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RESEARCH ARTICLE

How aerosol direct effects influence the source contributions to $PM_{2.5}$ concentrations over Southern Hebei, China in severe winter haze episodes

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HIGHLIGHTS

- The aerosol direct effects result in a 3%–9% increase in $PM_{2.5}$ concentrations over Southern Hebei.
- These impacts are substantially different under different $PM_{2.5}$ loadings.
- Industrial and domestic contributions will be underestimated if ignoring the feedbacks.

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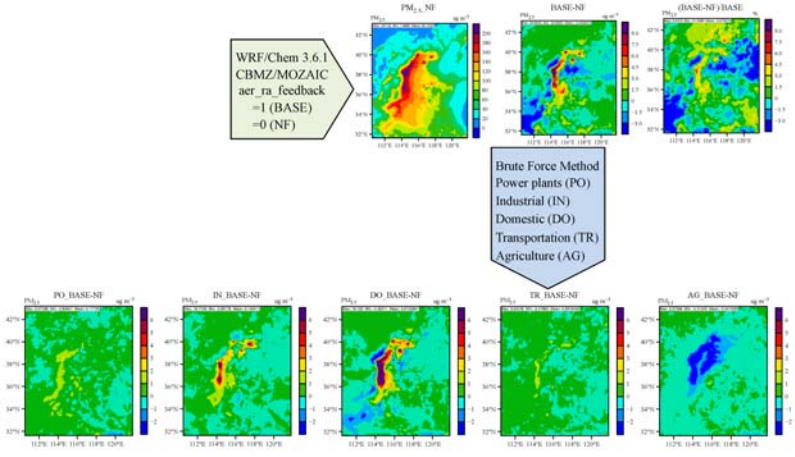
GRAPHIC ABSTRACT



ABSTRACT

Beijing-Tianjin-Hebei area is the most air polluted region in China and the three neighborhood southern Hebei cities, Shijiazhuang, Xingtai, and Handan, are listed in the top ten polluted cities with severe $PM_{2.5}$ pollution. The objective of this paper is to evaluate the impacts of aerosol direct effects on air quality over the southern Hebei cities, as well as the impacts when considering those effects on source apportionment using three dimensional air quality models. The WRF/Chem model was applied over the East Asia and northern China at 36 and 12 km horizontal grid resolutions, respectively, for the period of January 2013, with two sets of simulations with or without aerosol-meteorology feedbacks. The source contributions of power plants, industrial, domestic, transportation, and agriculture are evaluated using the Brute-Force Method (BFM) under the two simulation configurations. Our results indicate that, although the increases in $PM_{2.5}$ concentrations due to those effects over the three southern Hebei cities are only 3%–9% on monthly average, they are much more significant under high $PM_{2.5}$ loadings (~50 $\mu g \cdot m^{-3}$ when $PM_{2.5}$ concentrations are higher than 400 $\mu g \cdot m^{-3}$). When considering the aerosol feedbacks, the contributions of industrial and domestic sources assessed using the BFM will obviously increase (e.g., from 30%–34% to 32%–37% for industrial), especially under high $PM_{2.5}$ loadings (e.g., from 36%–44% to 43%–47% for domestic when $PM_{2.5}$>400 $\mu g \cdot m^{-3}$). Our results imply that the aerosol direct effects should not be ignored during severe pollution episodes, especially in short-term source apportionment using the BFM.

1 Introduction

Fine particulate matter ($PM_{2.5}$) pollution is now one of major air pollution problems in urban environment in

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China. The Ministry of Environmental Protection in China (MEP) set up the standard for the first time for $PM_{2.5}$, of 75 and 35 $\mu g \cdot m^{-3}$ for daily and annual average concentrations, respectively, in the National Ambient Air Quality Standards (NAAQS) in 2012 [1]. However, it is reported that only ~10% out of the monitored 161 cities in China could reach the new standard in 2014 [2]. In 2015, this number increased to 21.6%, out of the monitored 338 cities. The Beijing-Tianjin-Hebei area (BTH), the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and the Sichuan Basin are the most polluted regions in China [2,3].

Hebei has been reported as the top polluted province in China, as seven of the top ten polluted cities in China are within Hebei province in recent three years [2–4], due to its large quantities of industrial productions and huge amount of air pollutant emissions, especially in the southern area of Hebei [5]. The three most southern cities in Hebei, i.e., Shijiazhuang (the capital of Hebei), Xingtai, and Handan, listed in the top five polluted cities in 2013 and 2014, top eight cities in 2015, and back to top five in 2016, have attracted enormous scientific interests because of their extremely severe air pollution, with annual average $PM_{2.5}$ concentrations of 3–4 times of the new standard [2–4]. Quantifying the source contributions and understanding the formation mechanism of the severe $PM_{2.5}$ pollution are necessary and urgent to support the policymaking in air pollution controls in this region. Although there were several studies focusing on source apportionment of $PM_{2.5}$ over BTH or those southern Hebei cities [5–10], using receptor model or chemical transport model, or a hybrid method of those two, none of them involved the considerations of the impacts of aerosol-meteorology interactions under high concentration of $PM_{2.5}$ to the results of source apportionment. It is known that high loading of atmospheric aerosols will conversely impact the meteorology [11,12], e.g., reducing the downward solar radiation, thus decreasing the surface wind speed, temperature, and the planetary boundary layer (PBL) height, which is the so-called direct effect. This effect has been proved to be capable to impact the meteorology, and in turn change the air quality in America and Europe by several studies with online-coupled meteorology-chemistry models [13–16]. It has been reported that over the past decades, aerosol feedbacks have been significantly enhanced over the North China, which is part of the cause of enhancing haze pollution [17]. Gao et al. [18] evaluated the impact of aerosol direct effects on $PM_{2.5}$ concentrations in a haze event in 2010 over the North China Plain (NCP) using the Weather Research and Forecasting Model with Chemistry (WRF/Chem), a state-of-the-art 3-D chemical transport model, and found that this effect could induce about 278.2 m decrease in PBL height and more than 20 $\mu g \cdot m^{-3}$ increase in $PM_{2.5}$ concentrations in Shijiazhuang, and in the total feedbacks the contributions of black carbon (BC) absorption might be as significant as ~50%. They also concluded that during that event 64.5% of $PM_{2.5}$ in Beijing were contributed from sources outside Beijing, i.e., from southern Hebei, Tianjin, Shandong and Henan provinces. Wang et al. [19] applied the WRF and Community Multi-scale Air Quality Model (CMAQ) over the East Asia to evaluate the aerosol direct effects on $PM_{2.5}$ concentrations and concluded that the maximal decrease in daily solar radiation, and the maximal increase in $PM_{2.5}$ concentrations could reach as high as 53% and 140 $\mu g \cdot m^{-3}$ in Beijing during the extremely severe polluted month of January 2013. Lots of recent studies were reported on this month, focusing on the transport or chemical process during severe pollution episodes [8,19–21]. Zhang et al. [22] also simulated the period of January 2013 for eastern China using the WRF/Chem model to assess the aerosol-radiation-cloud feedbacks on meteorology and air quality. They concluded that aerosol direct and indirect effects resulted in reductions of downward shortwave flux at the surface (SWDOWN), 2 m temperature, 10 m wind speed, planetary boundary layer (PBL) height by up to 84.0 $W \cdot m^{-2}$, 3.2°C (3.2 K), 0.8 $m \cdot s^{-1}$, and 268 m, respectively, and the aerosol direct effects were dominant in modulating those factors [22]. Gao et al. [20] also analyzed the effects of aerosol feedbacks for the same period over the NCP and concluded that during the most polluted period of January 10–15, 2013, the aerosol feedbacks could induce an increase of 10–50 $\mu g \cdot m^{-3}$ (2%–30%) in surface $PM_{2.5}$ concentrations over Beijing, Tianjin, and south Hebei and the maximum increase in hourly $PM_{2.5}$ concentrations were around 50 (70%), 90 (60%) and 80 $\mu g \cdot m^{-3}$ (40%) over Beijing, Tianjin, and south Hebei, respectively. These recent studies reveal and demonstrate the importance of aerosol feedbacks on meteorology and air quality over the high aerosol loading regions. They also raise another question that, when we pursue $PM_{2.5}$ source apportionment for those regions using 3-D air quality model, whether the effects of aerosol-meteorology feedbacks should be fully considered and how much those effects will alter the source apportionment results, which are the key references for the policymakers to design air pollution control strategy, especially the emergency action plan during extremely haze episode.

Therefore, we evaluate the impact of aerosol radiative effects on regional meteorology and air quality, especially $PM_{2.5}$, over the southern Hebei cities in this study, which region have been thought to impact Beijing through pollution transport. This region has the highest loading of $PM_{2.5}$ and lower PBL due to its geophysical and meteorological characteristics [5]. Furthermore, source contributions from five emission sectors, i.e., power plants, industrial, domestic, transportation, and agriculture, to $PM_{2.5}$ concentrations are calculated by two simulations of considering and not considering the aerosol direct effects on air quality, to evaluate the impact of those effects to the results of $PM_{2.5}$ source apportionment. Those results will be helpful in further understanding the aerosol feedbacks

from different emission sources on meteorology and air quality and useful to policymakers to generate more effective emergency control plans for extreme haze events.

We also applied the WRF/Chem model in this study over the East Asia and northern China at 36 and 12 km grid resolution, respectively, for the period of January 2013, which is reported as the most polluted months in the past 60 years [23]. Note that the aforementioned three studies on this month over the NCP used a relatively coarse resolution (e.g., 36 km in [19] and 27 km in [22] and [20]) and focused more on Beijing and surrounding areas. A relatively finer resolution (12 km) focusing on southern Hebei cities in this study may present more precise information on aerosol feedbacks over Hebei. The impacts of aerosol feedbacks on meteorology including solar radiation, temperature, wind speed, and PBL height, as well as air quality, especially the $PM_{2.5}$ concentrations, are evaluated through the scenario simulations in this study.

The organizations of this paper are: Section 2 introduces the model configuration, inputs, and simulation scenarios; Section 3 presents the results discussions on the impact of aerosol direct effects to air quality, as well as to the source contributions from emission sectors; Section 4 summarized the major findings, policy implications, and limitations of this paper. The model performance evaluations on both meteorology and air quality, as well as the aerosol direct effects on meteorology are presented in supplementary file.

2 Methodology

2.1 Model configurations and simulation design

WRF/Chem version 3.6.1 is applied to simulate the period of January 2013. The two nested modeling domains are similar to the study of Wang et al. [24]. Domain 1 covers East Asia with a grid resolution of 36 km×36 km and Domain 2 encompasses Beijing, Tianjin and four provinces including Hebei, Henan, Shandong and Shanxi with a grid resolution of 12 km × 12 km. The vertical resolutions are 23 layers from the surface to the tropopause with the corresponding sigma levels of 1.000, 0.995, 0.988, 0.980, 0.970, 0.956, 0.938, 0.916, 0.893, 0.868, 0.839, 0.808, 0.777, 0.744, 0.702, 0.648, 0.582, 0.500, 0.400, 0.300, 0.200, 0.120, 0.052, and 0.000. A spin-up period of seven days (December 25–31, 2012) is used to minimize the influence of the initial conditions.

The model configurations and inputs are summarized in Table S1(see Supplementary material). The anthropogenic emission inventory is from the Multi-resolution Emission Inventory for China (MEIC) [25] for the base year 2010, following the study of Wang et al. [24]. Therefore, it should be noted that the total emissions in this study may be underestimated because of the economic increase during the period of 2010–2013. The national GDP increase 9.3% and 7.8% in 2011 and 2012, respectively [5]. The MEIC inventory, developed by Tsinghua University, includes all the anthropogenic emissions of sulfur dioxide (SO_2), nitrogen oxideds (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), ammonium (NH_3), carbon dioxide (CO_2), atmospheric particles with aerodynamic diameter less than or equal to 10 μm (PM_{10}), and $PM_{2.5}$ in China, and has been a reasonable estimation in terms of total emissions of cities but uncertainties may exist in the spatial allocations of emissions into fine grid [5,24]. The Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2 [26] is applied for online calculation of the biogenic emissions. Dust emissions are online calculated as well using the option of Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) and the Modal Aerosol Dynamics Model for Europe (MADE)/Seconday Organic Aerosol Model (SORGAM) [27].

The National Center for Environmental Prediction (NCEP) Final Analysis (FNL) reanalysis data sets are used for the meteorological initial (IC) and boundary conditions (BC) in this study. The chemical IC and BC for the domain 1 are from the default chemical profiles in WRF/Chem, and those for the domain 2 are nested down from the output of domain 1. No nudging is applied for the purpose of evaluating the aerosol-meteorological direct feedbacks on air quality. The Carbon-Bond Mechanism version Z (CBMZ) [28] is chosen as the gas-phase chemical chemisty. The MOSAIC that uses 8 volatility bins is used as aerosol module [29]. Other physical modules chosen in this study are described in detail in supplementary file.

The simulation design is as follow: first, the impact of aerosol-meteorology direct feedbacks on air quality is evaluated by comparing two simulations of turning on (baseline simualtion, referred to as BASE) and off (no-feedback simulation, referred to as NF) the aerosol radiative feedback option (aer_ra_feedback = 1 or 0) in the WRF/Chem model. Second, five major emission sectors are considered in evaluating the impacts of those effects on sector source contributions, including power plants (PO), industrial (IN), domestic (DO), transportation (TR), and agriculture (AG). It is calculated that those five sectors cover about 93% to 98% of total emissions of $PM_{2.5}$, PM_{10}, SO_2 and NO_x in the BTH area in winter [5]. The source emissions of each sector in the BTH area are zeroed out, in two simulations of turning on (BASE configuration) and off (NF configuration) the aerosol direct feedbacks in the model. The difference of each zero-out scenario to the base case represents the source contributions of each sector, which is so-called the Brute Force method (BFM) [30]. By comparing the differences under BASE and NF configuration, we can quantify the differences between considering and not considering the aerosol radiative feedbacks, thus get more understanding on which emission sectors are more effective in radiative feedbacks to contribute to the surface air pollution by

change the meteorological conditions, besides their direct emissions.

It should be noted that, to strictly assess the effects of a emission sector, we need to keep the BASE simulation, label the aerosols contributed by this source in each time step, and calculate out the pollutants concentrations increase led by their feedbacks during the simulation. It is technically unavailable at present. Besides the uncertainties of the BFM, we still pursued this study for two objectives. First, to give an estimation on the possible uncertainty ranges of the source apportionment studies using the BFM by 3D air quality models over the severe polluted BTH area without considering the aerosol-meteorology feedbacks, which were very common in recent years. Second, effectively reducing the peak concentrations during extremely severe polluted winter episodes to reduce the risk of public health damage, is one of the most important and challenging task in BTH in recent a couple of years. The local government will implement a set of emergency control activities in heavy pollution periods, including shutting down industries, restricting on road vehicles, etc. The method pursued in this study will give a prediction of when severe pollution appears, a large-scale reduction of which source may effectively cut down the peak concentrations. That information is important to the policy making of the local governments.

2.2 Evaluation database and protocols

The observational data used for model evaluation in this study include those used in Wang et al. [5,24] (see Table 1, Wang et al. [24]), and are described in detail in Wang et al. [5,24]. In summary, the National Climate Data Centre (NCDC) integrated surface database are used for evaluation of meteorological predictions, in terms of five major parameters: temperature at 2-m (T2), water vapor mixing ratio at 2-m (Q2), wind speed at 10-m (WS10), wind direction at 10-m (WD10), and daily precipitation. In total data at every 1- or 3-h (most at 3-h) at a total of 371 sites within our domains are used. To evaluate the aerosol radiative effects on meteorology, additional meteorological variables, i.e., SWDOWN and PBL height, are evaluated against the observations of the Clouds and Earth's Radiant Energy System (CERES) and the NCEP FNL PBL data.

Two data sets of chemical concentrations are used: the real-time database from the China National Environmental Monitoring Center (CNEMC) at 496 national monitoring stations in 74 major cities, and the observations at a site located in the Hebei University of Engineering (refer to as HEBEU) in Handan measured by the lead author's group since July 2012 [8]. The observations from CNEMC for the period of January 1–13 are unavailable for the model evaluation, due to the inaccessibility of the historic data of more than 24 h ago on the website of CNEMC [5]. Therefore, the $PM_{2.5}$ observations from Wang et al. [19] and Jiang et al. [31] for the whole month at the Shijiazhuang and Beijing are also used in this study in model evaluation and analysis of the aerosol direct effects in the two cities.

The meteorological evaluation is performed in terms of domainwide overall statistics, including the Mean Bias (MB), the Root Mean Square Error (RMSE), the Normalized Mean Bias (NMB), and the Normalized Mean Error (NME). A model performance having $MB \leqslant \pm 0.5$ K for T2, $MB \leqslant \pm 1$ g kg^{-1} for Q2, $MB \leqslant \pm 0.5$ m·s^{-1} for WS10, and $MB \leqslant \pm 10°$ for WD10 is seen as a satisfactory performance in this study according to the recommendations of Emery et al. [32] and Tesche et al. [33]. In addition to NMB and NME, the Mean Fractional Bias (MFB) and the Mean Fractional Error (MFE) are analyzed for the evaluation of chemical concentrations, following the guidance by US Environmental Protection Agency [34]. The criteria for chemical evaluation are refer to Boylan and Russell [35], that a model performance of $MFB \leqslant \pm 60\%$ and $MFE \leqslant 75\%$ for particulate matter (PM) is acceptable.

The evaluation on overall statistics for both meteorology and chemistry is described in Section S2, and the spatial distribution of some meteorological variables (e.g., SWDOWN and PBL) are presented in Section 3 in the supplementary file. The time series of $PM_{2.5}$ concentrations in the representative cities (i.e., Shijiazhuang, Xingtai, Handan, and Beijing) are discussed in section 3 below, to further assess the model performance in reproducing the temporal variations as well as the responses of those variables to aerosol-meteorology feedbacks.

3 Results and discussions

3.1 Impact of aerosol direct effects on $PM_{2.5}$ concentrations

The model performance evaluation against the NCDC observations (supplementary file) indicates that the model well reproduces humidity, wind direction and precipitations, but overpredicts the wind speed over both domains. The 2-m temperature is overpredicted over Doman 1 but is acceptable over Domain 2. The air quality predictions agree well with the observations from CNEMC, especially for the gaseous pollutants of NO_2 and SO_2. Although the WRF/Chem model tends to underpredict the concentrations of $PM_{2.5}$ and PM_{10}, which may partially due to the overprediction of wind speed, it still produces an acceptable prediction that meets the criteria proposed by US EPA [34].

The results of the BASE the NF simulations (supplementary file) indicate that the aerosol direct radiative effects will result in an average 11.5 W·m^{-2} decrease in SWDOWN, and an average 0.2°C(0.2 K) decrease in

Table 1 Impacts of aerosol direct effects on predicted hourly $PM_{2.5}$ concentrations (BASE-NF) in different $PM_{2.5}$ concentration ranges in Shijiazhuang, Xingtai, Handan, and Beijing

City	Predicted concentration range (BASE, $\mu g \cdot m^{-3}$)	No. of data pairs	Average prediction (BASE, $\mu g \cdot m^{-3}$)	Change in $\mu g \cdot m^{-3}$ (BASE-NF)	Change in % ((BASE-NF)/BASE)
Shijiazhuang	All	744	285.1	15.3	5.4
	⩽200	250	136.8	5.2	3.8
	200–400	327	282.8	11.8	4.2
	>400	167	511.5	37.3	7.3
Xingtai	All	744	251.5	9.3	3.7
	⩽200	307	139.2	–3.0	–2.2
	200–400	348	289.2	12.4	4.3
	>400	89	489.4	39.3	8.0
Handan	All	744	290.1	13.2	4.6
	⩽200	214	147.4	–3.5	–2.4
	200–400	387	281.1	7.5	2.7
	>400	143	527.9	53.5	10.1
Beijing	All	744	171.6	6.1	3.6
	⩽200	521	102.2	3	2.9
	200–400	171	280.1	9.1	3.2
	>400	52	510.4	26.6	5.2

temperature over Domain 1, and 13.1 $W \cdot m^{-2}$ and 0.2°C decrease over Domain 2. Over the southern Hebei area, the SWDOWN will be decreased by 20–27.5 $W \cdot m^{-2}$ (16%–24%), and the PBL height will be reduced by about 20–50 m (4%–18%). Considering the aerosol direct feedbacks will improve the model performance on SWDOWN with the NMBs reducing from 24% to 17% over Domain 1 and 46% to 33% over Domain 2 (against the CERES observations). And the predicted $PM_{2.5}$ concentraitons will increase around 1.8 $\mu g \cdot m^{-3}$ (~2%) in $PM_{2.5}$ and 2.2 $\mu g \cdot m^{-3}$ (~2%) in PM_{10} concentrations over Domain 2 at 12 km grid resolution. (supplementary file).

Figure 1 presents the simulated $PM_{2.5}$ concentrations over Domain 1 at 36 km grid resolution and Domain 2 at 12 km grid resolution, as well as the differences between the BASE and NF scenarios in these two domains in absolute value and precentage. Figure 1(a) shows that in Domain 1, the highest $PM_{2.5}$ concentrations appear in two regions, that one is from the southern Hebei, acrossing the middle and eastern Henan and Hubei, to the northern Hunan, and the other is the Sichuan Basin. Involving the aerosol feedbacks (BASE-NF) will not remarkably alter the spatial distribution of $PM_{2.5}$ concentrations, that the most obvious changes include the increase over the Sichuan Basin (at a level of 7.5–20 $\mu g \cdot m^{-3}$ and 7.5%–12%), the north-east and southern area of Hebei (1.5–7.5 $\mu g \cdot m^{-3}$ and 1.5%–7.5%), and in Beijing and Tianjin (1.5–8 $\mu g \cdot m^{-3}$ and 1.5%–8%). But over some regions, e.g., Shandong Province, the western area of Hubei and Hunan Province, the aerosol feedbacks will lead to an decrease of 1.5–6.7 $\mu g \cdot m^{-3}$ (1.5%–9.0%) in $PM_{2.5}$ concentrations. This can be explained by that the aerosol direct feedbacks will reduce the incoming solar radiation on the ground and thus result in lower PBL height which will increase the pollutant concentrations, but at the same time, the aerosol feedbacks may also lead to changes in wind field, which may decrease the pollutant concentrations [17,21]. And also, the reduction of radiation will lead to an decrease in temperature and concentrations of oxidant radicals and species, e.g., HO, HO_2, and O_3, which will both slow down the formation of secondary aerosol species, e.g., SO_2 to SO_4^{2-}, NO_2 to NO_3^-, and VOCs to seconday organic aerosols (see Figs. S5a and 5b), although these effects may not be the major PM increase pathway [17,21]. The net change of $PM_{2.5}$ concentrations depends on the relative significance of these effects.

When nesting to Domain 2 (Fig. 1(b)), higher concentrations of $PM_{2.5}$ occur over the areas from the urban Beijing, the central southern Hebei, to northern Henan. The comparison of two simulations indicate an increase in $PM_{2.5}$ concentrations by aerosol direcet effects in the urban area of the three cities, Beijing, Tianjin, Tangshan, and the linear shape area in central southern Hebei acrossing the cities of Baoding, Shijiazhuang, Xingtai, and Handan, at a level of 3–20 $\mu g \cdot m^{-3}$ and 3%–9%. The decrease in $PM_{2.5}$ concentrations appears in the areas along those increasing areas, at a level of less than 3%.

In summary, to the three southern Hebei cities which this paper focuses on, the aerosol direct effects will result in an

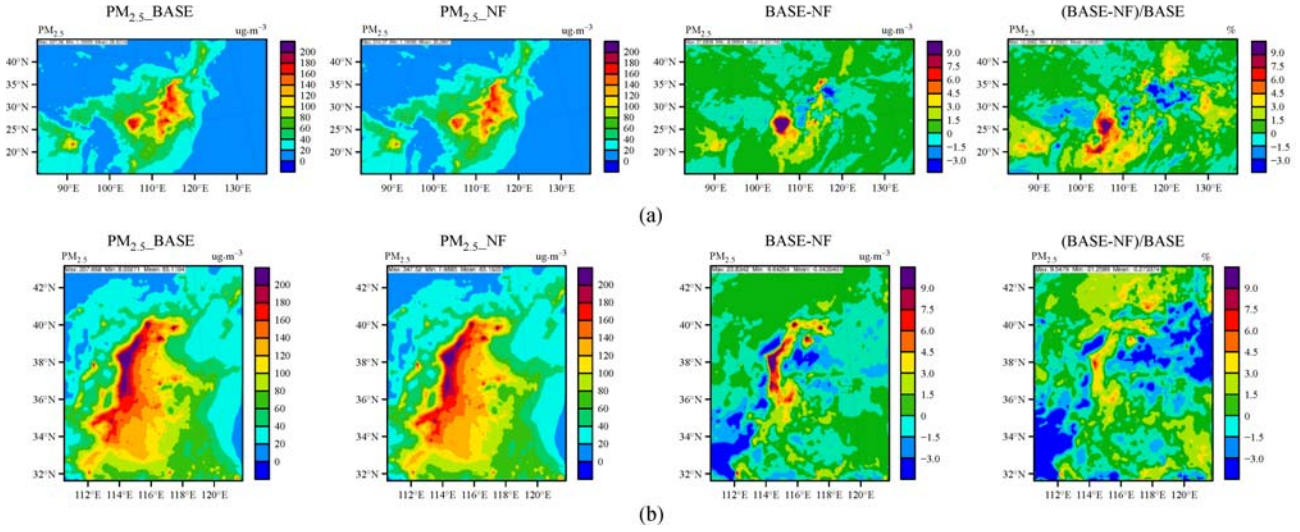

Fig. 1 Simulated $PM_{2.5}$ concentrations in the BASE and NF simulations, and their diferences in absolute value (BASE-NF) and percentage ((BASE-NF)/BASE) over Domain 1 (a) and Domain 2 (b)

increase in $PM_{2.5}$ concentrations by around 3%–9% in urban area during the severe polluted month of January 2013. This percentage is not extremely remarkable partially due to the monthly averaging of $PM_{2.5}$ concentration. Figure 2 presents the time series of the observed and predicted $PM_{2.5}$ concentrations by the BASE and NF simulations at the three cities in southern Hebei, and Beijing, as a comparison, to further understand the aerosol direct effects on the $PM_{2.5}$ concentrations under different pollution levels. It can be seen that comparing the differences between the BASE and the NF simulations, the aerosol direct effects are more obvious when $PM_{2.5}$ concentraions are on a high level. The model didn't reproduces the peak during January 12–14 in Shijiazhuang and Beijing (Fig. 2(a) and 2(d)), but considering the aerosol direct effects still reduce the NMBs from −16% (NF) to −11% (BASE) for Shijiazhuang, −15% to −12% for Beijing, respectively. For Xingtai city, the observations for the period of January 1–13 are not available, but considering the latter half month, the NMBs are reduced from −22% to −18% for Xingtai. As for Handan, the model captured the first $PM_{2.5}$ peak during January 6–8, and the first half peak in January 11–12, but failed in the following two days. The BASE predictions are much closer to the peak concentrations than the NF. The NMBs changes are from −6 (NF) to −1% (BASE).

To understand the aerosol effects in difference pollution level, we also summarize the differences between the two simulations under three $PM_{2.5}$ concentrations ranges, i.e., $\leqslant$200, 200–400, and >400 $\mu g \cdot m^{-3}$, in Table 1. First of all, the aerosol direct effects are more obvious in high $PM_{2.5}$ loading, e.g., when hourly $PM_{2.5}$ concentrations are higher than 400 $\mu g \cdot m^{-3}$, those effects for the three southern Hebei cities may reach as high as 8.2%–12.1% (39.3–59.5 $\mu g \cdot m^{-3}$), which cannot be ignored when considering the forecasting and emergent control plans of $PM_{2.5}$ pollution. When $PM_{2.5}$ concentrations are in the range of 200–400 $\mu g \cdot m^{-3}$, those effects could result in an increase around 4.0%–4.7% (12.4–14.0 $\mu g \cdot m^{-3}$). It is interesting that when the concentrations of $PM_{2.5}$ are lower than 200 $\mu g \cdot m^{-3}$, the aerosol direct feedbacks will result in a slight increase (2%, 3.7 $\mu g \cdot m^{-3}$) in $PM_{2.5}$ concentrations in Shijiazhuang, as well as in Beijing (2.6%, 3.0 $\mu g \cdot m^{-3}$), but will lead to a slight decrease in Xingtai (−2.3%, −2.9 $\mu g \cdot m^{-3}$) and Handan (−3.2%, −4.2 $\mu g \cdot m^{-3}$), which indicates that the decrease in $PM_{2.5}$ concentrations due to the change in wind field and decrease in atmospheric oxidant concentrations will offset the increase in $PM_{2.5}$ concentrations due to the lower PBL height in Handan and Xingtai under better air quality. Anyway, it can be summarized that the aerosol effects will enhance the urban $PM_{2.5}$ pollution during severe polluted period and increase the peak concentration of $PM_{2.5}$ (e.g., about 10% when $PM_{2.5}$ concentrations higher than 400 $\mu g \cdot m^{-3}$), but at the same time, when peak concentrations of $PM_{2.5}$ are reduced through effective control strategies, those effects will obviously decreased (e.g., about 4% when $PM_{2.5}$ concentrations in 200–400 $\mu g \cdot m^{-3}$).

3.2 Source contributions to $PM_{2.5}$

As mentioned above, the BFM is applied to assess the source contributions of five major emission sectors in the BTH, i.e., PO, IN, DO, TR, and AG to the $PM_{2.5}$ concentrations in the three southern Hebei cities, Shijiazhuang, Xingtai, and Handan, under the simulation scenarios of turning on and off aerosol direct feedbacks. The objective of those calculations is to get understanding

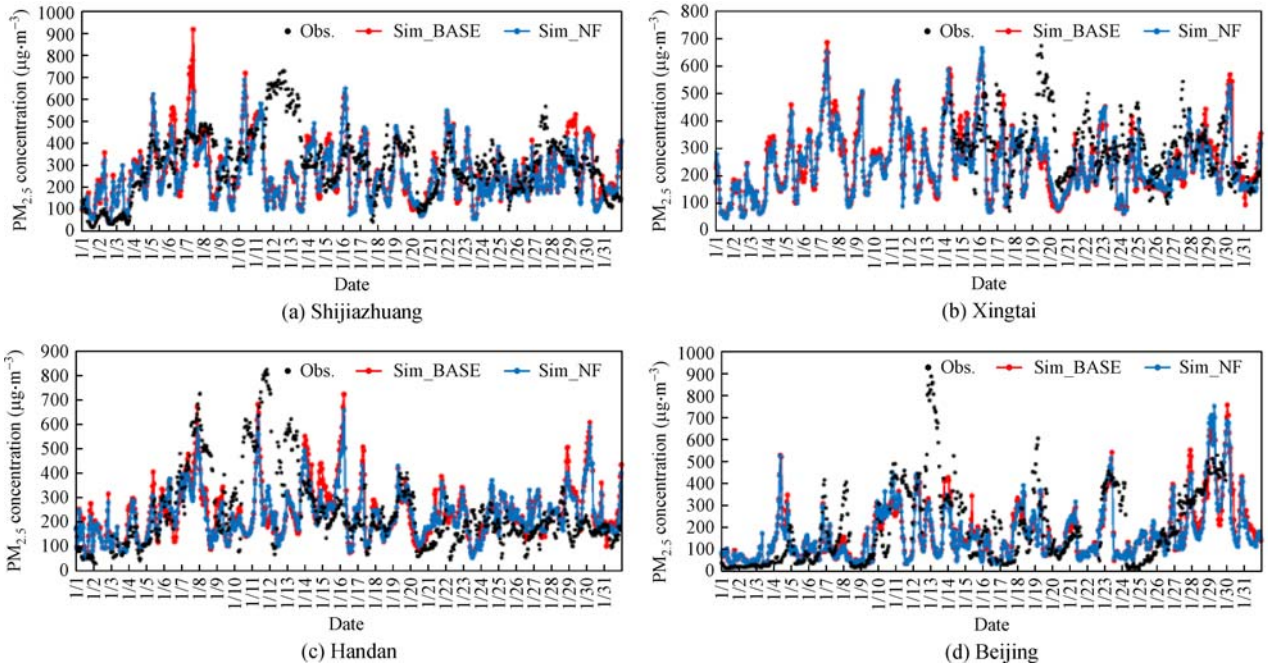

Fig. 2 Time series of observed and predicted $PM_{2.5}$ concentrations in Shijiazhuang, Xingtai, Handan, and Beijing, by the BASE and NF simulations

on the uncertainties introduced by not considering those effects in evaluating source contributions using 3-D air quality models.

Figure 3 presents the spatial distributions of the source contributions of the five emission sectors over Domain 2 predicated by BASE, NF scenario, and their differences (note the lengend scale of IN and DO is different to the others because of their large contributions). It can be found that the largest discrepancies between the two simulations appear over the southern Hebei area, Beijing, Tianjin, and some regions of northern Henan and western Shandong, where the most severe pollution occurrs (see Fig. 1). The contributions of PO, IN, DO, and TR will be increased when considering the aerosol direct feedback, but those of AG are on the contratry. The decrease in AG source contributions can be partially explained as the concentration decrease of the secondary inorganic species, i.e, $(NH_4)_2SO_4$ and NH_4NO_3, resulted by the decrease of atmospheric oxidizability (see Fig. S5) when considering the aerosol feedback, overcomes the increase lead by the meteorolgical changes (e.g., lower PBL, lower wind speed, higher humidity). The largest increase appears over the southern Hebei area, with the source contributions of PO, IN, DO, and TR increasing about 0.5–2.0, 1.0–18.7, 1.0–16.1, 1.0–3.2 $\mu g \cdot m^{-3}$, respectively. The decrease of AG over those area is about -4.0– -0.5 $\mu g \cdot m^{-3}$. That is to say, when we assess the source contributions using 3-D air quality models using the Brute Force method without considering the aerosol radiative feedbacks, the source contributions of PO, IN, DO, and TR will be most possibly underestimated and those of AG will be slightly overestimated over the Beijing-Tianjin-Hebei area.

To get more detailed understanding of those discrepancies under different pollution level, the source contribution ranges of the five emission sectors when $PM_{2.5}$ concentrations are under 200 $\mu g \cdot m^{-3}$, in 200–400 $\mu g \cdot m^{-3}$, and above 400 $\mu g \cdot m^{-3}$ by the two simuations for Shijiazhuang, Xingtai, and Handan are plotted in Fig. 4. And Table 2 summarizes the average contribtutions of the two simulation. First, the contributions of PO are small under all concentration levels for all the three cities, which are 0.6%, -0.4%, and -0.4% in BASE and 0.6%, -0.7%, and -0.7% in NF, for Shijiazhuang when $PM_{2.5}$ concentrations under 200 $\mu g \cdot m^{-3}$, in 200–400 $\mu g \cdot m^{-3}$, and above 400 $\mu g \cdot m^{-3}$, respectively. For Xingtai, the PO contributions change from -0.2%, -0.2%, and -0.2% of BASE, to -1.1%, -0.8%, and -0.5% of NF, and those numbers for Handan are 0.3%, -0.3%, and -0.3% of BASE, and 0.1%, -0.7%, and -0.8% of NF. It indicates that considering the aerosol feedbacks will lead to smaller negative contributions under higher $PM_{2.5}$ loadings (above 200 $\mu g \cdot m^{-3}$), but when $PM_{2.5}$ is lower than 200 $\mu g \cdot m^{-3}$, it will almost not change the contribution (Shijiazhuang), or lead to a larger positive contribuition (Handan), or alter a small negative contribution to a small positive contribution (Xingtai).

IN and DO are the two largest source contributors to $PM_{2.5}$ pollution in the three cities. For Shijiazhuang, although the contributions of IN predicted by BASE and

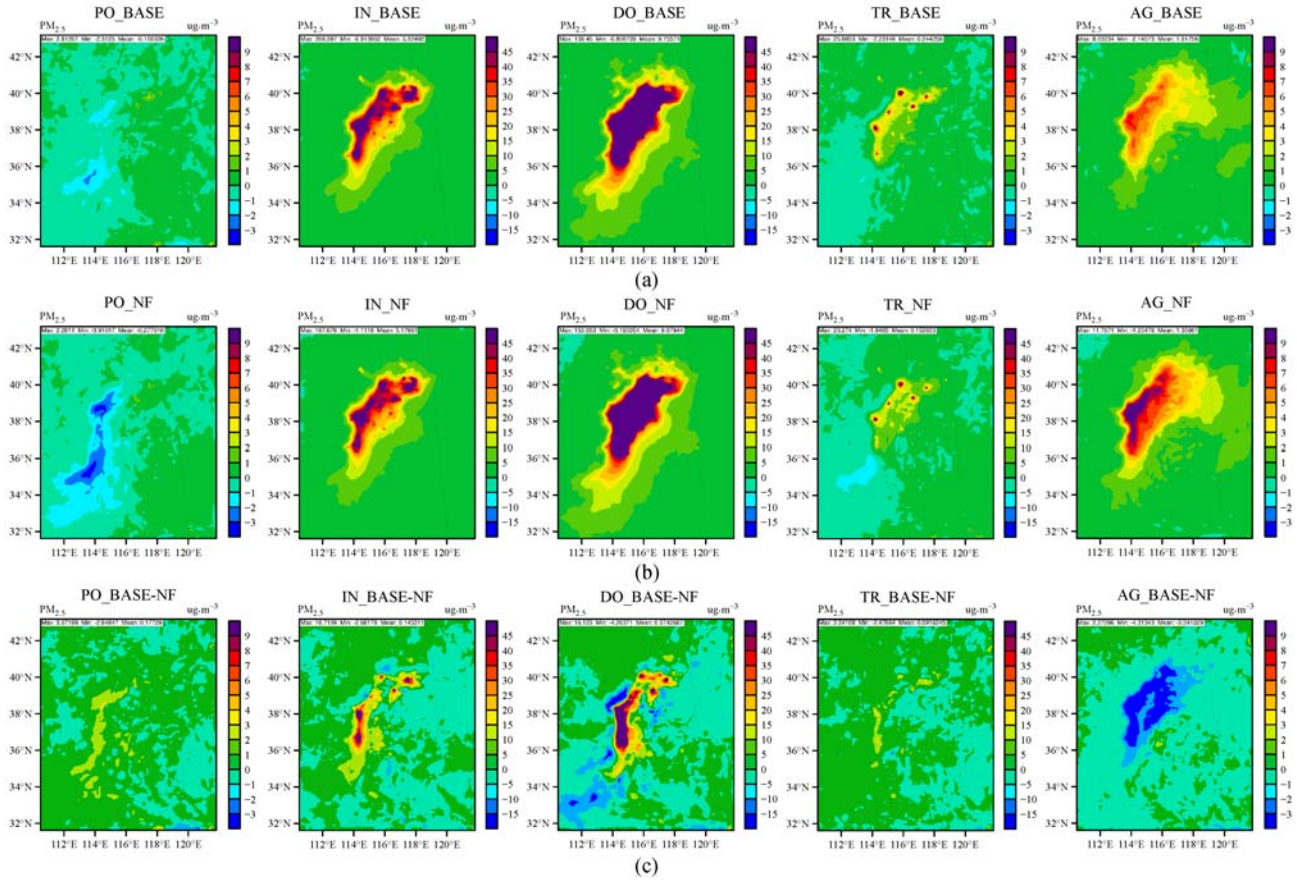

Fig. 3 Source contributions of PO, IN, DO, TR, and AG to $PM_{2.5}$ concentrations over Domain 2 calculated by the BASE (a) and NF (b) simulations, and their diferences in BASE-NF (c)

NF are only 2.7% in difference (36.7% vs. 34.0%), the contributions under different $PM_{2.5}$ pollution levels are 34.1%, 37.3%, and 39.4% of BASE, and 31.5%, 34.7%, and 36.4% of NF for $PM_{2.5} < 200$ $\mu g \cdot m^{-3}$, in 200–400 $\mu g \cdot m^{-3}$, and >400 $\mu g \cdot m^{-3}$, respectively. It indicates the facts that the discrepancies between considering aerosol feedbacks and not will obviously increase when $PM_{2.5}$ loadings grow (e.g., 2.6% for $PM_{2.5} < 200$ $\mu g \cdot m^{-3}$, and 3.0% for $PM_{2.5}$>400 $\mu g \cdot m^{-3}$). Those discrepancies for the other two cities are even more significant as 1.3%, 2.0%, and 5.3% for Xingtai, and 2.4%, 2.3%, and 5.5% for Handan when $PM_{2.5} < 200$ $\mu g \cdot m^{-3}$, in 200–400 $\mu g \cdot m^{-3}$, and >400 $\mu g \cdot m^{-3}$, respectively (see Table 2). That is to say, if aerosol radiative feedbacks are not considered in calculating the source contributions of IN, we may introduce nonnegligible underestimation especially under high $PM_{2.5}$ loadings, which are exactly the most important periods we concern. The contributions of DO have the similar pattern, that not considering the aerosol feedbacks will result in underestimations of its contributions by 2.7% and 3.2% for Shijiazhuang, 2.6% and 6.6% for Xingtai, and 1.1% and 7.4% in Handan, for $PM_{2.5}$ in 200–400 $\mu g \cdot m^{-3}$, and >400 $\mu g \cdot m^{-3}$, respectively. Note DO averagely contributed 41.4% and 38.3% of $PM_{2.5}$ in Xingtai and Handan. Therefore, the underestimations of 6.6% and 7.4% when $PM_{2.5}$>400 $\mu g \cdot m^{-3}$ are about 16% and 19% of the total contributions.

The source contributions of TR calculated by 3-D model using Brute Force method are generally low, as averagely 5.3%, 1.9%, and 3.4% for Shijiazhuang, Xingtai, and Handan, respectively, which is consistent to our previous study using CMAQ [5]. When considering aerosol feedbacks, the contributions of TR will increase by 0.7%–1.1% for Shijiazhuang, 0.2%–1.1% for Xingtai, and 0.4%–1.3% for Handan, respectively, under the three pollution levels. The average increases are 0.8%, 0.5%, and 0.8%, respectively.

The contributions of AG are quite different with other sources that its average contributions will decrease when considering aerosol feedbacks. Under lower $PM_{2.5}$ loadings, its contributions are relatively higher, as 3.5%, 4.7%, and 4.2% for Shijiazhuang, Xingtai, and Handan, respectively. When $PM_{2.5}$ concentrations increase to above 400 $\mu g \cdot m^{-3}$, its contributions decrease to 1.9%, 0.7%, and 1.1% for the three cities. Considering the aerosol feedbacks will lead to about −1.3%, −1.5%, and −1.3%

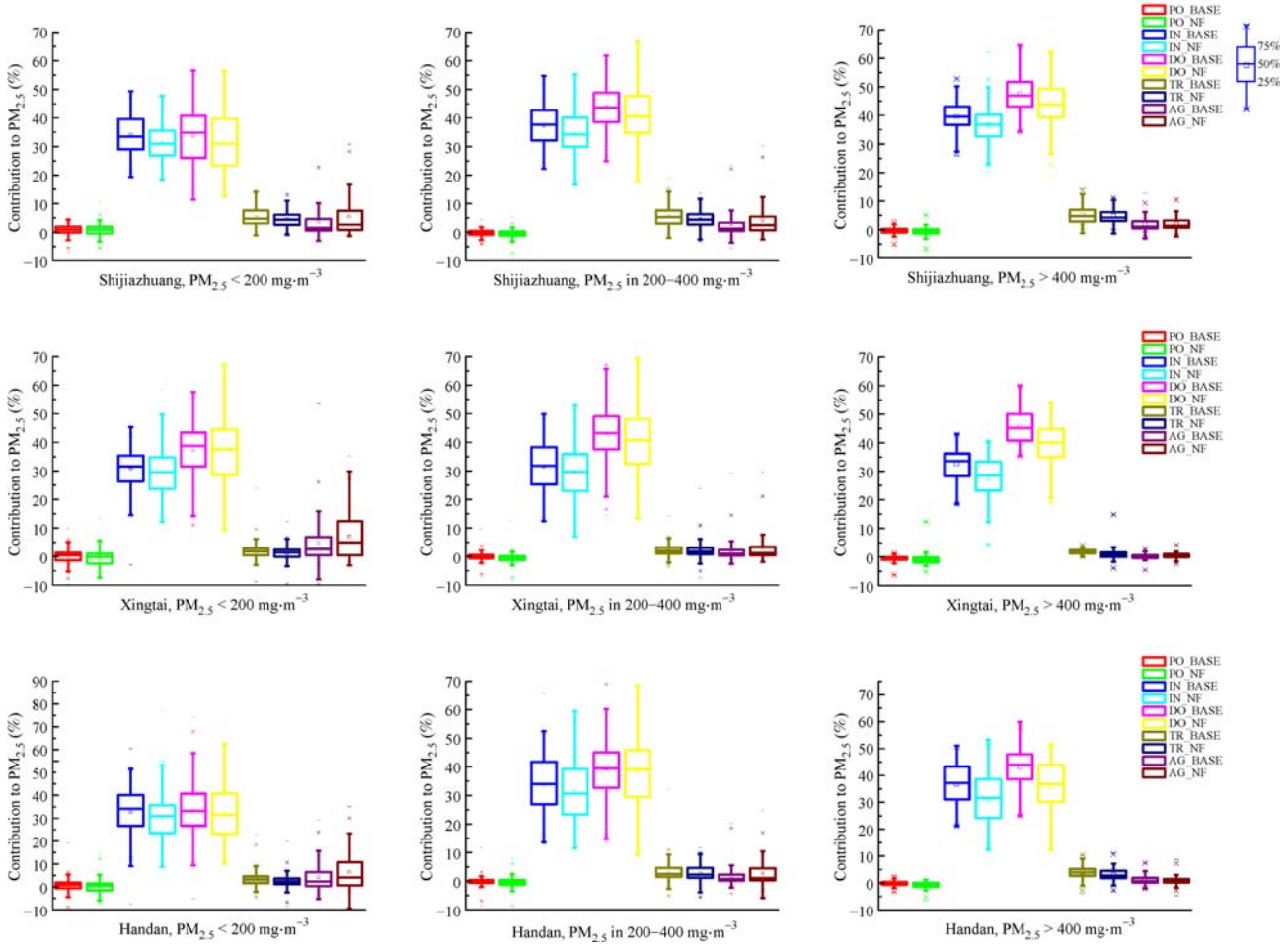

Fig. 4 The source contribution ranges of PO, IN, DO, TR, and AG to $PM_{2.5}$ concentrations in Shijiazhuang (row 1), Xingtai (row 2), and Handan (row 3) at different pollution level, calculated by the BASE and NF simulations

decrease in AG contributions. In summary, the contributions of AG under high $PM_{2.5}$ loadings are not as important as under lower $PM_{2.5}$ loadings.

To understand the relative significance of these source contributions during the pollution episode and their changes when considering or not the aerosol feedbacks, the simulated $PM_{2.5}$ concentrations and source contributions from PO, IN, DO, TR, and AG during the typical pollution episode from Jan. 5–8 are plotted in Fig. 5, for the BASE (left column) and the NF (right column). It can be seen that the contribution of IN and DO are obviously increased when considering the aerosol direct effects. In the most severe pollution episode from Jan. 6 to 7, the source contribution of IN and DO in Shijiazhuang are 33.8% and 42.4% in NF, respectively, but increase to 38.5% and 47.7%, respectively, when considering the aerosol direct effects. During the peak concentration period on Jan. 7, the contributions of IN and DO are 36.9% and 39.8% in NF, and 43.4% and 47.6% in BASE, respectively. The case is similar for Xingtai and Handan that during the severe pollution episode, the source contributions of IN and DO increase about 4.9%–8.8% when considering the aerosol direct effects. Furthermore, during the peak concentration hours of this episode, the contributions of IN and DO will increase about 8.6%–12.9% when considering the feedbacks. In Xingtai, the contribution of DO even exceed 50%. It reveals that when we consider short-term action plan to cut down the peak concentrations of $PM_{2.5}$ during winter haze episode in these cities, it is necessary to consider aerosol direct feedbacks, and the source apportionment results on monthly- or season-averaged are not enough to support the policy making. The episode based analysis is necessary, and for these southern Hebei cities, the industrial and domestic sources should be further considered because that their contributions might be more significant than the previous assessments during peak hours.

4 Conclusions

In this study, the WRF/Chem model was applied over the

Table 2 Impacts of aerosol direct effects on source contributions (%) of PO, IN, DO, TR, and AG to $PM_{2.5}$ concentrations (BASE vs. NF) in different $PM_{2.5}$ concentration ranges in Shijiazhuang, Xingtai, and Handan

City	Predicted concentration range (BASE, μg m^{-3})	PO_BASE	PO _NF	IN_BASE	IN _NF	DO_BASE	DO _NF	TR_BASE	TR _NF	AG_BASE	AG _NF
Shijiazhuang	Average	0.1	–0.3	36.7	34.0	41.2	38.7	5.3	4.5	2.8	4.1
	⩽200	0.6	0.6	34.1	31.5	33.7	31.9	5.2	4.5	3.5	5.5
	200-400	–0.4	–0.7	37.3	34.7	43.9	41.2	5.5	4.4	2.8	4.2
	>400	–0.4	–0.7	39.4	36.4	47.2	44.0	5.0	4.6	1.9	2.0
Xingtai	Average	–0.2	–0.9	31.6	29.5	41.4	39.5	1.9	1.4	2.9	4.4
	⩽200	–0.2	–1.1	30.6	29.3	37.3	37.4	1.6	0.9	4.7	7.0
	200-400	–0.2	–0.8	31.6	29.6	43.2	40.6	2.1	1.9	1.9	2.9
	>400	–0.2	–0.5	35.1	29.8	48.8	42.2	2.5	1.4	0.7	1.1
Handan	Average	–0.1	–0.5	34.1	31.2	38.3	36.1	3.4	2.6	2.3	3.6
	⩽200	0.3	0.1	32.7	30.3	33.3	32.6	3.5	2.2	4.2	6.5
	200-400	–0.3	–0.7	34.0	31.7	39.3	38.2	3.1	2.7	1.7	2.9
	>400	–0.3	–0.8	36.6	31.1	42.9	35.5	4.0	3.0	1.1	1.0

East Asia at 36 km horizontal grid resolution and northern China at 12 km grid resolution, for the severe polluted month of January 2013. Two sets of simulations including the baseline simulation with aerosol-meteorology feedbacks (BASE) and a sensitivity simulation without feedbacks (NF) were pursued to evaluate the aerosol direct radiative effects on air pollution in the southern Hebei cities, i.e., Shijiazhuang, Xingtai, and Handan. In each set, six emission sensitivity scenarios, i.e., the base case and the scenarios turning off the emissions from power plants (PO), industrial (IN), domestic (DO), transportation (TR), and agriculture (AG), separately, to evaluate their source contributions.

Considering the aerosol direct raditive effects will reduce the negative NMBs of the predicted $PM_{2.5}$ concentrations for the three cities (−16% to −11% for Shijiazhuang, −22% to −18% for Xingtai, and −6% to −1% for Handan). Those effects will result in an increase in $PM_{2.5}$ concentrations by around 3%–9% in urban area of the southern Hebei cities in January 2013. It is found that the increase in $PM_{2.5}$ concentrations are much more significant under high $PM_{2.5}$ loading (e.g., ~50 μg·m^{-3}, 10% when hourly $PM_{2.5}$ concentrations higher than 400 μg·m^{-3}), and lower in relatively better air quality (e.g., ~10 μg·m^{-3}, 4% when $PM_{2.5}$ concentrations in 200–400 μg·m^{-3}). When $PM_{2.5}$ concentrations are lower than 200 μg·m^{-3}, the aerosol direct feedbacks will result in a slight increase (~2%) in $PM_{2.5}$ concentrations in Shijiazhuang, but a slight decrease in Xingtai (~−2%) and Handan (~−3%). When applying the BFM to assess the source contributions, considering the aerosol feedbacks will result in increases in the calculated contributions from PO, IN, DO, and TR, but a decrease in the AG contribution, comparing to that not considering the feedbacks. Under high $PM_{2.5}$ loadings, the increases in IN and DO contributions are siginificant that cannot be neglected, e.g., from 44.0% to 47.2% for Shijiazhuang, 42.2% to 48.8% for Xingtai, and 35.5% to 42.9% for Handan for DO contribution under $PM_{2.5}$>400 μg·m^{-3}. It implies that those effects should not be ignored when considering the $PM_{2.5}$ forecasting or emergent control activity during severe pollution episodes, and the source contributions from IN and DO are more significant than the results from previous studies. But when peak concentrations of $PM_{2.5}$ are reduced through effective regional control strategies in the future, those effects will mitigate, which may reduce some difficulties to further cut down the episodic $PM_{2.5}$ concentrations in the three top polluted cities.

Several limitations remain in this study. First, as discussed in the supplementary file, uncertainties in the spatial allocation of the emissions may introduce uncertainties in the predictions of pollutants concentrations and the following source apportionment results. Second, although the BFM is capable of quantifying the direct and indirect affects due to interactions among PM and PM precursors, it cannot attribute 100% of atmospheric pollutants concentrations to source emissions as does a true source apportionment method. Third, one month simulation is not enough to fully evaluate the aerosol direct effects on air quality predictions and source contributions. The simulation period should be extended to get more understanding for different season and different year. Nevertheless, this study provides valuable scientific insights into the impacts of aerosol radiative feedbacks on air quality and source apportionment evaluation, which are the foundation for policymaking to effectively mitigate the severe air pollution over the BTH area and reduce the peak concentrations.

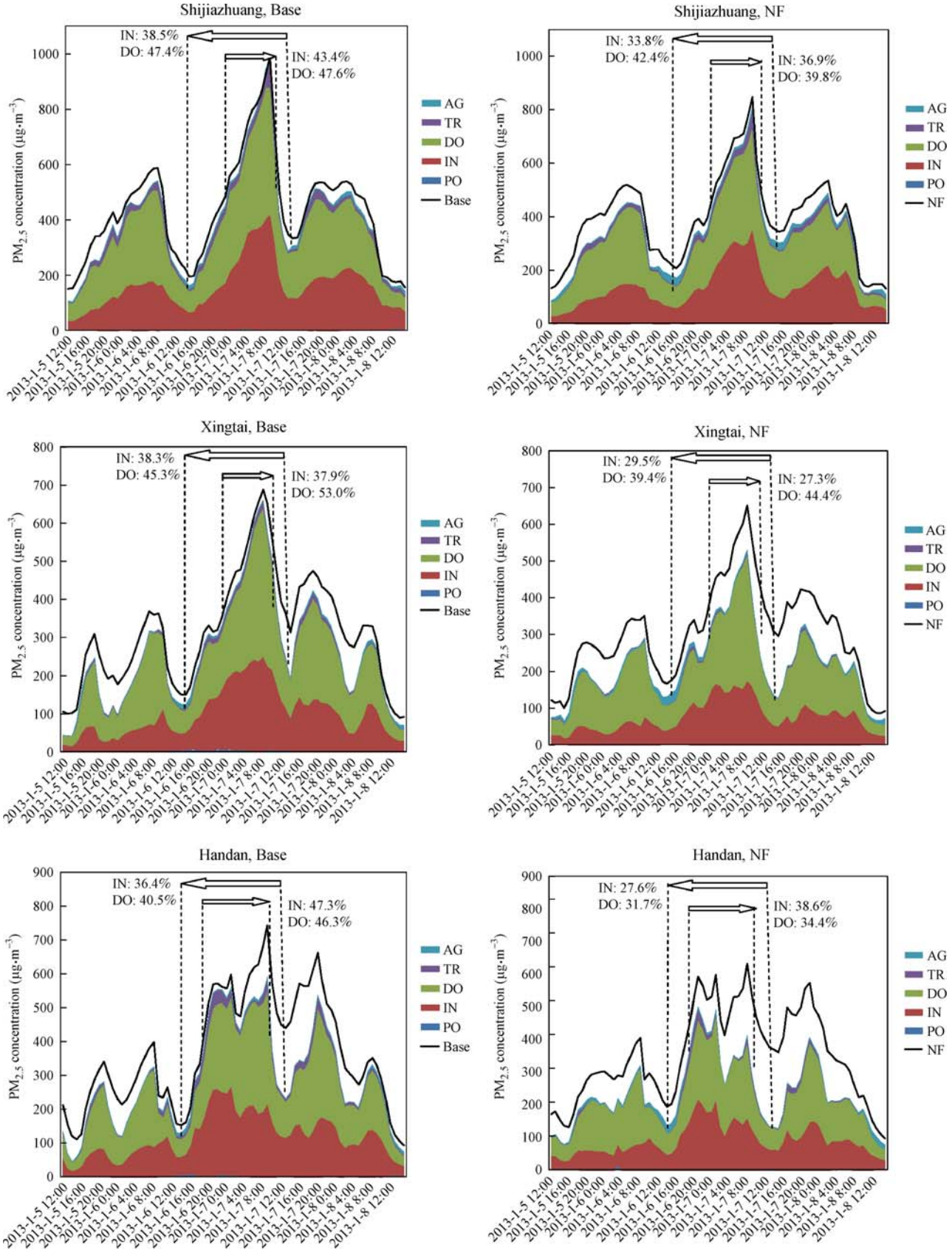

Fig. 5 Comparison of simulated $PM_{2.5}$ concentration and the source contributions of PO, IN, DO, TR, and AG in Shijiazhuang (row 1), Xingtai (row 2), and Handan (row 3) in BASE and NF for the extremely polluted episodes during Jan. 5–8, 2013. The two pairs of vertical dashed lines indicate a whole typical pollution episode and the extremely polluted hours during this episode, respectively. The arrows point to the average contributions of IN and DO during these two time intervals

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