

Response to Comments of Reviewer #1

(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

In this study, the authors examined the formation and evolution mechanisms of a haze event happened over Beijing–Tianjin–Hebei (BTH) in December 2015 using WRF-Chem model with a newly developed integrated process rate (IPR) analysis technique. They found that the $PM_{2.5}$ increase during aerosol accumulation stage was mainly attributed to strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes, and the restrained vertical mixing could be the primary reason for near-surface $PM_{2.5}$ increase when aerosol radiative feedback was considered. This study is interesting, and the results are solid. IPR technique provides a fundamental information of the physical/chemical processes of aerosol change. The manuscript is well written. I would suggest publication after addressing my comments below.

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. *Through IPR analysis, the authors found that $PM_{2.5}$ increase in the stage_1 was due to the increased aerosol chemical production and the decreased vertical mixing removal, but they did not explain why aerosol chemical production increased/vertical mixing removal decreased during this period.*

Response to why aerosol chemical production was increased:

Thanks for the reviewer's suggestion. In this manuscript, aerosol chemistry (AERC) process refers to microphysical nucleation, condensation, and coagulation, as well as the mass transfer between the gas phase and condensed phase.

The increased 24-h $PM_{2.5}$ change (23:00LST minus 00:00LST) due to AERC ($+29.6 \mu\text{g m}^{-3}$) over Beijing-Tianjin-Hebei (BTH) during the haze formation stage (December 16-22, Stage_1) was mainly attributed to the production of secondary inorganic aerosols, e.g., sulfate (SO_4^{2-} , $+2.7 \mu\text{g m}^{-3}$), nitrate (NO_3^- , $+16.8 \mu\text{g m}^{-3}$), and ammonium (NH_4^+ , $+9.8 \mu\text{g m}^{-3}$).

In order to explain why AERC was larger in Stage_1 than that in Stage_2 (December 23-27, aerosol dispersion stage), two metrics (sulfur oxidation ratio (SOR), $\text{SOR} = n\text{SO}_4^{2-} / (n\text{SO}_4^{2-} + n\text{SO}_2)$, and nitric oxidation ratio (NOR), $\text{NOR} = n\text{NO}_3^- / (n\text{NO}_3^- + n\text{NO}_2)$, n refers to the molar concentration) can be used to estimate the degree of secondary formation of SO_4^{2-} and NO_3^- (Sun et al., 2006; Zhao et al., 2013). Figure R1 shows the time series of calculated SOR and NOR averaged over the BTH region from 16 to 29 December 2015. When SOR and NOR are less than 0.1, SO_4^{2-} and NO_3^- mainly come from the primary source emissions, otherwise, high

oxidation rates of SOR and NOR can result in large fractions of SO_4^{2-} and NO_3^- in $\text{PM}_{2.5}$ (Fu et al., 2008). The mean SOR and NOR in Stage_1 (0.25 and 0.23, respectively) are larger than that in Stage_2 (0.23 and 0.19, respectively), indicating more aerosol particles can be produced in Stage_1.

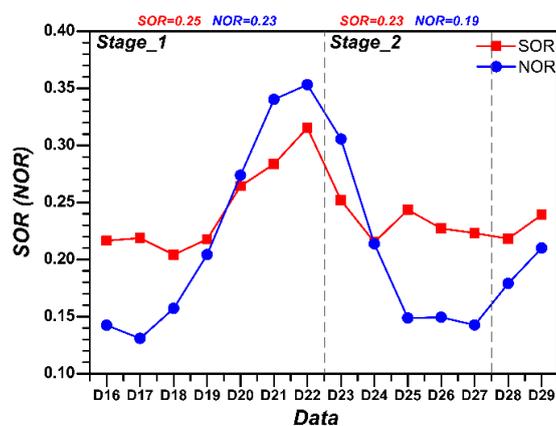


Figure R1. Time series of SOR (sulfur oxidation ratio, shown in red) and NOR (nitric oxidation ratio, shown in blue) averaged over the BTH (Beijing-Tianjin-Hebei) region from 16 to 29 December 2015.

Response to why vertical mixing removal was decreased:

In WRF-Chem, dry deposition (DRYD) is intermingled with vertical turbulent diffusion (DIFF), and the sum of DRYD and DIFF can reflect the role of vertical mixing (VMIX) in relocating the airborne pollutants vertically (Tao et al., 2015).

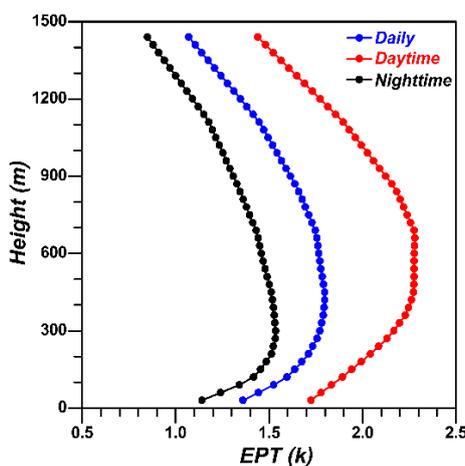


Figure R2. Differences in vertical profiles of daily equivalent potential temperature (EPT, shown in blue), EPT in the daytime (09:00LST-17:00LST, shown in red), and EPT at night (18:00LST-08:00LST, shown in black) between Stage_1 (16-22 December) and Stage_2 (23-27 December) averaged over the BTH region.

Under general conditions, VMIX can take place due to any inhomogeneous heating of the atmosphere (Li et al., 2017), which means VMIX is influenced by atmospheric stability, and the atmosphere stability can be exactly characterized by the profile of equivalent potential temperature (EPT) (Bolton, 1980; Zhao et al., 2013; Yang et al., 2016). If EPT rises with height, then the atmosphere is stable.

Figure R2 shows the differences in vertical profiles of EPTs between Stage_1 and Stage_2 averaged over the BTH region. The calculated daily EPT, ETP in the daytime (09:00LST-17:00LST) and EPT at night (18:00LST-08:00LST) in Stage_1 are all larger than that in Stage_2, and the differences are increased with the height (below ~400 m for the daily difference, below ~500 m in the daytime, and below ~300 m at night). All these indicate that the atmosphere is more stable during the pollution accumulation period, and the stabilized atmospheric layer will weaken VMIX to make more air pollutants accumulate in the near-surface layer in BTH.

2. *In Section 4.1, the authors used changes in synoptic conditions and atmospheric circulation to explain the aerosol variation, which seems not consistent with the IPR results.*

Response:

Thanks for the reviewer’s suggestion. The developed IPR (integrated process rate) analysis scheme adopted in this manuscript is used to quantify the contributions of each physical/chemical process to the variation in PM_{2.5} concentrations. We mainly focus on the pollution accumulation (dispersion) period to reveal important factors that cause the increase (decrease) in concentrations of aerosol particles. So firstly, we should reproduce the evolution of the daily PM_{2.5} concentrations during 16-29 December 2015, aiming to divide the severe haze event into aerosol accumulation stage and aerosol dispersion stage according to the average PM_{2.5} concentration in BTH, which can be significantly influenced by atmospheric circulation and synoptic conditions.

3. *The specific humidity did not have a visible change during Dec.20-22, so the aerosol chemical production was not due to the humidity. Probably it was due to increases in aerosol precursors from regional transport.*

Response:

We totally agree with the reviewer’s opinion. Figure R3 shows the diurnal variation of PM_{2.5} concentrations averaged over the BTH region during 20-22 December 2015. The PM_{2.5} concentration is increased by 50.4 μg m⁻³ (from 237.0 μg m⁻³ at 00:00LST to 287.4 μg m⁻³ at 23:00LST), and the contribution of AERC to the 24-h PM_{2.5} increase is +43.8 μg m⁻³, which contains the contributions from aerosol chemical productions of SO₄²⁻ (+2.6 μg m⁻³), NO₃⁻ (+27.0 μg m⁻³), and NH₄⁺ (+14.1 μg m⁻³).

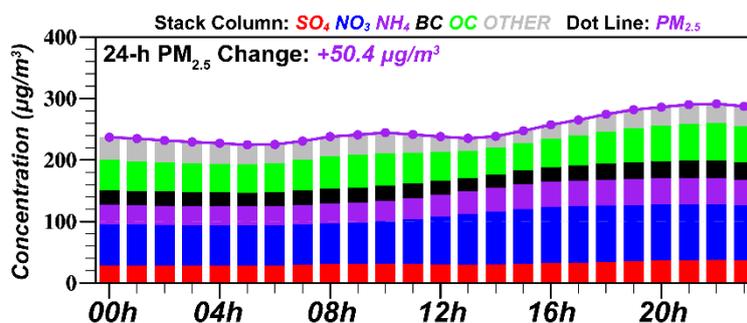


Figure R3. Diurnal variations of PM_{2.5} concentrations averaged over the BTH (Beijing-Tianjin-Hebei) region during 20-22 December 2015 (shown by purple dot line). The colored bars represent different components.

In this manuscript, AERC refers to microphysical nucleation, condensation, and coagulation, as well as the mass transfer between the gas phase and condensed phase. So the increased concentrations of SO_4^{2-} , NO_3^- and NH_4^+ due to AERC may mainly come from the gas-particle partition. As shown in Fig. R1, higher oxidation rates of SOR and NOR are calculated during December 20-22, indicating more SO_4^{2-} and NO_3^- can be produced from the gas-phase and/or liquid-phase oxidation of SO_2 (sulfur dioxide) and NO_2 (nitrogen dioxide).

4. *Although the model reproduced well the $\text{PM}_{2.5}$ variation during the haze event, it strongly underestimated concentrations of most aerosols, but overestimated nitrate concentration (Figure 5). However, during stage_1, it looks that most $\text{PM}_{2.5}$ increase is due to nitrate increase (Figure 8a). The authors need to discuss the potential influences of model biases on the results.*

Response:

Thanks for the reviewer's suggestion. The biases in BC (black carbon) may due to the uncertainty in primary emissions (Li et al., 2017). Secondary organic aerosols are not considered in the MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol module (Qiu et al., 2017), which makes the simulated OC (organic carbon) concentrations lower. Missing oxidation mechanisms (including gas-phase and aqueous-phase oxidation, as well as heterogeneous chemistry) of SO_2 may result in the under-predicted sulfate concentration, which also allows for excess nitrate in the presence of ammonia (Gao et al., 2014; Chen et al., 2016). Meanwhile, there may be an issue with NO_x partitioning and/or missing NO_x sink in current air quality models (Chen et al., 2019), making WRF-Chem overpredict the nitrate concentration. It should be noted that similar biases can also be found in other WRF-Chem studies (Zhang et al., 2015; Qiu et al., 2017).

Although underestimation/overestimation of aerosol compositions are simulated by the WRF-Chem model at the sites in Beijing (39.97 °N, 116.37 °E) and in Shijiazhuang (38.03 °N, 114.53 °E), the predicted total $\text{PM}_{2.5}$ surface concentrations in each city capture the observations pretty well (Fig. 4), especially the evolution characteristics during the whole simulation period. What's more, the main purpose of this manuscript is to investigate the evolution mechanisms of the total $\text{PM}_{2.5}$ concentrations during a haze event over BTH, including the contributions of each detailed physical or chemical process to the variations in the total $\text{PM}_{2.5}$ concentration.

According to the reviewer's suggestion, a discussion has been added in the revised manuscript as follows "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". (Page 20, Line 4-8)

Specific Comments:

1. *Page 1 Line 23: Delete 'happened'.*

Response:

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

2. *Page 1 Line 24 and following parts: Please change ‘outside transport’ to ‘regional transport’.*

Response:

Thanks for your suggestion. We have changed the expression in the whole revised manuscript.

3. *Page 3 Line 12: Delete ‘even’.*

Response:

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

4. *Page 4 Line 5: Change ‘contribution’ to ‘contributor’.*

Response:

According to the reviewer’s suggestion, we have changed the expression in the revised manuscript. **(Page 4, Line 4)**

5. *Page 4 Line 14: Recent studies found that black carbon-East Asian winter monsoon interactions and dust-wind interactions can also intensify winter haze in eastern China (e.g., Yang et al., 2017; Lou et al., 2019). The authors may would like to cite these studies.*

Response:

Thanks for the reviewer’s suggestion. Related references have been added in the revised manuscript.

“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)”. **(Page 4, Line 10-13)**

“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)”. **(Page 4, Line 17-19)**

6. *Page 8 Line 19: What does emission source mean and how it affects aerosol variation with 24-hr (Figure 8). Does the model include diurnal variation of emission? Why contributions of EMIS are different between stage_1 and stage_2?*

Response to what does emission source mean and how it affects aerosol variation with 24-hr (Figure 8):

Emission source (EMIS) means the primary emissions from anthropogenic emission, biogenic emission, biomass-burning emission, dust emission, sea-salt emission and so on. The emitted aerosols from primary emission sources can increase the near-surface PM_{2.5} concentrations significantly.

Response to does the model include diurnal variation of emission:

According to Chen et al. (2019), the diurnal and weekly variations of anthropogenic emission factors for each sector (power, industry, residential and transportation) are adopted in this

manuscript (Fig. R4).

Response to why contributions of EMIS are different between stage_1 and stage_2:

Thanks for your suggestion. The daily EMIS in Stage_1 and Stage_2 should be the same, and we have recalculated the 24-h values ($261.2 \mu\text{g m}^{-3}$) during the adjusted simulation period (Stage_1: December 16-22; Stage_2: December 23-27).

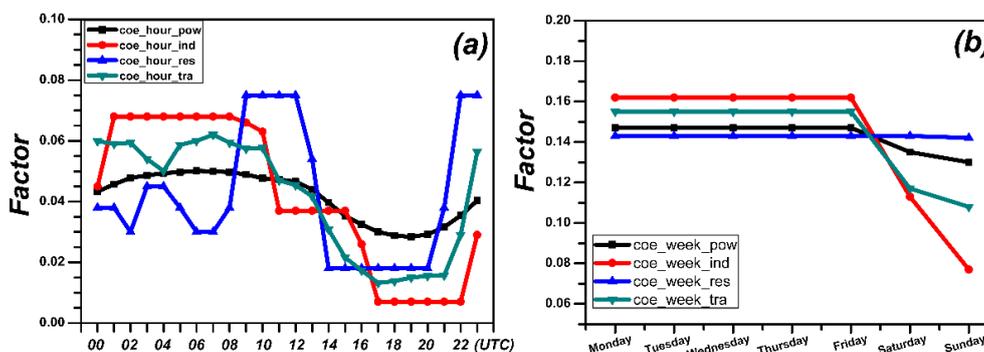


Figure R4. (a) Diurnal and (b) weekly variations of anthropogenic emission factors for power, industry, residential and transportation sectors.

7. Page 9 Line 19: Change 'closing' to 'turning off'.

Response:

According to the reviewer's suggestion, we have changed the expression in the revised manuscript. (Page 9, Line 18)

8. Page 11 Line 4: How these matrices calculated?

Response:

According to the reviewer's suggestion. The detailed calculation methods about the statistics are described in Table 3 in the revised manuscript. (Page 30)

9. Page 14 Line 14-22: Please rephrase this paragraph by illustrating absolute change first then percentage change (relative to what?). And what are the rest of contribution, from natural emission or emission outside the domain?

Response:

Thanks for your suggestion. In Fig.7, the unit of absolute contribution is " $\mu\text{g m}^{-3}$ ", and the absolute contribution of local emission (regional transport) to the $\text{PM}_{2.5}$ concentration in BTH can be expressed as the difference between OnlyBTH_Anth and NoAnth (the difference between NoBTH_Anth and NoAnth). For percentage contribution or relative contribution, the unit is "%", and it can be calculated by dividing the absolute contribution by the simulated $\text{PM}_{2.5}$ concentration from CTL. Meanwhile, according to the comments from Reviewer#2, the expressions of "absolute $\text{PM}_{2.5}$ concentrations" have been changed to " $\text{PM}_{2.5}$ concentrations" in the revised manuscript.

The rest of the contribution (represented by "Others" in Fig.7) to the $\text{PM}_{2.5}$ concentrations in

BTH may include the impacts from biogenic emission, biomass-burning emission, natural emission, aerosols outside the simulation domain, and the non-linear chemical formation for secondary aerosol.

According to the reviewer's suggestion, we have revised the paragraph as follows "As shown in Fig. 7(a), the PM_{2.5} concentration in BTH during Stage₁ 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 Stage₂, 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 Stage₂ 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 Stage₁ (1.7 g/kg) than that in Stage₂ (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." (Page 14-15, Line 19-28)

10. Page 15 Line 7: Please clarify that the dominant sources of surface-layer PM_{2.5} 'variation'.

Response:

Thanks for the reviewer's suggestion. Figure S3 shows the weighted contributions of each physical/chemical process to hourly PM_{2.5} changes in Stage₁ and Stage₂. The weighted contributions can be calculated using the equation $\%PC_i = \frac{PC_i}{\sum_{i=1}^n |PC_i|}$ (Goncalves et al., 2009), where PC_i is the absolute contribution (μg m⁻³) (i.e., the change in PM_{2.5} concentration induced by process *i*), and %PC_i is the weighted contribution (%) of process *i*. Note that the sum of %PC_i for all processes may not be 100%, but the sum of abs(%PC_i) is exactly 100%.

From Fig. S3, we can find that EMIS and AERC are the major contributors to make the PM_{2.5} concentration increase. But the processes of TRAN (advection), DRYD (dry deposition) and DIFF (turbulent diffusion) make the PM_{2.5} concentration decrease. The impacts of other processes (e.g., wet scavenging (WETP), cloud chemistry (CLDC)) on the hourly PM_{2.5} variation can be negligible ($-5\% < \%PC_i < 5\%$).

In the early morning (00:00LST-05:00LST) in Stage₁ and Stage₂, TRAN is the major negative contributor to make the PM_{2.5} concentration decrease. But at noon (10:00LST-13:00LST), the major negative contributor is DIFF. During 07:00LST-08:00LST and 16:00LST-21:00LST, EMIS is the major positive contributor to make the PM_{2.5} concentration increase. Generally, TRAN, EMIS and DIFF are the dominant processes which affect the surface-layer PM_{2.5} variation.

11. Figure 7: Change 'special' to 'specific' in caption and figure.

Response:

Thanks for your suggestion. We have revised it in the whole revised manuscript.

12. *Figure 11: Change '18h' to '20h'.*

Response:

According to the reviewer's suggestion, we have changed the label in the revised figure. (Page 41)

Reference:

- Bolton, D.: The computation of equivalent potential temperature. *Monthly Weather Review*, 108, 1046-1053, 10.1175/1520-0493(1980)108<1046:TCOEPT>2.0.CO;2, 1980.
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Pschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions, *Atmospheric Chemistry and Physics*, 15, 2969-2983, 10.5194/acp-15-2969-2015, 2015.

Thank you very much for your comments and suggestions.

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.

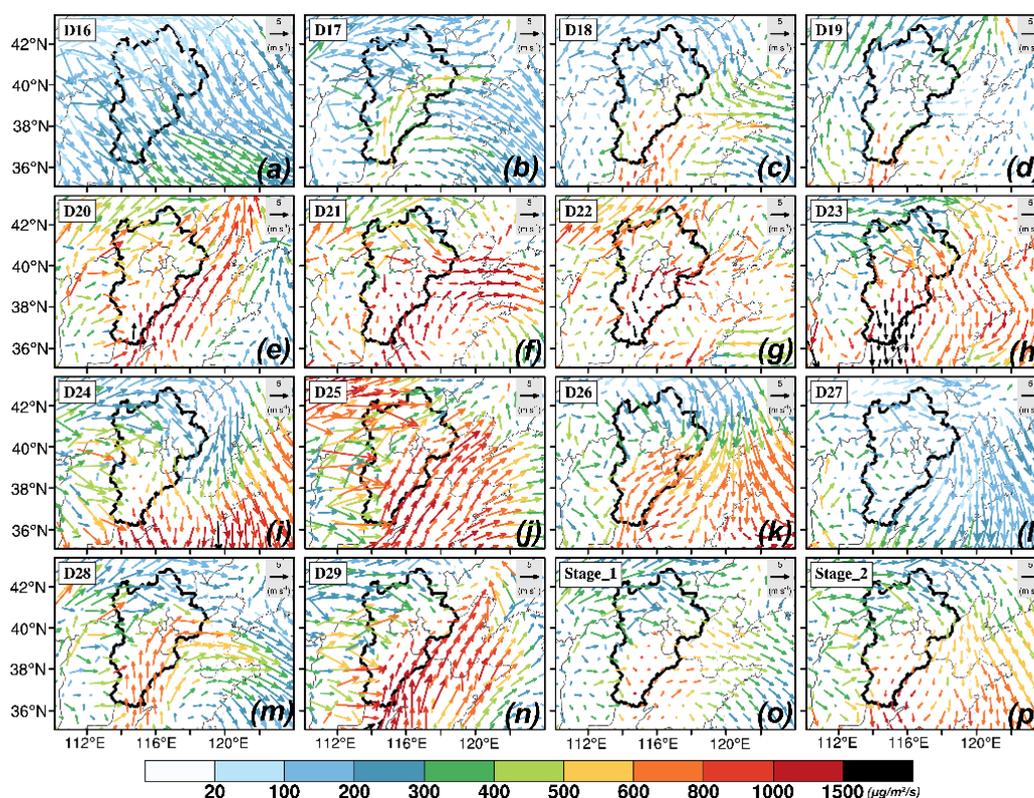


Figure R1. (a-n) Spatial distribution of simulated daily transport fluxes of $PM_{2.5}$ during 16-29 December 2015. The average $PM_{2.5}$ flux in Stage_1 (December 16-22) and Stage_2 (December 23-27) are also shown in (o) and (p).

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 %: PIL27. 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” PM_{2.5}. 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 PM_{2.5} 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., CO₂ and SO₂) 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).

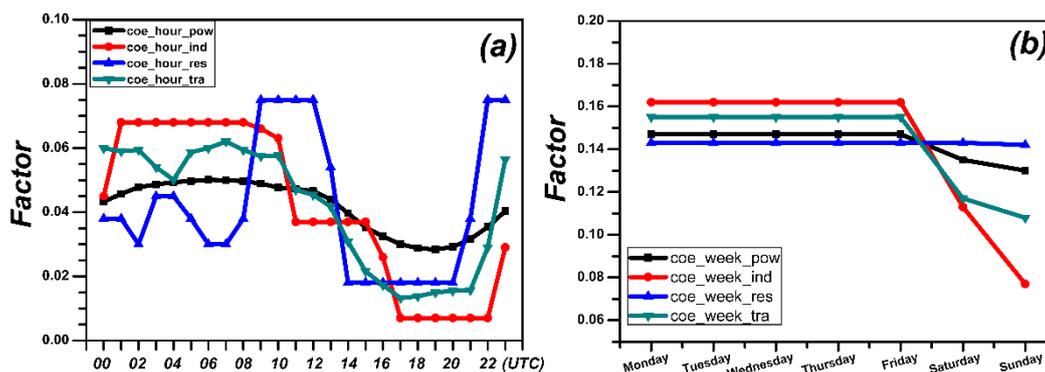


Figure R2. (a) Diurnal and (b) weekly variations of anthropogenic emission factors for power, industry, residential and transportation sectors.

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 $\text{NO}_2 + \text{OH}$.

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 $\text{NO}_2 + \text{OH} \rightarrow \text{HNO}_3$. At nighttime, N_2O_5 gas can react with vapor phase of H_2O (gas-phase reaction, the heterogeneous reaction of N_2O_5 ($\text{N}_2\text{O}_5 + \text{H}_2\text{O}(\text{l}) \rightarrow 2\text{HNO}_3(\text{l})$) 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 $\text{PM}_{2.5}$). This is why the calculated contributions of GASC to $\text{PM}_{2.5}$ 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 been revised as follows:

“Table 2 summarizes the experimental designs. To investigate the contributions of regional transport and local emission to the $\text{PM}_{2.5}$ 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 (T₂), 2 m relative humidity (RH₂), 10 m wind speed (WS₁₀) and 10 m wind direction (WD₁₀), 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 SO₄²⁻, NO₃⁻, NH₄⁺, 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 SO₄²⁻, NO₃⁻ and NH₄⁺. The biases in simulated concentrations of SO₄²⁻, NO₃⁻ and NH₄⁺ 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 drawn 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., O₃ 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)

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Thank you very much for your comments and suggestions.

Response to Comments of Reviewer #3

(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

General Comments:

This study investigated a haze event over BTH region in December 2015 with the process analysis method. The study examined the mechanisms underlying the event formation and evolution. They found the event was mainly controlled by the change of vertical mixing. In the end, the study also found that the vertical mixing and transport were two main processes that were responsible for the aerosol radiative feedback. The manuscript is well-written. However, the main point of processing analysis alone is not novel at all. Many previous studies have used this method in multiple air quality models, including WRF-Chem. The study only selected one event as the analysis case. Although the study found that the vertical mixing was the main contributor to the formation and evolution of the event, I didn't find anything new brought to the community. Besides these general comments, I also have some specific comments.

Response:

Thanks to the reviewer for the valuable comments and suggestions which are very helpful for us to improve our manuscript.

Process analysis techniques (i.e., integrated process rate (IPR) analysis) (Gipson, 1999) have already been fully implemented in CMAQ model (Community Multi-scale Air Quality model), and this air quality model with the IPR analysis has been widely used to analyze the formation and evolution mechanisms of ozone episodes (Goncalves et al., 2009; Khiem et al., 2010; Xing et al., 2017; Tang et al., 2017) and aerosol pollutions (Liu et al., 2011; Yang and Shiang-Yuh, 2013; Fan et al., 2014). However, the IPR analysis has not yet been officially adopted in the WRF-Chem model (Weather Research and Forecasting-Chemistry model) (Tao et al., 2015). Although several WRF-Chem model studies have used the method to investigate the impacts of physical/chemical processes on variations in O₃ concentrations (Jiang et al., 2012; Gao et al., 2018), few studies conducted the IPR analysis with WRF-Chem for aerosols.

Meanwhile, China has been suffering from serious haze pollutions, especially in the North China Plain in winter (Han et al., 2014; Gao et al., 2015; Sun et al., 2016). Extensive studies have been carried out to investigate the formation mechanisms underlying haze episodes (Liu et al., 2017; Wang et al., 2017; Kang et al., 2019; Wu et al., 2019), and try to explore possible solutions to improve air quality (Chen et al., 2019; Wang et al., 2019). But conclusions from previous studies can only reflect the combined effects of all physical and chemical processes. Detailed information of the impacts of individual process on haze events is usually unavailable.

Therefore, we develop an improved IPR analysis scheme in the WRF-Chem model to isolate nine physical/chemical processes impacting variations in aerosol concentrations and track their contributions quantitatively. The nine different processes are 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). We then use the IPR analysis to investigate the formation and evolution mechanisms of a severe haze event over Beijing-Tianjin-Hebei (BTH) during 16 to 29 December 2015, including analyzing the influence mechanisms of aerosol radiative forcing. Some results can be summarized as follows: (1) the $PM_{2.5}$ increase over BTH during the haze formation stage (December 16 to 22) may be mainly attributed to strong production by aerosol chemistry process and weak removal by advection and vertical mixing processes; (2) the restrained vertical mixing can be the primary reason for the enhancement in near-surface $PM_{2.5}$ increase when aerosol radiative forcing is considered.

In our future studies, multiple haze events during 2013 to 2018 will be further analyzed by using the IPR analysis to figure out the major physical/chemical process behind these pollutions.

Specific Comments:

- 1. The study selected the event of Dec. 20-29. Based on the result, it seems that the $PM_{2.5}$ concentration reached $200 \mu\text{g}/\text{m}^3$ on average on Dec. 20. To better show the formation of event, the simulation and analysis should start from the earlier date to demonstrate the concentration rising from a lower level such as $50 \mu\text{g}/\text{m}^3$ as shown in the end of this event.*

Response:

According to the reviewer's suggestion, simulation period has been extended from 13 to 29 December 2015, and the simulation results during December 16-29 are analyzed. Figure R1 shows the time series of simulated daily $PM_{2.5}$ concentrations averaged over the BTH region during December 16-29. The $PM_{2.5}$ concentration over BTH in December 16 is $24.2 \mu\text{g m}^{-3}$.

Due to the updated simulation period, all the results have been re-calculated in the revised manuscript. Here shows the revised 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 $24.2 \mu\text{g m}^{-3}$ to $289.8 \mu\text{g m}^{-3}$, 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 $107.9 \mu\text{g m}^{-3}$, 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 $+43.9 \mu\text{g m}^{-3}$ during Stage_1 and $-41.5 \mu\text{g m}^{-3}$ during Stage_2. Contributions of aerosol chemistry, advection and vertical mixing to the 24-h change were $+29.6 (+17.9) \mu\text{g m}^{-3}$, $-71.8 (-103.6) \mu\text{g m}^{-3}$ and $-177.3 (-221.6) \mu\text{g m}^{-3}$ 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 $4.8 \mu g m^{-3}$ during Stage_1, which could be mainly attributed to the contributions of vertical mixing process ($+22.5 \mu g m^{-3}$), advection process ($-19.6 \mu g m^{-3}$) and aerosol chemistry process ($+1.2 \mu g m^{-3}$). The restrained vertical mixing was the primary reason for the enhancement in near-surface $PM_{2.5}$ increase when aerosol radiative forcing was considered". (Page 1-2)

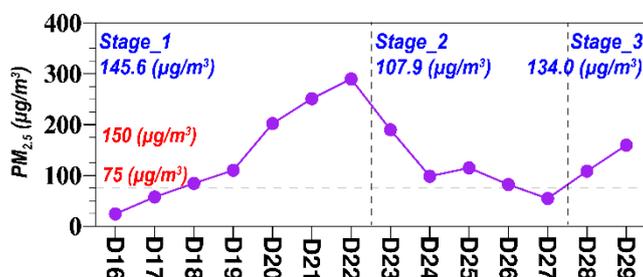


Figure R1. Time series of simulated daily $PM_{2.5}$ concentrations averaged over the Beijing-Tianjin-Hebei region from 16 to 29 December 2015.

- Line 19-21 of page 6, if the FDDA is turned on for the control and noARE experiments, I do expect some aerosol meteorological feedbacks can be diminished by the FDDA. Free runs without FDDA is preferred for studying aerosol feedback.

Response:

Thanks for the reviewer’s suggestion, and we totally agree with your opinion. Following Feng et al. (2016), Mar et al. (2016) and Werner et al. (2016), four-dimensional data assimilation (FDDA) is only applied to the first domain in this manuscript, and no analysis nudging is included for the inner second domain.

Here is the revised sentence “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)”. (Page 6, Line 21-23)

- Line 11-12 of page 7, the MOSAIC aerosol mechanism in WRF-Chem has not been coupled with the Shao dust emission scheme, at least in the publicly released version. If it was coupled in this study or any previous studies, please briefly introduce it and cite the related references.

Response:

Thanks for the reviewer’s suggestion. The Shao 2004 dust emission scheme (referred to as Shao_2004) is proposed by Shao (2004) and implemented in WRF-Chem by Kang et al. (2011) and Wu and Lin (2013). Previous studies have reported that Shao_2004 scheme shows a good performance in dust emission amounts over source areas, including the spatial distribution of dust particles over the downwind regions over East Asia (Wu and Lin, 2013; Kang et al., 2014; Su and Fung, 2015).

Three important parameters are used to calculate the dust emission amounts: (1) the threshold friction velocity, (2) the horizontal sand flux, and (3) the vertical dust flux. The threshold friction

velocity is defined as the minimum friction velocity to initiate soil particle movement. It can be parameterized by the cohesive force which is proportional to particle size (Shao and Lu, 2000), and it is strongly affected by soil moisture, salt concentrations in the soil, and roughness elements on the surface. The horizontal sand flux indicates the intensity of dust saltation, defined as a vertical integral of the streamwise saltating particle flux density when the friction velocity exceeds the threshold friction velocity (White, 1979). The vertical dust flux is defined as the emitted dust mass concentration per unit area per unit time, and it is calculated using the equation proposed by Shao (2004). More detailed descriptions of the dust parameterization can be found in Shao (2004) and Kang et al. (2011).

Although in the publicly released version of the WRF-Chem model, the MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol mechanism has not been coupled with the Shao_2004 scheme, we have tried to make several modifications in the script of module_mosaic_addemiss.F (i.e., add a new subroutine to process the Shao dust emission scheme), aiming to equip the sectional aerosol mechanism with the new Shao_2004 scheme. Following Zhao et al. (2010), similar parameters are used to partition the total dust masses into eight bins in the MOSAIC module.

Analyzing the simulation results from Chen et al. (2018), the WRF-Chem model (the MOSAIC aerosol mechanism has been coupled with the Shao dust emission scheme) can successfully reproduce the spatiotemporal evolution of a dust storm happened during 14-17 April 2015.

4. *Line 18-24 of page 9, this part is confusing. In WRF-Chem, the aerosol-cloud interaction is linked with wet deposition and cloud aqueous chemistry. If aerosol-cloud interaction is turned off in this study, then the CTL and noARE experiments should use different chemistry mechanisms, i.e., noARE likely used the one without wet deposition and cloud chemistry. Please provide more details about this. If this is the case, the difference between the control and noARE should include not only the aerosol feedback but also the difference in chemical processes. Based on the results, it seems that there were little cloud and precipitation during the period. The major aerosol feedback is from aerosol-radiation interaction, therefore, it makes more sense that in noARE experiment only the aerosol radiative feedback is turned off with aerosol-cloud interaction not touched. Furthermore, we generally do not call aerosol-cloud interaction as aerosol radiative effects.*

Response:

We totally agree with the reviewer's comments that aerosol-cloud interactions include the cloud chemistry and wet scavenging.

In the WRF-Chem model, aerosol indirect effects (AIEs) (including the first and the second indirect effects) are implemented by linking simulated droplet number to radiation schemes (e.g., Rapid Radiative Transfer Model for Global Circulation Models (RRTMG) shortwave radiation scheme) and microphysics schemes (e.g., Purdue Lin Scheme) (Skamarock et al., 2008). The Lin scheme can predict the cloud droplet number (Chen and Sun, 2002), and the auto-conversion is dependent on it, following Liu et al. (2005). Aerosol particles acting as cloud condensation nuclei are coupled with the cloud physics portion of the model. This coupling allows for fully interactive feedbacks: aerosols affect cloud droplet number and cloud radiative properties, and clouds alter aerosol size and composition via aqueous processes and wet scavenging (Gustafson et al., 2007).

In order to access the impacts of aerosol-cloud interactions, the most common approach is to design a hypothetical scenario (i.e., NoAIE) with a prescribed distribution of cloud droplet number (Zhao et al., 2017), and the aerosol indirect effect (AIE) can be quantified by comparing the simulations results between the baseline scenario and NoAIE.

Following the suggestion provided by the WRF-Chem user's guide (https://ruc.noaa.gov/wrf/wrf-chem/Users_guide.pdf), AIE is turned off in this manuscript by using a prescribed vertically uniform cloud droplet number, which is calculated from the control case during the whole simulation period. Similar processing method can also be found in many other studies (Forkel et al., 2015; Kong et al., 2015; Zhang et al., 2015; Zhao et al., 2017).

5. *Line 11 of page 10, please provide the full name of NOAA READY GDAS. In addition, please provide more information about the PBL data from this dataset. Is it retrieval or direct observation? If it is retrieval, what is the method used for the retrieval?*

Response:

Thanks for your suggestion. The full names of NOAA, READY and GDAS are National Oceanic and Atmospheric Administration (NOAA), Real-time Environmental Applications and Display sYstem (READY), and Global Data Assimilation System (GDAS), respectively.

The meteorological data of planetary boundary layer height (PBLH) is extracted from the National Oceanic and Atmospheric Administration's (NOAA) Global Data Assimilation System (GDAS) data. As shown by Huang et al. (2012), PBL heights of GDAS agree well with the vertical lidar observations in Shanghai.

The National Weather Service's National Centers for Environmental Prediction (NCEP) runs a series of computer analyses and forecasts operationally. One of the operational systems is GDAS (Nemuc et al., 2012). GDAS can be assimilated by surface observations, balloon data, wind profiler data, aircraft reports, buoy observations, radar observations, and satellite observations (Rolph et al., 2017). GDAS is run four times a day at 00:00, 06:00, 12:00, and 18:00UTC. Model outputs include the analyses and forecast fields at three hours after each analysis (Wang et al., 2014). NCEP post-processing of the GDAS converts the data from spectral coefficient form to 1 degree latitude-longitude grids and from sigma levels to 23 pressure layers. More detailed information can be found at <https://www.ready.noaa.gov/gdas1.php>.

Generally, PBLH is calculated every 3-hour each day by the NOAA's READY Archived Meteorology online calculating program (<http://ready.arl.noaa.gov/READYamet.php>). This program can plot a time-series of calculated boundary layer depth using the chosen meteorological data. The calculations use the same equations as the NOAA HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) transport and dispersion model.

In HYSPLIT, there are two options to estimate the boundary layer stability. The preferred method is to use the fluxes of heat and momentum provided by the meteorological model, if available. Otherwise the temperature and wind gradients of each grid-point sounding are used to estimate stability. More information about the calculation method is described by Draxler and Rolph (2003).

According to the reviewer's suggestion, we have revised the sentence as follows "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>". (Page 10, Line 11-15)

6. *In the figures of hourly timeseries such as Fig. 3 or 4, please specify whether it is local time or UTC time? In Fig. 3, is there a low limit from PBL retrieval? It seems the values are limited to 50 m. The same is applied to the simulations. Any specific reason?*

Response:

Thanks for the reviewer's suggestion. The UTC time has been converted to China Standard Time by adding 8 h to Beijing Time in China. Related descriptions have been added in the revised figures (Fig. 3 and 4). (Page 33-34)

At night, the PBL height collapses into a shallow stable boundary layer, retrieved and simulated PBLHs are close to ~50 m and ~30 m, respectively. The low limit of the PBLH is chosen to correspond to the minimum height resolution (Draxler and Hess, 2004).

7. *Fig. 5 shows the aerosol components at the station of Shijiazhuang? Why not show the total PM2.5 surface concentration at this station as a reference? In addition, since the simulation seems capturing the hourly PM2.5 variation well, why not show the hourly component comparison instead of period average only? It would be interesting and provide useful information.*

Response:

Thanks for the reviewer's suggestion. The daily measurements of mass concentrations of SO_4^{2-} , NO_3^- , NH_4^+ , BC and OC are collected at the observation site (38.03 N, 114.53 E) in the city of Shijiazhuang, and these observations are provided by the Campaign on Atmospheric Aerosol Research network of China (CARE-China). More information about these chemical observations can be found in Huang et al. (2017) and Liu et al. (2018).

We totally agree with the reviewer's opinion, but we can only get the daily concentrations of these aerosol chemical compositions (SO_4^{2-} , NO_3^- , NH_4^+ , BC and OC) at the two sites ((39.97 N, 116.37 E) and (38.03 N, 114.53 E)). What's more, similar evaluation method can also be found in other studies (Gao et al., 2016; Qiu et al., 2017).

8. *What does the black line represent in Fig. 10?*

Response:

The black line in Fig. 10 means the zero contour line, which can be used to clearly distinguish the positive and negative differences. According to the reviewer's suggestion, we have revised the caption of Fig. 10 as follows "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) $\text{PM}_{2.5}$ concentration ($\mu\text{g m}^{-3}$) 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". (Page 40)

9. *In Fig. 11, the process analysis showed the averaged 24-h change of PM_{2.5} during the period. What does this mean? Why is the averaged 24-h change important? I think that the change through each stage of the event would be more interesting. Please clarify.*

Response to the question of “In Fig. 11, the process analysis showed the averaged 24-h change of PM_{2.5} during the period. What does this mean?”

Simulation results from 16 to 29 December 2015 are analyzed in this manuscript. According to the daily PM_{2.5} concentrations averaged over the BTH region (Fig. 6(l)), the life cycle of the haze event typically consists of the two stages: (1) aerosol accumulation stage (December 16-22, Stage_1), and (2) aerosol dispersion stage (December 23-27, Stage_2).

Take the first stage as an example, simulated hourly PM_{2.5} concentrations in BTH during December 16-22 are averaged into one day to show the mean diurnal variation of the near-surface PM_{2.5} concentrations, as listed in Figs. 11(a) and (b).

Analyzing Figs. 11(a) and (b), we can find that the PM_{2.5} concentration is increased by 39.1 $\mu\text{g m}^{-3}$ in the NoARE case, from 127.4 $\mu\text{g m}^{-3}$ at 00:00LST to 166.5 $\mu\text{g m}^{-3}$ at 23:00LST. However, the 24-h change is larger (+43.9 $\mu\text{g m}^{-3}$) in CTL case, from 136.5 $\mu\text{g m}^{-3}$ at 00:00LST to 180.4 $\mu\text{g m}^{-3}$ at 23:00LST. All these indicate that when aerosol radiative effects (ARE) are considered, the diurnal evolution of the simulated near-surface PM_{2.5} concentrations is enhanced (+39.1 $\mu\text{g m}^{-3}$ vs. +43.9 $\mu\text{g m}^{-3}$), which means more suspended aerosol particles will deteriorate the air quality in the next day in BTH.

In order to explain the enhancement of 4.8 $\mu\text{g m}^{-3}$ (12%) induced by ARE, the IPR analysis is used to track the contributions of each physical/chemical process, as shown in Fig. 11(c), and the restrained vertical mixing can be the primary reason for this enhancement.

Response to the question of “Why is the averaged 24-h change important? I think that the change through each stage of the event would be more interesting. Please clarify.”

Thanks for the reviewer’s suggestion. If the change of the PM_{2.5} concentrations through the entire aerosol accumulation stage is used to investigate the evolution mechanism of the haze event, only the values at the beginning time (00:00LST in December 16) and the finish time (23:00LST in December 22) are analyzed. Many useful information will be discarded, especially the daily evolution characteristics of the simulated PM_{2.5} concentrations in BTH.

In this manuscript, hourly PM_{2.5} concentrations during each stage are averaged into one day to show the mean diurnal variation of the near-surface PM_{2.5} concentration. Similar analytical method can also be found in many other studies (Fan et al., 2015; Tao et al., 2015; Xing et al., 2017).

10. *In Fig. 12b, using height (m) instead of model levels as the y-axis makes more sense.*

Response:

According to the reviewer’s suggestion, we have re-plotted the figure in the revised manuscript.
(Page 42)

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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 2016–22, 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 +50.443.9 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 +43.829.6 (+17.9) μg m⁻³, -71.8 (-103.6) μg m⁻³ and -161.6177.3 (-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 9.64.8 μg m⁻³ during Stage_1, which could be mainly attributed to the contributions of vertical mixing process (+39.822.5 μg m⁻³), advection process (-38.6-19.6 μg m⁻³) and aerosol chemistry process (+5.41.2 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 SO₂ to SO₄²⁻ was strongly associated with high relative humidity, and NO₃⁻ 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. (2018ba) 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 $\mu\text{g m}^{-3}$ 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 between 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, and so on) is necessary for fully understanding 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 2009–2013, 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 $\mu\text{g m}^{-3}$. 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 $\mu\text{g m}^{-3}$. 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.

5 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

10 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 15 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.

20 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 SO₂ (sulfur dioxide), NO_x (nitrogen oxides), CO (carbon monoxide), CO₂ (carbon dioxide), NMVOC (non-methane volatile organic compounds), NH₃ (ammonia), BC (black carbon), OC (organic carbon), PM_{2.5} and PM₁₀. 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 (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), 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.

5 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).

10 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 (O₃) 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 O₃ concentrations. Gao et al. (2018) investigated the impacts of BC-PBL interactions on O₃ 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 O₃ formation by using a simplified IPR analysis scheme.

20 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.,

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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.

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

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turned off by using ~~thea 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 13⁷ 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/READYmet.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 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)).~~

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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 ~~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 0.873 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 29.48% 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 –4.31% 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 0.768 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 evolutionary characteristics of the observed PM_{2.5} concentrations in the nine cities ($R_s=0.5857-0.8890$). 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 $\mu\text{g m}^{-3}$ 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 RH₂ and overestimated WS₁₀); (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 $\pm 30\%$ and MFE $\leq -50\%$) proposed by Boylan and Russel (2006).

Figure 5 compares the simulated and observed surface-layer concentrations of BC, OC, SO₄²⁻, NO₃⁻, and NH₄⁺ in Beijing and Shijiazhuang averaged during 2016-29 December 2015. WRF-Chem model underestimates the concentrations of SO₄²⁻, NH₄⁺ and BC, OC, NH₄⁺ and SO₄²⁻ concentrations in Beijing OC in Beijing (Shijiazhuang) by 3.6% (33.1%), 34.919% (38.640%), 24.14% (37.59%), and 32.521% (44.641%), respectively, and but overestimates the concentrations of NO₃⁻ concentration by 22.229% (51.844%). Due to the low reactivity of BC in the atmosphere, the uncertainty in BC emission may cause this underestimation the 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 SO₂ gas-phase and aqueous-phase oxidation, as well as heterogeneous chemistry may explain the underestimation of SO₄²⁻. 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)). 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 occurred gradually formed.

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

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whole simulation period into three stages: (1) aerosol accumulation stage (December ~~16-20~~, 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.

5 In Stage_1, the daily mean PM_{2.5} concentrations averaged over BTH increased from ~~209.024.2~~ $\mu\text{g m}^{-3}$ to 289.8 $\mu\text{g m}^{-3}$, and the average PM_{2.5} concentration was ~~250.0145.6~~ $\mu\text{g m}^{-3}$ (Fig. 7(a)), ~~far beyond close to~~ the air quality threshold value of “heavily polluted” (PM_{2.5} 24-h average concentration > 150 $\mu\text{g m}^{-3}$). The WS₁₀ 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>),
10 indicating that the near surface circulation was insufficient to disperse accumulated air pollutants. The decreased PBLH (from ~~448.6701.6~~ 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 $\mu\text{g m}^{-3}$, still exceeding the Grade II standard (75 $\mu\text{g m}^{-3}$) defined by the National Ambient Air Quality Standards of China.

15 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
20 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 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 ($107.9 \mu g m^{-3}$) 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 510.9% 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 0.9486.

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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 $50.443.9 \mu g m^{-3}$ (from $237.0136.5 \mu g m^{-3}$ at 00:00LST to $287.4180.4 \mu g m^{-3}$ at 23:00LST) during the period of particulate accumulation (Stage_1), but it decreased by $41.5 \mu g m^{-3}$ during the period of particulate elimination (Stage_2).

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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.

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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) $\mu g m^{-3}$, -71.8 (-103.6) $\mu g m^{-3}$ and -177.3 (-221.6) $\mu g m^{-3}$ for Stage_1 (Stage_2), respectively. ~~AERC and VMIX process to 24 h $PM_{2.5}$ changes were +43.8 (+17.9) $\mu g m^{-3}$ and -161.6 (-221.6) $\mu g m^{-3}$ for~~ Stage_1 (Stage_2), respectively. Small differences were found for contributions from other processes between Stage_1 and Stage_2 (differences smaller than ~~40.5~~ $\mu g m^{-3}$). 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

~~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.

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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_a-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 ~~36.528%~~ (~~31.623.9~~ W m⁻²) in Stage_1, ~~18.3%~~ (16.6 W m⁻²) in Stage_2, and ~~24.123%~~ (~~21.519.9~~ 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 ~~84.765%~~ (~~25.519.1~~ W m⁻²) in Stage_1, ~~37.4%~~ (10.8 W m⁻²) in Stage_2, and ~~53.951%~~ (~~15.714.7~~ W m⁻²) during the whole period, respectively.

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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_a-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_a-surface PM_{2.5} concentrations during December 23_a-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 $40.839.1 \mu g m^{-3}$ was simulated in NoARE case. When aerosol radiative forcing was considered, the 24-h increase of $PM_{2.5}$ concentration was $50.443.9 \mu g m^{-3}$. The enhancement of $9.64.8 \mu g m^{-3}$ (23.512%) 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 $+22.5 \mu g m^{-3}$ to the enhancement in 24-h $PM_{2.5}$ increase ($+4.8 \mu g m^{-3}$) for Stage_1. The TRAN process, however, contributed $-19.6 \mu g m^{-3}$. 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 $+1.2 \mu g m^{-3}$. 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 10:00 to 17:00LST in Stage_1 were $-0.5 \mu g m^{-3}$, $+5.91.3 \mu g m^{-3}$, and $+2.90.8 \mu g m^{-3}$, 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 $+6.34.1 \mu g m^{-3}$, 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 $\sim 400\text{--}300$ 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 3400 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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5 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. eoncentration. 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 evolutions 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 $\mu g m^{-3}$.

far beyond the threshold value of “heavily polluted”. The contributions of local emission and regional transport to the PM_{2.5} concentrations averaged over BTH were comparable (42.349% and 36.632%, respectively), meaning the combined effect together effect resulted in the high PM_{2.5} concentration in BTHs. 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 50.951% 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.

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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} concentrations. 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 50.443.9 μg m⁻³ (23:00LST minus 00:00LST) during Stage_1, but it decreased by 41.541.5 μg m⁻³ during Stage_2. Contributions of AERC, TRAN and VMIX (vertical mixing, the sum of DRYD and DIFF) process to the 24-h PM_{2.5} changes were +43.829.6 (+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.

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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 impact effects.

When aerosol radiative feedback was considered, the 24-h PM_{2.5} increase was enhanced by 9.64.8 μg m⁻³ (23.512%) during

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

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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 the 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 $\text{PM}_{2.5}$ 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.

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As Zheng et al. (2018) pointed out that the $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$) 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

5 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.

10 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) ^a	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}^b	\overline{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	-12.1	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) ^a	59	173.6	168.2	-3.1	132.7	47.3	0.869	0.768

带格式表格

^aT₂: temperature at 2 m (k); RH₂: relative humidity at 2 m (%); WS₁₀: wind speed at 10 m (m s⁻¹); WD₁₀: wind direction at 10 m (°).

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

带格式的：上标

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

带格式的：上标

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

带格式的：上标

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

10 ⁶IOA is the IOA; index of agreement; $IOA = 1 - \frac{\sum_{i=1}^{nstd} (SIM_i - OBS_i)^2}{\sum_{i=1}^{nstd} ((OBS_i - OBS_i) + (SIM_i - SIM_i))^2}$

⁷R is the R; correlation coefficient; $R = \frac{\sum_{i=1}^{nstd} (OBS_i - \overline{OBS}) \times (SIM_i - \overline{SIM})}{\sqrt{\sum_{i=1}^{nstd} (OBS_i - \overline{OBS})^2 + \sum_{i=1}^{nstd} (SIM_i - \overline{SIM})^2}}$

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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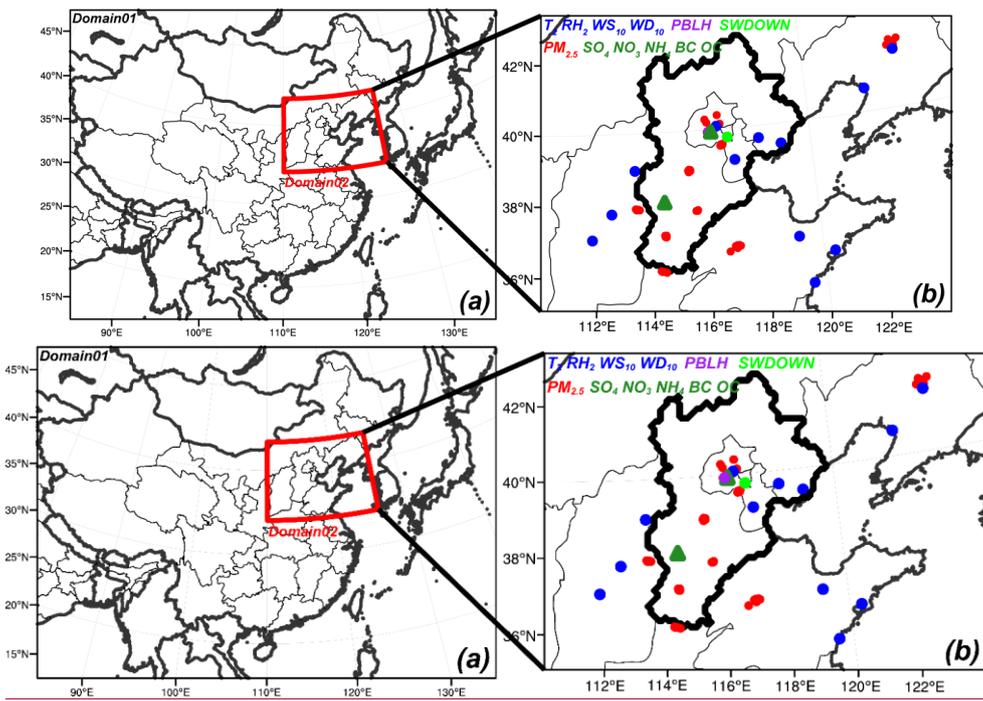
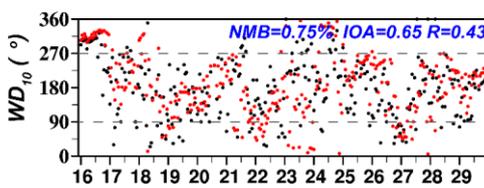
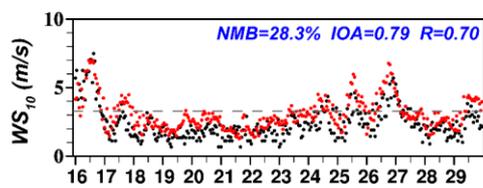
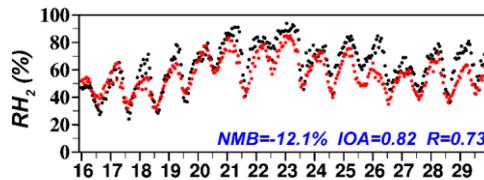
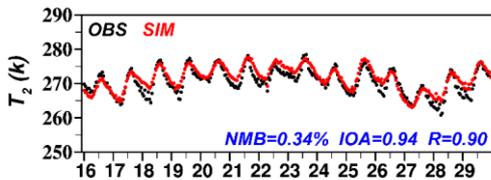
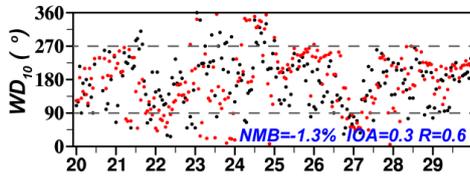
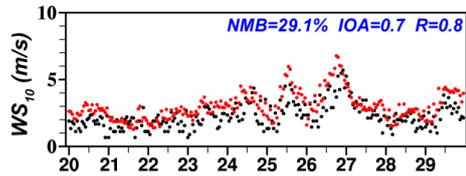
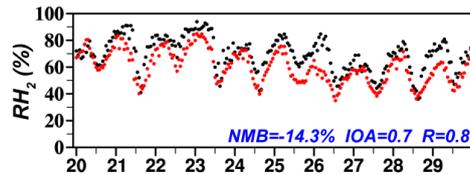
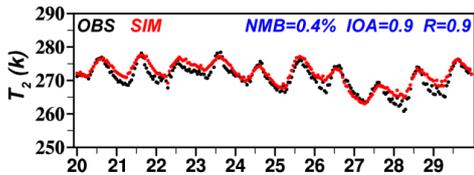


Figure 1. (a) Map of the two nested model domains. (b) Locations of the observations used for model evaluation.



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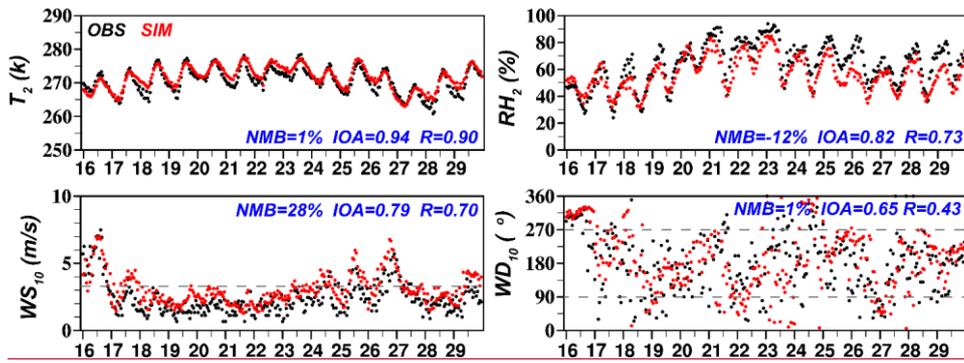


Figure 2. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly 2 m temperature (T_2 , K), 2 m relative humidity (RH_2 , %), 10 m wind speed (WS_{10} , $m s^{-1}$), and 10 m wind direction (WD_{10} , $^\circ$) averaged over the 12 stations during ~~2016~~-29 December 2015.

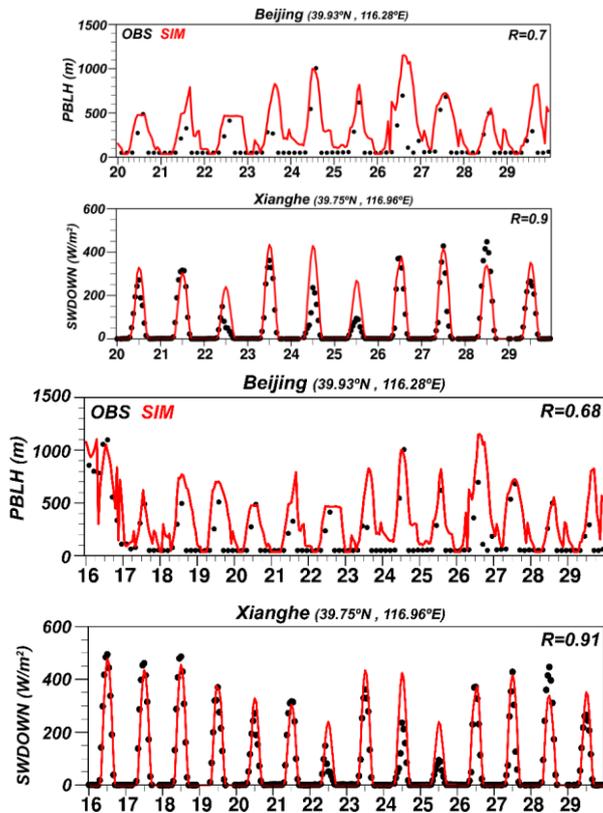
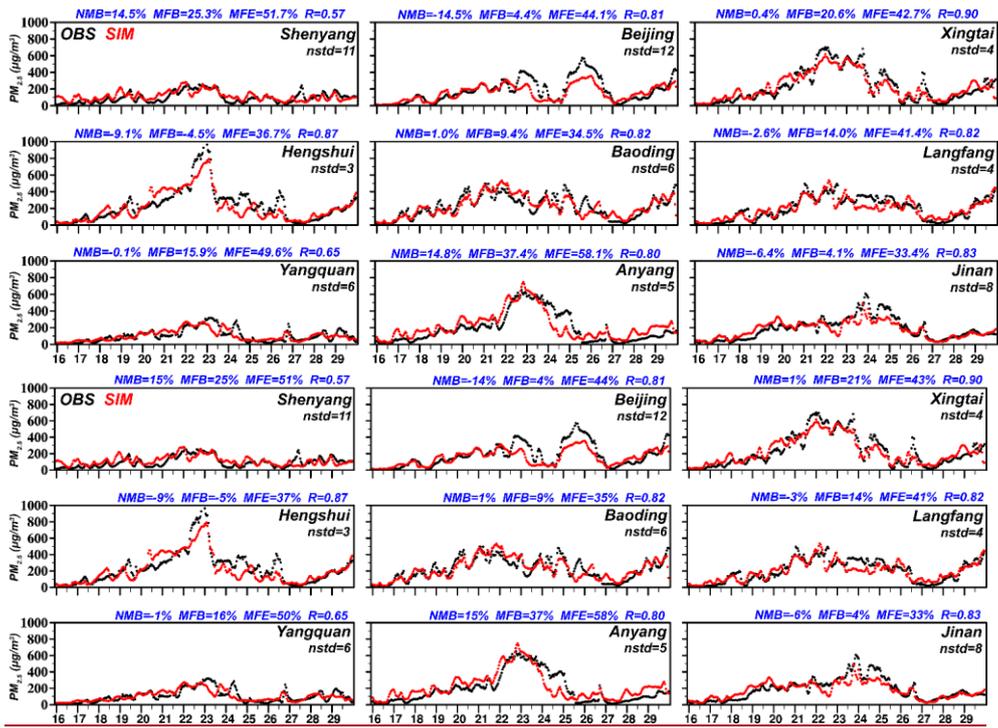
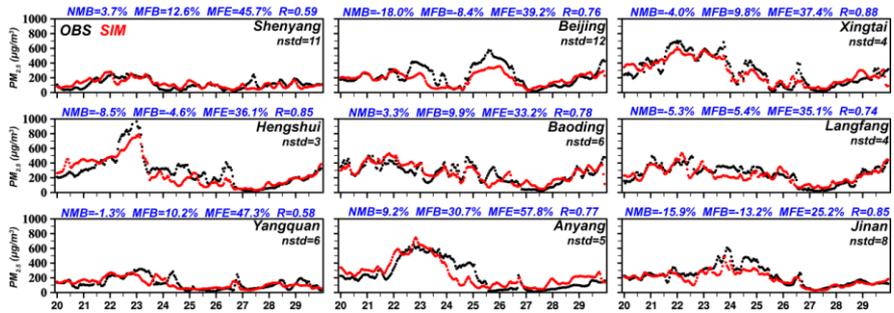


Figure 3. Time series of observed (shown in black dots) and simulated (shown in red lines) hourly planetary boundary layer height (PBLH, m) at the site of (39.93 N, 116.28 E) in Beijing, and shortwave downward radiation flux (SWDOWN, $W m^{-2}$) at the Xianghe Station (39.75 N, 116.96 E) from 16~~20~~ 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).

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Figure 4. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly PM_{2.5} concentrations ($\mu\text{g m}^{-3}$) 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.

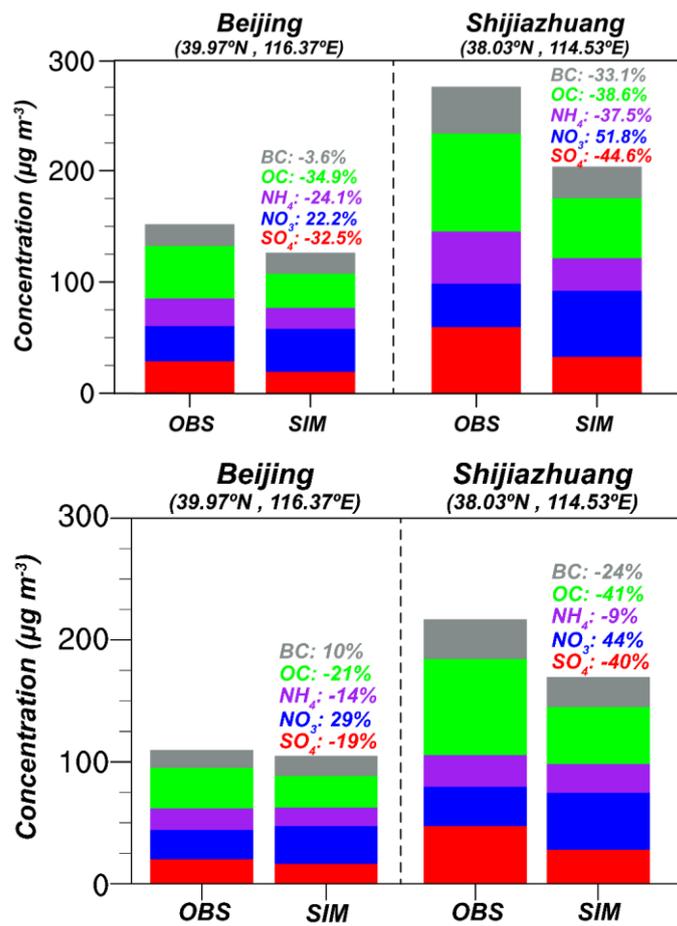
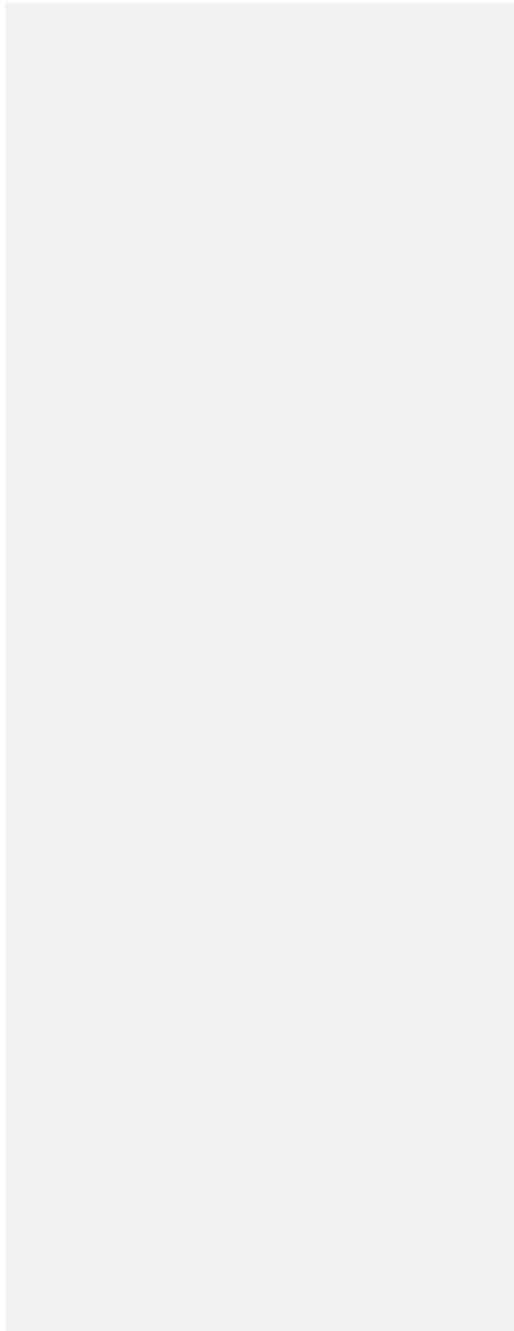
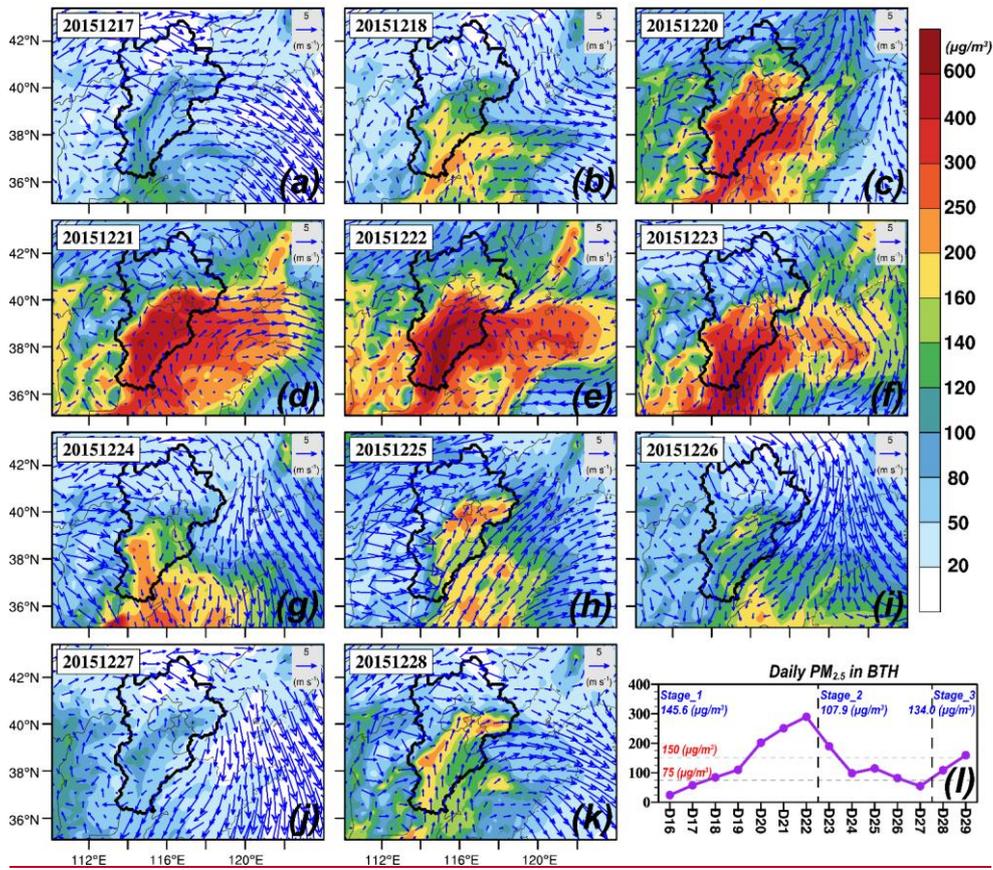


Figure 5. Comparison of observed and simulated surface-layer mass concentrations ($\mu\text{g m}^{-3}$) of SO_4^{2-} (red), NO_3^- (blue), NH_4^+ (purple), OC (green), and BC (gray) in the sites of (a) (39.97°N, 116.37°E) in Beijing, and (b) (38.03°N, 114.53°E) in Shijiazhuang averaged over 2016–29 December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

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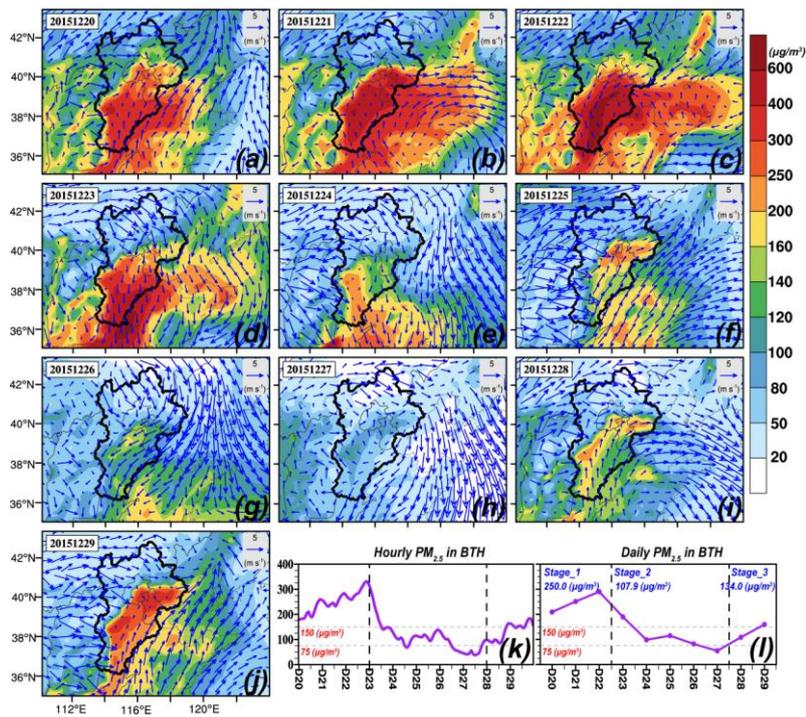
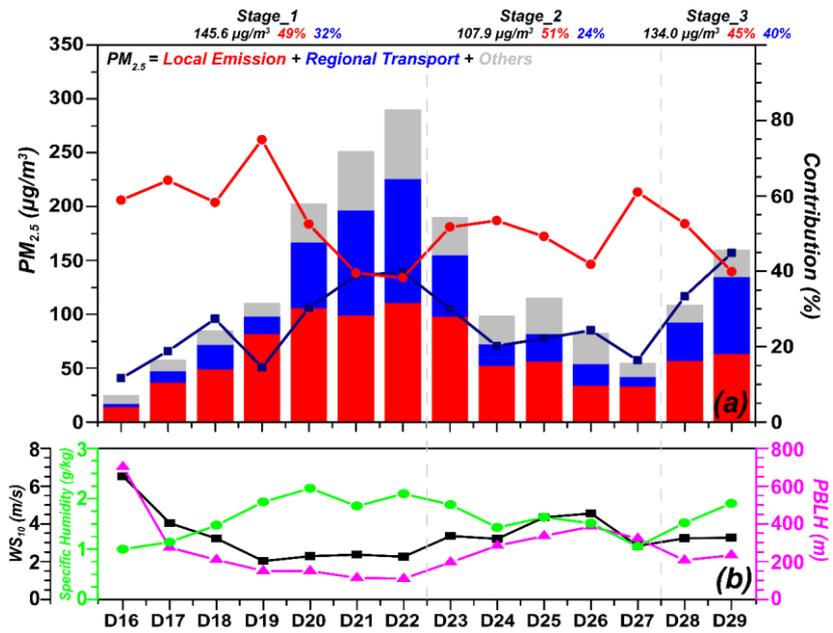
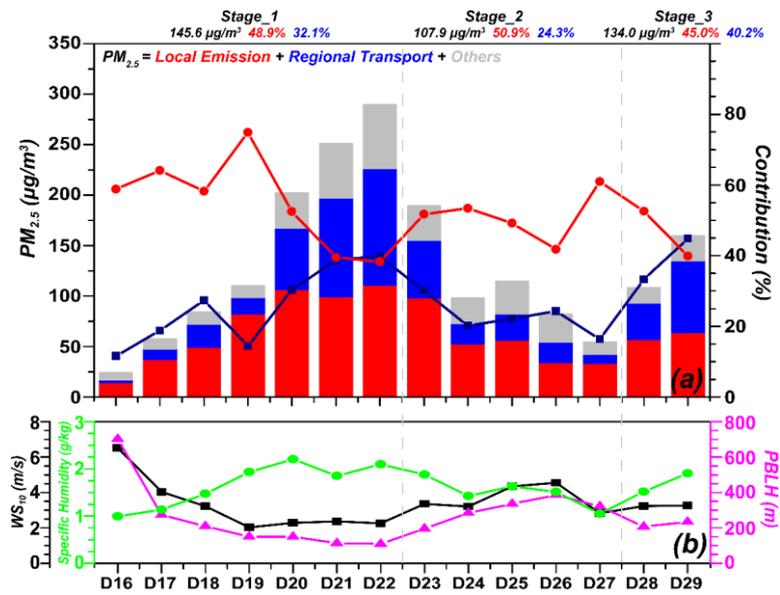


Figure 6. (a-k) Spatial distributions of simulated daily PM_{2.5} concentrations (shaded, $\mu\text{g m}^{-3}$) and wind vectors (arrows, m s^{-1}) from 20 to 29 December 2015. Time series of simulated hourly and daily PM_{2.5} concentrations averaged over the Beijing-Tianjin-Hebei region are also shown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





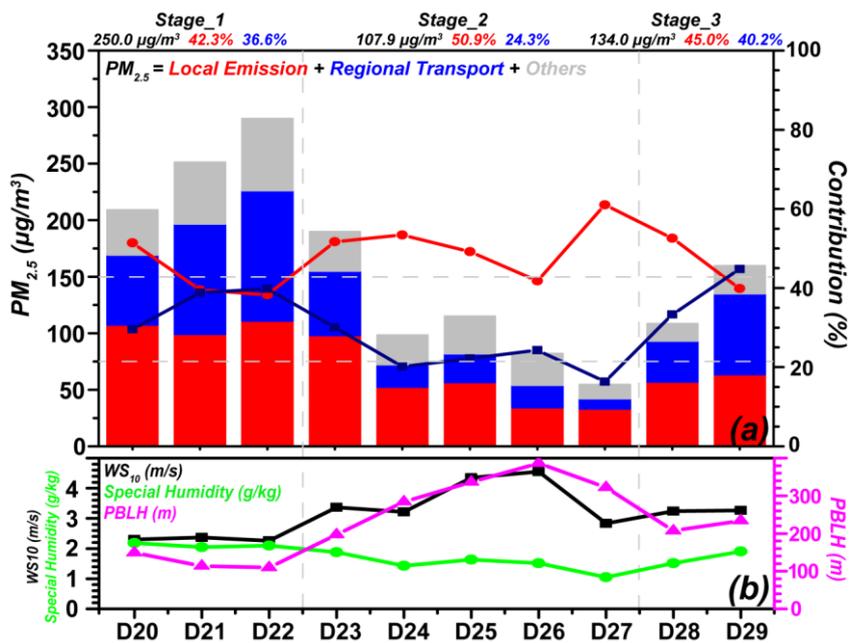
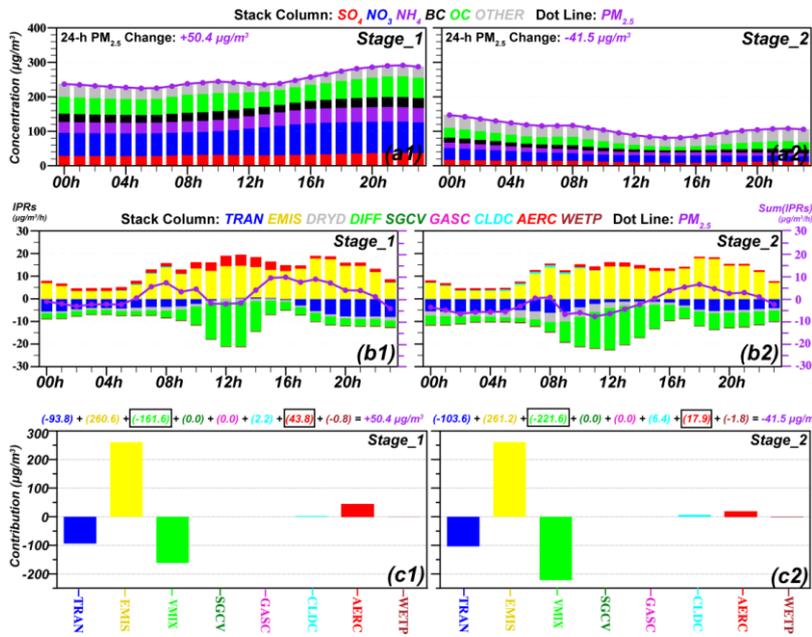


Figure 7. (a) Contributions of local emission (shown in red) and regional transport (shown in blue) to the near-surface PM_{2.5} concentrations averaged over the Beijing-Tianjin-Hebei region from 20-16 to 29 December 2015. The absolute contributions ($\mu\text{g m}^{-3}$) are shown in bars, ($\mu\text{g m}^{-3}$) 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 (WS₁₀, 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).



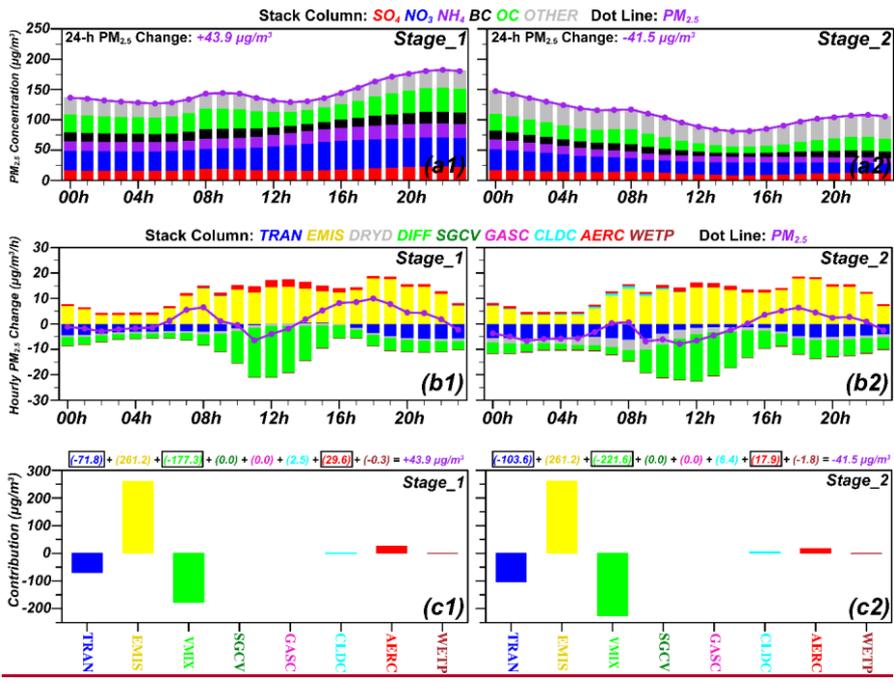


Figure 8. (a1-a2) Diurnal variations of PM_{2.5} concentrations averaged over Beijing-Tianjin-Hebei during Stage_1 and Stage_2 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is the 24-h change in PM_{2.5} concentration (23:00LST 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.

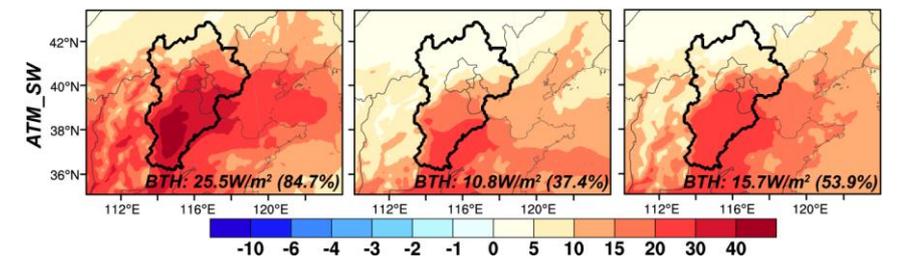
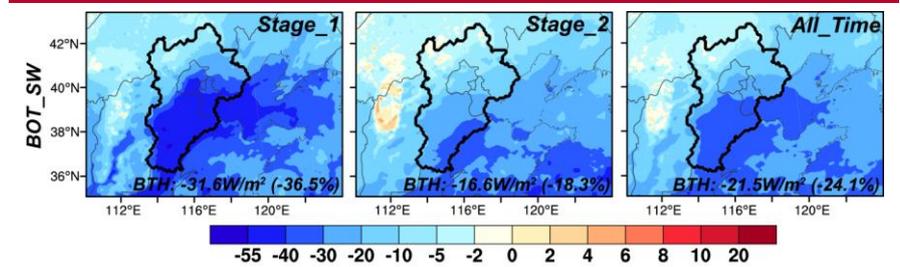
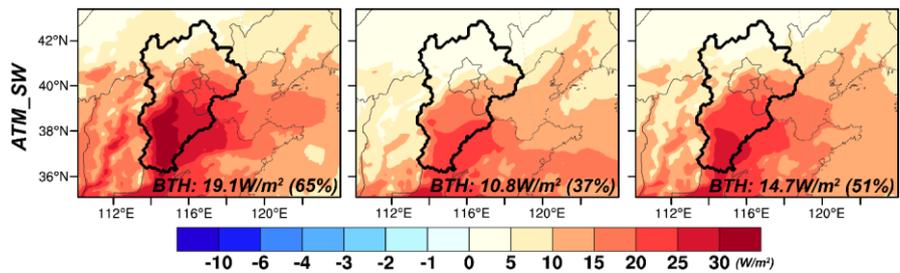
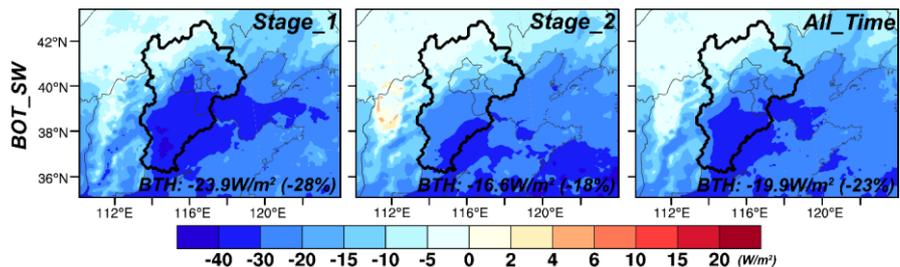
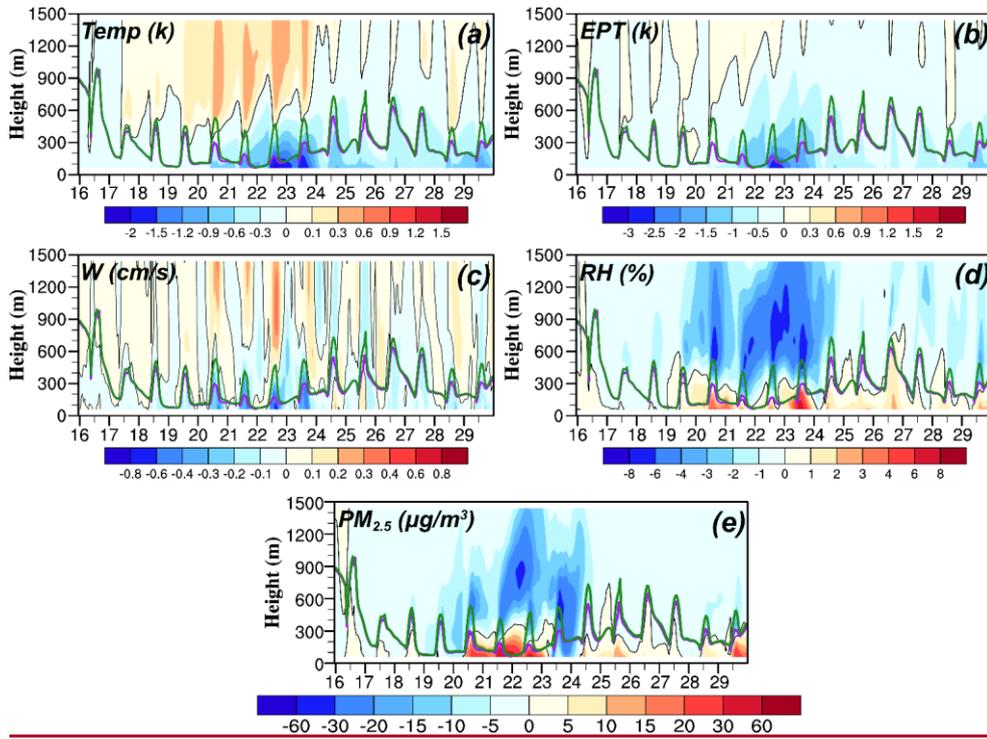


Figure 9. The differences in simulated all-sky radiative forcing ($W m^{-2}$) between CTL and NoARE cases (CTL minus NoARE) averaged over Stage_1, Stage_2, and the whole simulation period. “BOT_SW” and “ATM_SW” denote the downward shortwave radiative flux at the surface and in the atmosphere, respectively. 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.



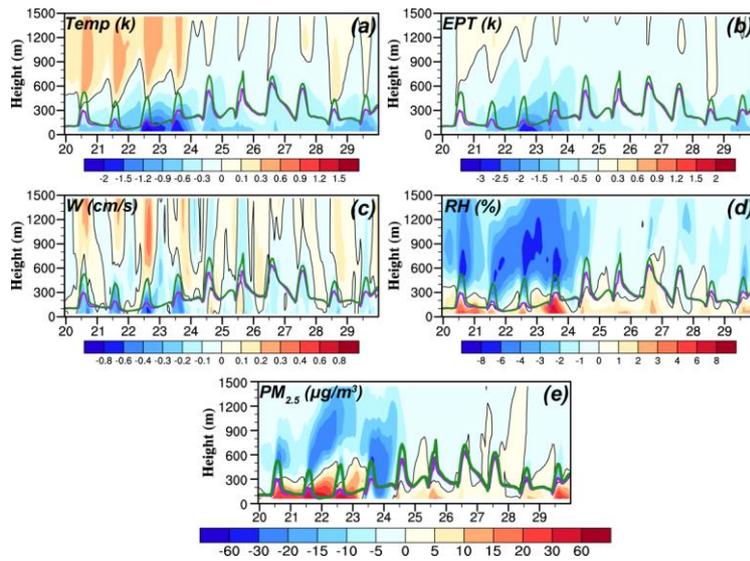
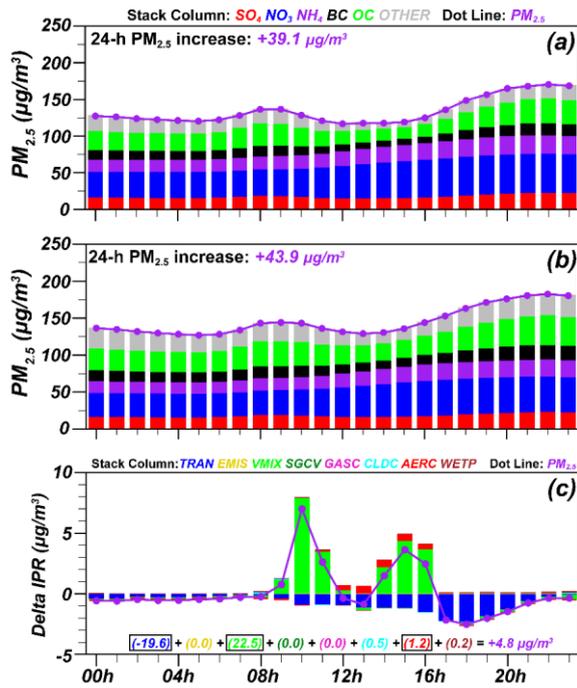


Figure 10. Time series of differences in (a) temperature (k), (b) equivalent potential temperature (k), (c) vertical wind speed (cm s^{-1}), (d) relative humidity (%), and (e) $\text{PM}_{2.5}$ concentration ($\mu\text{g m}^{-3}$) between CTL and NoARE cases (CTL minus NoARE) averaged over the Beijing-Tianjin-Hebei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

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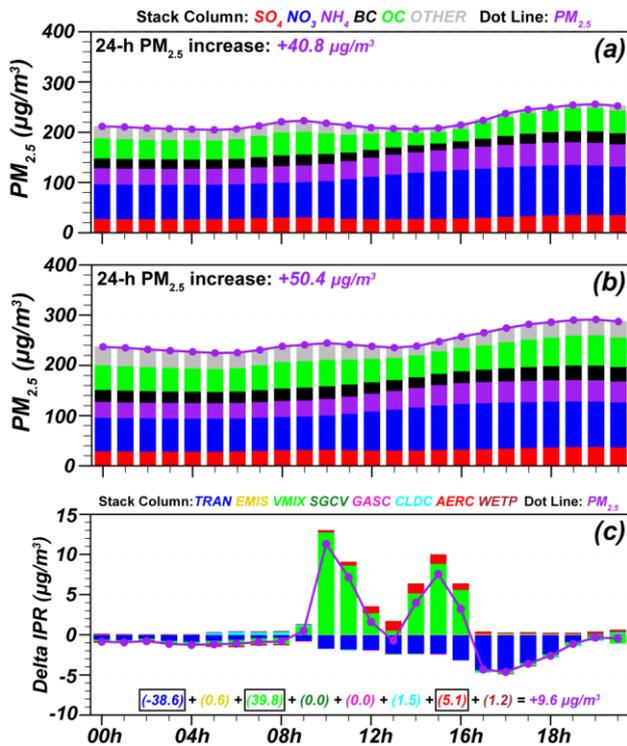
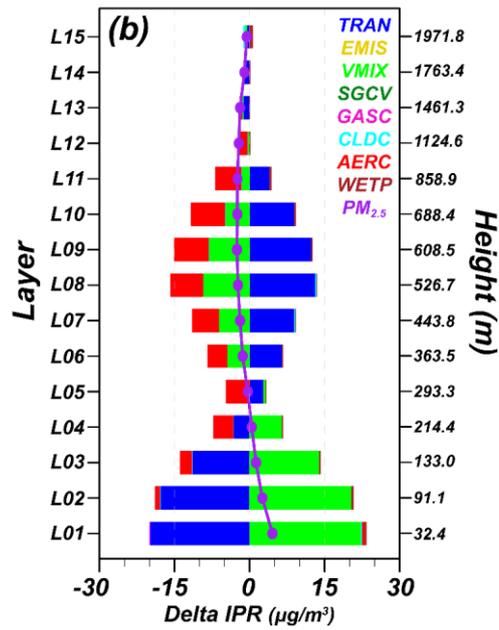
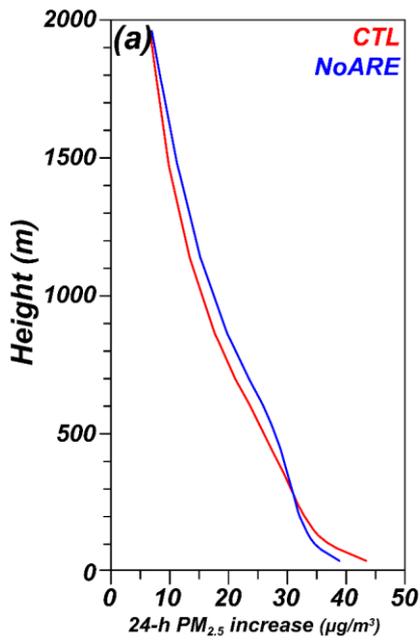


Figure 11. Diurnal variations of the near-surface PM_{2.5} concentrations in (a) NoARE and (b) CTL simulations averaged over the Beijing-Tianjin-Hebei region during Stage_1 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is 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.



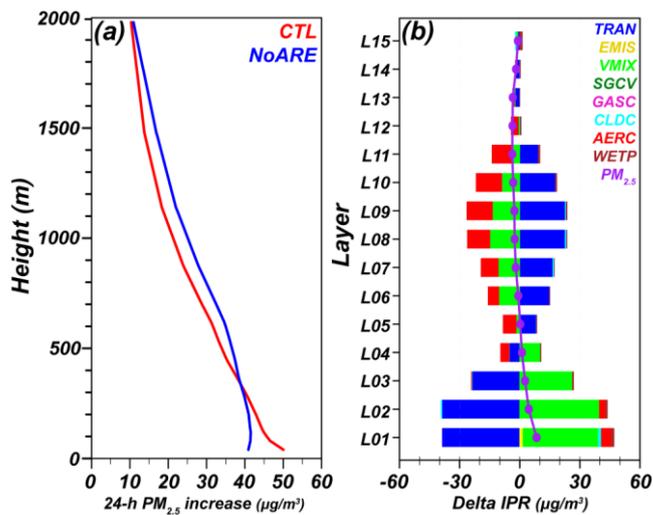


Figure 12. (a) Vertical profiles of the 24-h increases in $PM_{2.5}$ concentrations (23:00LST minus 00:00LST) averaged over Beijing-Tianjin-Hebei during Stage_1 in CTL and NoARE cases. (b) Vertical profiles of the differences in the 24-h $PM_{2.5}$ increases caused by aerosol radiative effect (CTL minus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

5

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
25 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 $63 \mu\text{g m}^{-3}$ 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 $150 \mu\text{g m}^{-3}$. 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 $1000 \mu\text{g m}^{-3}$. 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) \times 159 (south-north) and 150 (west-east) \times 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 $1^\circ \times 1^\circ$. 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 SO₂ (sulfur dioxide), NO_x (nitrogen oxides), CO (carbon monoxide), CO₂ (carbon dioxide), NMVOC (non-methane volatile organic compounds), NH₃ (ammonia), BC (black carbon), OC (organic carbon), PM_{2.5} and PM₁₀. 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 (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), 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 (O₃) 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 O₃ concentrations. Gao et al. (2018) investigated the impacts of BC-PBL interactions on O₃ 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 O₃ 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.

5 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

10 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 15 $PM_{2.5}$ 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 20 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.

5 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

10 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 15 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

5 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
10 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
15 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.

20 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
5 evolutionary characteristics of the observed PM_{2.5} concentrations in the nine cities ($R_s=0.57-0.90$). 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 $\mu\text{g m}^{-3}$ 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
10 (e.g., underestimated RH₂ and overestimated WS₁₀); (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 $\pm 30\%$ and MFE $\leq 50\%$) proposed by Boylan and Russel (2006).

Figure 5 compares the simulated and observed surface-layer concentrations of BC, OC, SO₄²⁻, NO₃⁻, and NH₄⁺ in
15 Beijing and Shijiazhuang averaged during 16-29 December 2015. WRF-Chem model underestimates the concentrations of SO₄²⁻, NH₄⁺ and OC in Beijing (Shijiazhuang) by 19% (40%), 14% (9%), and 21% (41%), respectively, but overestimates the NO₃⁻ 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 SO₂ gas-phase and
20 aqueous-phase oxidation, as well as heterogeneous chemistry may explain the underestimation of SO₄²⁻. 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 $200 \mu\text{g m}^{-3}$. 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 $600 \mu\text{g m}^{-3}$ 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 $\mu\text{g m}^{-3}$ to 289.8 $\mu\text{g m}^{-3}$, and the average PM_{2.5} concentration was 145.6 $\mu\text{g m}^{-3}$ (Fig. 7(a)), close to the air quality threshold value of “heavily polluted” (PM_{2.5} 24-h average concentration > 150 $\mu\text{g m}^{-3}$). The WS₁₀ 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 $\mu\text{g m}^{-3}$, still exceeding the Grade II standard (75 $\mu\text{g m}^{-3}$) 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 $\mu\text{g m}^{-3}$) 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.

5 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 $43.9 \mu\text{g m}^{-3}$ (from $136.5 \mu\text{g m}^{-3}$ at 00:00LST to $180.4 \mu\text{g m}^{-3}$ at 23:00LST) during the period of particulate accumulation (Stage_1), but it decreased by $41.5 \mu\text{g m}^{-3}$ during the period of particulate elimination (Stage_2).

10 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.

15 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
20 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 $+29.6 (+17.9) \mu\text{g m}^{-3}$, $-71.8 (-103.6) \mu\text{g m}^{-3}$ and $-177.3 (-221.6) \mu\text{g m}^{-3}$ for Stage_1 (Stage_2), respectively. Small differences were found for contributions from other processes between Stage_1 and Stage_2 (differences smaller than $5 \mu\text{g m}^{-3}$). 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

5 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
10 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
15 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.

20 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 $-0.5 \mu\text{g m}^{-3}$, $+1.3 \mu\text{g m}^{-3}$, and $+0.8 \mu\text{g m}^{-3}$, 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 $\text{PM}_{2.5}$ increase caused by ARE was $+4.1 \mu\text{g m}^{-3}$, and the restrained vertical mixing could be the primary reason for near-surface $\text{PM}_{2.5}$ increase when aerosol radiative forcing was considered.

Figure 12(a) shows the vertical profiles of the 24-h increases in $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ concentrations during the haze event, while IPR analysis is used to quantify the contributions of each physical/chemical process to the variation in $\text{PM}_{2.5}$ 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 $145.6 \mu g m^{-3}$. 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 $107.9 \mu g m^{-3}$. 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 $43.9 \mu g m^{-3}$ (23:00LST minus 00:00LST) during Stage_1, but it decreased by $41.5 \mu g m^{-3}$ 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 $+29.6 (+17.9) \mu g m^{-3}$, $-71.8 (-103.6) \mu g m^{-3}$ and $-177.3 (-221.6) \mu g m^{-3}$ 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 $43.2 \mu g m^{-3}$. 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 $4.8 \mu g$

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

5 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 $\text{PM}_{2.5}$ 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.

15 As Zheng et al. (2018) pointed out that the $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$) 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

- 5 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.

10 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	$\overline{\text{OBS}}^1$	$\overline{\text{SIM}}^2$	NMB ³	MFB ⁴	MFE ⁵	IOA ⁶	R ⁷
T₂ (k) ^a	12	270.7	271.6	1	1	1	0.94	0.90
RH₂ (%) ^a	12	63.8	56.1	-12	-12	22	0.82	0.73
WS₁₀ (m s ⁻¹) ^a	12	2.5	3.2	28	32	58	0.79	0.70
WD₁₀ (°) ^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

^aT₂: temperature at 2 m (k); RH₂: relative humidity at 2 m (%); WS₁₀: wind speed at 10 m (m s⁻¹); WD₁₀: wind direction at 10 m (°).

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

³NMB is the normalized mean bias, $\text{NMB} = \frac{1}{\text{nstd}} \times \sum_{i=1}^{\text{nstd}} \frac{\text{SIM}_i - \text{OBS}_i}{\text{OBS}_i} \times 100\%$.

5 ⁴MFB is the mean fractional bias, $\text{MFB} = \frac{2}{\text{nstd}} \times \sum_{i=1}^{\text{nstd}} \frac{\text{SIM}_i - \text{OBS}_i}{\text{SIM}_i + \text{OBS}_i} \times 100\%$.

⁵MFE is the mean fractional error, $\text{MFE} = \frac{2}{\text{nstd}} \times \sum_{i=1}^{\text{nstd}} \frac{|\text{SIM}_i - \text{OBS}_i|}{\text{SIM}_i + \text{OBS}_i} \times 100\%$.

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

⁷R is the correlation coefficient, $\text{R} = \frac{\sum_{i=1}^{\text{nstd}} (\text{OBS}_i - \overline{\text{OBS}}) \times (\text{SIM}_i - \overline{\text{SIM}})}{\sqrt{\sum_{i=1}^{\text{nstd}} (\text{OBS}_i - \overline{\text{OBS}})^2 + \sum_{i=1}^{\text{nstd}} (\text{SIM}_i - \overline{\text{SIM}})^2}}$.

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.

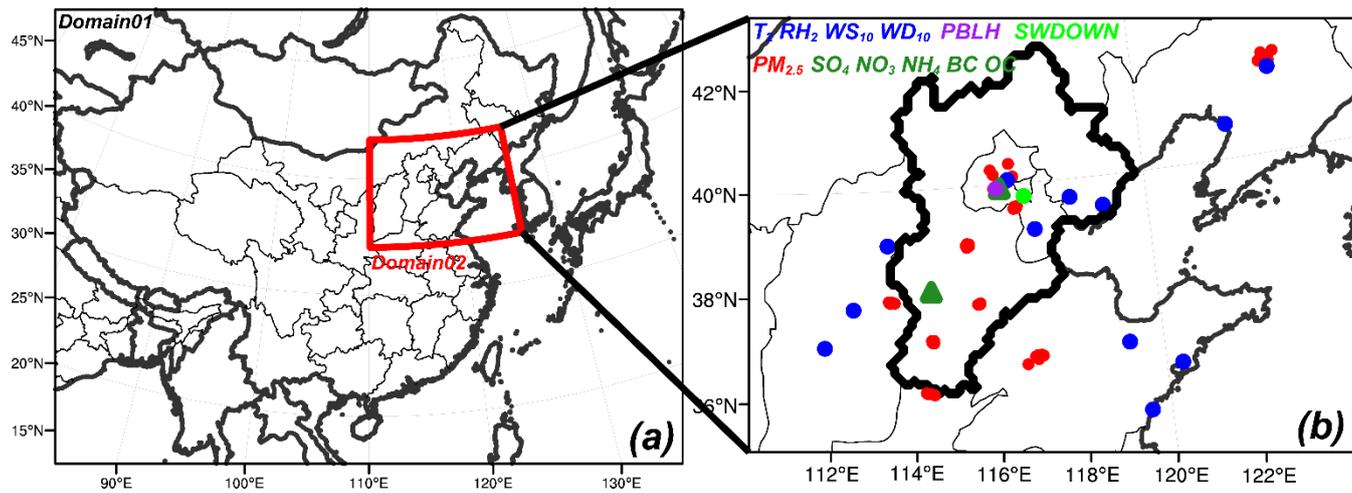


Figure 1. (a) Map of the two nested model domains. (b) Locations of the observations used for model evaluation.

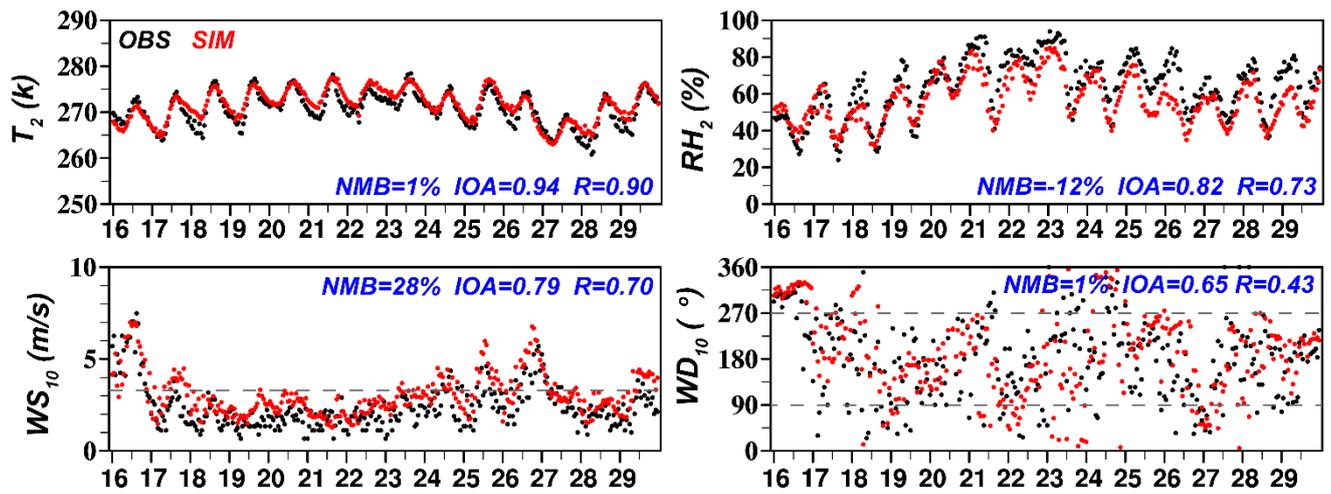


Figure 2. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly 2 m temperature (T_2 , k), 2 m relative humidity (RH_2 , %), 10 m wind speed (WS_{10} , $m\ s^{-1}$), and 10 m wind direction (WD_{10} , $^\circ$) averaged over the 12 stations during

5 16-29 December 2015.

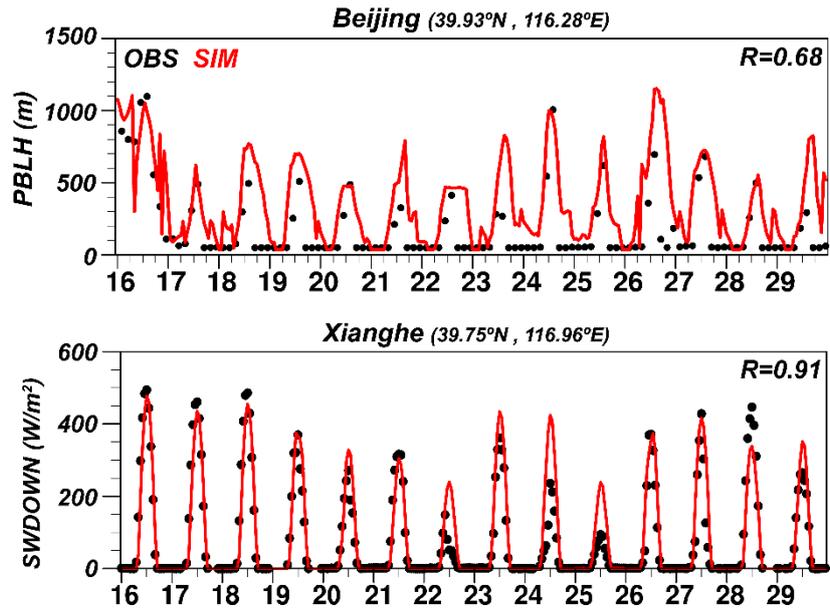


Figure 3. Time series of observed (shown in black dots) and simulated (shown in red lines) hourly planetary boundary layer height (PBLH, m) at the site of (39.93°N, 116.28°E) in Beijing, and shortwave downward radiation flux (SWDOWN, $W\ m^{-2}$) at the Xianghe Station (39.75°N, 116.96°E) from 16 to 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).

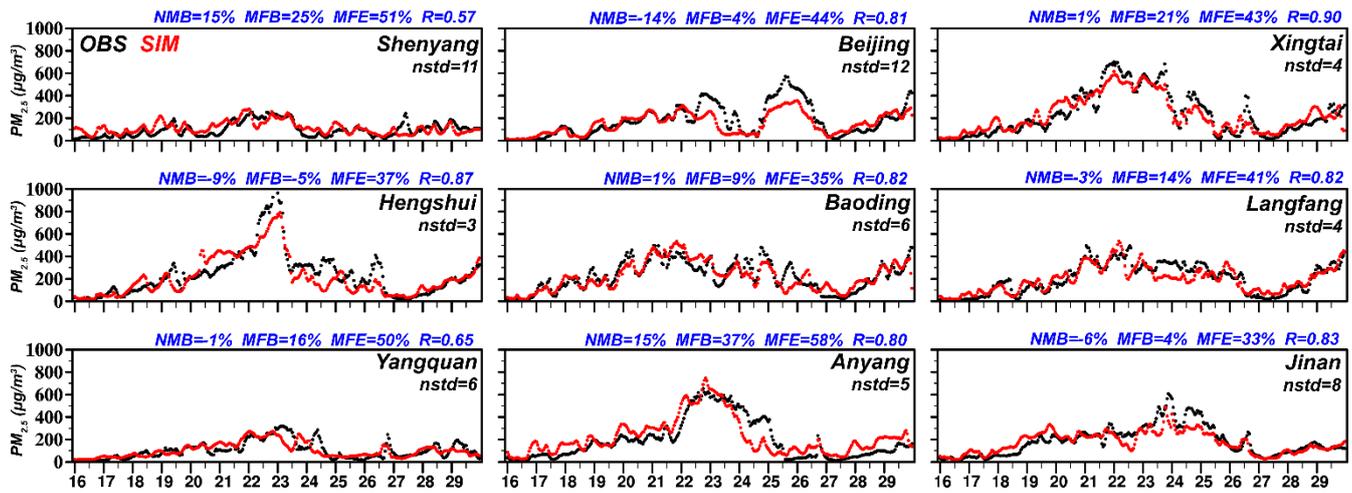


Figure 4. Time series of observed (shown in black dots) and simulated (shown in red dots) hourly $PM_{2.5}$ concentrations ($\mu g m^{-3}$) in the nine cities (Shengyang, Beijing, Xingtai, Hengshui, Baoding, Langfang, Yangquan, Anyang, and Jinan) from 16 to 29 December 2015. The nstd in each panel represents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

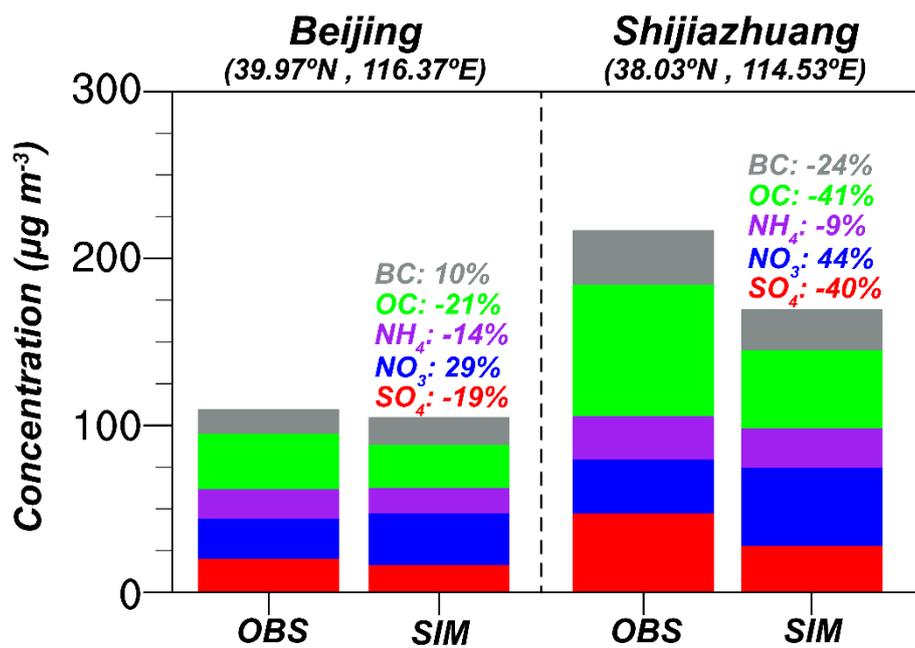


Figure 5. Comparison of observed and simulated surface-layer mass concentrations ($\mu\text{g m}^{-3}$) of SO_4^{2-} (red), NO_3^- (blue), NH_4^+ (purple), OC (green), and BC (gray) in the sites of (a) (39.97°N, 116.37°E) in Beijing, and (b) (38.03°N, 114.53°E) in Shijiazhuang

5 averaged over 16-29 December 2015. Also listed in colored numbers are normalized mean biases (NMBs) for each species.

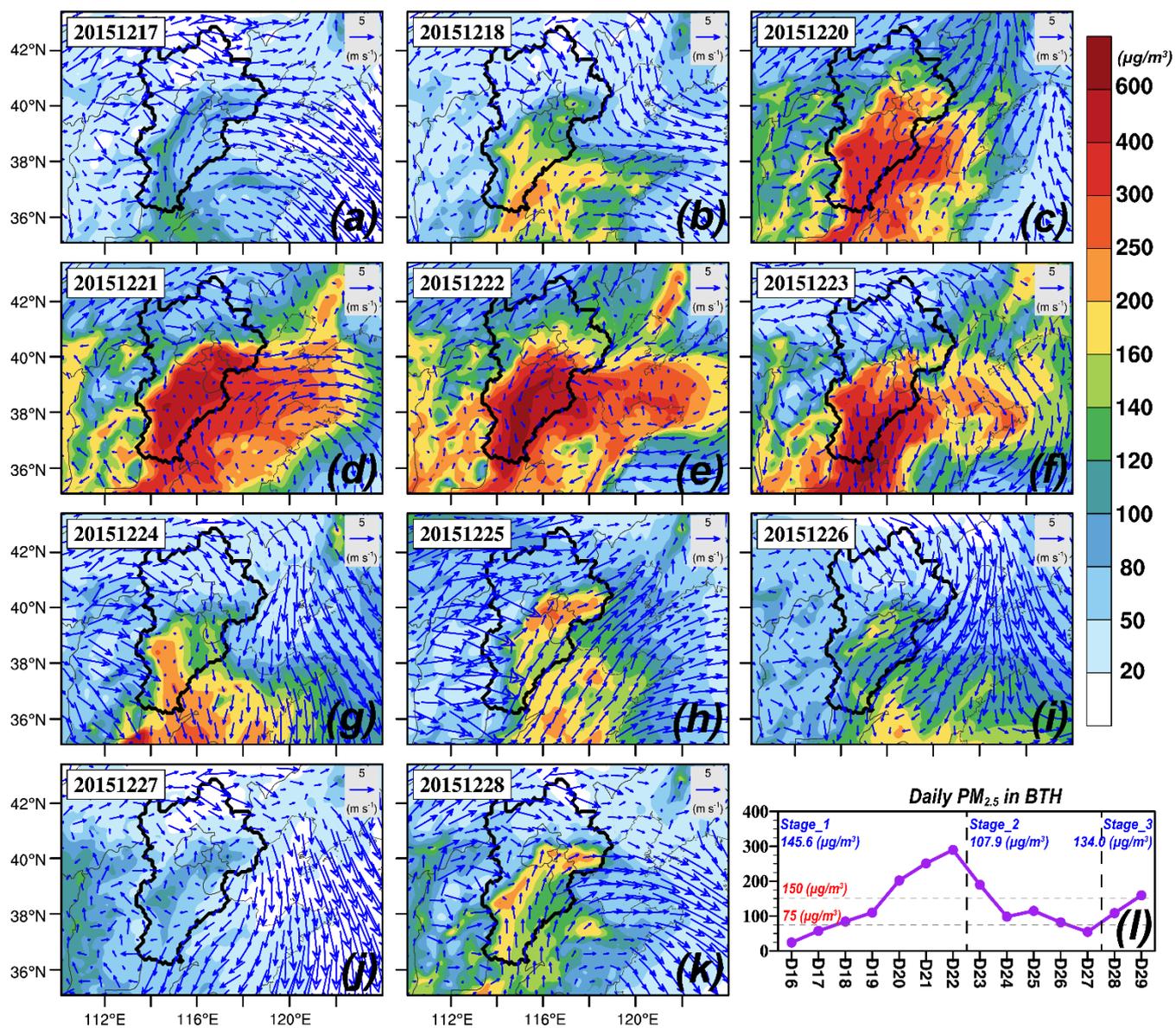


Figure 6. (a-k) Spatial distributions of simulated daily PM_{2.5} concentrations (shaded, $\mu\text{g m}^{-3}$) and wind vectors (arrows, m s^{-1}).

Time series of simulated daily PM_{2.5} concentrations averaged over the Beijing-Tianjin-Hebei region are also shown in (l).

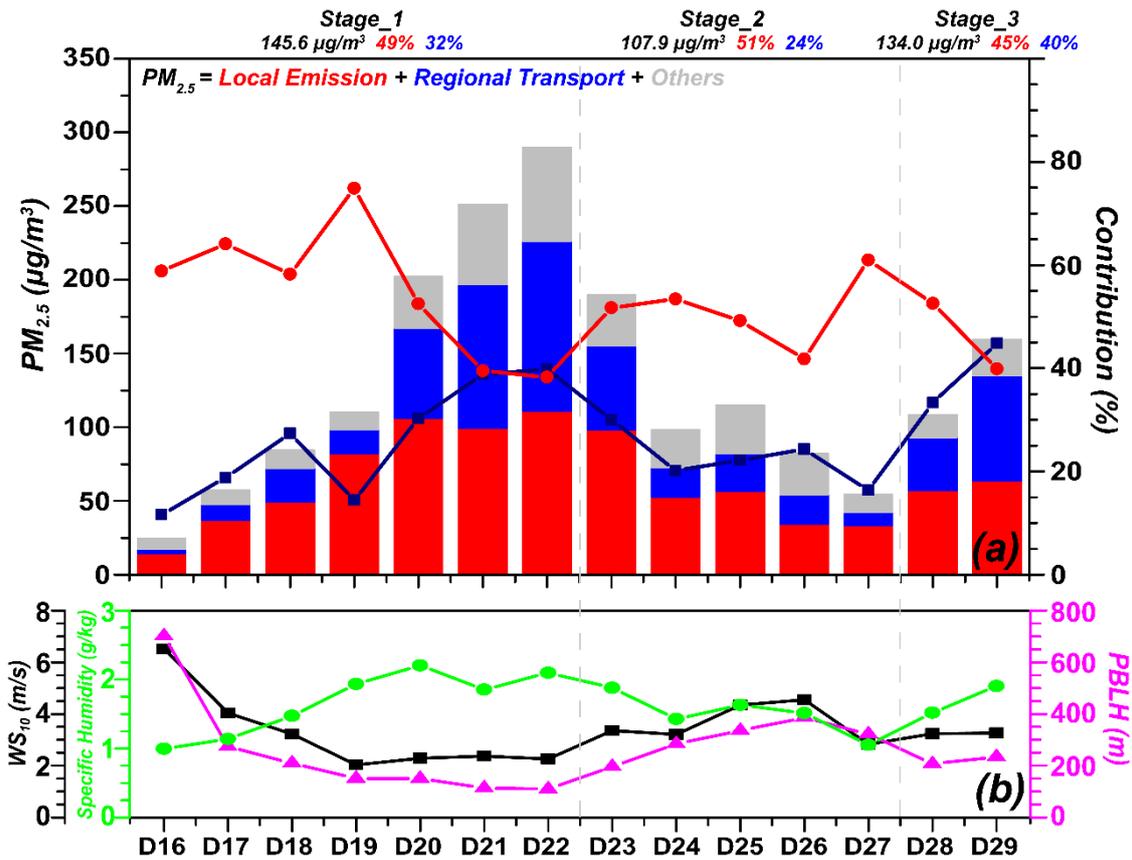


Figure 7. (a) Contributions of local emission (shown in red) and regional transport (shown in blue) to the near-surface $PM_{2.5}$ concentrations averaged over the Beijing-Tianjin-Hebei region from 16 to 29 December 2015. The absolute contributions ($\mu g m^{-3}$) are shown in bars, and the percentage 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).

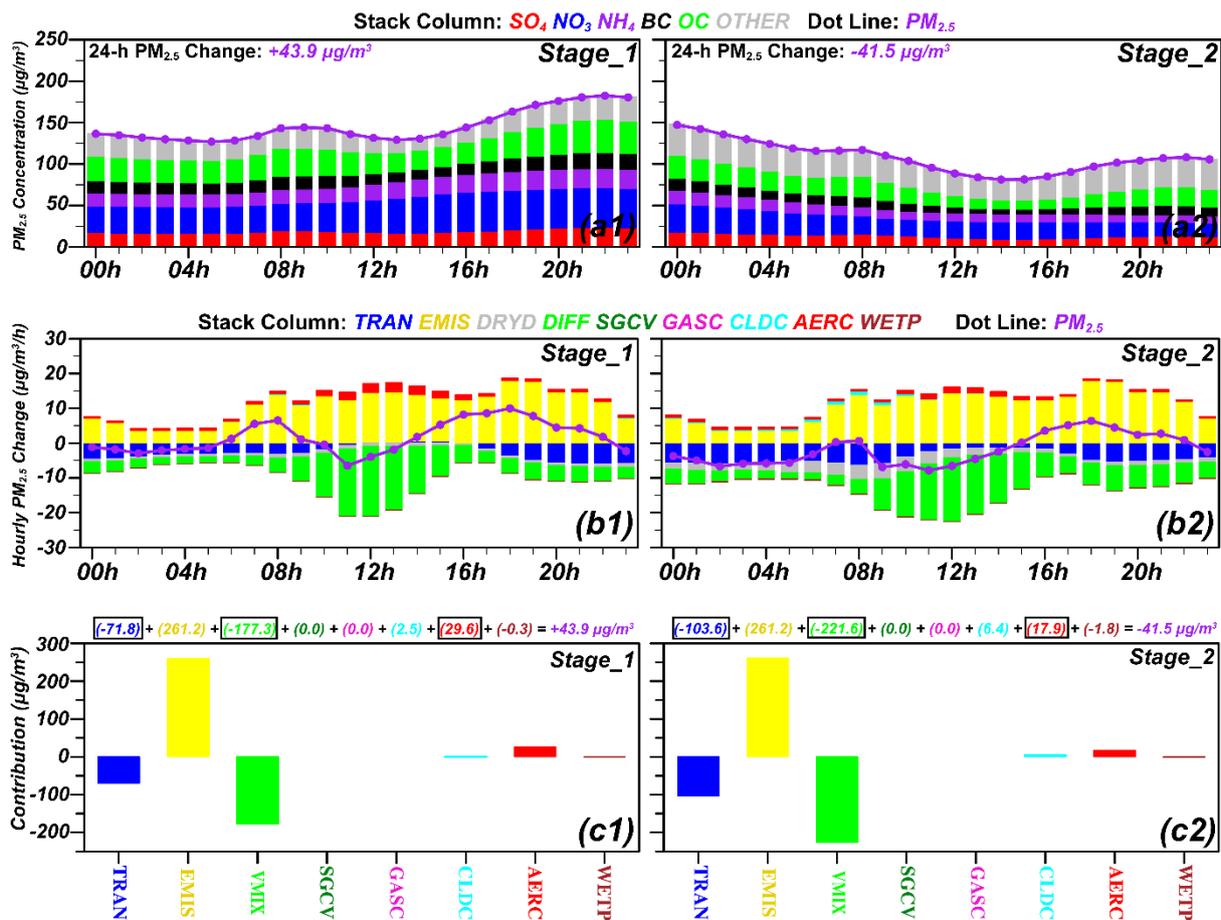


Figure 8. (a1-a2) Diurnal variations of PM_{2.5} concentrations averaged over Beijing-Tianjin-Hebei during Stage_1 and Stage_2 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is the 24-h change in PM_{2.5} concentration (23:00LST 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.

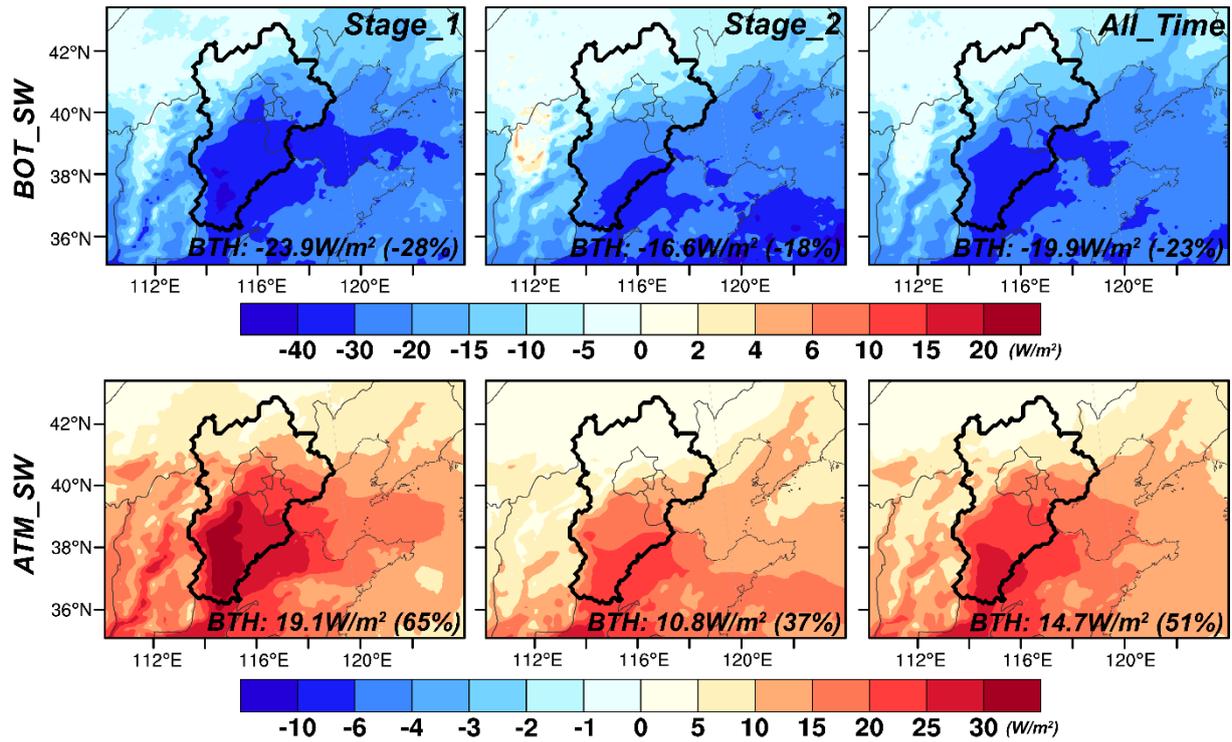
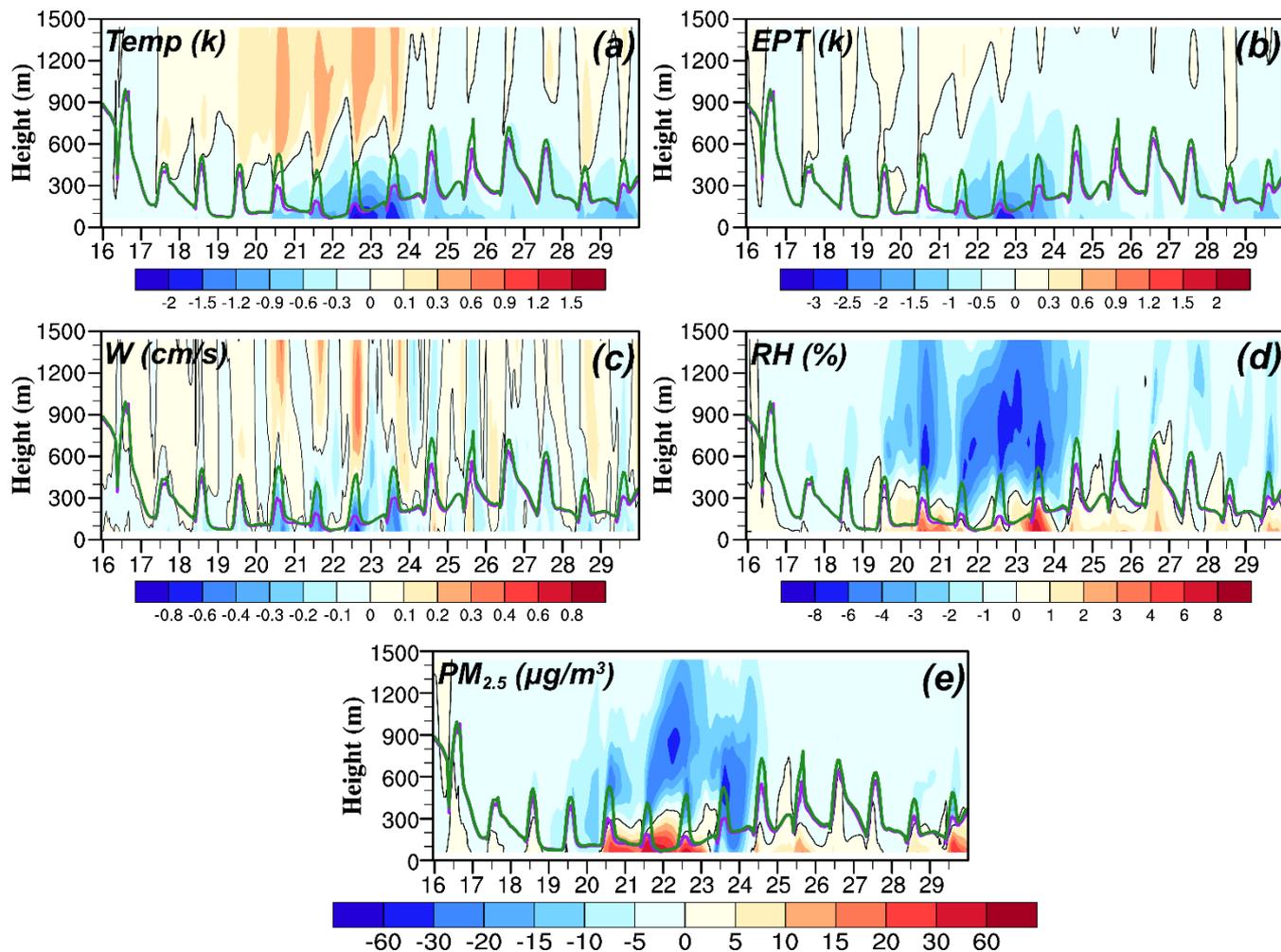


Figure 9. The differences in simulated all-sky radiative forcing ($W m^{-2}$) between CTL and NoARE cases (CTL minus NoARE) averaged over Stage_1, Stage_2, and the whole simulation period. “BOT_SW” and “ATM_SW” denote the downward shortwave radiative flux at the surface and in the atmosphere, respectively. 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.



5 **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.

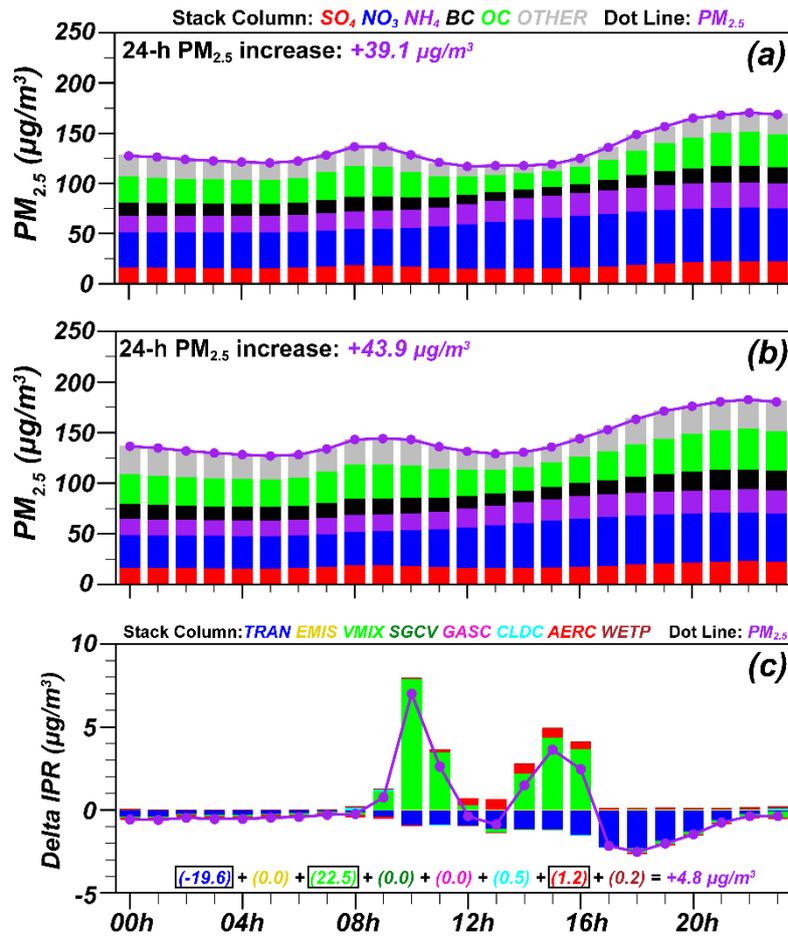


Figure 11. Diurnal variations of the near-surface PM_{2.5} concentrations in (a) NoARE and (b) CTL simulations averaged over the Beijing-Tianjin-Hebei region during Stage_1 (shown by purple dot lines). The colored bars represent different components. Also shown at the top left corner of each panel is 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.

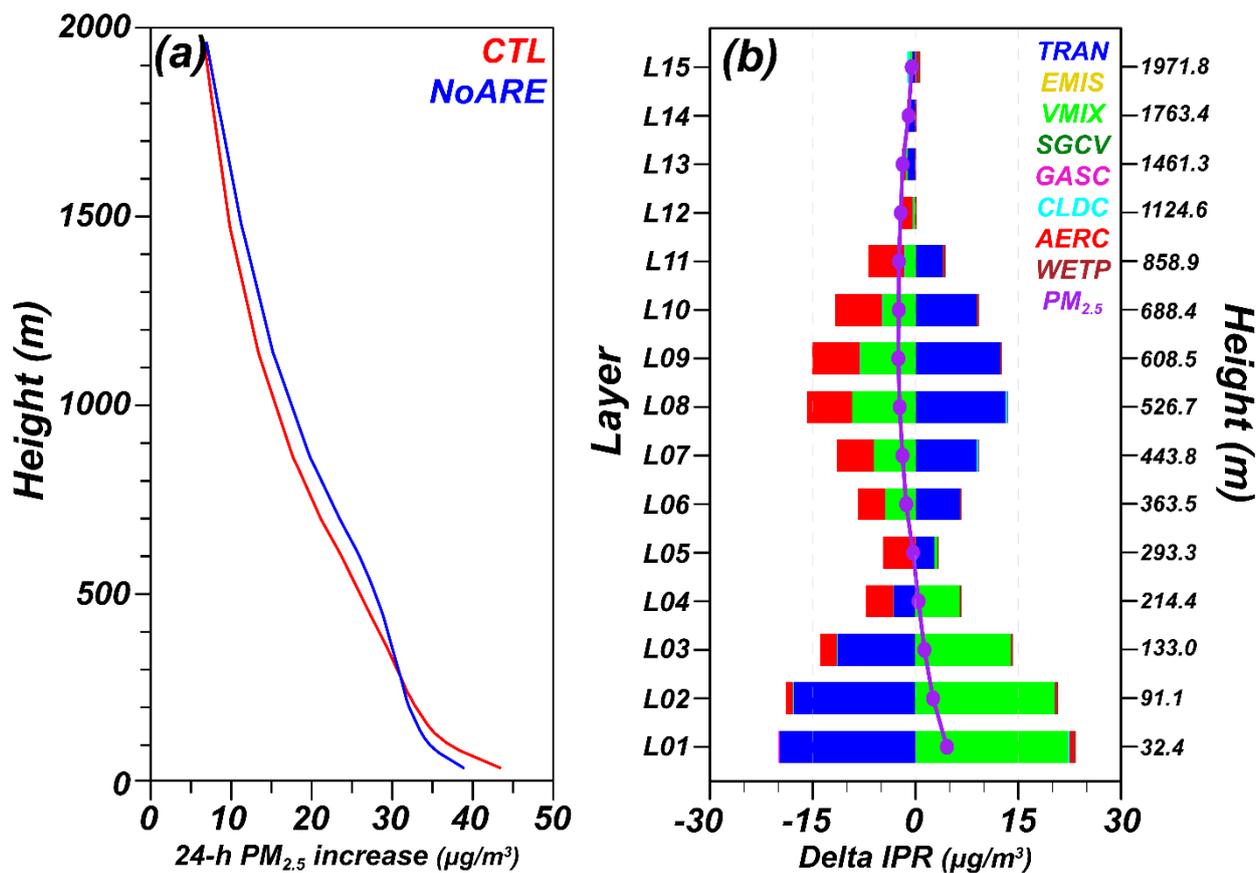


Figure 12. (a) Vertical profiles of the 24-h increases in PM_{2.5} concentrations (23:00LST minus 00:00LST) averaged over Beijing-Tianjin-Hebei during Stage_1 in CTL and NoARE cases. (b) Vertical profiles of the differences in the 24-h PM_{2.5} increases caused by aerosol radiative effect (CTL minus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