Reply to Anonymous Referee #1

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestion, as described below.

Source attribution of air pollution is of great importance in emission control policy making. This work developed a source-oriented method in WRF-Chem regional model and applied it in the source appointment of fine particle pollution in the highly-pollution North China Plain region. Based on a one-month simulation using this source-oriented version of the WRF-Chem model, the authors indicated different contributions from local and non-local emissions for Beijing, Tianjin and other provinces and highlight the cooperation among provinces. Overall, this work is well structured but still needs more clarification and some in-depth analysis. Here are some issues that are suggested to be addressed for further improving this work.

1 Comment: Some detailed configurations of model need to be clarified and updated. The definition of source regions needs to be provided in the method. There is no information or figure of the model domain and designation of source regions. See Fig.1 in Hu et al. (2015). Also, technically, the simulations are performed based on an emission inventory for the year of 2006. It is well known that China has made great effort in emission control in the past decade. For instance, SO_2 has been dropping sharply since 2006, and the SO_2 emission are estimated to be drop by over 70% in NCP in past 5 years due to the implementation of the toughest-ever clean air policy in China (Zhang et al., 2019). Is the emission inventory for the year of 2006 can represent the current emission scenario since the emissions underwent dramatic changes in both magnitude and spatial distribution in recent years?

Response: We have added figure S1 to show designation of the source region and clarified in Section 3.2: "*We have marked the emitted precursors in six provinces, including Beijing, Tianjin, Hebei, Henan, Shandong, and Shanxi in simulations of the source-oriented WRF-Chem model (Figure S1).*".

We have clarified in Section 2.1: "*It is worth noting that the emission inventory used in this study is developed by Zhang et al. (2009) and Li et al. (2017) with the base year of 2012. Considering that the great changes in emission inventory due to implementation of the toughest-ever clean air policy in China (Zhang et al., 2019), the emission inventory has been adjusted according to the trends from 2012 to 2015 proposed by Zheng et al. (2018).*".

2 Comment: Besides of photochemistry and heterogeneous chemistry, chemical production in cloud water is also an important contributor to secondary aerosol like sulfate and SOA. Why not track it in the SA calculation?

Response: We have explained in Section 2.2: "*It is worth noting that, although it is lack of precipitation during the simulated episode, the SA of sulfate formed in cloud water is also considered. The SO_2 in cloud water is oxidized mainly by H_2O_2, O_3, NO_2, formic acid, and O_2* catalyzed *by Fe^{3+} and Mn^{2+}*.". We have considered the heterogeneous SOA formation from glyoxal and methyglyoxal on aerosol or cloud droplet surfaces with a reactive uptake coefficient of 3.7×10^{-3}.

3 Comment: The descriptions on the model modifications need more detailed information and supporting references. 1. The yield value is vital for the simulating SOA but most references cited in this work is too outdated. Please specify the yield values from different VOCs to S/IVOC used in this simulation. 2. Line148-150, how the heterogenous oxidation of SO_2 in the aerosol water are parameterized? The aerosol water is not an ideal solution and thus all the classic reaction rate is not applicable here, and how the effect of ionic strength and aerosol water acidity that would significantly influence mass transfer are considered. 3. As mentioned, ISORRPIA is calculating aerosol thermodynamical equilibrium. How does the model attribute the chemical production from different sources since they are interacting with each other? The authors' writing style makes it quite hard to follow or repeat.

Response:

1. We have clarified in Section 2.2: "*The SOA yield from VOCs is NOx dependent (Li et al., 2011a). The high-NOx and low-NOx yields are listed in the Table S1 and parameters used to treat partitioning of POA emissions are listed in Table S2.*".

2. We have clarified in Section 2.2:"*In this study, a SO_2 heterogeneous reaction parameterization associated with aerosol water is used, in which the SO_2 oxidation in aerosol water by O_2 catalyzed by Fe^{3+} is limited by mass transfer resistances in the gas-phase and the gas-particle interface. Considering the effect of ionic strength and aerosol water acidity, the sulfate heterogeneous formation from SO_2 is therefore parameterized as a first-order irreversible uptake by aerosols, with a reactive uptake coefficient of 0.5×10^{-4}, assuming that there is enough alkalinity to maintain the high iron-catalyzed reaction rate (Li et al., 2017). The detailed description of the parameterization of the heterogeneous oxidation of SO_2 involving aerosol water can be seen in Supplement.*". We have provided the detailed description in Section S1.

3. We have explained in Section 2.2: "*Therefore, as a bulk method, the ISORROPIA cannot be applied to distribute the gas and aerosol phase for the inorganic aerosol from each source separately because of the interaction among various sources.* ", and "*The SA for nitrate and ammonium aerosols follows the mass conversion of $N(+VI)$ and $N(-III)$ from each source, respectively, when the total ammonia and nitrate are distributed between the gas and aerosol phases by the ISORROPIA after one time step integration, as shown in Figure 3.*"

4 Comment: Another, the discussion of the results is a little descriptive, and more in-depth analysis and political implications are suggested here. For instance, is there any difference in source attributions at different altitude, and why? To control the air pollution in a more cost-effective way, which kinds of emission sectors, like residential combustion and transportation, should be given priority over any other.

Response:

1. We have added in Section 3.2: "*Figure S20 also provides the vertical profiles of the average $PM_{2.5}$ contribution from local and non-local emissions in Beijing, Tianjin, Hebei, Henan, Shandong, and Shanxi during the episode. Generally, the $PM_{2.5}$ contribution of local emissions in the six provinces in the NCP declines rapidly with altitude due to the efficient advection in the upper PBL. The local contribution decreases to less than 20% in the upper PBL in Beijing and Tianjin and is generally more than 25% in the other four provinces. In Shandong, the $PM_{2.5}$ concentration is mainly dominated by local emissions*

in the lower PBL, but the local contribution presents a significant decreasing trend in the upper PBL."

2. We have added in Summary and conclusions: "*In this study, the source-oriented WRF-Chem model is also used to mark the precursors emitted from residential, transportation, industry, power, and agriculture sectors, respectively, to evaluate the contribution of anthropogenic emissions to the $PM_{2.5}$ concentration in the NCP. The average contribution of residential emissions to the $PM_{2.5}$ level is the most significant, with a maximum exceeding 100 μg m^{-3} during the study episode (Figure S21). In addition, the contribution of industry emissions to $PM_{2.5}$ concentration in the NCP also varies from 10 from 100 μg m^{-3} during the study episode. Therefore, more attention should be paid to residential and industry sectors to control the air pollution in a more cost-effective way.*" and "*The contribution of residential and industry emissions to the $PM_{2.5}$ concentration in Hebei, Shandong, and Henan is the most obvious (Figure S21). Therefore, efficient emission mitigations of air pollutants in the three provinces need to be carried out continuously to lower PM levels.*".

5 Comment: This work aims to discuss the contribution of local emissions and trans-boundary transport in NCP. Recent studies have demonstrated that the aerosol from cross-regional transport could exert substantial impacts on local meteorological condition in North China Plain, thereby deteriorating the $PM_{2.5}$ pollution in this region. Such interaction has been also identified to be an important process in trans-boundary pollution (Huang et al., 2020). Can this source-oriented model resolve such kind process and quantify the relative contribution.

Response: We have clarified in Summary and conclusions: "*The developed source-oriented model is mainly used in this study to quantitatively evaluate the local and non-local contributions to the PM pollution in the NCP. A recent study (Huang et al., 2020) has demonstrated that, absorption aerosols contributed by cross-regional transport from the Yangtze River Delta (YRD) to the upper PBL in the NCP induce the aerosol-PBL interaction and further lead to the suppressed PBL height, notable reduction of temperature and a substantial enhancement of relative humidity, favoring secondary aerosol production and aggravation of air pollution in the NCP. In this study, a sensitivity study without BC transported from the south of 32°N is conducted to analyze the contribution of the effect of*

cross-regional transport of air pollutants on local meteorological conditions during the selected simulated episode. The temperature and PBL height decrease in the NCP caused by the BC transported from the south are not significant, with a maximum of 0.04 ℃ and 1.6%, and the increase of relative humidity just varies from -0.2% to 0.1% (Figure S22). Therefore, the aerosol-PBL interaction induced by the trans-boundary transport of absorption aerosols can not be observed in this study. In the future, more typical air pollution episodes need to be simulated to quantify the impact of regional transport of absorption aerosols on meteorological conditions."

References:

Hu, J. L., Wu, L., Zheng, B., Zhang, Q., He, K. B., Chang, Q., Li, X. H., Yang, F. M., Ying, Q., and Zhang, H. L.: Source contributions and regional transport of primary particulate matter in China, Environ. Pollut., 207, 31-42, doi: 10.1016/j.envpol.2015.08.037, 2015.

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