (2015). The differences between CTL and NoARE (i.e., CTL minus NoARE) represent the impacts of aerosol radiative forcing.~~

~~The IPR analysis method is then applied to CTL and NoARE cases, respectively, to investigate the detailed influence mechanisms (i.e., the prominent physical or chemical process responsible for the aerosol radiative impacts on the haze episode).~~

All the five simulations are conducted for the period from 1~~3~~7 to 29 December 2015, and the initial three days are discarded as the model spin-up to minimize the impacts of initial conditions. Simulation results from the CTL case during ~~20~~ 16 to 29 December 2015 are used to evaluate the model performance.

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2.4 Observational data

Simulated meteorological parameters in CTL case, including 2 m temperature (T_2), 2 m relative humidity (RH_2), 10 m wind speed (WS_{10}) and 10 m wind direction (WD_{10}), are compared with hourly observations at twelve stations, which are collected from NOAA's National Climatic Data Center (https://gis.ncdc.noaa.gov/maps/ncei/cdo/hourly). Due to limited observations of PBL height in BTH, the retrieved PBLH in 3-hour intervals obtained from the GDAS (Global Data Assimilation System) (https://ready.arl.noaa.gov/READYamet.php) in Beijing (39.93 °N, 116.28 °E) is also used to evaluate the model performance. More detailed information about the GDAS meteorological dataset (1 °×1 °) can be found in Rolph et al. (2013), Kong et al. (2015) and https://www.ready.noaa.gov/gdas1.php. ~~**The meteorological observation sites are marked in blue dots in Fig. 1(b).** PBL height (PBLH) in 3-hour intervals provided by NOAA READY archived meteorological GDAS data () in Beijing (39.93 °N, 116.28 °E) (marked in purple dot in Fig. 1(b)) is also used to evaluate the model performance.~~ Hourly shortwave downward radiation flux (SWDOWN) at the Xianghe station (39.75 °N, 116.96 °E)~~, marked in light green dot in Fig. 1(b),~~ is taken from WRMC–BSRN (World Radiation Monitoring Center–Baseline Surface Radiation Network, http://bsrn.awi.de) for the energy budget evaluation. The hourly observed surface–layer $PM_{2.5}$ concentrations at the 59 stations~~ (marked in red dots in Fig. 1(b))~~ are obtained from the CNEMC (China National Environmental Monitoring Center, http://www.cnemc.cn/). The daily measurements of mass concentrations of SO_4^{2-}, NO_3^-, NH_4^+, ~~BC~~ BC and OC ~~OC~~ are ~~also~~ collected at the ~~two sites~~sites of (39.97 °N, 116.37 °E) ~~((39.97 °N, 116.37 °E) and (38.03 °N, 114.53 °E))~~ in Beijing and (38.03 °N, 114.53 °E) in Shijiazhuang (Huang et al., 2017; Liu et al., 2018)~~, respectively~~. Detailed locations of these observations are shown in Fig. 1(b).

~~(marked in dark green triangles in Fig. 1(b)).~~

3. Model evaluation

Accurate representations of observed meteorological fields and pollutant concentrations provide foundations for haze analysis with the WRF–Chem model. Detailed comparisons between observed and simulated meteorological parameters (T_2, RH_2, WS_{10}, WD_{10}, PBLH, and SWDOWN) and pollutant concentrations ($PM_{2.5}$, BC, OC, SO_4^{2-}, NO_3^-, and NH_4^+) are

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presented in this section.

3.1 Meteorological parameters

Figure 2 shows the time series of observed and simulated hourly meteorological variables averaged over the 12 stations during 2016–29 December 2015. Corresponding statistical metrics, including mean value, mean bias (MB), gross error (GE), normalized mean bias (NMB), root mean square error (RMSE), mean fractional bias (MFB), mean fractional error (MFE), index of agreement (IOA), and correlation coefficient (R) are presented in Table 3. As shown in Fig. 2, simulated T_2, RH_2, WS_{10} and WD_{10} agree well with the observational data. For temperature, the WRF-Chem model can perfectly depict its diurnal and daily variations with both R and IOA of 0.90 and 0.94, respectively, but slightly overestimates the low values at night, with the NMB of 10.4%. Observed relative humidity can be reasonably reproduced by the model with R and IOA of 0.8 73 and 0.782, respectively. But a persistent underestimation is found with the NMB of −14.32%. Different surface layer and boundary layer options parameterizations may have influence on the simulated near-surface moisture fluxes, and the settings of these schemes can partially explain the biases of RH_2 between observations and simulations (Qian et al., 2016). This negative bias of RH_2 was can also reported be simulated in by other studies (Zhang et al., 2009; Gao et al., 2015). WRF-Chem can capture the observed low values of wind speed during 2019–22 23 December and high values of wind speed during 16-17 and 25–27 December. The positive NMB of 29.18% may probably result from unresolved topographical features in surface drag parameterization and the coarse resolution used in the nested domain (Yahya et al., 2015; Zheng et al., 2015). For wind direction, the calculated NMB is =1.31% and the R IOA is 0.65, indicating that the WRF-Chem model can generally reproduce the varied wind direction during the simulation period.

Simulated hourly PBLH and SWDOWN are also compared with observations in Fig. 3. It is noted that PBLH provided by GDAS of NOAA are in 3-hour intervals. The simulations in CTL case agree well with the observations, including capturing the daily maximum in the daytime and the low values at night. The correlation coefficients are 0.7 68 and 0.91 for PBLH and SWDOWN, respectively.

3.2 $PM_{2.5}$ and its components

Observed hourly surface-layer $PM_{2.5}$ concentrations from 20-16 to 29 December 2015 in the nine cities (Shengyang, Beijing, Xingtai, Hengshui, Baoding, Langfang, Yangquan, Anyang, and Jinan) are compared with the model results from CTL case (Fig. 4). The statistical metrics are shown in Table 3. Generally, WRF-Chem model can reasonably reproduce the evolutional characteristics of the observed $PM_{2.5}$ concentrations in the nine cities (Rs=0.5857-0.8890). Both the observed and simulated $PM_{2.5}$ concentrations exhibit a growth trend during December 2016-22 and 28-29, and a decreasing tendency during December 23-27. However, an obvious underestimation is found in Beijing from 25 to 26 December when a maximum hourly concentration of 600 μg m^{-3} was observed. Thise negative bias is also simulated by previous studies (Chen et al., 2018; Zhang et al., 2018b), and the possible reasons for the underestimation are (1) the bias in simulated meteorological conditions (e.g., underestimated RH_2 and overestimated WS_{10}); (2) the missing mechanisms of some gas-aerosol phase partitioning and heterogeneous reactions which may produce secondary inorganic aerosol (Huang et al., 2014a; Wang et al., 2014); (3) the lack of SOA simulation in MOSAIC mechanism (Gao et al., 2016). Generally, the performance statistics of $PM_{2.5}$ in almost all cities meet the model performance goal (MFB within ±30% and MFE≤ ≤50%) proposed by Boylan and Russel (2006).

Figure 5 compares the simulated and observed surface-layer concentrations of BC, OC, SO_4^{2-}, NO_3^-, and NH_4^+ in Beijing and Shijiazhuang averaged during 2016-29 December 2015. WRF-Chem model underestimates the concentrations of SO_4^{2-}, NH_4^+ andBC, OC, NH_4^+ and SO_4^{2-} concentrations in Beijing OC in Beijing (Shijiazhuang) by 3.6% (33.1%), 34.919% (38.640%), 24.114% (37.59%), and 32.521% (44.641%), respectively, and but overestimates the concentrations of NO_3^- concentration by 22.229% (51.844%). Due to the low reactivity of BC in the atmosphere, the uncertainty in BC emission may cause this underestimationthe biases in Beijing (NMB=+10%) and Shijiazhuang (NMB=-24%). (Li et al., 2017b). For OC, the underestimation may result from the lack of SOA in the MOSAIC aerosol module (Qiu et al., 2017). Missing some mechanisms of SO_2 gas-phase and aqueous-phase oxidation, as well as heterogeneous chemistry may explain the underestimation of SO_4^{2-}. It is noted that similar biases of the aerosol components were also reported by other WRF-Chem studies (Zhang et al., 2015a; Qiu et al., 2017).

4. Formation and evolution mechanisms of the haze episode

In this section, we first reproduce the evolution of the severe haze episode, and then investigate the formation and evolution mechanisms, including examining contributions of local emission and ~~outside~~ regional transport to the ~~absolute~~ $PM_{2.5}$ concentration in BTH, and the contributions of each detailed physical/chemical process to the variations in the $PM_{2.5}$ concentration.

4.1 Spatial-~~-~~temporal evolutions of surface-~~-~~layer $PM_{2.5}$ concentrations

Figures 6(a-~~-~~jk) show the spatial distributions of simulated daily mean surface-~~-~~layer $PM_{2.5}$ concentrations from ~~20~~ 17 to ~~29~~ 28 December 2015. From December 17, aerosol particles started to accumulate in the near-surface layer in BTH under a prevailing southerly air flow. On December 20, the BTH region was under a uniform pressure field (Fig. S2(a)). The regional average wind speed was less than 3 m s^{-1}, and the boundary layer became stable, which constrained aerosols within a low mixing layer. Meanwhile, a low-~~-~~pressure center situated to the north of BTH, where air pollutants from south, southwest, and southeast converged. Consequently, the daily mean $PM_{2.5}$ concentration averaged over BTH was over 200 μg m^{-3}. On December 21, a weak low-~~-~~pressure center was formed near the Bohai Bay and a weak high-~~-~~pressure center moved to Shandong Peninsula (Fig. S2(b)). The synoptic conditions brought more air masses from south to north, and worsened air quality in BTH. On December 22, a weak high pressure system moved within Inner Mongolia (Fig. S2(c)), which could bring cold air to the BTH region. Meanwhile, the polluted air could also be transported back to the BTH, leading to a continuous increase in the $PM_{2.5}$ concentration, with the maximum daily mean value exceeding 600 μg m^{-3} in BTH (Fig. 6(~~e~~e)). Due to the enhanced anticyclone originated from Siberian (Fig. S2(d)), the accumulation of aerosol particles in BTH was terminated with the incursion of a strong cold front from 23 to 27 December. But frequent transitions between high and low pressure systems over BTH accompanying with the shifting wind directions resulted in a quick $PM_{2.5}$ variation, especially on December 24 and 25, when a low-~~-~~pressure system developed northeast of BTH (Fig. S2(e)). The air mass in BTH was influenced by the pollutants from south, resulting in a temporary increase in the concentration of $PM_{2.5}$ on December 25. After December 27~~On December 28 and 29~~, another haze episode ~~occurred~~gradually formed.

According to the trends in simulated $PM_{2.5}$ concentrations averaged over the BTH region (Fig~~s~~. 6(~~k-l~~)), we divide the

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whole simulation period into three stages: (1) aerosol accumulation stage (December 1620--22, Stage_1); (2) aerosol dispersion stage (December 23--27, Stage_2); (3) formation stage for another haze event (December 28--29, Stage_3). In this manuscript, we mainly focus on the first two stages to reveal important factors that cause the accumulation and dispersion of particulate matters.

In Stage_1, the daily mean $PM_{2.5}$ concentrations averaged over BTH increased from 209.024.2 μg m^{-3} to 289.8 μg m^{-3}, and the average $PM_{2.5}$ concentration was 250.0145.6 μg m^{-3} (Fig. 7(a)), far beyond close to the air quality threshold value of "heavily polluted" ($PM_{2.5}$ 24-h average concentration > 150 μg m^{-3}). The WS_{10} was low (Fig. 7(b)), especially during the heavy pollution period (20-22 December), and the mean wind speed was 2.3-3 m s^{-1} (Fig. 7(b)), less than 3.2 m s^{-1} (one of the indicators used to define air stagnation by NOAA, https://www.ncdc.noaa.gov/societal-impacts/air-stagnation/overview), indicating that the near surface circulation was insufficient to disperse accumulated air pollutants. The decreased PBLH (from 148.6701.6 m to 109.9 m) could compress air pollutants into a shallow layer, resulting in an elevated pollution level. During Stage_2, the $PM_{2.5}$ concentration decreased gradually with the increased wind speed and PBLH. The $PM_{2.5}$ concentration averaged during Stage_2 was 107.9 μg m^{-3}, still exceeding the Grade II standard (75 μg m^{-3}) defined by the National Ambient Air Quality Standards of China.

4.2 Contributions of local emission and regional transport to absolute $PM_{2.5}$ concentrations

Previous studies have reported that anthropogenic emission was the internal dominant cause of haze events in China (Jiang et al., 2013; Sun et al., 2014; Gu and Liao, 2016; Yang et al., 2016b). Emission control measures have been taken to ensure good air quality for major events (e.g., APEC) or to mitigate the severity of coming pollution episodes (Zhou et al., 2018). Other studies, such as Sun et al. (2017) and Wang et al. (2017b), pointed out that outside regional transport contributed more than 50% of the particulate concentrations in BTH during haze events. This section discusses the contributions of local anthropogenic emission and regional transport to the $PM_{2.5}$ concentration in BTH, aiming to reveal the relative importance during this haze episode.

As shown in Fig. 7(a), the $PM_{2.5}$ concentration in BTH during Stage_1 was mainly contributed by the combined effects

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of local emission and regional transport. The contributions of local emission and regional transport to the $PM_{2.5}$ concentration were comparable (49% and 32%, respectively), especially during the heavy pollution period (December 20-22, 43% vs. 37%). ~~(42.3% and 36.6%, respectively). In~~During Stage_2, the contributions of ~~outside~~ regional transport decreased from 30.~~0~~% to 16.~~3~~%. The relative high $PM_{2.5}$ concentration (107.9 μg m^{-3}) was principally caused by the local emission. On average, the contributions of local emission and regional transport to the $PM_{2.5}$ concentration ~~during~~ in Stage_2 were 5~~10.9~~% and 24.~~3~~%, respectively. The impact of ~~outside~~ regional transport could be qualitatively expressed by specific humidity, which was treated as an indicator for the origin of air masses (Jia et al., 2008). Air masses from the south were usually warmer and wetter than those from the north, so the specific humidity averaged over the BTH was higher in Stage_1 (1.7 g/kg) than that in Stage_2 (1.4 g/kg) (Fig. 7(b)). The evolution of $PM_{2.5}$ nicely followed the trend of specific humidity with a high correlation coefficient of 0.~~91~~86.

4.3 Contributions of each physical/chemical process to variations in $PM_{2.5}$ concentrations

Figures 8(a1-~~-~~a2) show the diurnal variations of $PM_{2.5}$ concentrations averaged over the BTH region during Stage_1 and Stage_2, respectively. The $PM_{2.5}$ concentration increased by ~~50.4~~43.9 μg m^{-3} (from ~~237.0~~136.5 μg m^{-3} at 00:00LST to ~~287.4~~180.4 μg m^{-3} at 23:00LST) during the period of particulate accumulation (Stage_1), but it decreased by 41.5 μg m^{-3} during the period of particulate elimination (Stage_2).

The hourly $PM_{2.5}$ changes induced by each and all physical/chemical processes during Stage_1 and Stage_2 by using the IPR analysis method are shown in Figs. 8(b1-~~-~~b2). During both stages, the dominant sources of surface-layer $PM_{2.5}$ were EMIS and AERC, while the main sinks were TRAN, DIFF, and DRYD. The maximum positive contribution of EMIS could be found during the rush hours (07:00-~~-~~08:00LST and 16:00-~~-~~19:00LST) (Fig. S3). The maximum negative contributions of TRAN and DIFF appeared at late night (0~~1~~0:00-~~-~~05:00LST) and at noon (11:00-~~-~~14:00LST), respectively.

To explain the reason for 24-~~-~~h $PM_{2.5}$ increase during Stage_1 and 24-~~-~~h $PM_{2.5}$ decrease during Stage_2 (Figs. 8(a1-~~-~~a2)), we quantify the contributions of each physical/chemical process to 24-~~-~~h $PM_{2.5}$ changes for both stages (Figs. 8(c1-~~-~~c2)), which are calculated by integrating hourly $PM_{2.5}$ changes induced by each process from 00:00LST to 23:00LST (Figs. 8(b1-~~-~~b2)). In WRF-~~-~~Chem, DRYD is intermingled with vertical diffusion, so changes in the column burden during

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vertical mixing can be attributed to DRYD (Tao et al., 2015). Following Tao et al. (2015), we define vertical mixing (VMIX) as the sum of DIFF and DRYD. As shown in Figs. 8(c1~~,~~ c2), contributions of AERC, TRAN and VMIX processes to 24-h $PM_{2.5}$ changes were +29.6 (+17.9) μg m^{-3}, -71.8 (-103.6) μg m^{-3} and -177.3 (-221.6) μg m^{-3} for Stage_1 (Stage_2), respectively. ~~AERC and VMIX process to 24-h $PM_{2.5}$ changes were +43.8 (+17.9) μg m^{-3} and -161.6 (-221.6) μg m^{-3} for Stage_1 (Stage_2), respectively.~~ Small differences were found for contributions from other processes between Stage_1 and Stage_2 (differences smaller than ~~10~~ 5 μg m^{-3}). Therefore, the $PM_{2.5}$ increase over the BTH region during haze formation stage was mainly attributed to strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes. On the contrary, during haze elimination stage (Stage_2), more aerosols in BTH were transported out of BTH or dispersed to the upper atmosphere or subsided to the ground. What's more, the dry cold air from the north decreased the specific humidity (as shown in Fig. 7(b)) in BTH, leading to weaker production of secondary aerosols by aerosol chemistry process.

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5 Aerosol radiative effects (ARE) on the haze episode

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~~Suspended aerosol particulates can perturb the earth-atmosphere radiation balance, alter meteorological fields, and further affect air quality (Wang et al., 2017a). Previous studies have demonstrated the significance of aerosol-radiation feedbacks on air quality in BTH, especially during winter haze periods.~~ Previous studies have demonstrated that ~~T~~the aerosol radiative forcing ~~was reported to~~could increase the near-surface $PM_{2.5}$ concentrations by about 12%-29%~~by 11.9%-28.7% of the near-surface $PM_{2.5}$ concentrations~~ (Gao et al., 2015; Gao et al., 2016; Qiu et al., 2017; Zhou et al., 2018). However, the detailed influence mechanisms (i.e., the prominent physical or chemical process responsible for the aerosol radiative impacts on $PM_{2.5}$ concentrations) are still unclear. In this section, we examine the effects of aerosol radiative forcing on meteorological parameters and $PM_{2.5}$ levels during the haze episode, with a special focus on the detailed influence mechanism by using the IPR analysis.

5.1 Effects of aerosol radiative forcing on meteorological parameters and $PM_{2.5}$ concentrations

Figure 9 illustrates the impacts of aerosols on the downward shortwave radiative flux (SW) at the surface (BOT_SW)

and in the atmosphere (ATM_SW), calculated by subtracting the model results of NoARE from those of CTL, during Stage_1, Stage_2, and the whole simulation period. Downward SW at the surface was strongly decreased when ARE was considered, especially over high aerosol loading regions during heavily polluted periods. ~~It was known that aerosols could scatter and absorb incoming solar radiation and lead to surface dimming. Besides, in-cloud particles could change the lifetime and albedo of cloud and influence the shortwave radiation at the ground.~~ Generally, the shortwave radiation fluxes at the surface averaged over BTH were reduced by ~~36.5~~28% (~~31.6~~23.9 W m^{-2}) in Stage_1, 18~~.3~~% (16.6 W m^{-2}) in Stage_2, and ~~24.1~~23% (~~21.5~~19.9 W m^{-2}) during the whole simulation period, respectively. Contrary to the significant negative effects at the surface, as a result of ARE, the downward SW fluxes in the atmosphere averaged over BTH were increased by ~~84.7~~65% (~~25.5~~19.1 W m^{-2}) in Stage_1, 37~~.4~~% (10.8 W m^{-2}) in Stage_2, and ~~53.9~~51% (~~15.7~~14.7 W m^{-2}) during the whole period, respectively.

The impacts of ARE (including aerosol direct and indirect effects) on meteorological parameters and $PM_{2.5}$ concentrations are analyzed in Fig. 10. Because less SW could reach the ground, near surface temperature was decreased over BTH (Fig. 10(a)), especially during ~~stage_1 when $PM_{2.5}$ concentrations were higher~~heavy pollution periods, and the largest decrease was up to 2 k. Meanwhile, the increased SW in the atmosphere could warm the upper air. As a result, a more stable atmosphere was expected. It is known that the atmospheric stability can be exactly characterized by the profile of equivalent potential temperature (EPT) (Bolton, 1980; Zhao et al., 2013; Yang et al., 2016a). If EPT rises with height, the atmosphere is stable. As shown in Fig. 10(b), the EPT was decreased in the lower atmosphere (below ~1000 m) with the largest decrease of 3 k on December 22, but increased in the upper atmosphere (above ~1200 m). The change in the EPT profile indicated that ARE could lead to a more stable atmosphere, which further weakened vertical movement in BTH (Fig. 10(c)). As a result of ARE, the PBLH was decreased and the relative humidity in the lower atmosphere was increased (Fig. 10(d)). All the changes in meteorological variables were beneficial for $PM_{2.5}$ accumulation in the lower atmosphere (Fig. 10(e)). The daily maximum increase of $PM_{2.5}$ concentration was 43.2 μg m^{-3} due to ARE. It was noticed that ARE had a negative impact on the near surface $PM_{2.5}$ concentrations during December 23 24, which could be explained that absorbing aerosols (i.e., BC) induced anomalous northeasterlies, and the relatively clean air transported from the northeastern regions to BTH (Fig. S4).

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5.2 Influence mechanism of aerosol radiative effects

Since variations in $PM_{2.5}$ concentrations are directly caused by physical and chemical processes (Zhu et al., 2015), the IPR method is then used to investigate the detailed influence mechanisms (i.e., the prominent physical or chemical processes responsible for the aerosol radiative impacts on haze episodes). Figs. 11(a~~-~~b) show the diurnal variations of $PM_{2.5}$ concentrations in NoARE and CTL cases averaged over the BTH region in Stage_1. A 24~~-~~h increase of ~~40.8~~39.1 μg m^{-3} was simulated in NoARE case. When aerosol radiative forcing was considered, the 24~~-~~h increase of $PM_{2.5}$ concentration was ~~50.4~~43.9 μg m^{-3}. The enhancement of ~~9.6~~4.8 μg m^{-3} (~~23.5~~12%) induced by ARE could be mainly attributed to the contributions of VMIX, TRAN, and AERC processes, as shown in Fig. 11(c). The vertical mixing was strongly restrained by ARE, therefore fewer particles diffused from the surface to the upper layer, resulting in the accumulation of $PM_{2.5}$ in a lower atmospheric boundary layer. The changes induced by ARE in contributions of VMIX process exhibited positive values in the lower layers and negative values in the upper layers (Fig. S5(a)). Generally, the VMIX process contributed +22.5 μg m^{-3} to the enhancement in 24~~-~~h $PM_{2.5}$ increase (+4.8 μg m^{-3}) for Stage_1. The TRAN process, however, contributed -19.6 μg m^{-3}. Constrained vertical mixing due to ARE could increase aerosol precursors and water vapor in the thin boundary layer to enhance the formation of secondary particles. Generally, the AERC process contributed +1.2 μg m^{-3}. The positive contribution of AERC was mainly distributed over the high polluted regions in BTH (Fig. S5(b)). Detailedly, the average changes in concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ during the daytime from ~~10~~11:00 to ~~16~~17:00LST in Stage_1 were -0.5 μg m^{-3}, +~~5.9~~1.3 μg m^{-3}, and +~~2.9~~0.8 μg m^{-3}, respectively. The decreased near~~-~~surface temperature caused by ARE may suppress the chemical formation of SO_4^{2-}. Generally, the total contribution of VMIX, TRAN, and AERC processes to the change in 24~~-~~h $PM_{2.5}$ increase caused by ARE was +~~6.3~~4.1 μg m^{-3}, and the restrained vertical mixing could be the primary reason for near~~-~~surface $PM_{2.5}$ increase when aerosol radiative forcing was considered.

Figure 12(a) shows the vertical profiles of the 24~~-~~h increases in $PM_{2.5}$ concentrations (23:00LST minus 00:00LST) averaged over BTH during Stage_1 in CTL and NoARE cases. Below ~~400~~ 300 m (between L01 and L04), the 24~~-~~h increase simulated by CTL was larger than that in NoARE, which could be mainly explained by that the positive contributions of VMIX ~~and AERC~~ exceeded the negative contributions of TRAN in the lower atmosphere when aerosol radiative effect was considered (Fig. 12(b)). However, in the upper layers (from 3~~4~~00 to 2000 m ~~or L05 to L15~~), aerosol

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radiative forcing weakened the 24-h $PM_{2.5}$ increase during Stage_1. When aerosol radiative effect was considered, fewer particulate matters, precursors and water vapor were diffused from the surface to the upper layers, and therefore fewer particles were formed in the upper layers. Despite of the positive contributions of TRAN, the net contributions of VMIX, AERCTRAN, and TRAN AERC to $PM_{2.5}$ changes caused by ARE in the upper atmosphere were negative.

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6. Conclusions and discussions

In this study, an online coupled mesoscale meteorology-chemistry model (WRF-Chem) with an improved integrated process rate (IPR) analysis (i.e., process analysis) scheme is applied to investigate the formation and evolution mechanisms of a severe haze episode happened in the BTH region during 2016-29-29 December 2015. Sensitivity experiments are conducted to examine the contributions of local emission and outside regional transport to the absolute $PM_{2.5}$ concentrations during the haze episodeevent, while the IPR analysis is used to quantify the contributions of each detailed physical/chemical process to the variations in the $PM_{2.5}$ concentration. concentration. The impacts of aerosol radiative forcing (, including direct and indirect effects) , on meteorological parameters and $PM_{2.5}$ levels during the haze episode are also quantified, with a special focus on the detailed influence mechanism (i.e., the prominent physical or chemical processes responsible for the aerosol radiative impacts on the the haze episodeevent). An integrated comparison between observations and simulations demonstrates good performance for

The measurements from NOAA and WRMC-BSRN are used to evaluate the simulated meteorological parameters; the observations from CNEMC are used to evaluate the simulated $PM_{2.5}$ concentrations. Generally, good agreements between observations and simulations are achieved for both both meteorological and chemical variables, indicating that the WRF-Chem model has the capability to reproduce the haze episode.

Spatial-temporal evolutionss of surface layerthe near-surface $PM_{2.5}$ concentrations, and the contributions of local emission and outside regional transport to the absolute $PM_{2.5}$ concentrationthe severe haze even in BTHs, were firstly analyzed. During the aerosol accumulation stage (December 2016-22, Stage_1), the daily near-surface $PM_{2.5}$ concentrations in BTH experienced a consistent increase, with the average $PM_{2.5}$ concentrationmean value reaching of 250.0145.6 μg m^{-3}. ,

~~far beyond the threshold value of "heavily polluted".~~ The contributions of local emission and regional transport to the $PM_{2.5}$ concentration~~s averaged over BTH~~ were comparable (~~42.3~~49% and ~~36.6~~32%, respectively), meaning the combined ~~effect together~~effect resulted in the high $PM_{2.5}$ concentration in BTH~~s~~. During the aerosol dispersion stage (December 23~~–~~27, Stage_2), the average $PM_{2.5}$ concentration in BTH was ~~the near-surface $PM_{2.5}$ concentrations in BTH underwent a consistent decrease, and the average $PM_{2.5}$ concentration was~~ 107.9 μg m^{-3}. The contributions of local emission and regional transport were ~~50.9~~51% and 24~~.3~~%, respectively. Therefore, the relatively high $PM_{2.5}$ concentration during Stage_2 was principally caused by local emission. During December 28~~–~~29 (Stage_3), another haze event was formed and developed.

The IPR analysis was then used to explain the reason for $PM_{2.5}$ increase during Stage_1 and ~~$PM_{2.5}$~~ decrease during Stage_2, by quantifying the contributions of each physical/chemical process to variations in $PM_{2.5}$ concentration~~s~~. During both stages, the dominant sources ~~of surface layer $PM_{2.5}$~~ were emission (EMIS) and aerosol chemistry (AERC) ~~process~~, while the main sinks were turbulent diffusion (DIFF), advection (TRAN), and dry deposition (DRYD) ~~process~~. The $PM_{2.5}$ concentration increased by ~~50.4~~43.9 μg m^{-3} (23:00LST minus 00:00LST) during Stage_1, but it decreased by ~~41.5~~41.5 μg m^{-3} during Stage_2. Contributions of AERC, TRAN and ~~and~~ VMIX (vertical mixing, the sum of DRYD and DIFF) ~~process~~ to the 24~~–~~h $PM_{2.5}$ changes were +~~43.8~~29.6 (+17.9) μg m^{-3}, ~~–71.8 (-103.6) μg m^{-3} and -177.3 (-221.6) μg m^{-3} and –161.6 (–221.6) μg m^{-3}~~ for Stage_1 (Stage_2), respectively. Small differences in contributions from other processes were found between Stage_1 and Stage_2. Therefore, the $PM_{2.5}$ increase over BTH during the haze formation stage ~~(Stage_1)~~ was attributed to strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes.~~.~~

When aerosol radiative forcing was considered, the equivalent potential temperature was decreased in the lower layers but increased in the upper layers, leading to a more stable atmosphere. ~~As a result of ARE~~Meanwhile, the decreased PBLH and increased relative humidity were also beneficial for $PM_{2.5}$ accumulation. The daily maximum increase of the near~~–~~surface $PM_{2.5}$ concentration in BTH was 43.2 μg m^{-3}. ~~.~~

The IPR method was also used to investigate the detailed influence mechanism of aerosol radiative ~~impacts~~effects. When aerosol radiative feedback was considered, the 24~~–~~h $PM_{2.5}$ increase was enhanced by ~~9.6~~4.8 μg m^{-3} (~~23.5~~12%) during

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Stage_1, which could be mainly attributed to the contributions of VMIX (+22.5 μg m^{-3}), TRAN (-19.6 μg m^{-3}), and AERC (+1.2 μg m^{-3}) processes. Generally, the VMIX, TRAN, and AERC processes contributed +39.8 μg m^{-3}, −38.6 μg m^{-3}, and +5.1 μg m^{-3} to the enhancement in 24 h $PM_{2.5}$ increase (+9.6 μg m^{-3}), respectively. The restrained vertical mixing could be the primary reason for near-surface $PM_{2.5}$ increase when aerosol radiative forcing was considered.

There are some limitations in this work.

The uncertainty of the MIX anthropogenic emission inventory, the lack of secondary organic aerosols, and the missing mechanisms of some heterogeneous reactions may lead to theresult in large uncertainties in the final simulation results, especially the predicted aerosol chemical compositions, such as SO_4^{2-}, NO_3^- and NH_4^+. The biases in simulated concentrations of SO_4^{2-}, NO_3^- and NH_4^+ may have impacts on the contributions of AERC and CLDC processes to the air pollution variation. Uncertainties should be quantitatively analyzed in future studies. .

Besides, conclusions draw from a case study in BTH cannot represent a full view of the underlying mechanisms of haze formation and elimination. Better understanding will be attained by conducting multiple-case simulations in future.

What's more, an anomalous northeasterly induced by absorbing aerosols was observed, leading to a decrease in the near-surface $PM_{2.5}$ concentrations during December 23–24 2015 in BTH, which was different from previous studies that reported light-absorbing aerosols could worsen air quality (Li et al., 2016; Huang et al., 2018; Gao et al., 2018). More experiments should be designed in future to examine the changes in atmospheric thermal and atmospheric dynamic caused by absorbing aerosol radiative forcing and their impacts on haze episodes.

As Zheng et al. (2018) pointed out that the $PM_{2.5}$ concentration in China has been decreasing in recent years, but the decreased fine particulate matter could stimulate ozone production (Li et al., 2019a; Zhu et al., 2019). Multi-pollutant mixture may be a hot topic in the future, and the IPR analysis can be a useful method to provide a quantitative analysis about the formation mechanism of the complex air pollutions, including figuring out the major physical/chemical process behind these events. Meanwhile, significant differences between model predictions (e.g., O_3 and $PM_{2.5}$) are found among current multi-scale air quality models (Chen et al., 2019b; Li et al., 2019b), even though the same inputs are used. These different

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performances can be associated with the differences in model formulations, including parameterizations and numerical methods (Carmichael et al., 2008). In order to acquire a quantitative attribution of the cause of differences between simulation results, process analysis method should be developed and implemented in these models, and the IPR analysis will be easier to draw conclusions about the fundamental problems that cause the differences between model predictions.

Data availability

Observational datasets and simulation results are available upon request to the corresponding author (hongliao@nuist.edu.cn).

Author contributions

HL and LC conceived the study and designed the experiments. LC and JZ performed the simulations and carried out the data analysis. YG, MZ, YQ, ZL, ~~and~~ NL and YW ~~–~~provided useful comments on the paper. LC prepared the manuscript with contributions from all co~~–~~authors.

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Competing interests

The authors declare that they have no conflict of interest.

Special issue statement

This study is part of the special issue "Regional transport and transformation of air pollution in eastern China". It is not associated with a conference.

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Table 1. Parameterizations used in the WRF-Chem model

Options	WRF-Chem
Microphysics option	Purdue Lin scheme
Longwave radiation option	RRTMG scheme
Shortwave radiation option	RRTMG scheme
Surface layer option	Revised MM5 Monin-Obukhov scheme
Land surface option	Unified Noah land-surface model
Urban canopy model	Single-layer UCM scheme
Boundary layer option	YSU scheme
Cumulus option	Grell 3D ensemble scheme
Photolysis scheme	Fast-J
Dust scheme	Shao_2004
Chemistry option	CBMZ
Aerosol option	MOSAIC
Analysis nudging	On

Table 2. Experimental design

Case Description	Anthropogenic Emission	Aerosol Direct Effect	Aerosol Indirect Effect
CTL	Y	Y	Y
NoAnth	Without emission in the whole domain	Y	Y
NoBTH_Anth	Without emission in BTH	Y	Y
OnlyBTH_Anth	Only emission in BTH	Y	Y
NoARE	Y	N	N

Table 3. Statistical metrics between observations and simulations

Variables	nstd[b]	OBS[b]	SIM[b]	MB[b]	GE[b]	NMB[b]	RMSE[b]	MFB[b]	MFE[b]	IOA[b]	R[b]
T_2 (k)[a]	12	271.0	272.0	1.1	2.1	0.4	2.6	0.4	0.8	0.9	0.9
RH_2 (%)[a]	12	69.6	59.6	-10.0	14.0	-14.3	18.1	-15.2	22.6	0.7	0.8
WS_{10} (m s^{-1})[a]	12	2.4	3.1	0.7	1.4	29.1	1.8	33.3	58.0	0.7	0.8
WD_{10} (°)[a]	12	181.7	179.4	-2.3	89.4	-1.3	135.6	-4.6	59.6	0.3	0.6
$PM_{2.5}$ (μg m^{-3})	59	210.0	194.3	-15.7	79.2	-7.5	110.0	2.8	44.3	0.7	0.8

带格式表格

Variables	nstd[b]	$\overline{OBS}$[1]OBS[b]	$\overline{SIM}$[2]SIM[b]	NMB[3b]	MFB[4b]	MFE[5b]	IOA[6b]	R[7b]
T_2 (k)[a]	12	270.7	271.6	10.3	10.3	10.7	0.94	0.90
RH_2 (%)[a]	12	63.8	56.1	-12.1	-121.8	22.2	0.82	0.73
WS_{10} (m s^{-1})[a]	12	2.5	3.2	28.3	32.4	587.5	0.798	0.707
WD_{10} (°)[a]	12	190.8	192.2	10.8	-21.6	554.8	0.657	0.43
$PM_{2.5}$ (μg m^{-3})	59	173.6	168.2	-3.1	132.7	47.3	0.869	0.768

带格式表格

[a]T_2: temperature at 2 m (k); RH_2: relative humidity at 2 m (%); WS_{10}: wind speed at 10 m (m s^{-1}); WD_{10}: wind direction at 10 m (°).

[b]nstd: the number of observation sites; [1,2]$\overline{OBS}$ and $\overline{SIM}$ represent the average observations and simulations, respectively. OBS: the average observations; $\overline{OBS} = \frac{1}{nstd} \times \sum_{i=1}^{nstd} OBS_i$, SIM: the average simulations $\overline{SIM} = \frac{1}{nstd} \times \sum_{i=1}^{nstd} SIM_i$;

MB: mean bias; GE: gross error; [3]NMB is the NMB: normalized mean bias (%), $NMB = \frac{1}{nstd} \times \sum_{i=1}^{nstd} \frac{SIM_i - OBS_i}{OBS_i} \times 100\%$;

RMSE: root mean square error; [4]MFB is the MFB: mean fractional bias (%), $MFB = \frac{2}{nstd} \times \sum_{i=1}^{nstd} \frac{SIM_i - OBS_i}{SIM_i + OBS_i} \times 100\%$;

[5]MFE is the MFE: mean fractional error (%), $MFE = \frac{2}{nstd} \times \sum_{i=1}^{nstd} \frac{|SIM_i - OBS_i|}{SIM_i + OBS_i} \times 100\%$;

[6]IOA is the IOA: index of agreement, $IOA = 1 - \frac{\sum_{i=1}^{nstd}(SIM_i - OBS_i)^2}{\sum_{i=1}^{nstd}(|OBS_i - \overline{OBS}| + |SIM_i - \overline{SIM}|)^2}$;

[7]R is the R: correlation coefficient, $R = \frac{\sum_i^{nstd}|(OBS_i - \overline{OBS}) \times (SIM_i - \overline{SIM})|}{\sqrt{\sum_i^{nstd}(OBS_i - \overline{OBS})^2 + \sum_i^{nstd}(SIM_i - \overline{SIM})^2}}$;

Where OBS_i and SIM_i mean observations and model predictions, respectively. i refers to a given station, and nstd is the total number of stations.

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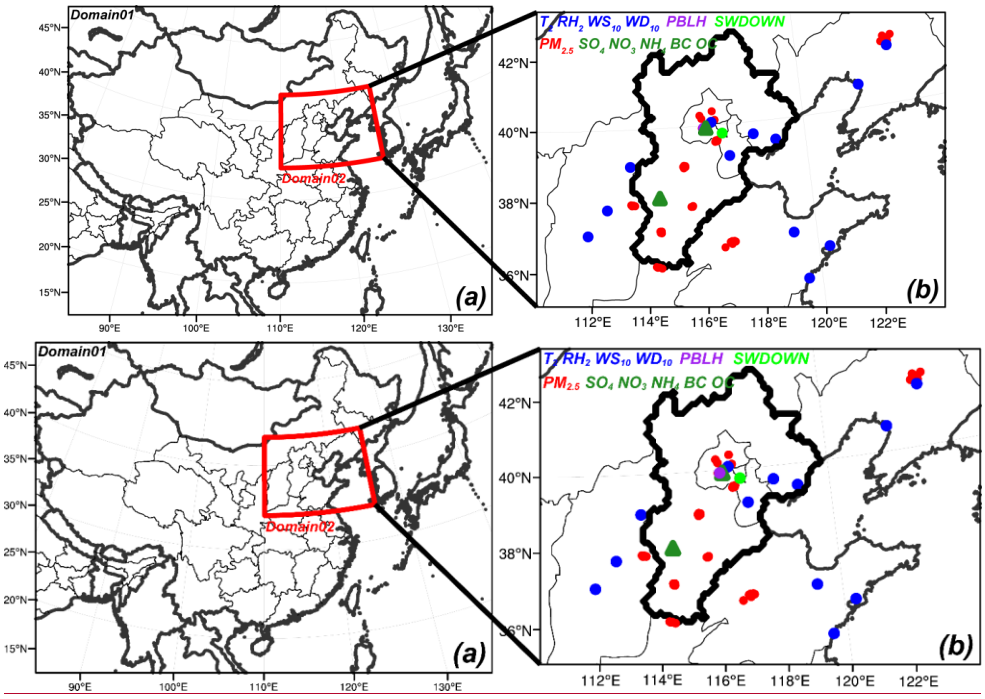
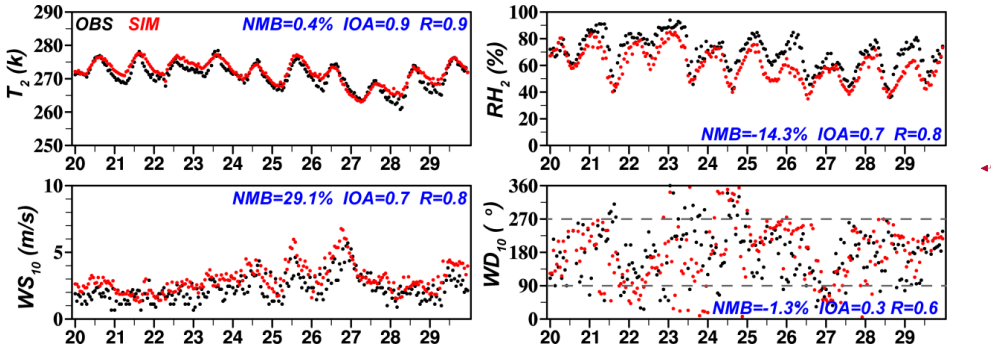
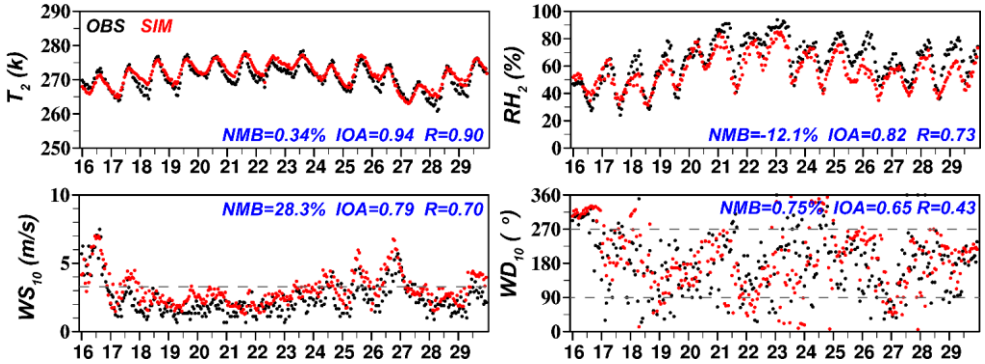


Figure 1. (a) Map of the two nested model domains. (b) Locations of the observations used for model evaluation.


OBS SIM
NMB=0.4% IOA=0.9 R=0.9
T2 (k)
RH2 (%)
NMB=-14.3% IOA=0.7 R=0.8
NMB=29.1% IOA=0.7 R=0.8
WS10 (m/s)
WD10 (°)
NMB=-1.3% IOA=0.3 R=0.6


OBS SIM
NMB=0.34% IOA=0.94 R=0.90
T2 (k)
RH2 (%)
NMB=-12.1% IOA=0.82 R=0.73
NMB=28.3% IOA=0.79 R=0.70
WS10 (m/s)
WD10 (°)
NMB=0.75% IOA=0.65 R=0.43

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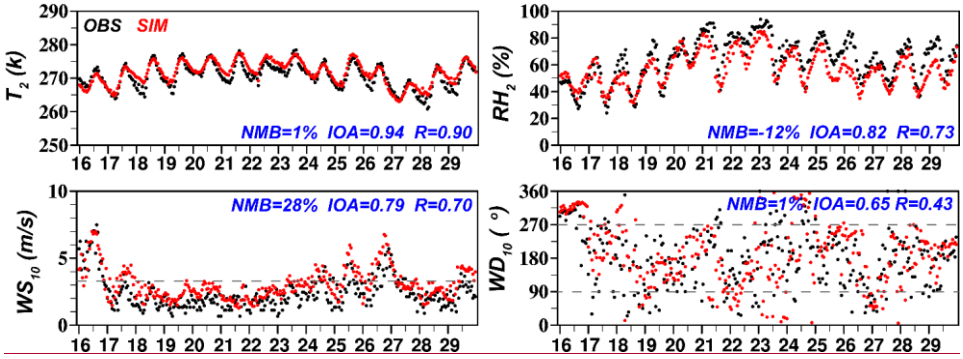


Figure 2. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly 2 m temperature (T_2, k), 2 m relative humidity (RH_2, %), 10 m wind speed (WS_{10}, m s^{-1}), and 10 m wind direction (WD_{10}, °) averaged over the 12 stations during ~~20~~16 – 29 December 2015.

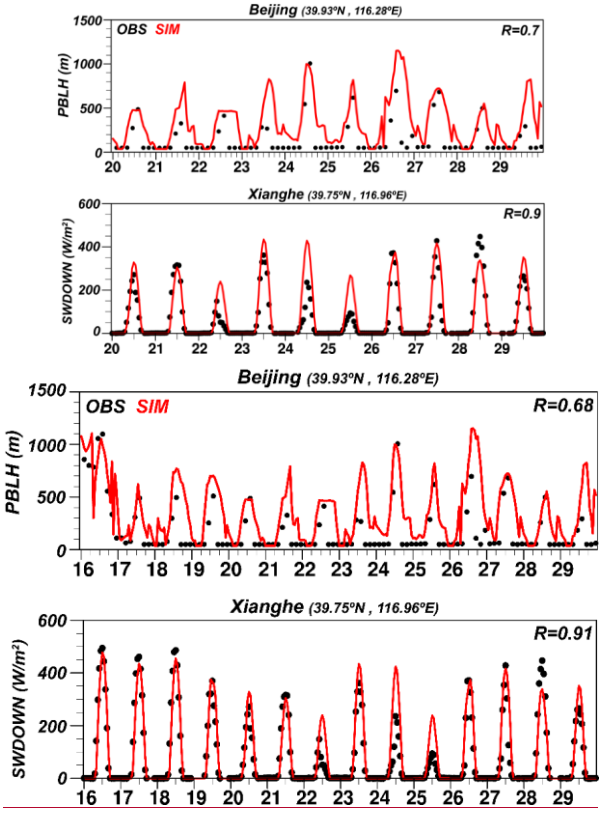


Figure 3. Time series of observed (shown in black dots) and simulated (shown in red lines) hourly planetary boundary layer height (PBLH, m) at the site of (39.93 °N, 116.28 °E) in Beijing, and shortwave downward radiation flux (SWDOWN, W m^{-2}) at the Xianghe Station (39.75 °N, 116.96 °E) from 16~~20~~ to 29 December 2015. Notably, PBLH provided by Global Data Assimilation System (GDAS)~~GDAS of NOAA~~ are in 3-hour intervals. All the time is converted to China Standard Time (Beijing Time).

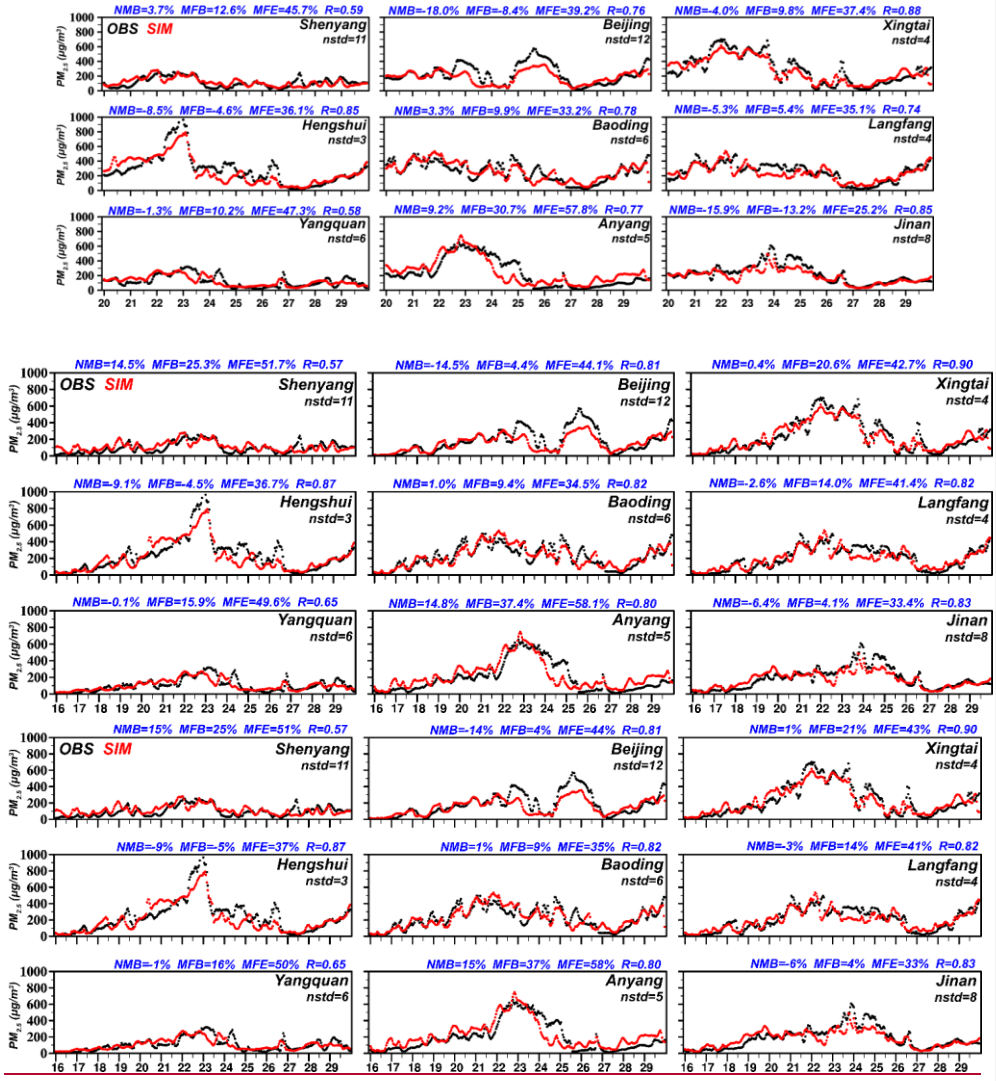

NMB=3.7% MFB=12.6% MFE=45.7% R=0.59
OBS SIM
Shenyang
nstd=11
NMB=-18.0% MFB=-8.4% MFE=39.2% R=0.76
Beijing
nstd=12
NMB=-4.0% MFB=9.8% MFE=37.4% R=0.88
Xingtai
nstd=4
NMB=-8.5% MFB=-4.6% MFE=36.1% R=0.85
Hengshui
nstd=3
NMB=3.3% MFB=9.9% MFE=33.2% R=0.78
Baoding
nstd=6
NMB=-5.3% MFB=5.4% MFE=35.1% R=0.74
Langfang
nstd=4
NMB=-1.3% MFB=10.2% MFE=47.3% R=0.58
Yangquan
nstd=6
NMB=9.2% MFB=30.7% MFE=57.8% R=0.77
Anyang
nstd=5
NMB=-15.9% MFB=-13.2% MFE=25.2% R=0.85
Jinan
nstd=8
PM2.5 (μg/m3)
20 21 22 23 24 25 26 27 28 29
NMB=14.5% MFB=25.3% MFE=51.7% R=0.57
OBS SIM
Shenyang
nstd=11
NMB=-14.5% MFB=4.4% MFE=44.1% R=0.81
Beijing
nstd=12
NMB=0.4% MFB=20.6% MFE=42.7% R=0.90
Xingtai
nstd=4
NMB=-9.1% MFB=-4.5% MFE=36.7% R=0.87
Hengshui
nstd=3
NMB=1.0% MFB=9.4% MFE=34.5% R=0.82
Baoding
nstd=6
NMB=-2.6% MFB=14.0% MFE=41.4% R=0.82
Langfang
nstd=4
NMB=-0.1% MFB=15.9% MFE=49.6% R=0.65
Yangquan
nstd=6
NMB=14.8% MFB=37.4% MFE=58.1% R=0.80
Anyang
nstd=5
NMB=-6.4% MFB=4.1% MFE=33.4% R=0.83
Jinan
nstd=8
PM2.5 (μg/m3)
16 17 18 19 20 21 22 23 24 25 26 27 28 29
NMB=15% MFB=25% MFE=51% R=0.57
OBS SIM
Shenyang
nstd=11
NMB=-14% MFB=4% MFE=44% R=0.81
Beijing
nstd=12
NMB=1% MFB=21% MFE=43% R=0.90
Xingtai
nstd=4
NMB=-9% MFB=-5% MFE=37% R=0.87
Hengshui
nstd=3
NMB=1% MFB=9% MFE=35% R=0.82
Baoding
nstd=6
NMB=-3% MFB=14% MFE=41% R=0.82
Langfang
nstd=4
NMB=-1% MFB=16% MFE=50% R=0.65
Yangquan
nstd=6
NMB=15% MFB=37% MFE=58% R=0.80
Anyang
nstd=5
NMB=-6% MFB=4% MFE=33% R=0.83
Jinan
nstd=8
PM2.5 (μg/m3)
16 17 18 19 20 21 22 23 24 25 26 27 28 29

Figure 4. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly $PM_{2.5}$ concentrations (μg m^{-3}) in the nine cities (Shengyang, Beijing, Xingtai, Hengshui, Baoding, Langfang, Yangquan, Anyang, and Jinan) from ~~20~~ 16 to 29 December 2015. The nstd in each panel represents the number of observation sites in each city. Beijing Time is used for these hourly time series.

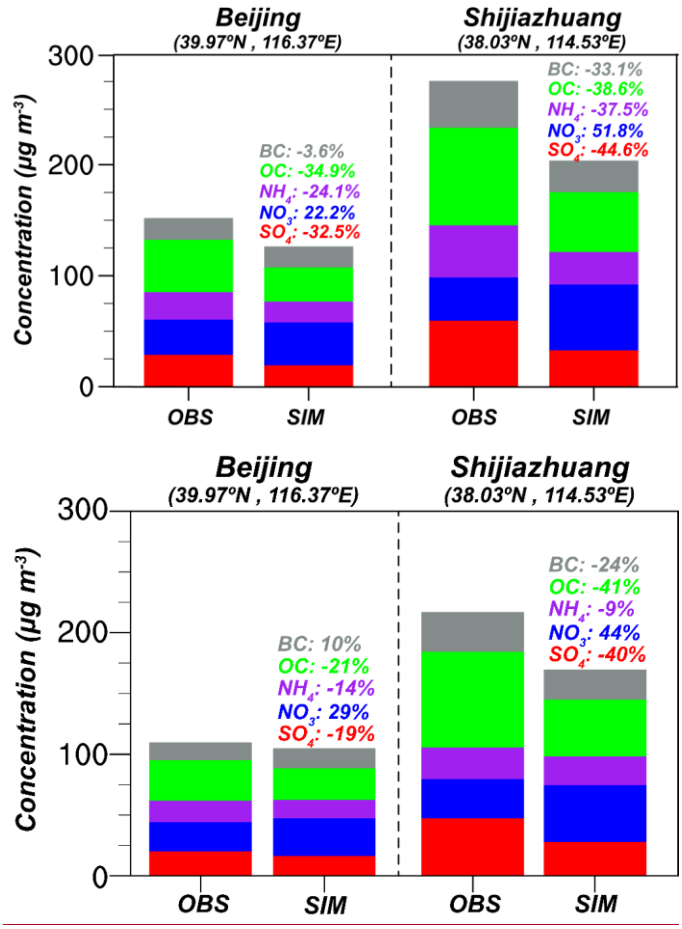
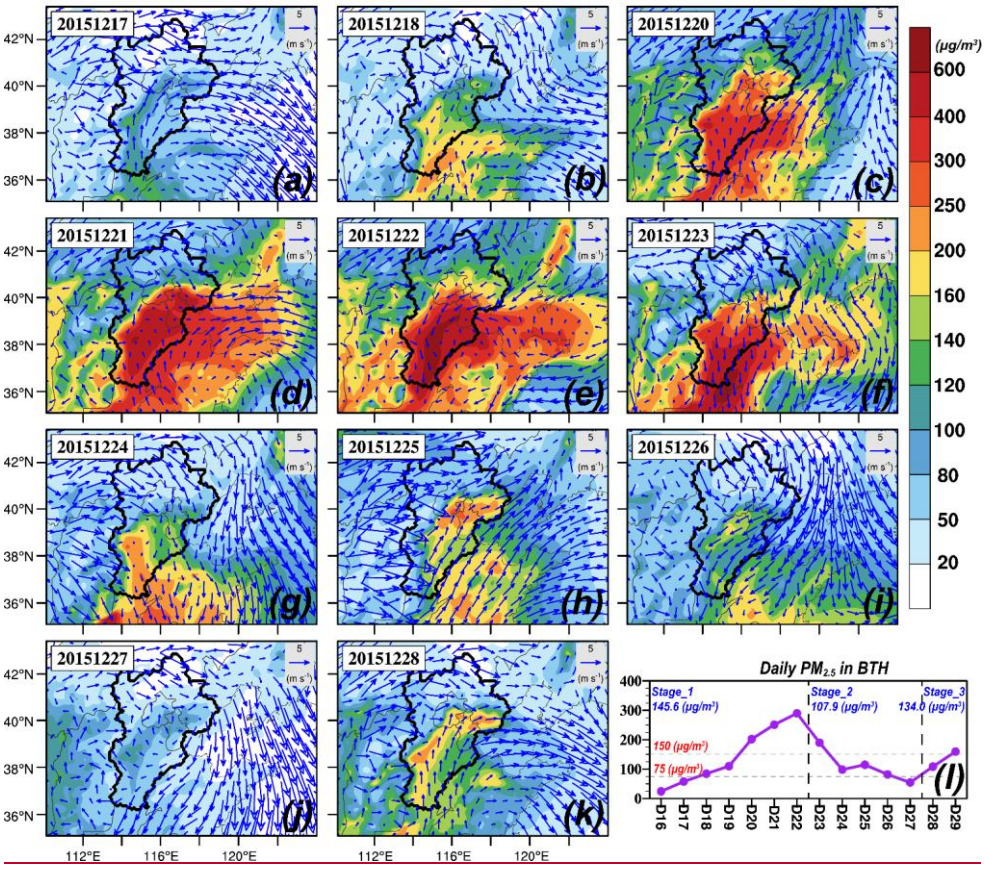


Figure 5. Comparison of observed and simulated surface–layer mass concentrations (μg m^{-3}) of SO_4^{2-} (red), NO_3^- (blue), NH_4^+ (purple), OC (green), and BC (gray) in the sites of (a) (39.97 °N, 116.37 °E) in Beijing, and (b) (38.03 °N, 114.53 °E) in Shijiazhuang averaged over ~~2016~~ 29 December 2015. Also listed in colored numbers are normalized mean biases (NMBs) for each species.


20151217
20151218
20151220
20151221
20151222
20151223
20151224
20151225
20151226
20151227
20151228
(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)
(i)
(j)
(k)
(l)
42°N
40°N
38°N
36°N
112°E
116°E
120°E
(μg/m³)
600
400
300
250
200
160
140
120
100
80
50
20
Daily PM2.5 in BTH
Stage_1
145.6 (μg/m³)
Stage_2
107.9 (μg/m³)
Stage_3
134.0 (μg/m³)
150 (μg/m³)
75 (μg/m³)
D16
D17
D18
D19
D20
D21
D22
D23
D24
D25
D26
D27
D28
D29

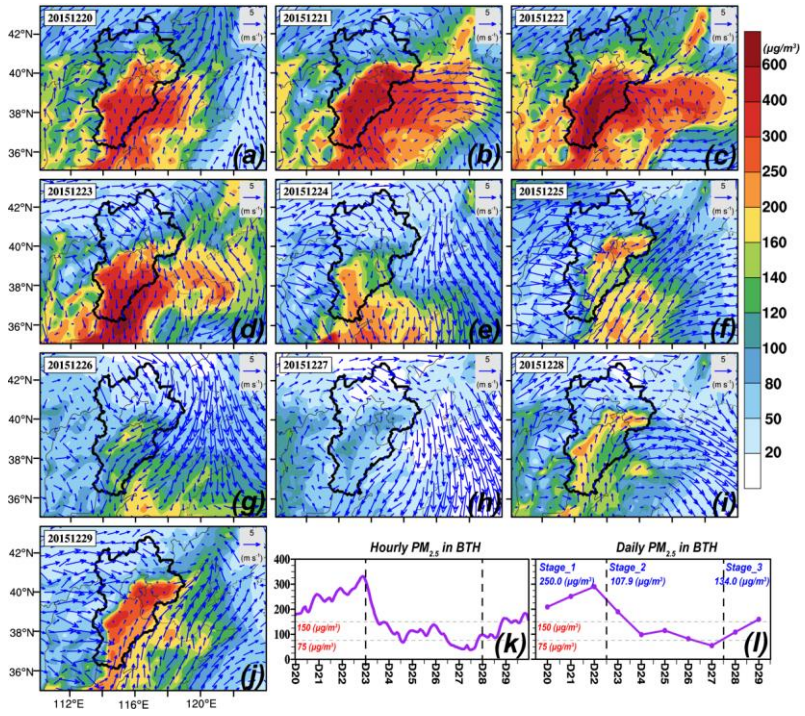
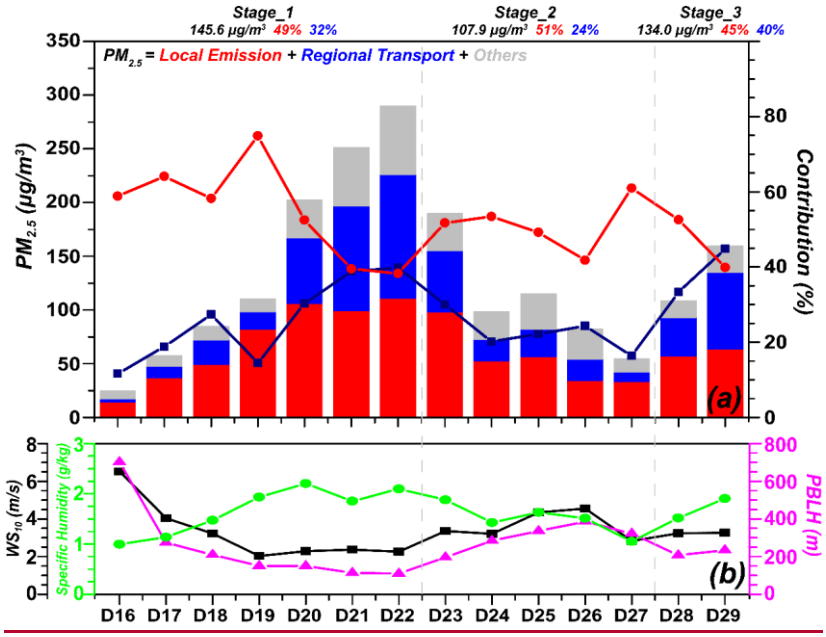
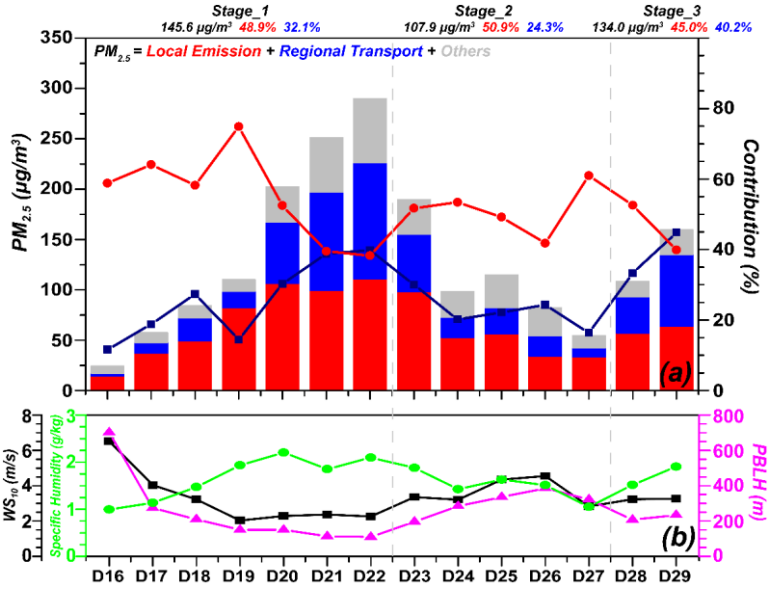


Figure 6. (a-~~k~~j) Spatial distributions of simulated daily $PM_{2.5}$ concentrations (shaded, μg m^{-3}) and wind vectors (arrows, m s^{-1}) ~~from 20 to 29 December 2015~~. Time series~~s~~ of simulated ~~hourly and~~ daily $PM_{2.5}$ concentrations averaged over the Beijing-Tianjin-Hebei region are also shown in ~~(k) and~~ (l~~), respectively.~~.


Stage_1
145.6 μg/m³ 49% 32%
Stage_2
107.9 μg/m³ 51% 24%
Stage_3
134.0 μg/m³ 45% 40%
PM2.5 = Local Emission + Regional Transport + Others
PM2.5 (μg/m³)
Contribution (%)
(a)
WS10 (m/s)
Specific Humidity (g/kg)
PBLH (m)
(b)
D16 D17 D18 D19 D20 D21 D22 D23 D24 D25 D26 D27 D28 D29


Stage_1
145.6 μg/m³ 48.9% 32.1%
Stage_2
107.9 μg/m³ 50.9% 24.3%
Stage_3
134.0 μg/m³ 45.0% 40.2%
PM2.5 = Local Emission + Regional Transport + Others
PM2.5 (μg/m³)
Contribution (%)
(a)
WS10 (m/s)
Specific Humidity (g/kg)
PBLH (m)
(b)
D16 D17 D18 D19 D20 D21 D22 D23 D24 D25 D26 D27 D28 D29

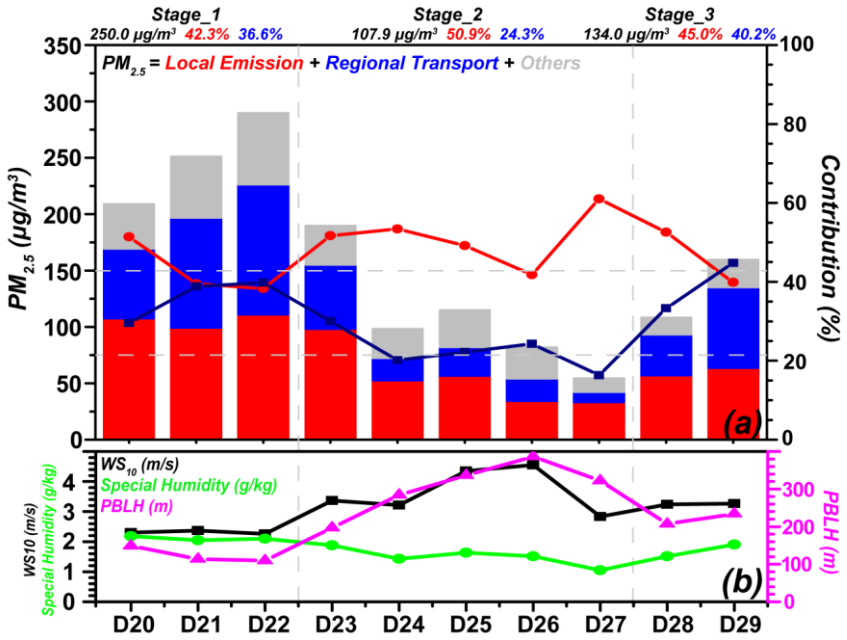
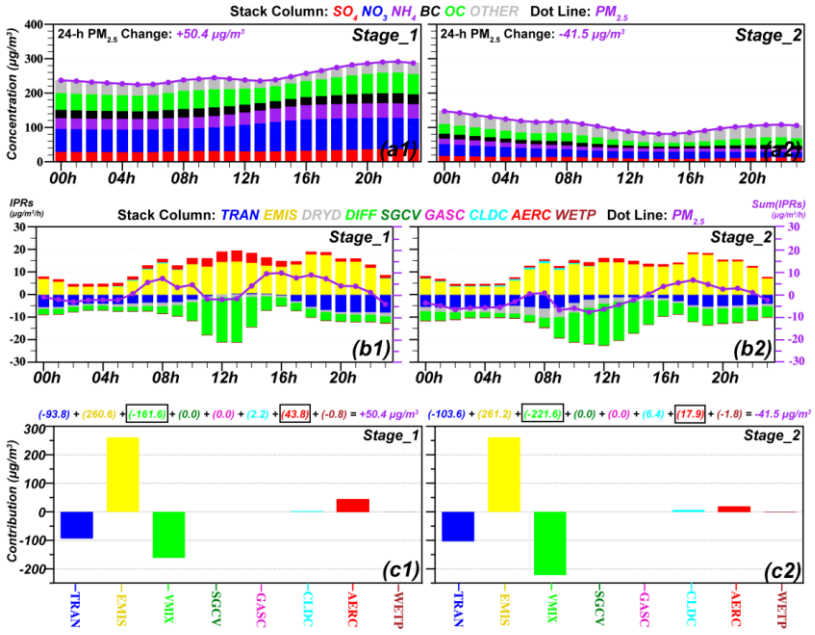


Figure 7. (a) Contributions of local emission (shown in red) and regional transport (shown in blue) to the near-surface $PM_{2.5}$ concentrations averaged over the Beijing-Tianjin-Hebei region from ~~20~~ 16 to 29 December 2015. The absolute contributions (μg m^{-3}) are shown in bars, ~~(μg m^{-3})~~and the percentage contributions (%) are shown in lines ~~(%)~~. The $PM_{2.5}$ concentration and the percentage contributions averaged over each stage are listed at the top of (a). Simulated daily 10 m wind speed (WS_{10}, m s^{-1}, shown in black dot line), ~~special~~ specific humidity (g kg^{-1}, shown in green dot line), and PBLH (m, shown in purple dot line) averaged over Beijing-Tianjin-Hebei are also shown in (b).


Stack Column: SO4 NO3 NH4 BC OC OTHER Dot Line: PM2.5
24-h PM2.5 Change: +50.4 μg/m³
Stage_1
24-h PM2.5 Change: -41.5 μg/m³
Stage_2
Concentration (μg/m³)
(a1)
(a2)
00h 04h 08h 12h 16h 20h
IPRs (μg/m³/h)
Stack Column: TRAN EMIS DRYD DIFF SGCV GASC CLDC AERC WETP Dot Line: PM2.5
Sum(IPRs) (μg/m³/h)
(b1)
(b2)
(-93.8) + (260.6) + (-161.6) + (0.0) + (0.0) + (2.2) + (43.8) + (-0.8) = +50.4 μg/m³
(-103.6) + (261.2) + (-221.6) + (0.0) + (0.0) + (6.4) + (17.9) + (-1.8) = -41.5 μg/m³
Contribution (μg/m³)
(c1)
(c2)
TRAN EMIS VMIX SGCV GASC CLDC AERC WETP

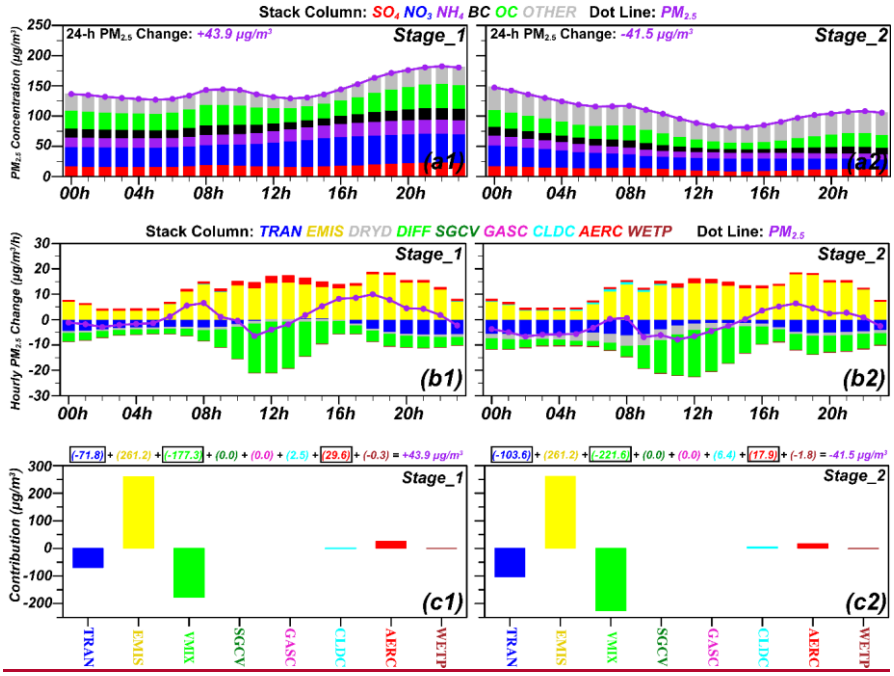


Figure 8. (a1–a2) Diurnal variations of $PM_{2.5}$ concentrations averaged over Beijing-Tianjin-Hebei during Stage_1 and Stage_2 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is the 24-h change in $PM_{2.5}$ concentration (23:00LST minus 00:00LST). (b1–b2) The hourly $PM_{2.5}$ changes induced by each physical/chemical process by using the IPR analysis method (shown by colored bars). The purple dot lines represent hourly $PM_{2.5}$ changes induced by all processes, also indicating the differences between current and previous-hour $PM_{2.5}$ concentrations. (c1–c2) Contributions of each physical/chemical process to 24-h $PM_{2.5}$ changes.

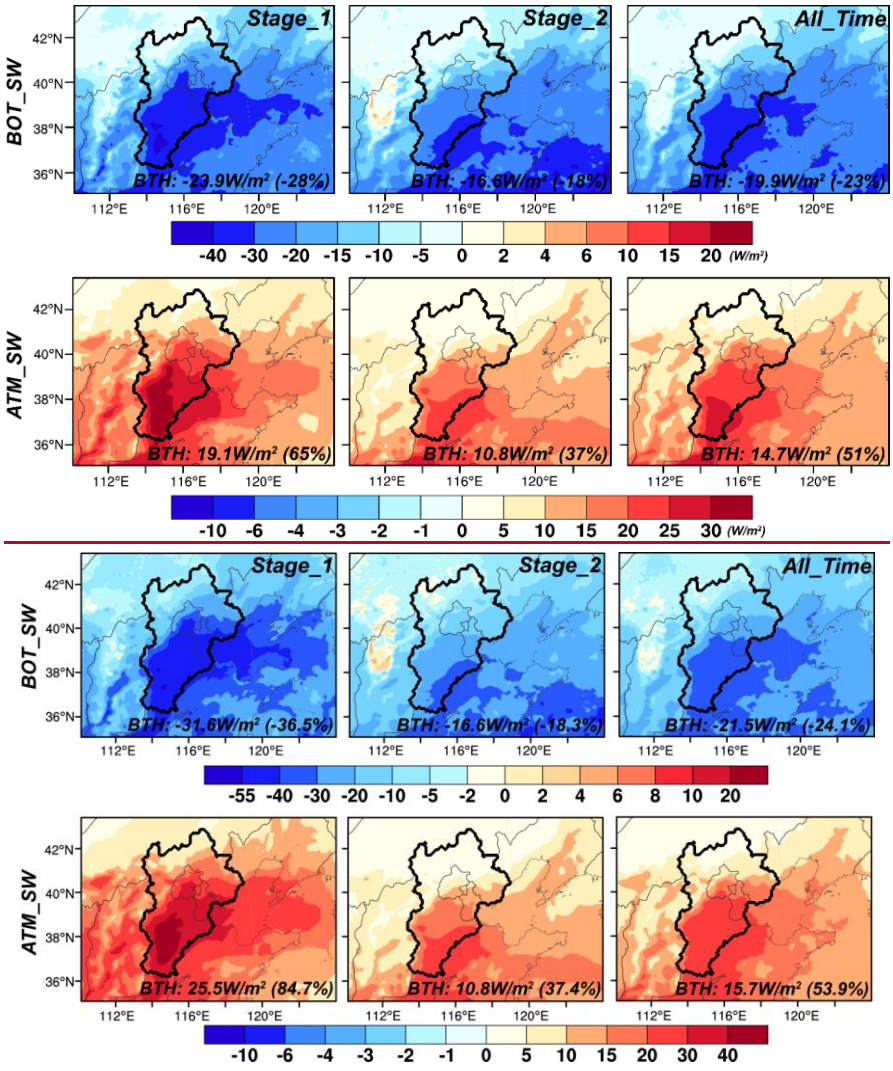
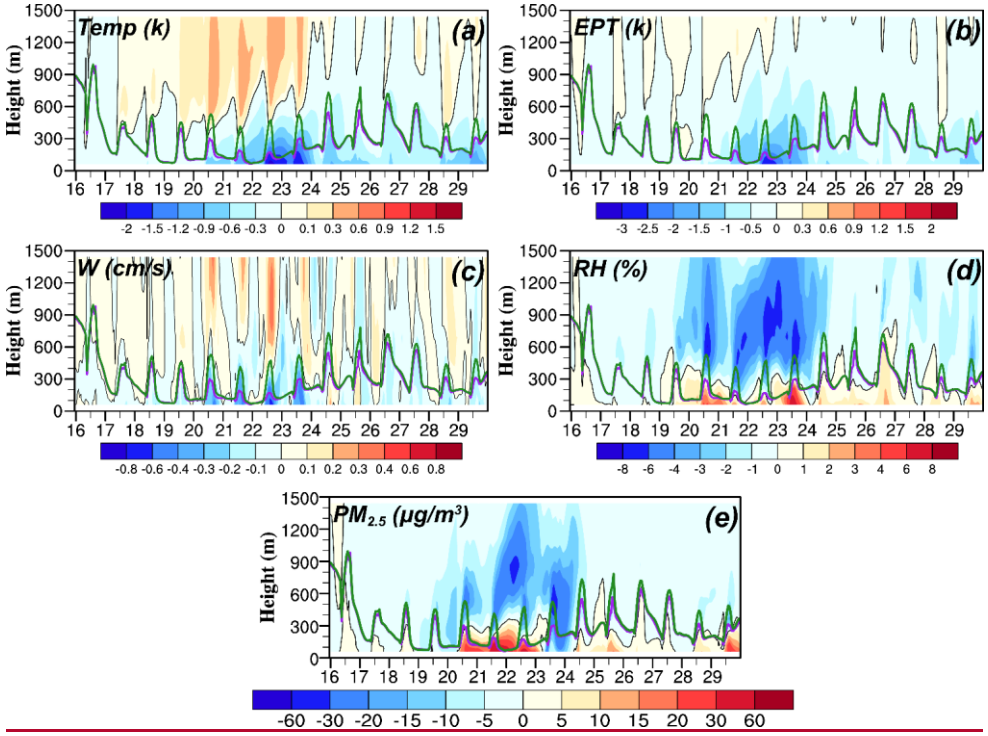

Stage_1
Stage_2
All_Time
BOT_SW
ATM_SW
BTH: -23.9W/m² (-28%)
BTH: -16.6W/m² (-18%)
BTH: -19.9W/m² (-23%)
BTH: 19.1W/m² (65%)
BTH: 10.8W/m² (37%)
BTH: 14.7W/m² (51%)
BTH: -31.6W/m² (-36.5%)
BTH: -16.6W/m² (-18.3%)
BTH: -21.5W/m² (-24.1%)
BTH: 25.5W/m² (84.7%)
BTH: 10.8W/m² (37.4%)
BTH: 15.7W/m² (53.9%)
42°N
40°N
38°N
36°N
112°E
116°E
120°E
-40 -30 -20 -15 -10 -5 0 2 4 6 10 15 20 (W/m²)
-10 -6 -4 -3 -2 -1 0 5 10 15 20 25 30 (W/m²)
-55 -40 -30 -20 -10 -5 -2 0 2 4 6 8 10 20
-10 -6 -4 -3 -2 -1 0 5 10 15 20 30 40

Figure 9. The differences in simulated all-sky radiative forcing (W m^{-2}) between CTL and NoARE cases (CTL minus NoARE) averaged over Stage_1, Stage_2, and the whole simulation period. "BOT_SW" and "ATM_SW" denote the downward shortwave radiative flux at the surface and in the atmosphere, respectively. The calculated differences in the simulated radiative forcing averaged over Beijing-Tianjin-Hebei for each stage are also shown at the bottom of each panel.


Temp (k)
(a)
EPT (k)
(b)
W (cm/s)
(c)
RH (%)
(d)
PM2.5 (μg/m3)
(e)
Height (m)
1500
1200
900
600
300
0
16 17 18 19 20 21 22 23 24 25 26 27 28 29
-2 -1.5 -1.2 -0.9 -0.6 -0.3 0 0.1 0.3 0.6 0.9 1.2 1.5
-3 -2.5 -2 -1.5 -1 -0.5 0 0.3 0.6 0.9 1.2 1.5 2
-0.8 -0.6 -0.4 -0.3 -0.2 -0.1 0 0.1 0.2 0.3 0.4 0.6 0.8
-8 -6 -4 -3 -2 -1 0 1 2 3 4 6 8
-60 -30 -20 -15 -10 -5 0 5 10 15 20 30 60

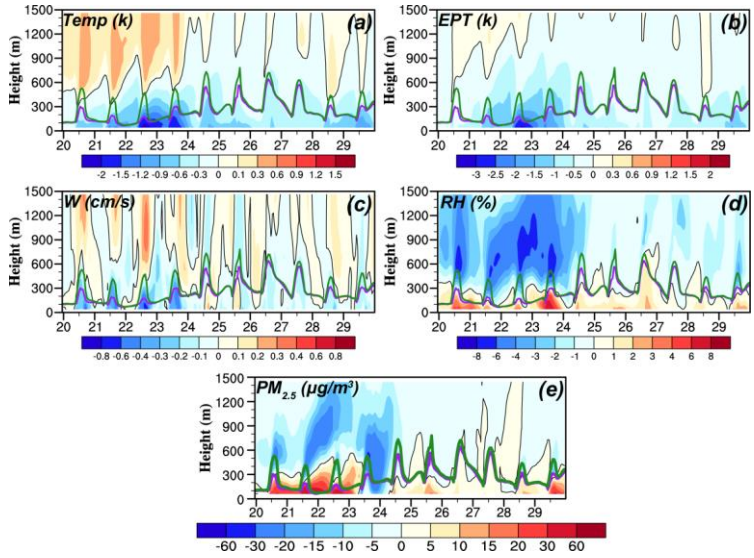
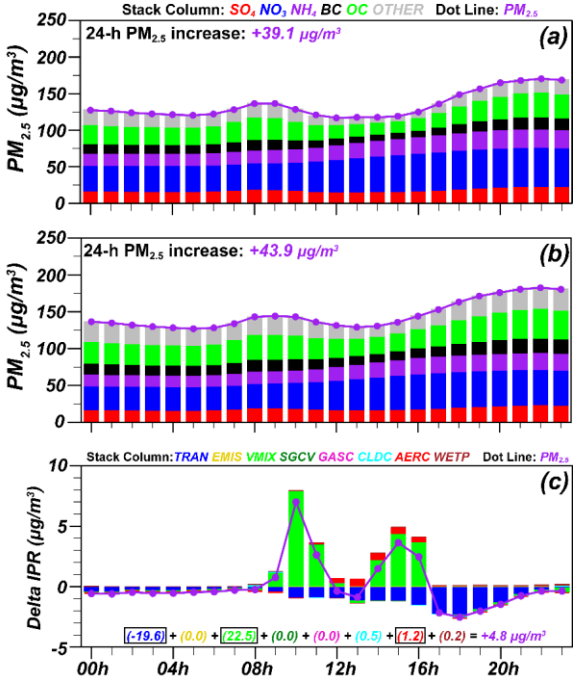


Figure 10. Time series of differences in (a) temperature (k), (b) equivalent potential temperature (k), (c) vertical wind speed (cm s^{-1}), (d) relative humidity (%), and (e) $PM_{2.5}$ concentration (μg m^{-3}) between CTL and NoARE cases (CTL minus NoARE) averaged over the Beijing-Tianjin-Hebei region. The purple and green lines denote the simulated PBLH in CTL and NoARE cases, respectively. The black line represents the zero contour line.


Stack Column: SO4 NO3 NH4 BC OC OTHER Dot Line: PM2.5
24-h PM2.5 increase: +39.1 µg/m³
(a)
PM2.5 (µg/m³)
250
200
150
100
50
0
24-h PM2.5 increase: +43.9 µg/m³
(b)
Stack Column: TRAN EMIS VMIX SGCV GASC CLDC AERC WETP Dot Line: PM2.5
(c)
Delta IPR (µg/m³)
10
5
0
-5
(-19.6) + (0.0) + (22.5) + (0.0) + (0.0) + (0.5) + (1.2) + (0.2) = +4.8 µg/m³
00h
04h
08h
12h
16h
20h

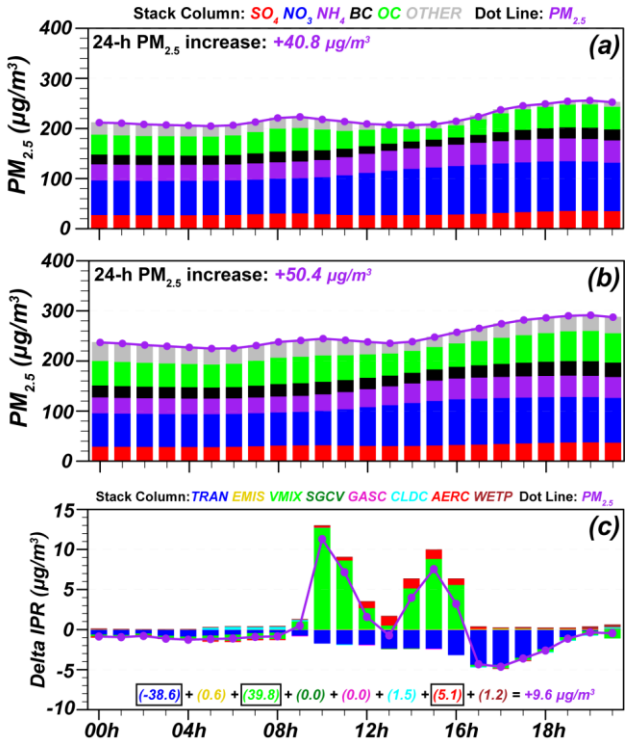
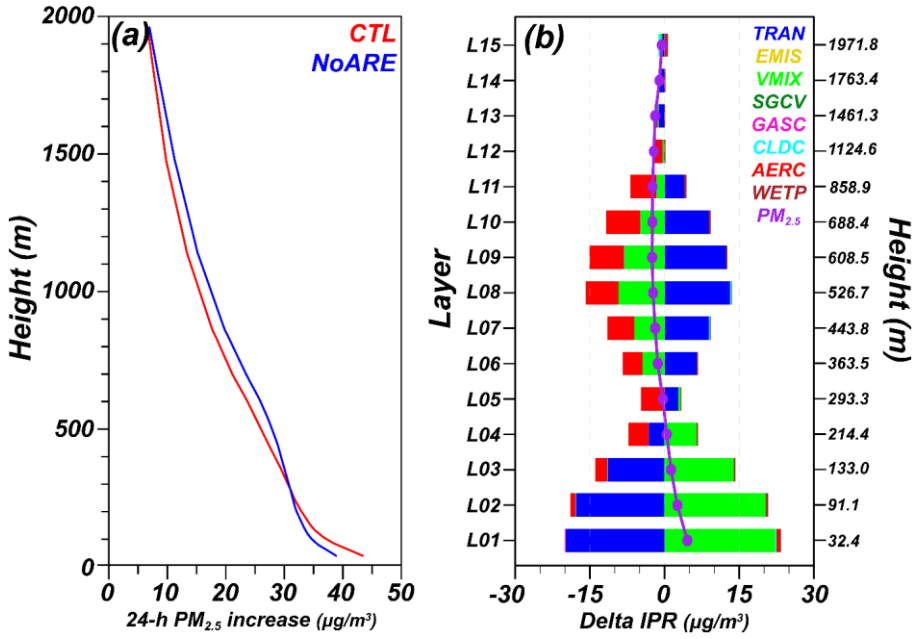


Figure 11. Diurnal variations of the near-surface $PM_{2.5}$ concentrations in (a) NoARE and (b) CTL simulations averaged over the Beijing-Tianjin-Hebei region during Stage_1 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is the 24-h increase in $PM_{2.5}$ concentration (23:00LST minus 00:00LST). (c) Differences in hourly IPRs caused by aerosol radiative forcing (CTL minus NoARE). The numbers listed in (c) represent the contributions of each process to the change in 24-h $PM_{2.5}$ increase caused by aerosol radiative forcing.


(a)
CTL
NoARE
Height (m)
2000
1500
1000
500
0
0
10
20
30
40
50
24-h PM2.5 increase (μg/m³)
(b)
TRAN
EMIS
VMIX
SGCV
GASC
CLDC
AERC
WETP
PM2.5
Layer
L15
L14
L13
L12
L11
L10
L09
L08
L07
L06
L05
L04
L03
L02
L01
1971.8
1763.4
1461.3
1124.6
858.9
688.4
608.5
526.7
443.8
363.5
293.3
214.4
133.0
91.1
32.4
Height (m)
-30
-15
0
15
30
Delta IPR (μg/m³)

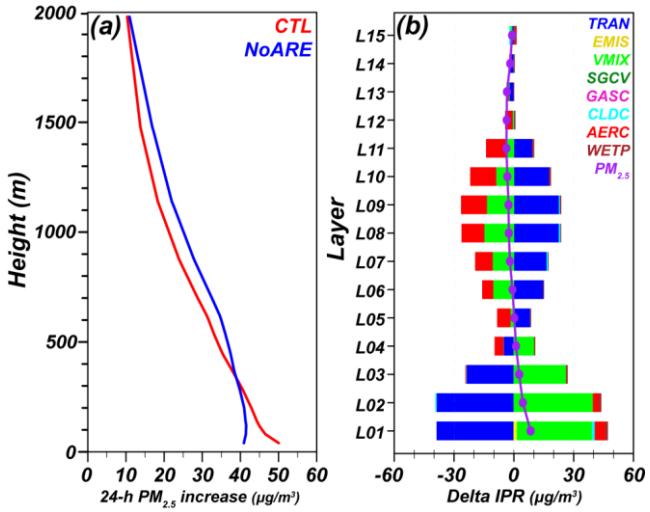


Figure 12. (a) Vertical profiles of the 24-h increases in $PM_{2.5}$ concentrations (23:00LST minus 00:00LST) averaged over Beijing-Tianjin-Hebei during Stage_1 in CTL and NoARE cases. (b) Vertical profiles of the differences in the 24-h $PM_{2.5}$ increases caused by aerosol radiative effect (CTL minus NoARE, as show in purple dot line), and the contributions of each physical/chemical process (as shown in colored bars).

Final Revised Manuscript:

Assessing the formation and evolution mechanisms of severe haze pollution in Beijing-Tianjin-Hebei region by using process analysis

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Abstract. Fine-particle pollution associated with haze threatens human health, especially in the North China Plain, where extremely high $PM_{2.5}$ concentrations were frequently observed during winter. In this study, the WRF-Chem model coupled with an improved integrated process analysis scheme was used to investigate the formation and evolution mechanisms of a haze event over Beijing-Tianjin-Hebei (BTH) in December 2015, including examining the contributions of local emission and regional transport to the $PM_{2.5}$ concentration in BTH, and the contributions of each detailed physical or chemical process to the variations in the $PM_{2.5}$ concentration. The influence mechanisms of aerosol radiative forcing (including aerosol direct and indirect effects) were also examined by using the process analysis. During the aerosol accumulation stage (December

16-22, Stage_1), the near-surface $PM_{2.5}$ concentration in BTH was increased from 24.2 μg m^{-3} to 289.8 μg m^{-3}, with the contributions of regional transport increased from 12% to 40%, while the contributions of local emission were decreased from 59% to 38%. During the aerosol dispersion stage (December 23-27, Stage_2), the average concentration of $PM_{2.5}$ was 107.9 μg m^{-3}, which was contributed by local emission of 51% and regional transport of 24%. The 24-h change (23:00LST minus 00:00LST) in the near-surface $PM_{2.5}$ concentration was +43.9 μg m^{-3} during Stage_1 and -41.5 μg m^{-3} during Stage_2. Contributions of aerosol chemistry, advection and vertical mixing to the 24-h change were +29.6 (+17.9) μg m^{-3}, -71.8 (-103.6) μg m^{-3} and -177.3 (-221.6) μg m^{-3} during Stage_1 (Stage_2), respectively. Small differences in contributions of other processes were found between Stage_1 and Stage_2. Therefore, the $PM_{2.5}$ increase over BTH during haze formation stage was mainly attributed to the strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes. When aerosol radiative feedback was considered, the 24-h $PM_{2.5}$ increase was enhanced by 4.8 μg m^{-3} during Stage_1, which could be mainly attributed to the contributions of vertical mixing process (+22.5 μg m^{-3}), advection process (-19.6 μg m^{-3}) and aerosol chemistry process (+1.2 μg m^{-3}). The restrained vertical mixing was the primary reason for the enhancement in near-surface $PM_{2.5}$ increase when aerosol radiative forcing was considered.

1 Introduction

Anthropogenic activities associated with rapidly developed industrialization and urbanization have been leading to a sustained increase in the amounts of atmospheric pollutants, especially in the fast-developing countries (IPCC, 2013). As one of the largest emission sources of aerosols and their precursors, China has been suffering from serious air pollution for years (Lei et al., 2011; Li et al., 2011; Liu et al., 2018a), with severe haze events frequently occurring in winter, especially over large urban agglomerations, such as the North China Plain (NCP) (Han et al., 2014; Gao et al., 2015), the Yangtze River Delta area (YRD) (Ding et al., 2016; Wang et al., 2016a), and the Sichuan Basin (SCB) (Zhao et al., 2018; Zhang et al., 2019). During severe haze events, the observed maximum hourly surface-layer $PM_{2.5}$ (fine particulate matter with aerodynamic diameter of 2.5 μm or less) concentration exceeded 1000 μg m^{-3} (Wang et al., 2013b; Sun et al., 2016; Li et al., 2017a), which could significantly influence visibility (Li et al., 2014), radiation budget (Steiner et al., 2013), atmospheric circulation (Jiang et al., 2017), cloud properties (Unger et al., 2009), and human health (Hu et al., 2014; Guo et al., 2017).

Extensive studies have been carried out in recent years to analyze the formation mechanisms of haze episodes in China. Wang et al. (2013a) used a synergy of ground-based observations, satellite, and lidar measurements to study a long-lasting and severe haze episode occurred in eastern China in January 2013, and concluded that stagnant meteorological conditions, which could be generally characterized by weak wind speed, high relative humidity, intense inversion, and low mixing layer height, were tightly associated with severe haze episodes. Based on National Center for Environmental Prediction (NCEP) reanalysis data, Shu et al. (2017) identified five typical synoptic patterns, and pointed out that each synoptic pattern exerted different impacts on particle pollution over YRD. By analyzing the simulation results from a large ensemble climate model (MIROC5), Li et al. (2018a) investigated the contributions of anthropogenic influence to severe haze events happened over eastern China in January 2013 and December 2015, and found that anthropogenic forcing (i.e., increased emissions of greenhouse gases) could modify atmospheric circulation pattern, and these human-induced circulation changes were conducive to the occurrence of severe haze events. Zhang et al. (2015a) used a global 3-D chemical transport model (GEOS-Chem) to quantify the local source contributions to wintertime surface-layer $PM_{2.5}$ concentrations over North China from 2013 to 2015, and reported that emissions from residential and industrial sources and transportation contributed most to the high concentrations of atmospheric aerosols in Beijing. Many studies reported that regional transport of aerosols also

played an important role in haze episodes (Wang et al., 2013b; Jiang et al., 2015; Li et al., 2018b). Wang et al. (2013b) reported that the cross-city clusters transport outside BTH (Beijing, Tianjin, and Hebei) and transport among cities inside BTH contributed 20%-35% and 26%-35% of $PM_{2.5}$ concentrations over BTH, respectively. Secondary aerosol formation and their hygroscopic growth were also confirmed to be a large contributor to severe haze episodes (Huang et al., 2014b; Han et al., 2015; Chen et al., 2019a). The conversion of SO_2 to SO_4^{2-} was strongly associated with high relative humidity, and NO_3^- was found to be produced mainly by photochemical and heterogeneous reactions (Chen et al., 2016; Zhang et al., 2018a).

It is well known that aerosols can scatter and absorb solar radiation to alter the radiative balance of the atmosphere and surface (direct radiative effect), and can serve as cloud condensation nuclei or ice nuclei to affect cloud properties (indirect radiative effect) (Twomey, 1974). These impacts are coupled with atmospheric dynamics to produce a chain of interactions with a large range of meteorological variables that influence both weather and climate (Ramanathan et al., 2001; Huang et al., 2006; Li et al., 2017c; Yang et al., 2017), which will further induce feedbacks on aerosol production, accumulation, and even severe haze pollutions (Petaja et al., 2016; Li et al., 2017d; Zhao et al., 2017; Gao et al., 2018; Lou et al., 2019). Based on multi-year measurements (from 2010 to 2016), Huang et al. (2018) found that aerosol radiative effects led to a significant heating in the upper planetary boundary layer (PBL) and a substantial dimming at the surface over North China. This is because high concentrations of light-absorbing aerosols were observed, and the aerosol-meteorology interactions depressed the development of PBL, and therefore aggravated the haze pollution (Su et al., 2018). The light-absorbing aerosols can also amplify haze in NCP by weakening East Asian winter monsoon wind speeds through ocean and cloud feedbacks (Lou et al., 2019). By using the WRF-Chem model, Gao et al. (2015) analyzed the feedbacks between aerosols and meteorological fields over NCP in January 2013, and found that aerosols caused a significant negative (positive) radiative forcing at the surface (in the atmosphere), resulting in a weaker surface-layer wind speed and lower PBL height (PBLH). The average surface-layer $PM_{2.5}$ concentration was increased by 10-50 $\mu g\ m^{-3}$ as a result of the more stable atmosphere. By analyzing the observations from a comprehensive field experiment and simulation results from WRF-Chem model, Liu et al. (2018b) concluded that the decreased PBLH associated with increased aerosol concentrations could enhance surface-layer relative humidity by weakening the vertical transport of water vapor, and the increased relative humidity at the surface accelerated the formation

of secondary particulate matters through heterogeneous reactions, leading to the increase of the $PM_{2.5}$ concentration by 63 μg m^{-3} averaged over the NCP during 15-21 December, 2016.

All these studies discussed above revealed that the formation of haze episode was caused by the synergy impacts of local emissions, regional transport, meteorological conditions, and chemical production. Nevertheless, only the net combined effects on the concentrations of pollutants were provided, without the capabilities of understanding and isolating the atmospheric physical and chemical processes involved. The quantitative assessment of the contributions from each detailed physical/chemical process (e.g., vertical mixing process, advection process, emission source process, aerosol chemistry process, cloud chemistry process) is necessary for fully understanding of the formation and evolution mechanisms of haze episodes (Goncalves et al., 2009; Xing et al., 2017; Kang et al., 2019). What's more, although many previous studies have identified the positive feedback effects of aerosol radiative forcing on particulate accumulation, the detailed influence mechanisms of the forcing-response relationship at each process chain remain largely elusive (i.e., the prominent physical or chemical processes responsible for the aerosol radiative impacts on haze episodes). Since 2013, substantial efforts have been taken to improve air quality in China, including emission reduction and energy transition. However, haze events continued to occur frequently all over the country. For example, a severe, long-lasting, and wide-ranging haze episode was observed in December 2015 over the central and eastern China, with the regional average $PM_{2.5}$ concentration exceeding 150 μg m^{-3}. For BTH, a red alert for haze (the most serious level) was issued for the period from 20 to 22 December 2015, with the maximum hourly $PM_{2.5}$ concentration exceeding 1000 μg m^{-3}. The formation and evolution mechanisms, and the aerosol radiative feedbacks of this severe haze episode have not been fully estimated yet.

In this study, we develop an improved online integrated process rate (IPR) analysis scheme (i.e., process analysis) in the fully coupled online Weather Research and Forecasting-Chemistry (WRF-Chem) model, to investigate the formation and evolution mechanisms of the severe haze episode happened over NCP from 16 to 29 December 2015. Sensitivity experiments are conducted to examine the contributions of local emission and regional transport to the $PM_{2.5}$ concentrations during the haze episode, while the IPR analysis is used to quantify the contributions of each detailed physical/chemical process to the variations in the $PM_{2.5}$ concentrations. The effects of aerosol radiative forcing, including direct and indirect effects, on meteorological parameters and $PM_{2.5}$ levels during the haze episode are also quantified, with a special focus on

the detailed influence mechanism. We hope that the results concluded in this study may provide better understanding of the formation mechanisms for severe haze events, and help policy makers take targeted measures to improve air quality over North China.

This manuscript is arranged as follows. Model configuration, integrated process rate (IPR) analysis (i.e., process analysis), numerical experiments, and observations are presented in Section 2. Model evaluation is conducted in Section 3. The formation and evolution mechanisms of the haze episode are investigated in Section 4. Section 5 provides the impacts of aerosol radiative forcing. Summaries and discussions are presented in Section 6.

2 Methods

2.1 Model configuration

A fully coupled online Weather Research and Forecasting-Chemistry model (WRF-Chem v3.7) is used to simulate meteorological fields and concentrations of gases and aerosols simultaneously (Skamarock et al., 2008; Grell et al., 2005). The WRF-Chem model is designed with two domains using 219 (west-east) × 159 (south-north) and 150 (west-east) × 111 (south-north) grid points at the horizontal resolutions of 27 and 9 km, respectively (Fig. 1). The outer domain covers nearly the whole East Asia, and the inner domain is located in the NCP. In order to minimize the impacts from IBCs (lateral boundary conditions), we only analyze the simulation results from the inner region of the second domain (i.e., BTH), following Chen et al. (2018) and Wu et al. (2012). The vertical dimension is resolved by 29 full sigma levels, with 15 layers located in the lowest 2 km for finer resolution in the planetary boundary layer, and the height of the first layer averaged in BTH is about 30 m.

Meteorological initial and lateral boundary conditions used in the WRF-Chem model are taken from the NCEP (National Center for Environmental Prediction) Final Operational Global Analysis data with the spatial resolution of 1 ° × 1 °. Four-dimensional data assimilation (FDDA) with the nudging coefficient of 3.0×10^{-4} for wind (in and above PBL), temperature (above PBL) and water vapor mixing ratio (above PBL) is adopted to improve the accuracy of simulation results (no analysis nudging is included for the inner domain) (Lo et al., 2008; Otte, 2008; Wang et al., 2016b; Werner et al., 2016).

The forecasts from the global chemical transport model MOZART-4 are processed to provide the chemical initial and boundary conditions for the WRF-Chem model (Emmons et al., 2010).

Anthropogenic emission data are obtained from the MIX Asian emission inventory (http://www.meicmodel.org/dataset-mix.html), with a horizontal resolution of 0.25 degree (Li et al., 2017b). It is developed to support the MICS-Asia III (Model Inter-Comparison Study for Asia Phase III) and the TF HTAP (Task Force on Hemispheric Transport of Air Pollution) projects. This inventory includes SO_2 (sulfur dioxide), NO_x (nitrogen oxides), CO (carbon monoxide), CO_2 (carbon dioxide), NMVOC (non-methane volatile organic compounds), NH_3 (ammonia), BC (black carbon), OC (organic carbon), $PM_{2.5}$ and PM_{10}. All these species are from several sectors, such as agriculture, industry, power, transportation and residential, and the emission rate of each species for each hour is based on Gao et al. (2015). The biogenic emissions are calculated online using the MEGANv2.04 (Model of Emission of Gases and Aerosol from Nature v2.04) model (Guenther, 2006). Biomass-burning emissions are obtained from the GFEDv3 (Global Fire Emissions Database v3) (Randerson et al., 2005). Dust emissions and sea salt emissions are calculated online by using algorithms proposed by Shao (2004) and Gong et al. (1997), respectively.

The Carbon-Bond Mechanism version Z (CBMZ) (Zaveri and Peters, 1999) is selected to simulate the gas phase chemistry, and the 8-bin sectional aerosol module, MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) (Zaveri et al., 2008), with some aqueous chemistry, is used to simulate aerosol evolution. All major aerosol species are considered in the MOSAIC scheme, including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), chloride (Cl), sodium (Na), BC, primary organic mass, liquid water, and other inorganic mass (Zaveri et al., 2008). The aerosol size distribution is divided into discrete size bins defined by their lower and upper dry particle diameters (Zhao et al., 2010). In the current CBMZ/MOSAIC scheme, the formation of SOA (secondary organic aerosol) is not included (Zhang et al., 2012; Gao et al., 2016). Aerosol optical properties, including extinction efficiency, single scatter albedo, and asymmetry factor are computed by Mie theory, based on aerosol composition, mixing state, and size distribution (Barnard et al., 2010). The impacts of aerosols on photolysis rates are calculated using the Fast-J photolysis scheme (Wild et al., 2010). Aerosol radiation is simulated by RRTMG (Rapid Radiative Transfer Model for GCMs) for both shortwave (SW) and longwave (LW) radiation (Zhao et al., 2011). More information about the parameterizations used in this study can be found in Table 1.

2.2 Integrated process rate (IPR) analysis

Most air quality models are configured to output only the pollutant concentrations that reflect the combined effects of all physical and chemical processes. Quantitative information of the impacts of individual process is usually unavailable. Process analysis techniques (i.e., integrated process rate (IPR) analysis) can be used in grid-based Eulerian models (e.g., WRF-Chem) to obtain contributions of each physical/chemical process to variations in pollutant concentrations. Eulerian models utilize the numerical technique of operator splitting to solve continuity equations for each species into several simple ordinary differential equations or partial differential equations that only contain the influence of one or two processes (Gipson, 1999).

The IPR analysis method has been fully implemented in Community Multi-scale Air Quality (CMAQ) model, and has been widely applied to study regional photochemical ozone (O_3) pollution (Goncalves et al., 2009; Khiem et al., 2010; Xing et al., 2017; Tang et al., 2017). Several WRF-Chem model studies used the IPR analysis to investigate the impacts of physical/chemical process on variations in O_3 concentrations. Gao et al. (2018) investigated the impacts of BC-PBL interactions on O_3 concentrations by analyzing the contributions from photochemistry, vertical mixing, and advection processes. Jiang et al. (2012) calculated the contributions of photochemical reactions and physical processes to O_3 formation by using a simplified IPR analysis scheme.

Applying the IPR analysis to diagnose the contributions of each physical or chemical process to variations in aerosol concentrations in WRF-Chem model is more complex technically, and therefore few studies conducted the IPR analysis for aerosols. In this study, we developed an improved IPR analysis scheme in the WRF-Chem model to isolate the processes impacting variations in aerosol concentrations into nine different processes, namely advection (TRAN), emission source (EMIS), dry deposition (DYRD), turbulent diffusion (DIFF), sub-grid convection (SGCV), gas-phase chemistry (GASC), cloud chemistry (CLDC), aerosol chemistry (AERC), and wet scavenging (WETP). TRAN includes horizontal and vertical advection, which is highly related to wind and aerosol concentration gradients from upwind regions to downwind areas (Gao et al., 2018). DRYD is based on resistance models for trace gases (Wesely, 1989) and aerosol particles (Ackermann et al., 1998). SGCV refers to the scavenging within the sub-grid wet convective updrafts. CLDC refers to aqueous-phase photolytic and radical chemistry reactions in clouds, including the activation processes. AERC means microphysical nucleation,

condensation, and coagulation, as well as the mass transfer between the gas phase and condensed phase. WETP contains in-cloud rainout and below-cloud washout during grid-scale precipitation. The contribution of individual process can be calculated as the difference of aerosol concentrations before and after the corresponding operator.

Based on the principle of mass balance, IPR can be verified by comparing the variations in aerosol concentrations (the concentration at the current time minus the concentration at the previous time) with the sum of the contributions from the nine processes during each time step. As shown in Fig. S1, the net contributions of all processes match the variations in aerosol concentrations pretty well.

2.3 Numerical experiments

Table 2 summarizes the experimental designs. To investigate the contributions of regional transport and local emission to the $PM_{2.5}$ concentrations in BTH, four simulations with different anthropogenic emission categories are conducted: (1) CTL: The control simulation with all anthropogenic emissions considered; (2) NoAnth: No anthropogenic emission is considered in the whole domain; (3) NoBTH_Anth: Same as CTL, but anthropogenic emissions in BTH are excluded; (4) OnlyBTH_Anth: Contrary to the NoBTH_Anth case, anthropogenic emissions are only considered in BTH. All the physical and chemical schemes used in these cases are identical. The contributions of regional transport and local emission to the $PM_{2.5}$ concentration in BTH can be identified by comparing the simulation results of NoBTH_Anth and NoAnth (i.e., NoBTH_Anth minus NoAnth) and OnlyBTH_Anth and NoAnth (i.e., OnlyBTH_Anth minus NoAnth), respectively.

To quantify the aerosol radiative effects (ARE) on haze pollution, another sensitivity experiment (referred to as NoARE case) is designed by turning off the feedbacks between aerosols and meteorological variables, including eliminating the aerosol direct effect (ADE) and aerosol indirect effect (AIE) in the model. The ADE is turned off by removing the mass of aerosol species from the calculation of aerosol optical properties as did in Qiu et al. (2017). The AIE is turned off by using a prescribed vertically uniform cloud droplet number, which is calculated from the CTL case during the whole simulation period, following Gao et al. (2015) and Zhang et al., (2015a). The differences between CTL and NoARE (i.e., CTL minus NoARE) represent the impacts of aerosol radiative forcing.

The IPR analysis method is applied to all the designed experiments. Comparing the contributions of each detailed

process between pollution accumulation stage and dissipation stage in CTL can quantitatively explain the reason for the variation of the $PM_{2.5}$ concentrations in BTH. Meanwhile, the prominent physical or chemical process responsible for the aerosol radiative impacts on the haze episode can also be investigated by analyzing the IPR analysis method used in CTL and NoARE cases.

All the five simulations are conducted for the period from 13 to 29 December 2015, and the initial three days are discarded as the model spin-up to minimize the impacts of initial conditions. Simulation results from the CTL case during 16 to 29 December 2015 are used to evaluate the model performance.

2.4 Observational data

Simulated meteorological parameters in CTL case, including 2 m temperature (T_2), 2 m relative humidity (RH_2), 10 m wind speed (WS_{10}) and 10 m wind direction (WD_{10}), are compared with hourly observations at twelve stations, which are collected from NOAA's National Climatic Data Center (https://gis.ncdc.noaa.gov/maps/ncei/cdo/hourly). Due to limited observations of PBL height in BTH, the retrieved PBLH in 3-hour intervals obtained from the GDAS (Global Data Assimilation System) (https://ready.arl.noaa.gov/READYamet.php) in Beijing (39.93 °N, 116.28 °E) is also used to evaluate the model performance. More detailed information about the GDAS meteorological dataset (1 °×1 °) can be found in Rolph et al. (2013), Kong et al. (2015) and https://www.ready.noaa.gov/gdas1.php. Hourly shortwave downward radiation flux (SWDOWN) at the Xianghe station (39.75 °N, 116.96 °E) is taken from WRMC-BSRN (World Radiation Monitoring Center-Baseline Surface Radiation Network, http://bsrn.awi.de) for the energy budget evaluation. The hourly observed surface-layer $PM_{2.5}$ concentrations at the 59 stations are obtained from the CNEMC (China National Environmental Monitoring Center, http://www.cnemc.cn/). The daily measurements of mass concentrations of SO_4^{2-}, NO_3^-, NH_4^+, BC and OC are collected at the sites of (39.97 °N, 116.37 °E) in Beijing and (38.03 °N, 114.53 °E) in Shijiazhuang (Huang et al., 2017; Liu et al., 2018). Detailed locations of these observations are shown in Fig. 1(b).

3. Model evaluation

Accurate representations of observed meteorological fields and pollutant concentrations provide foundations for haze

analysis with the WRF-Chem model. Detailed comparisons between observed and simulated meteorological parameters (T_2, RH_2, WS_{10}, WD_{10}, PBLH, and SWDOWN) and pollutant concentrations ($PM_{2.5}$, BC, OC, SO_4^{2-}, NO_3^-, and NH_4^+) are presented in this section.

3.1 Meteorological parameters

Figure 2 shows the time series of observed and simulated hourly meteorological variables averaged over the 12 stations during 16-29 December 2015. Corresponding statistical metrics, including mean value, normalized mean bias (NMB), mean fractional bias (MFB), mean fractional error (MFE), index of agreement (IOA), and correlation coefficient (R) are presented in Table 3. As shown in Fig. 2, simulated T_2, RH_2, WS_{10} and WD_{10} agree well with the observational data. For temperature, the WRF-Chem model can perfectly depict its diurnal and daily variations with R and IOA of 0.90 and 0.94, respectively, but slightly overestimates the low values at night, with the NMB of 1%. Observed relative humidity can be reasonably reproduced by the model with R and IOA of 0.73 and 0.82, respectively. But a persistent underestimation is found with the NMB of −12%. Different surface layer and boundary layer parameterizations may have influence on the simulated near-surface moisture fluxes, and the settings of these schemes can partially explain the biases of RH_2 between observations and simulations (Qian et al., 2016). This negative bias of RH_2 can also be simulated by other studies (Zhang et al., 2009; Gao et al., 2015). WRF-Chem can capture the observed low values of wind speed during 19-23 December and high values of wind speed during 16-17 and 25-27 December. The positive NMB of 28% may probably result from unresolved topographical features in surface drag parameterization and the coarse resolution used in the nested domain (Yahya et al., 2015; Zheng et al., 2015). For wind direction, the calculated NMB is 1% and the IOA is 0.65, indicating that the WRF-Chem model can generally reproduce the varied wind direction during the simulation period.

Simulated hourly PBLH and SWDOWN are also compared with observations in Fig. 3. It is noted that PBLH provided by GDAS of NOAA are in 3-hour intervals. The simulations in CTL case agree well with the observations, including capturing the daily maximum in the daytime and the low values at night. The correlation coefficients are 0.68 and 0.91 for PBLH and SWDOWN, respectively.

3.2 $PM_{2.5}$ and its components

Observed hourly surface-layer $PM_{2.5}$ concentrations from 16 to 29 December 2015 in the nine cities (Shengyang, Beijing, Xingtai, Hengshui, Baoding, Langfang, Yangquan, Anyang, and Jinan) are compared with the model results from CTL case (Fig. 4). The statistical metrics are shown in Table 3. Generally, WRF-Chem model can reasonably reproduce the evolutional characteristics of the observed $PM_{2.5}$ concentrations in the nine cities (Rs=0.57-0.90). Both the observed and simulated $PM_{2.5}$ concentrations exhibit a growth trend during December 16-22 and 28-29, and a decreasing tendency during December 23-27. However, an obvious underestimation is found in Beijing from 25 to 26 December when a maximum hourly concentration of 600 μg m^{-3} was observed. This negative bias is also simulated by previous studies (Chen et al., 2018; Zhang et al., 2018b), and the possible reasons for the underestimation are (1) the bias in simulated meteorological conditions (e.g., underestimated RH_2 and overestimated WS_{10}); (2) the missing mechanisms of some gas-aerosol phase partitioning and heterogeneous reactions which may produce secondary inorganic aerosol (Huang et al., 2014a; Wang et al., 2014); (3) the lack of SOA simulation in MOSAIC mechanism (Gao et al., 2016). Generally, the performance statistics of $PM_{2.5}$ in almost all cities meet the model performance goal (MFB within ±30% and MFE≤50%) proposed by Boylan and Russel (2006).

Figure 5 compares the simulated and observed surface-layer concentrations of BC, OC, SO_4^{2-}, NO_3^-, and NH_4^+ in Beijing and Shijiazhuang averaged during 16-29 December 2015. WRF-Chem model underestimates the concentrations of SO_4^{2-}, NH_4^+ and OC in Beijing (Shijiazhuang) by 19% (40%), 14% (9%), and 21% (41%), respectively, but overestimates the NO_3^- concentration by 29% (44%). Due to the low reactivity of BC in the atmosphere, the uncertainty in BC emission may cause the biases in Beijing (NMB=+10%) and Shijiazhuang (NMB=-24%). For OC, the underestimation may result from the lack of SOA in the MOSAIC aerosol module (Qiu et al., 2017). Missing some mechanisms of SO_2 gas-phase and aqueous-phase oxidation, as well as heterogeneous chemistry may explain the underestimation of SO_4^{2-}. It is noted that similar biases of aerosol components were also reported by other WRF-Chem studies (Zhang et al., 2015a; Qiu et al., 2017).

4. Formation and evolution mechanisms of the haze episode

In this section, we first reproduce the evolution of the severe haze episode, and then investigate the formation and

evolution mechanisms, including examining contributions of local emission and regional transport to the $PM_{2.5}$ concentration in BTH, and the contributions of each detailed physical/chemical process to the variations in the $PM_{2.5}$ concentration.

4.1 Spatial-temporal evolutions of surface-layer $PM_{2.5}$ concentrations

Figures 6(a-k) show the spatial distributions of simulated daily mean surface-layer $PM_{2.5}$ concentrations from 17 to 28 December 2015. From December 17, aerosol particles started to accumulate in the near-surface layer in BTH under a prevailing southerly air flow. On December 20, the BTH region was under a uniform pressure field (Fig. S2(a)). The regional average wind speed was less than 3 m s^{-1}, and the boundary layer became stable, which constrained aerosols within a low mixing layer. Meanwhile, a low-pressure center situated to the north of BTH, where air pollutants from south, southwest, and southeast converged. Consequently, the daily mean $PM_{2.5}$ concentration averaged over BTH was over 200 μg m^{-3}. On December 21, a weak low-pressure center was formed near the Bohai Bay and a weak high-pressure center moved to Shandong Peninsula (Fig. S2(b)). The synoptic conditions brought more air masses from south to north, and worsened air quality in BTH. On December 22, a weak high pressure system moved within Inner Mongolia (Fig. S2(c)), which could bring cold air to the BTH region. Meanwhile, the polluted air could also be transported back to the BTH, leading to a continuous increase in the $PM_{2.5}$ concentration, with the maximum daily mean value exceeding 600 μg m^{-3} in BTH (Fig. 6(e)). Due to the enhanced anticyclone originated from Siberian (Fig. S2(d)), the accumulation of aerosol particles in BTH was terminated with the incursion of a strong cold front from 23 to 27 December. But frequent transitions between high and low pressure systems over BTH accompanying with the shifting wind directions resulted in a quick $PM_{2.5}$ variation, especially on December 24 and 25, when a low-pressure system developed northeast of BTH (Fig. S2(e)). The air mass in BTH was influenced by the pollutants from south, resulting in a temporary increase in the concentration of $PM_{2.5}$ on December 25. After December 27, another haze episode gradually formed.

According to the trends in simulated $PM_{2.5}$ concentrations averaged over the BTH region (Fig. 6(l)), we divide the whole simulation period into three stages: (1) aerosol accumulation stage (December 16-22, Stage_1); (2) aerosol dispersion stage (December 23-27, Stage_2); (3) formation stage for another haze event (December 28-29, Stage_3). In this manuscript, we mainly focus on the first two stages to reveal important factors that cause the accumulation and dispersion of particulate

matters.

In Stage_1, the daily mean $PM_{2.5}$ concentrations averaged over BTH increased from 24.2 μg m^{-3} to 289.8 μg m^{-3}, and the average $PM_{2.5}$ concentration was 145.6 μg m^{-3} (Fig. 7(a)), close to the air quality threshold value of "heavily polluted" ($PM_{2.5}$ 24-h average concentration > 150 μg m^{-3}). The WS_{10} was low (Fig. 7(b)), especially during the heavy pollution period (20-22 December), and the mean wind speed was 2.3 m s^{-1}, less than 3.2 m s^{-1} (one of the indicators used to define air stagnation by NOAA, https://www.ncdc.noaa.gov/societal-impacts/air-stagnation/overview), indicating that the near surface circulation was insufficient to disperse accumulated air pollutants. The decreased PBLH (from 701.6 m to 109.9 m) could compress air pollutants into a shallow layer, resulting in an elevated pollution level. During Stage_2, the $PM_{2.5}$ concentration decreased gradually with the increased wind speed and PBLH. The $PM_{2.5}$ concentration averaged during Stage_2 was 107.9 μg m^{-3}, still exceeding the Grade II standard (75 μg m^{-3}) defined by the National Ambient Air Quality Standards of China.

4.2 Contributions of local emission and regional transport to $PM_{2.5}$ concentrations

Previous studies have reported that anthropogenic emission was the dominant cause of haze events in China (Jiang et al., 2013; Sun et al., 2014; Gu and Liao, 2016; Yang et al., 2016b). Emission control measures have been taken to ensure good air quality for major events (e.g., APEC) or to mitigate the severity of coming pollution episodes (Zhou et al., 2018). Other studies, such as Sun et al. (2017) and Wang et al. (2017), pointed out that regional transport contributed more than 50% of the particulate concentrations in BTH during haze events. This section discusses the contributions of local anthropogenic emission and regional transport to the $PM_{2.5}$ concentration in BTH, aiming to reveal the relative importance during this haze episode.

As shown in Fig. 7(a), the $PM_{2.5}$ concentration in BTH during Stage_1 was mainly contributed by the combined effects of local emission and regional transport. The contributions of local emission and regional transport to the $PM_{2.5}$ concentration were comparable (49% and 32%, respectively), especially during the heavy pollution period (December 20-22, 43% vs. 37%). During Stage_2, the contributions of regional transport decreased from 30% to 16%. The relative high $PM_{2.5}$ concentration (107.9 μg m^{-3}) was principally caused by the local emission. On average, the contributions of local emission and regional transport to the $PM_{2.5}$ concentration in Stage_2 were 51% and 24%, respectively. The impact of regional

transport could be qualitatively expressed by specific humidity, which was treated as an indicator for the origin of air masses (Jia et al., 2008). Air masses from the south were usually warmer and wetter than those from the north, so the specific humidity averaged over the BTH was higher in Stage_1 (1.7 g/kg) than that in Stage_2 (1.4 g/kg) (Fig. 7(b)). The evolution of $PM_{2.5}$ nicely followed the trend of specific humidity with a high correlation coefficient of 0.86.

4.3 Contributions of each physical/chemical process to variations in $PM_{2.5}$ concentrations

Figures 8(a1-a2) show the diurnal variations of $PM_{2.5}$ concentrations averaged over the BTH region during Stage_1 and Stage_2, respectively. The $PM_{2.5}$ concentration increased by 43.9 μg m^{-3} (from 136.5 μg m^{-3} at 00:00LST to 180.4 μg m^{-3} at 23:00LST) during the period of particulate accumulation (Stage_1), but it decreased by 41.5 μg m^{-3} during the period of particulate elimination (Stage_2).

The hourly $PM_{2.5}$ changes induced by each and all physical/chemical processes during Stage_1 and Stage_2 by using the IPR analysis method are shown in Figs. 8(b1-b2). During both stages, the dominant sources of surface-layer $PM_{2.5}$ were EMIS and AERC, while the main sinks were TRAN, DIFF, and DRYD. The maximum positive contribution of EMIS could be found during the rush hours (07:00-08:00LST and 16:00-19:00LST) (Fig. S3). The maximum negative contributions of TRAN and DIFF appeared at late night (01:00-05:00LST) and at noon (11:00-14:00LST), respectively.

To explain the reason for 24-h $PM_{2.5}$ increase during Stage_1 and 24-h $PM_{2.5}$ decrease during Stage_2 (Figs. 8(a1-a2)), we quantify the contributions of each physical/chemical process to 24-h $PM_{2.5}$ changes for both stages (Figs. 8(c1-c2)), which are calculated by integrating hourly $PM_{2.5}$ changes induced by each process from 00:00LST to 23:00LST (Figs. 8(b1-b2)). In WRF-Chem, DRYD is intermingled with vertical diffusion, so changes in the column burden during vertical mixing can be attributed to DRYD (Tao et al., 2015). Following Tao et al. (2015), we define vertical mixing (VMIX) as the sum of DIFF and DRYD. As shown in Figs. 8(c1-c2), contributions of AERC, TRAN and VMIX processes to 24-h $PM_{2.5}$ changes were +29.6 (+17.9) μg m^{-3}, -71.8 (-103.6) μg m^{-3} and -177.3 (-221.6) μg m^{-3} for Stage_1 (Stage_2), respectively. Small differences were found for contributions from other processes between Stage_1 and Stage_2 (differences smaller than 5 μg m^{-3}). Therefore, the $PM_{2.5}$ increase over the BTH region during haze formation stage was mainly attributed to strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes. On the contrary,

during haze elimination stage (Stage_2), more aerosols in BTH were transported out of BTH or dispersed to the upper atmosphere or subsided to the ground. What's more, the dry cold air from the north decreased the specific humidity (as shown in Fig. 7(b)) in BTH, leading to weaker production of secondary aerosols by aerosol chemistry process.

5 Aerosol radiative effects (ARE) on the haze episode

Previous studies have demonstrated that the aerosol radiative forcing could increase the near-surface $PM_{2.5}$ concentrations by about 12%-29% (Gao et al., 2015; Gao et al., 2016; Qiu et al., 2017; Zhou et al., 2018). However, the detailed influence mechanisms (i.e., the prominent physical or chemical process responsible for the aerosol radiative impacts on $PM_{2.5}$ concentrations) are still unclear. In this section, we examine the effects of aerosol radiative forcing on meteorological parameters and $PM_{2.5}$ levels during the haze episode, with a special focus on the detailed influence mechanism by using the IPR analysis.

5.1 Effects of aerosol radiative forcing on meteorological parameters and $PM_{2.5}$ concentrations

Figure 9 illustrates the impacts of aerosols on the downward shortwave radiative flux (SW) at the surface (BOT_SW) and in the atmosphere (ATM_SW), calculated by subtracting the model results of NoARE from those of CTL, during Stage_1, Stage_2, and the whole simulation period. Downward SW at the surface was strongly decreased when ARE was considered, especially over high aerosol-loading regions during heavily polluted periods. Generally, the shortwave radiation fluxes at the surface averaged over BTH were reduced by 28% (23.9 W m^{-2}) in Stage_1, 18% (16.6 W m^{-2}) in Stage_2, and 23% (19.9 W m^{-2}) during the whole simulation period, respectively. Contrary to the significant negative effects at the surface, as a result of ARE, the downward SW fluxes in the atmosphere averaged over BTH were increased by 65% (19.1 W m^{-2}) in Stage_1, 37% (10.8 W m^{-2}) in Stage_2, and 51% (14.7 W m^{-2}) during the whole period, respectively.

The impacts of ARE (including aerosol direct and indirect effects) on meteorological parameters and $PM_{2.5}$ concentrations are analyzed in Fig. 10. Because less SW could reach the ground, near-surface temperature was decreased over BTH (Fig. 10(a)), especially during heavy pollution periods, and the largest decrease was up to 2 k. Meanwhile, the increased SW in the atmosphere could warm the upper air. As a result, a more stable atmosphere was expected. It is known

that the atmospheric stability can be exactly characterized by the profile of equivalent potential temperature (EPT) (Bolton, 1980; Zhao et al., 2013; Yang et al., 2016a). If EPT rises with height, the atmosphere is stable. As shown in Fig. 10(b), the EPT was decreased in the lower atmosphere (below ~1000 m) with the largest decrease of 3 k on December 22, but increased in the upper atmosphere (above ~1200 m). The change in the EPT profile indicated that ARE could lead to a more stable atmosphere, which further weakened vertical movement in BTH (Fig. 10(c)). As a result of ARE, the PBLH was decreased and the relative humidity in the lower atmosphere was increased (Fig. 10(d)). All the changes in meteorological variables were beneficial for $PM_{2.5}$ accumulation in the lower atmosphere (Fig. 10(e)). The daily maximum increase of $PM_{2.5}$ concentration was 43.2 μg m^{-3} due to ARE. It was noticed that ARE had a negative impact on the near-surface $PM_{2.5}$ concentrations during December 23-24, which could be explained that absorbing aerosols (i.e., BC) induced anomalous northeasterlies, and the relatively clean air transported from the northeastern regions to BTH (Fig. S4).

5.2 Influence mechanism of aerosol radiative effects

Since variations in $PM_{2.5}$ concentrations are directly caused by physical and chemical processes (Zhu et al., 2015), the IPR method is then used to investigate the detailed influence mechanisms (i.e., the prominent physical or chemical processes responsible for the aerosol radiative impacts on haze episodes). Figs. 11(a-b) show the diurnal variations of $PM_{2.5}$ concentrations in NoARE and CTL cases averaged over the BTH region in Stage_1. A 24-h increase of 39.1 μg m^{-3} was simulated in NoARE case. When aerosol radiative forcing was considered, the 24-h increase of $PM_{2.5}$ concentration was 43.9 μg m^{-3}. The enhancement of 4.8 μg m^{-3} (12%) induced by ARE could be mainly attributed to the contributions of VMIX, TRAN, and AERC processes, as shown in Fig. 11(c). The vertical mixing was strongly restrained by ARE, therefore fewer particles diffused from the surface to the upper layer, resulting in the accumulation of $PM_{2.5}$ in a lower atmospheric boundary layer. The changes induced by ARE in contributions of VMIX process exhibited positive values in the lower layers and negative values in the upper layers (Fig. S5(a)). Generally, the VMIX process contributed +22.5 μg m^{-3} to the enhancement in 24-h $PM_{2.5}$ increase (+4.8 μg m^{-3}) for Stage_1. The TRAN process, however, contributed -19.6 μg m^{-3}. Constrained vertical mixing due to ARE could increase aerosol precursors and water vapor in the thin boundary layer to enhance the formation of secondary particles. Generally, the AERC process contributed +1.2 μg m^{-3}. The positive contribution of AERC

was mainly distributed over the high polluted regions in BTH (Fig. S5(b)). Detailedly, the average changes in concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ during the daytime from 11:00 to 17:00LST in Stage_1 were -0.5 μg m^{-3}, +1.3 μg m^{-3}, and +0.8 μg m^{-3}, respectively. The decreased near-surface temperature caused by ARE may suppress the chemical formation of SO_4^{2-}. Generally, the total contribution of VMIX, TRAN, and AERC processes to the change in 24-h $PM_{2.5}$ increase caused by ARE was +4.1 μg m^{-3}, and the restrained vertical mixing could be the primary reason for near-surface $PM_{2.5}$ increase when aerosol radiative forcing was considered.

Figure 12(a) shows the vertical profiles of the 24-h increases in $PM_{2.5}$ concentrations (23:00LST minus 00:00LST) averaged over BTH during Stage_1 in CTL and NoARE cases. Below ~300 m (between L01 and L04), the 24-h increase simulated by CTL was larger than that in NoARE, which could be mainly explained by that the positive contributions of VMIX exceeded the negative contributions of TRAN in the lower atmosphere when aerosol radiative effect was considered (Fig. 12(b)). However, in the upper layers (from 300 to 2000 m), aerosol radiative forcing weakened the 24-h $PM_{2.5}$ increase during Stage_1. When aerosol radiative effect was considered, fewer particulate matters, precursors and water vapor were diffused from the surface to the upper layers, and therefore fewer particles were formed in the upper layers. Despite of the positive contributions of TRAN, the net contributions of VMIX, TRAN, and AERC to $PM_{2.5}$ changes caused by ARE in the upper atmosphere were negative.

6. Conclusions and discussions

In this study, an online coupled mesoscale meteorology-chemistry model (WRF-Chem) with an improved integrated process rate (IPR) analysis (i.e., process analysis) scheme is applied to investigate the formation and evolution mechanisms of a severe haze episode happened in the BTH region during 16-29 December 2015. Sensitivity experiments are conducted to examine the contributions of local emission and regional transport to the $PM_{2.5}$ concentrations during the haze event, while IPR analysis is used to quantify the contributions of each physical/chemical process to the variation in $PM_{2.5}$ concentration. The impacts of aerosol radiative forcing (including direct and indirect effects) are also quantified, with a special focus on the detailed influence mechanism (i.e., prominent process responsible for the aerosol radiative impacts on the haze event). An integrated comparison between observations and simulations demonstrates good performance for both meteorological and

chemical variables, indicating that the WRF-Chem model has the capability to reproduce the haze episode.

Spatial-temporal evolutions of the near-surface $PM_{2.5}$ concentration, and the contributions of local emission and regional transport to the severe haze even in BTH, were firstly analyzed. During the aerosol accumulation stage (December 16-22, Stage_1), the daily $PM_{2.5}$ concentration in BTH experienced a consistent increase, with the mean value of 145.6 μg m^{-3}. The contributions of local emission and regional transport to the $PM_{2.5}$ concentration were comparable (49% and 32%, respectively), meaning the combined effect resulted in the high $PM_{2.5}$ concentration in BTH. During the aerosol dispersion stage (December 23-27, Stage_2), the average $PM_{2.5}$ concentration in BTH was 107.9 μg m^{-3}. The contributions of local emission and regional transport were 51% and 24%, respectively. Therefore, the relatively high $PM_{2.5}$ concentration during Stage_2 was principally caused by local emission. During December 28-29 (Stage_3), another haze event was formed and developed.

The IPR analysis was then used to explain the reason for $PM_{2.5}$ increase during Stage_1 and decrease during Stage_2, by quantifying the contributions of each physical/chemical process to variations in $PM_{2.5}$ concentration. During both stages, the dominant sources were emission (EMIS) and aerosol chemistry (AERC), while the main sinks were turbulent diffusion (DIFF), advection (TRAN), and dry deposition (DRYD). The $PM_{2.5}$ concentration increased by 43.9 μg m^{-3} (23:00LST minus 00:00LST) during Stage_1, but it decreased by 41.5 μg m^{-3} during Stage_2. Contributions of AERC, TRAN and VMIX (vertical mixing, the sum of DRYD and DIFF) to the 24-h $PM_{2.5}$ changes were +29.6 (+17.9) μg m^{-3}, -71.8 (-103.6) μg m^{-3} and -177.3 (-221.6) μg m^{-3} for Stage_1 (Stage_2), respectively. Small differences in contributions from other processes were found between Stage_1 and Stage_2. Therefore, the $PM_{2.5}$ increase over BTH during the haze formation stage was attributed to strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes.

When aerosol radiative forcing was considered, the equivalent potential temperature was decreased in the lower layers but increased in the upper layers, leading to a more stable atmosphere. Meanwhile, the decreased PBLH and increased relative humidity were also beneficial for $PM_{2.5}$ accumulation. The daily maximum increase of the near-surface $PM_{2.5}$ concentration in BTH was 43.2 μg m^{-3}. The IPR method was also used to investigate the detailed influence mechanism of aerosol radiative effects. When aerosol radiative feedback was considered, the 24-h $PM_{2.5}$ increase was enhanced by 4.8 μg

m^{-3} (12%) during Stage_1, which could be mainly attributed to the contributions of VMIX (+22.5 μg m^{-3}), TRAN (-19.6 μg m^{-3}), and AERC (+1.2 μg m^{-3}). The restrained vertical mixing could be the primary reason for near-surface $PM_{2.5}$ increase when aerosol radiative forcing was considered.

There are some limitations in this work. The uncertainty of the MIX anthropogenic emission inventory, the lack of secondary organic aerosols, and the missing mechanisms of some heterogeneous reactions may result in large uncertainties in the final simulation results, especially the predicted aerosol chemical compositions, such as SO_4^{2-}, NO_3^- and NH_4^+. The biases in simulated concentrations of SO_4^{2-}, NO_3^- and NH_4^+ may have impacts on the contributions of AERC and CLDC processes to the air pollution variation. Uncertainties should be quantitatively analyzed in future studies. Besides, conclusions draw from a case study in BTH cannot represent a full view of the underlying mechanisms of haze formation and elimination. Better understanding will be attained by conducting multiple-case simulations in future. What's more, an anomalous northeasterly induced by absorbing aerosols was observed, leading to a decrease in the near-surface $PM_{2.5}$ concentrations during December 23-24 2015 in BTH, which was different from previous studies that reported light-absorbing aerosols could worsen air quality (Li et al., 2016; Huang et al., 2018; Gao et al., 2018). More experiments should be designed in future to examine the changes in atmospheric thermal and atmospheric dynamic caused by absorbing aerosol radiative forcing and their impacts on haze episodes.

As Zheng et al. (2018) pointed out that the $PM_{2.5}$ concentration in China has been decreasing in recent years, but the decreased fine particulate matter could stimulate ozone production (Li et al., 2019a; Zhu et al., 2019). Multi-pollutant mixture may be a hot topic in the future, and the IPR analysis can be a useful method to provide a quantitative analysis about the formation mechanism of the complex air pollutions, including figuring out the major physical/chemical process behind these events. Meanwhile, significant differences between model predictions (e.g., O_3 and $PM_{2.5}$) are found among current multi-scale air quality models (Chen et al., 2019b; Li et al., 2019b), even though the same inputs are used. These different performances can be associated with the differences in model formulations, including parameterizations and numerical methods (Carmichael et al., 2008). In order to acquire a quantitative attribution of the cause of differences between simulation results, process analysis method should be developed and implemented in these models, and the IPR analysis will be easier to draw conclusions about the fundamental problems that cause the differences between model predictions.

Data availability

Observational datasets and simulation results are available upon request to the corresponding author (hongliao@nuist.edu.cn).

Author contributions

HL and LC conceived the study and designed the experiments. LC and JZ performed the simulations and carried out the data analysis. YG, MZ, YQ, ZL, NL and YW provided useful comments on the paper. LC prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

Special issue statement

This study is part of the special issue "Regional transport and transformation of air pollution in eastern China". It is not associated with a conference.

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Table 1. Parameterizations used in the WRF-Chem model

Options	WRF-Chem
Microphysics option	Purdue Lin scheme
Longwave radiation option	RRTMG scheme
Shortwave radiation option	RRTMG scheme
Surface layer option	Revised MM5 Monin-Obukhov scheme
Land surface option	Unified Noah land-surface model
Urban canopy model	Single-layer UCM scheme
Boundary layer option	YSU scheme
Cumulus option	Grell 3D ensemble scheme
Photolysis scheme	Fast-J
Dust scheme	Shao_2004
Chemistry option	CBMZ
Aerosol option	MOSAIC
Analysis nudging	On

Table 2. Experimental design

Case Description	Anthropogenic Emission	Aerosol Direct Effect	Aerosol Indirect Effect
CTL	Y	Y	Y
NoAnth	Without emission in the whole domain	Y	Y
NoBTH_Anth	Without emission in BTH	Y	Y
OnlyBTH_Anth	Only emission in BTH	Y	Y
NoARE	Y	N	N

Table 3. Statistical metrics between observations and simulations

Variables	nstd	$\overline{OBS}$[1]	$\overline{SIM}$[2]	NMB[3]	MFB[4]	MFE[5]	IOA[6]	R[7]
T_2 (k)[a]	12	270.7	271.6	1	1	1	0.94	0.90
RH_2 (%)[a]	12	63.8	56.1	-12	-12	22	0.82	0.73
WS_{10} (m s^{-1})[a]	12	2.5	3.2	28	32	58	0.79	0.70
WD_{10} (°)[a]	12	190.8	192.2	1	-2	55	0.65	0.43
$PM_{2.5}$ (μg m^{-3})	59	173.6	168.2	-3	13	47	0.86	0.76

[a]T_2: temperature at 2 m (k); RH_2: relative humidity at 2 m (%); WS_{10}: wind speed at 10 m (m s^{-1}); WD_{10}: wind direction at 10 m (°).

[1,2]$\overline{OBS}$ and $\overline{SIM}$ represent the average observations and simulations, respectively. $\overline{OBS} = \frac{1}{nstd} \times \sum_{i=1}^{nstd} OBS_i$, $\overline{SIM} = \frac{1}{nstd} \times \sum_{i=1}^{nstd} SIM_i$.

[3]NMB is the normalized mean bias, $NMB = \frac{1}{nstd} \times \sum_{i=1}^{nstd} \frac{SIM_i - OBS_i}{OBS_i} \times 100\%$.

[4]MFB is the mean fractional bias, $MFB = \frac{2}{nstd} \times \sum_{i=1}^{nstd} \frac{SIM_i - OBS_i}{SIM_i + OBS_i} \times 100\%$.

[5]MFE is the mean fractional error, $MFE = \frac{2}{nstd} \times \sum_{i=1}^{nstd} \frac{|SIM_i - OBS_i|}{SIM_i + OBS_i} \times 100\%$.

[6]IOA is the index of agreement, $IOA = 1 - \frac{\sum_{i=1}^{nstd} (SIM_i - OBS_i)^2}{\sum_{i=1}^{nstd} (|OBS_i - \overline{OBS}| + |SIM_i - \overline{SIM}|)^2}$.

[7]R is the correlation coefficient, $R = \frac{\sum_{i}^{nstd} |(OBS_i - \overline{OBS}) \times (SIM_i - \overline{SIM})|}{\sqrt{\sum_{i}^{nstd} (OBS_i - \overline{OBS})^2 + \sum_{i}^{nstd} (SIM_i - \overline{SIM})^2}}$.

Where OBS_i and SIM_i mean observations and model predictions, respectively. i refers to a given station, and nstd is the total number of stations.

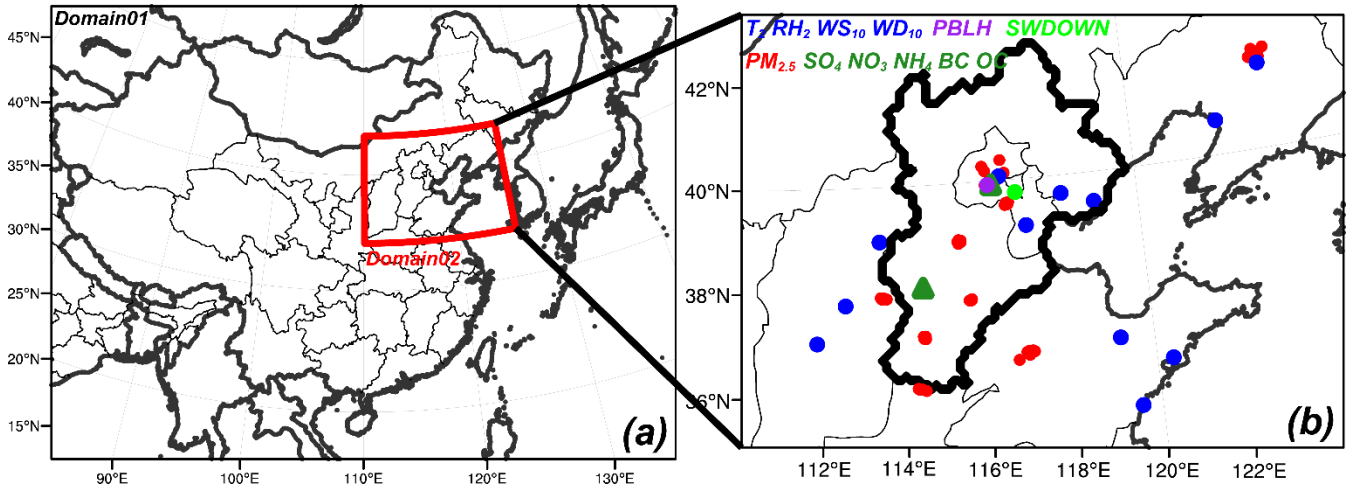

Figure 1. (a) Map of the two nested model domains. (b) Locations of the observations used for model evaluation.

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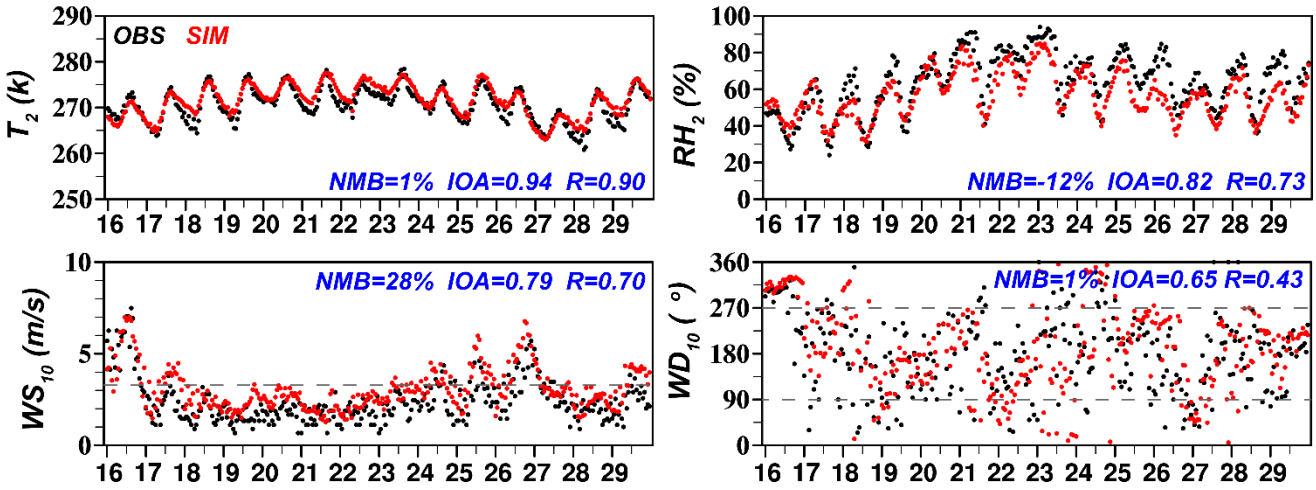

Figure 2. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly 2 m temperature (T_2, k), 2 m relative humidity (RH_2, %), 10 m wind speed (WS_{10}, m s^{-1}), and 10 m wind direction (WD_{10}, °) averaged over the 12 stations during 16-29 December 2015.

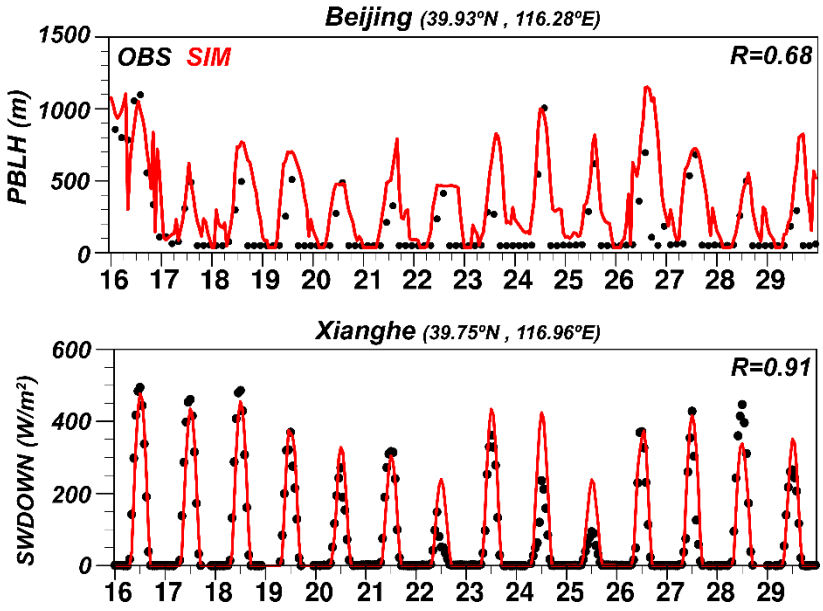

Figure 3. Time series of observed (shown in black dots) and simulated (shown in red lines) hourly planetary boundary layer height (PBLH, m) at the site of (39.93 °N, 116.28 °E) in Beijing, and shortwave downward radiation flux (SWDOWN, W m^{-2}) at the Xianghe Station (39.75 °N, 116.96 °E) from 16 to 29 December 2015. Notably, PBLH provided by Global Data Assimilation System (GDAS) are in 3-hour intervals. All the time is converted to China Standard Time (Beijing Time).

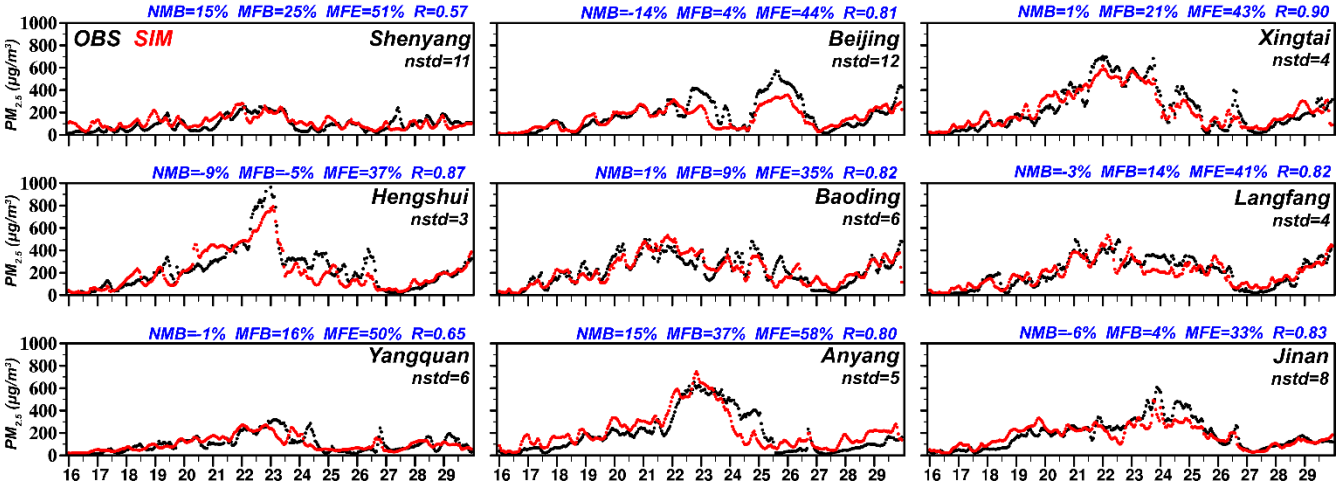

Figure 4. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly $PM_{2.5}$ concentrations (μg m^{-3}) in the nine cities (Shengyang, Beijing, Xingtai, Hengshui, Baoding, Langfang, Yangquan, Anyang, and Jinan) from 16 to 29 December 2015. The nstd in each panel represents the number of observation sites in each city. Beijing Time is used for these hourly time series.

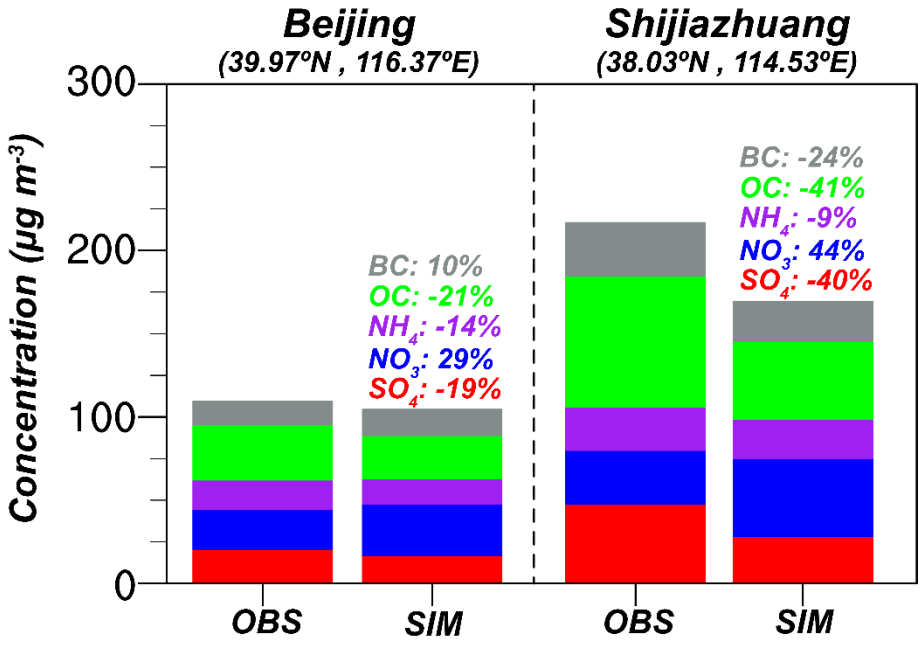

Figure 5. Comparison of observed and simulated surface-layer mass concentrations (μg m^{-3}) of SO_4^{2-} (red), NO_3^- (blue), NH_4^+ (purple), OC (green), and BC (gray) in the sites of (a) (39.97 ℃N, 116.37 ℃E) in Beijing, and (b) (38.03 ℃N, 114.53 ℃E) in Shijiazhuang averaged over 16-29 December 2015. Also listed in colored numbers are normalized mean biases (NMBs) for each species.

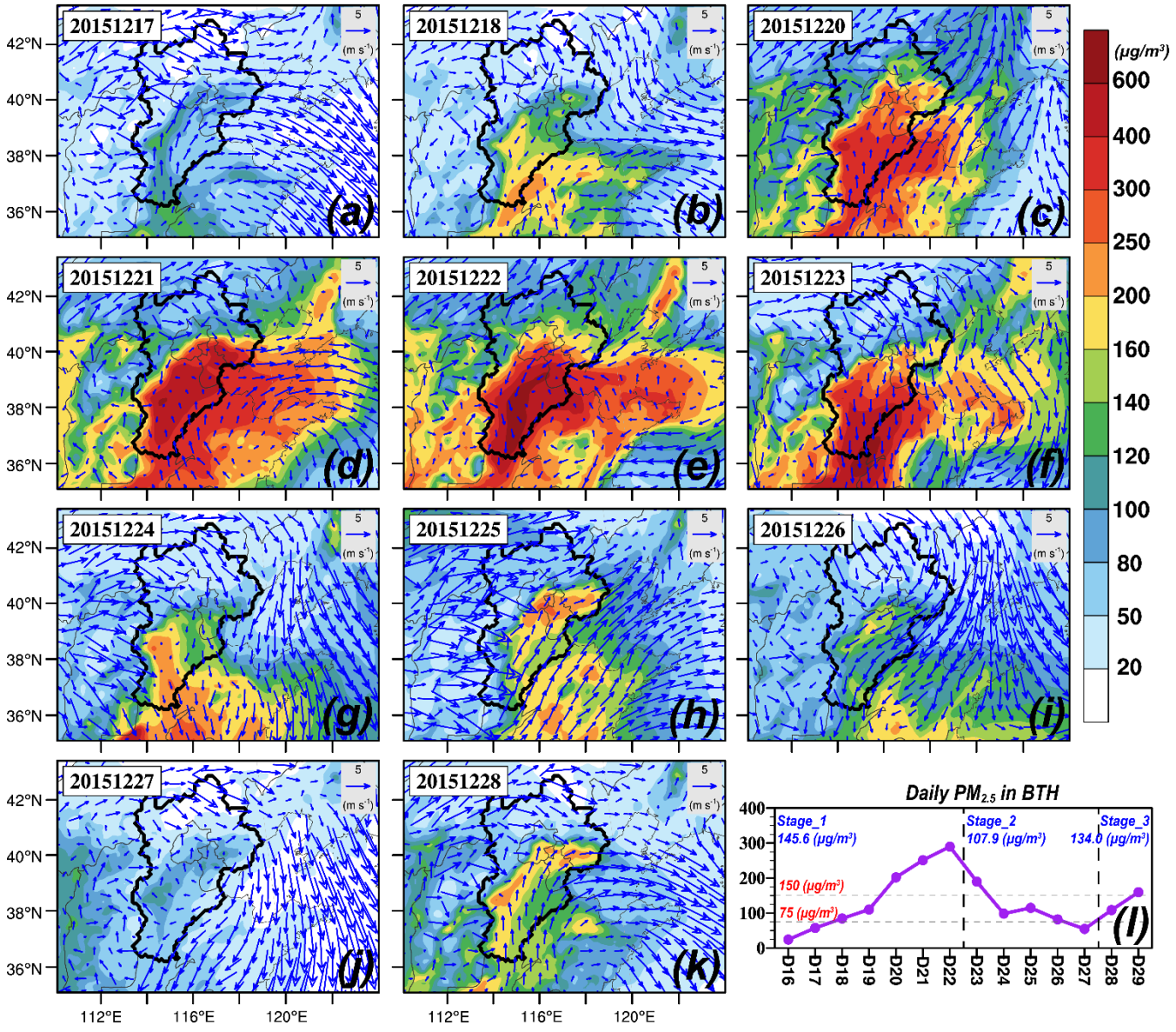

Figure 6. (a-k) Spatial distributions of simulated daily $PM_{2.5}$ concentrations (shaded, μg m^{-3}) and wind vectors (arrows, m s^{-1}). Time series of simulated daily $PM_{2.5}$ concentrations averaged over the Beijing-Tianjin-Hebei region are also shown in (l).

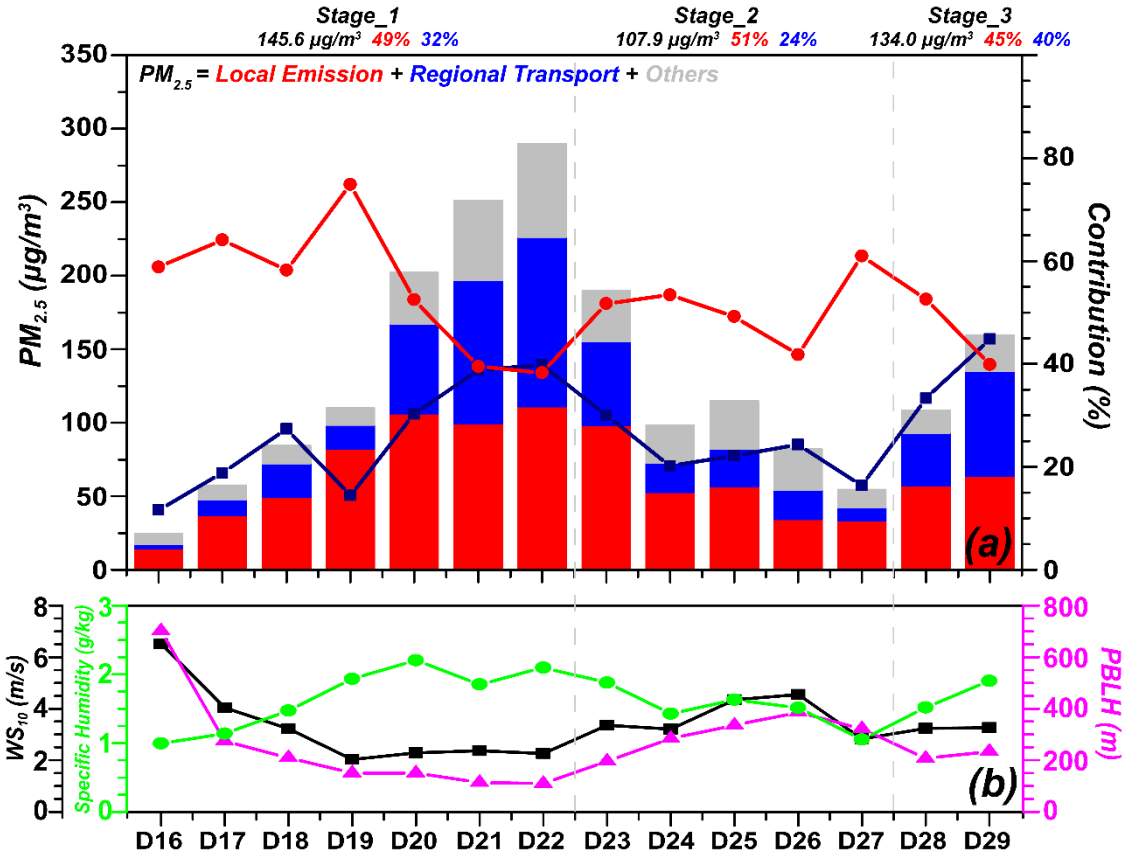

Figure 7. (a) Contributions of local emission (shown in red) and regional transport (shown in blue) to the near-surface $PM_{2.5}$ concentrations averaged over the Beijing-Tianjin-Hebei region from 16 to 29 December 2015. The absolute contributions (μg m^{-3}) are shown in bars, and the percentage contributions (%) are shown in lines. The $PM_{2.5}$ concentration and the percentage contributions averaged over each stage are listed at the top of (a). Simulated daily 10 m wind speed (WS_{10}, m s^{-1}, shown in black dot line), specific humidity (g kg^{-1}, shown in green dot line), and PBLH (m, shown in purple dot line) averaged over Beijing-Tianjin-Hebei are also shown in (b).

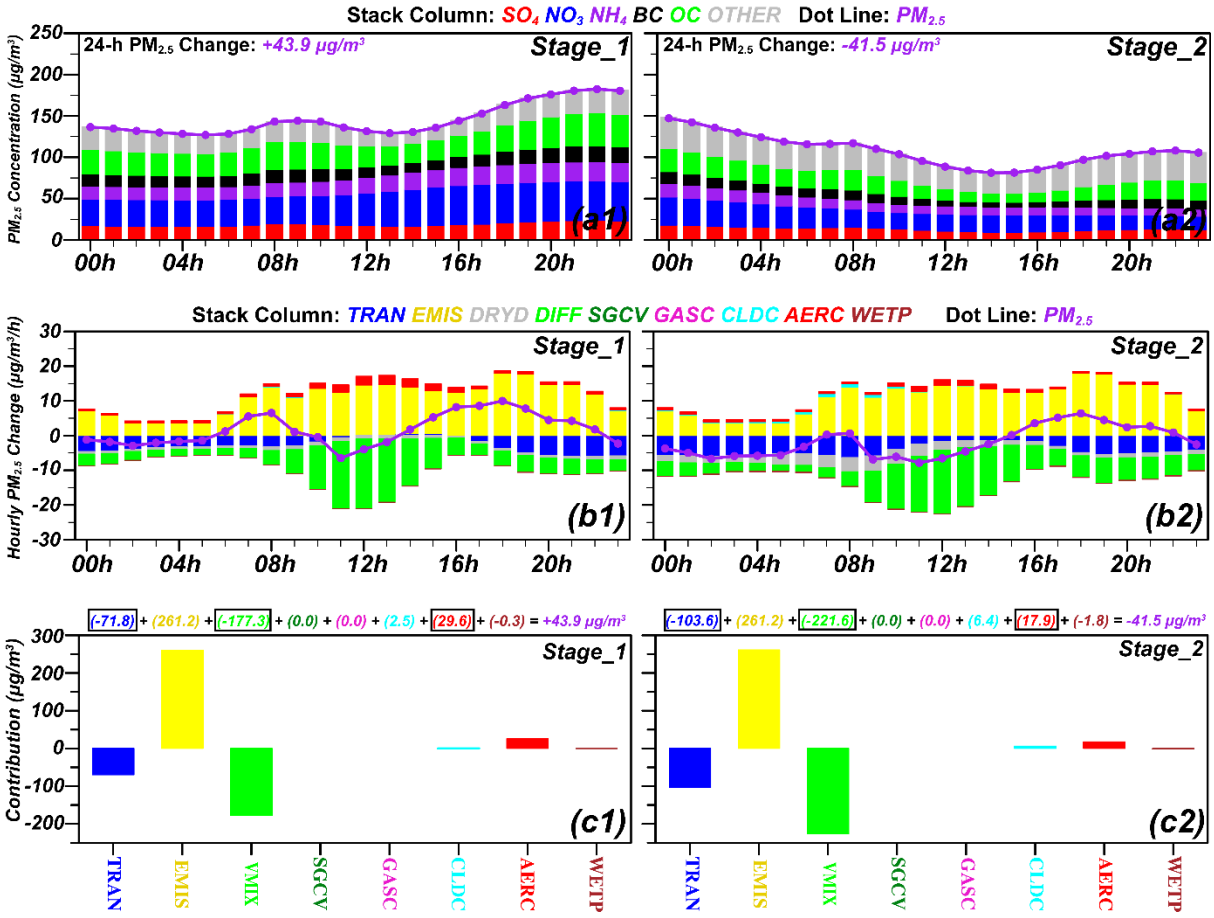

Figure 8. (a1-a2) Diurnal variations of $PM_{2.5}$ concentrations averaged over Beijing-Tianjin-Hebei during Stage_1 and Stage_2 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is the 24-h change in $PM_{2.5}$ concentration (23:00LST minus 00:00LST). (b1-b2) The hourly $PM_{2.5}$ changes induced by each physical/chemical process by using the IPR analysis method (shown by colored bars). The purple dot lines represent hourly $PM_{2.5}$ changes induced by all processes, also indicating the differences between current and previous-hour $PM_{2.5}$ concentrations. (c1-c2) Contributions of each physical/chemical process to 24-h $PM_{2.5}$ changes.

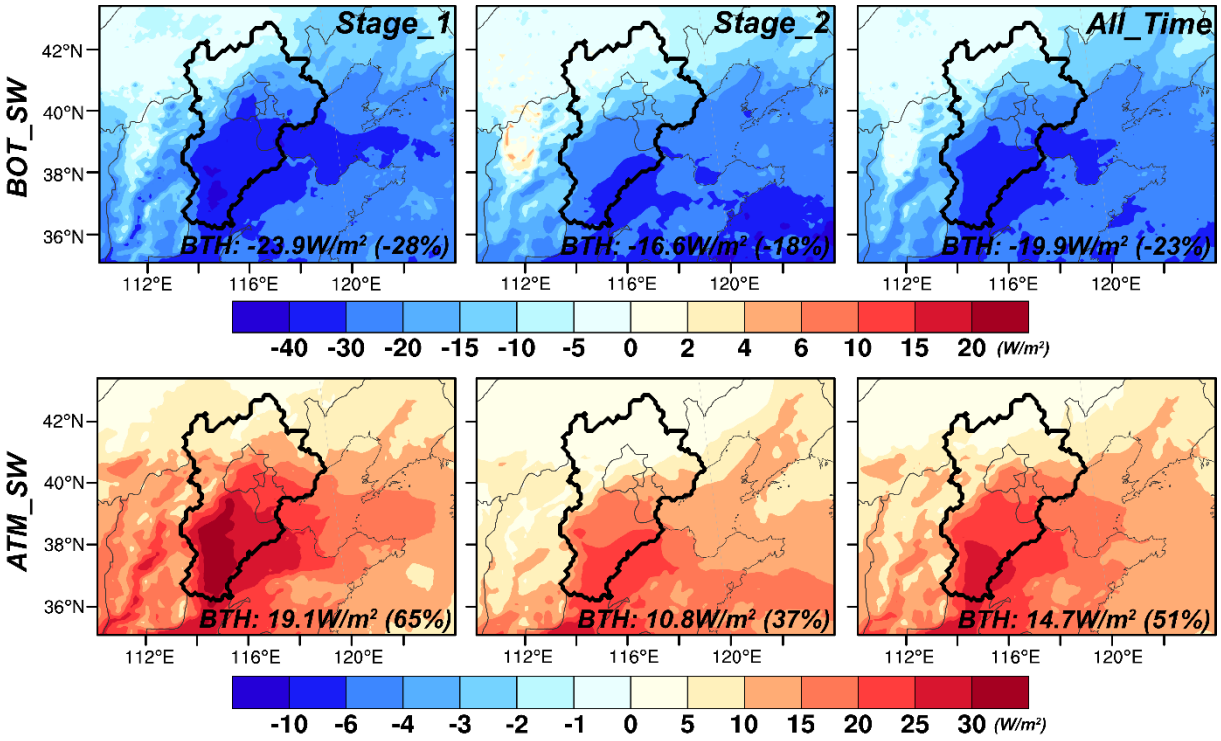

Figure 9. The differences in simulated all-sky radiative forcing (W m^{-2}) between CTL and NoARE cases (CTL minus NoARE) averaged over Stage_1, Stage_2, and the whole simulation period. "BOT_SW" and "ATM_SW" denote the downward shortwave radiative flux at the surface and in the atmosphere, respectively. The calculated differences in the simulated radiative forcing averaged over Beijing-Tianjin-Hebei for each stage are also shown at the bottom of each panel.

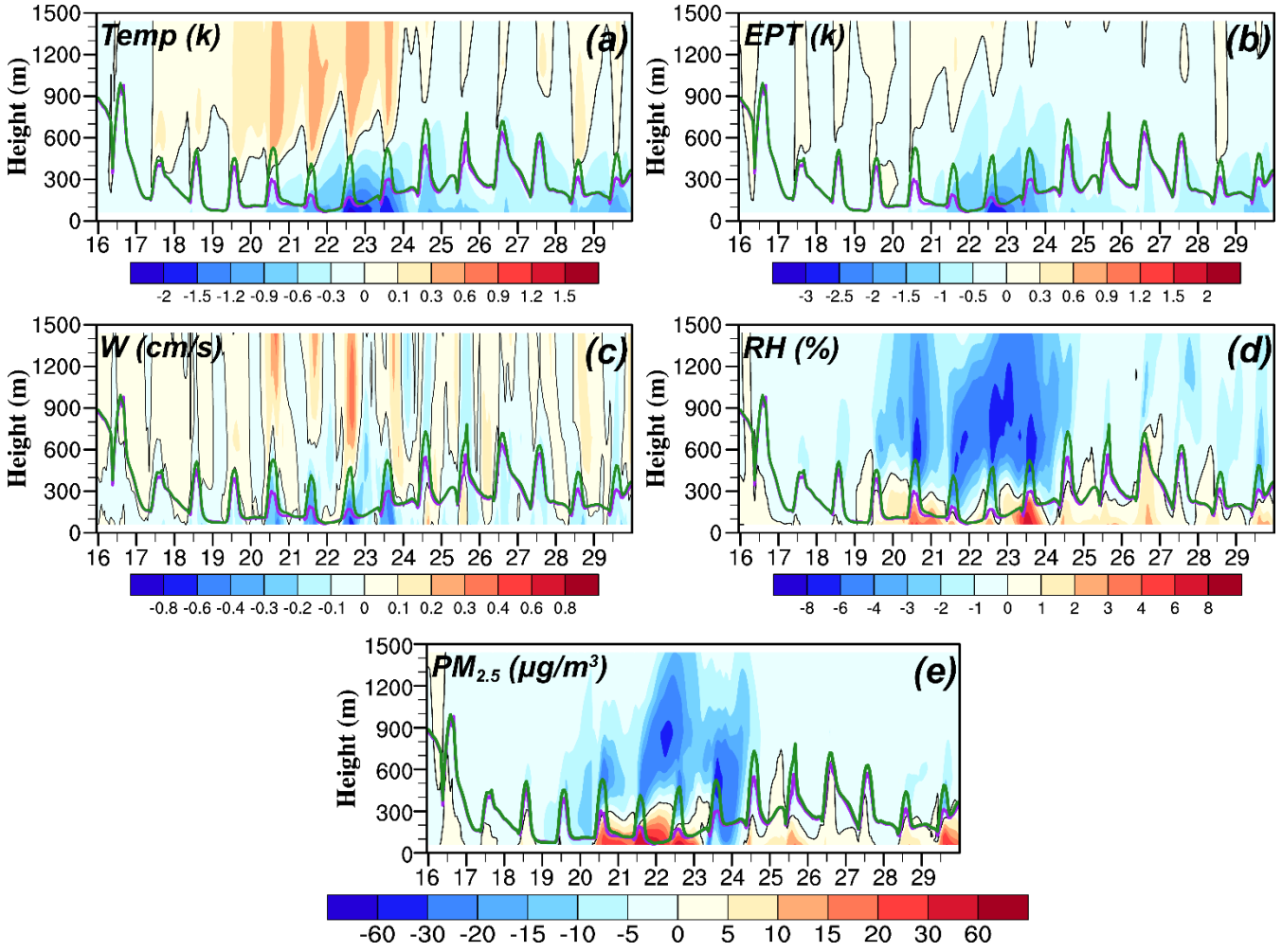

Figure 10. Time series of differences in (a) temperature (k), (b) equivalent potential temperature (k), (c) vertical wind speed (cm s^{-1}), (d) relative humidity (%), and (e) $PM_{2.5}$ concentration (μg m^{-3}) between CTL and NoARE cases (CTL minus NoARE) averaged over the Beijing-Tianjin-Hebei region. The purple and green lines denote the simulated PBLH in CTL and NoARE cases, respectively. The black line represents the zero contour line.

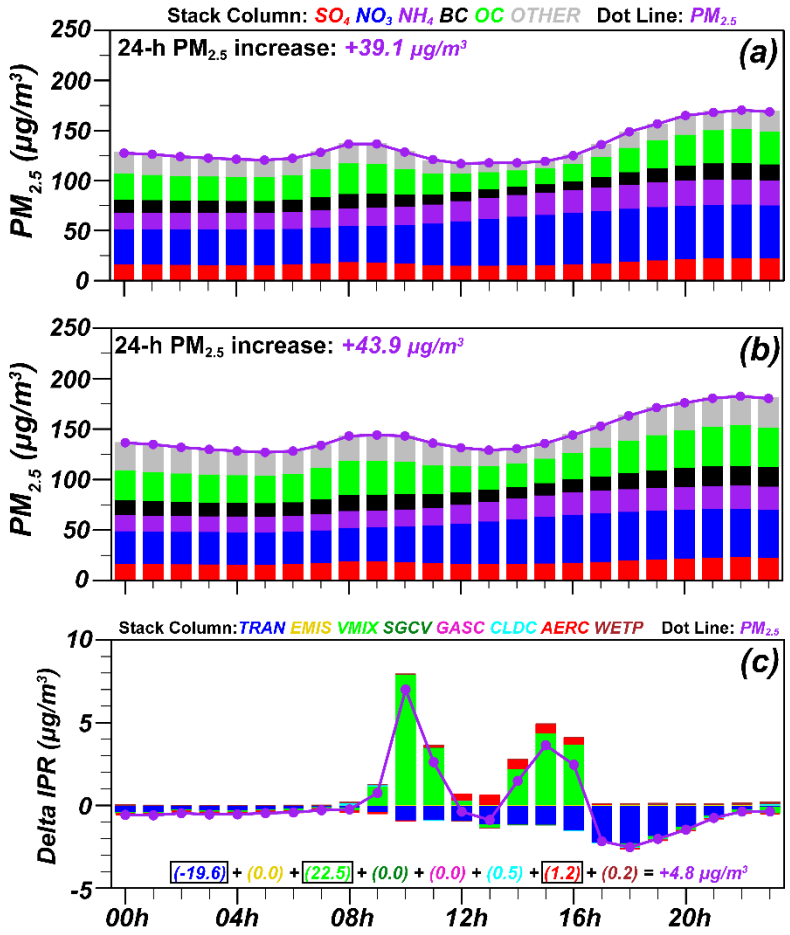

Figure 11. Diurnal variations of the near-surface $PM_{2.5}$ concentrations in (a) NoARE and (b) CTL simulations averaged over the Beijing-Tianjin-Hebei region during Stage_1 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is the 24-h increase in $PM_{2.5}$ concentration (23:00LST minus 00:00LST). (c) Differences in hourly IPRs caused by aerosol radiative forcing (CTL minus NoARE). The numbers listed in (c) represent the contributions of each process to the change in 24-h $PM_{2.5}$ increase caused by aerosol radiative forcing.

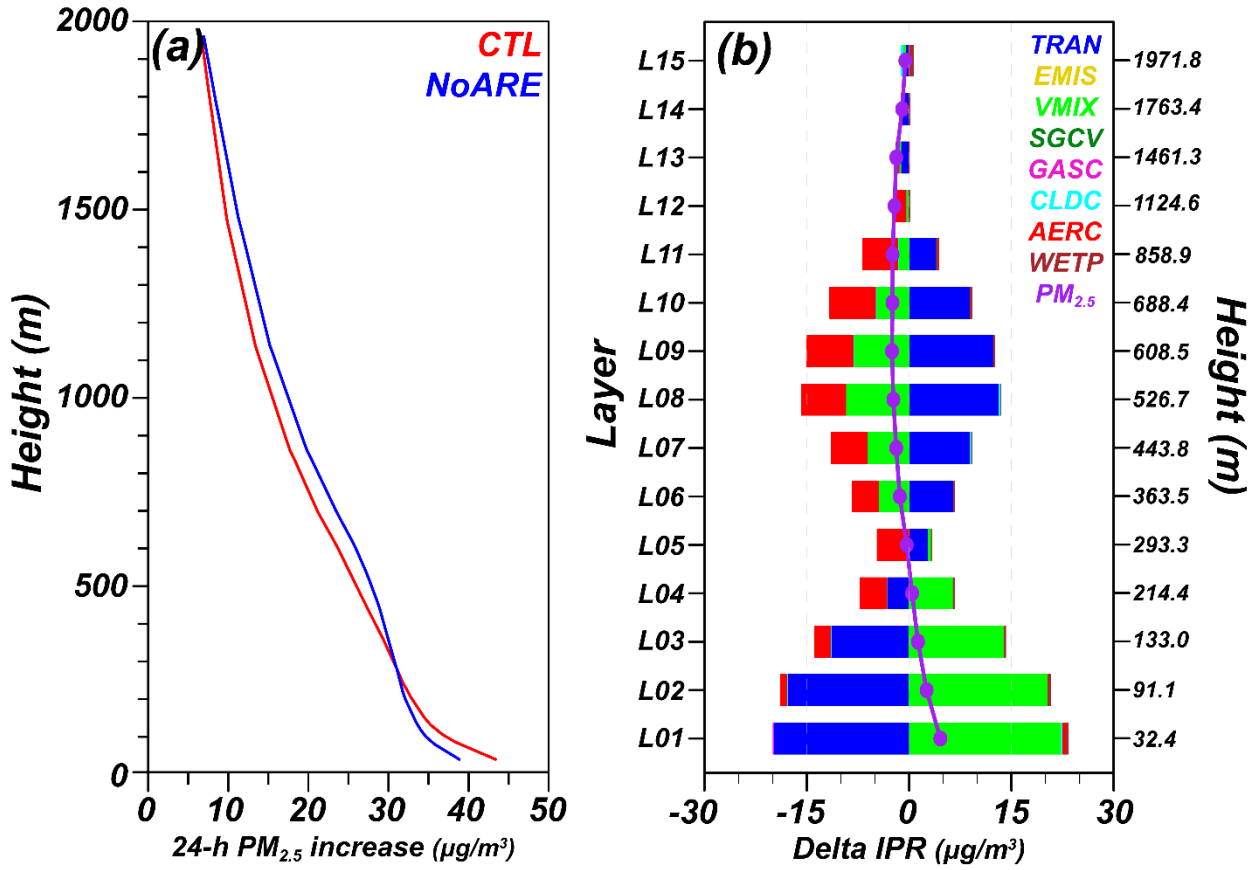

Figure 12. (a) Vertical profiles of the 24-h increases in $PM_{2.5}$ concentrations (23:00LST minus 00:00LST) averaged over Beijing-Tianjin-Hebei during Stage_1 in CTL and NoARE cases. (b) Vertical profiles of the differences in the 24-h $PM_{2.5}$ increases caused by aerosol radiative effect (CTL minus NoARE, as show in purple dot line), and the contributions of each physical/chemical process (as shown in colored bars).