Reply to Anonymous Referee #2

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.

Using a modified version of the WRF-Chem model, this paper utilizes a source apportionment method to examine particulate matter characteristics during a wintertime pollution event in China. The authors first compare the model output to chemistry and aerosol observations from ground-based sites and conclude that the model generally performs well. Then, the authors aim to explore the relative contributions of local and non-local emissions on air quality in various regions of China - a topic that is very important for emission regulations. Their findings suggest that for Beijing and Tianjin, local emissions tend to dominate when the air quality is excellent and good; however, the impact of non-local emissions becomes more pronounced as air quality decreases. I think that the results stemming from this work are interesting and worthy of publication. In general, the paper is well written and the authors do a satisfactory job explaining their findings. However, I do have a few comments regarding several topics on which the authors could further elaborate. Overall, I recommend that the paper be accepted for publication once the authors address my comments. My major and minor concerns are described below, and my grammatical recommendations are provided in an associated PDF document.

Major/general comments:

1 Comment: Pollution event meteorology: In the context of this study, the transport of pollution is strongly dependent upon regional meteorology (e.g., advection of particulate matter by the mean wind). However, the authors do not put nearly enough emphasis on this topic. How do the large-scale meteorological conditions evolve over the course of the month? Surely, there was some variability; even just looking at Fig. 5a, one can hypothesize that there is some synoptic-scale influence. Please add a figure showing this evolution, perhaps

near-surface pressure, winds, and temperatures at various snapshots during the event that correspond to the peaks and valleys in Fig. 5a. Moreover, the only discussion of wind flow is surrounding Fig. 6 and some other brief sections in the text. In Fig. 6, it appears as though the authors plot mean wind arrows for the pollution event. How do you calculate average wind direction during the time period? Do you think that it is valid to show a planview of average winds over a month-long period? Many of the regions show calm winds, but there is likely much variability over the course of the entire event. Showing a time series here of observed and/or modeled winds should help clarify. Additionally, regarding L210-212: Do you hypothesize that this is going on here? Are you able to use the surface measurements to determine if the modeled wind field is a major issue for this particular case? Again, perhaps a time series of wind speed/direction would help. Regarding L272-275: Did this occur during the case study examined here? The wind arrows in Fig. 6 suggest not, but it is difficult to tell since they are averages.

Response: We have added the description of the meteorological data in Section 2.3: "*The meteorological parameters including surface pressure, temperature, wind speed and direction with a 3-hour interval are obtained from the website http://www.meteomanz.com, including the observation sites at Beijing, Tianjin, Shijiazhuang, Jinan, Zhengzhou, Hefei, and Nanjing (Figure S1). Furthermore, the reanalysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) are used to analyze the synoptic patterns during the study episode.*".

We have clarified in Section 3.1: "*Generally, the accumulation and trans-boundary transport of air pollutants is mainly dependent on regional meteorological conditions. Figure S2 shows the average geopotential heights at 500hPa and the mean sea level pressures with wind vectors during the study episode. During the simulated episode, the NCP is situated behind the trough at 500 hPa. The NCP is controlled by the high pressure system at the surface on a large scale due to the upper level trough, ranging from 1026 to 1030 hPa, and the prevailing wind over the NCP is weak or calm, which is unfavorable for dissipation of air pollutants. Figure S3 shows the diurnal profiles of observed and simulated near-surface pressure, temperature, wind speed, and wind direction averaged at monitoring sites in the NCP from 05 December 2015 to 04 January 2016. The WRF-Chem model performs well in reproducing the diurnal variability of near surface pressure, surface temperature (TSFC), wind speed, and wind direction, with IOAs of 0.63, 0.84, 0.75, and 0.54, respectively. During the study episode, the simulated and observed of near surface pressures are 1024.0hPa and 1028.5hPa,*

indicating that a high pressure system controlling the NCP (Figure S2). The southerly wind prevails over the NCP during the study episode, with the simulated and observed wind direction of 180.6° and 175.1°. Moreover, the simulated and observed wind speed is approximately 2 m s^{-1} over the NCP during the simulated episode. Therefore, the air pollutants are subject to being transported from south to north, and the weak or calm wind also appears in some regions, which is favorable for the accumulation of air pollutants. For example, from 16 to 24 December 2015, the wind speed in the NCP decreases and the wind direction turns to be southerly, facilitating accumulation of air pollutants, and meanwhile a serious PM pollution episode with high $PM_{2.5}$ concentrations occurs.".

We have clarified the calculation of average of wind direction in Supplement Section S3: *"The wind direction simulated in this study is calculated using the U (the velocity toward east) and V (the velocity toward north) component at a specific grid point over the simulation domain and the average wind direction is calculated based on the average U and V."*.

2 Comment: Figure 5: Because there are so many sites, it would be nice to see the spread among sites. Is the model doing well at all of the sites? Or are many sites under- and over-predicted to "average out" and make it look like they are doing well? Does the model do well in one region over another? I suggest that you add a figure with panels showing scatter plots of these chemical species that compare observations and model for all sites and color by region.

Response: We have clarified in Section S2.1 *Air pollutants simulations in different cities in the NCP: "Considering that there are many monitoring sites in the NCP, scatter plots of observed and simulated $PM_{2.5}$, O_3, NO_2, SO_2 and CO concentrations for all sites in Beijing, Tianjin, Hebei, Henan, Shandong, Shanxi, Jiangsu, and Anhui from 05 December 2015 to 04 Janurary 2016 have also been provided in Figures S4 to S8, respectively. Except Anhui, the correlation coefficients between observed and simulated $PM_{2.5}$ concentrations are generally larger than 0.70 (Figure S4). The model also performs well in simulating the O_3 concentration in the NCP, with correlation coefficients generally larger than 0.80 (Figure S5). The NO_2 concentration in the NCP is also simulated reasonably, with correlation coefficients generally ranging from 0.70 to 0.80 (Figure S6). Considering that the SO_2 is mainly emitted from point sources, which is more sensitive to meteorological conditions, the model has difficulties in simulating the SO_2 concentration, with correlation coefficients generally less*

than 0.60 (Figure S7). In addition to Tianjin and Shanxi, the CO concentration is also reasonably reproduced, with correlation coefficient larger than 0.70 (Figure S8).".

3 Comment: Source apportionment uncertainty: Are you able to quantify the uncertainty in your source apportionment calculations? For instance, in Tables 2-5 and Figs. 9 and 10, can you add some information that helps understand the error in your estimates? For instance, add ranges in the tables and error bars on the bar plot figures.

Response: We have added uncertainty in the tables and error bars on the bar plot figures in Tables 2-5 and Figs. 9 and 10.

Minor/Specific comments:

1 Comment: L97-100: At the end of section 1, please provide a brief description of what you will present in the following sections.

Response: We have added at the end of Section 1: "*The model and methodology are described in Section 2. The results and discussions are presented in Section 3, and summary and conclusions are given in Section 4.*".

2 Comment: Figure 1: I do not see any "blue circles", maybe you mean to say "circles".

Response: We have revised the figure caption of Figure 1"T*he circles represent centers of cities with ambient monitoring sites, and the size of blue circles denotes the number of ambient monitoring sites of cities.*" as "*The circles represent centers of cities with ambient monitoring sites, and the size of circles denotes the number of ambient monitoring sites of cities.*".

3 Comment: Figure 1: What is the total number of sites considered in the analysis? This would be important to know also for Fig. 5.

Response: We have added in the Section 2.3: "*The model performance in simulating $PM_{2.5}$, O_3, NO_2, SO_2, and CO is validated using the hourly observations released by Ministry of Ecology and Environment of China (China MEP), with 389 observation sites in the NCP.*".

4 Comment: L199-200: Do you allow for model spin-up? Table 1 says that the model start time is 05 December 2015, but you show results starting on this day.

Response: Yes, we have allowed for model spin-up. The spin-up time is 4 days and 4 hours, and the simulation period starts from 05 December 2015. We have updated Table 1.

5 Comment: Figures 6 and 8: What about the diurnal variability in the spatial distributions?

Response: We have added the diurnal variability in the spatial distributions and clarified in Section 3.1: "*The diurnal variability in the spatial distribution of simulated and observed air pollutants is shown in Figures S9 to S12. The spatial patterns of air pollutants at different time are generally similar to those of the episode average. The $PM_{2.5}$ pollution in the NCP is more severe during nighttime and early morning, especially at 08:00 and 20:00 BJT due to the rush hour.*" and Section 3.2: "*The diurnal variations in the spatial distribution of average $PM_{2.5}$ contributions from the six provinces during the study episode are also shown in Figures S14 to S19. There is no significant difference among the spatial distribution of $PM_{2.5}$ contributions from the six provinces at different time, but the higher $PM_{2.5}$ contribution of emissions from the source region generally occurs at 08:00 and 20:00 BJT. ".*

6 Comment: L225: Do you have evidence of cloud coverage during this event?

Response: We have clarified in Section S2.2 Cloud properties: "*Clouds are one of the most important factors affecting the solar radiation reaching the ground. The daily cloud fraction (CF) used in this study was retrieved from Terra- and Aqua- Moderate Resolution Imaging Spectroradiometer (MODIS) level 2 products. Figure S13 presents the scatter plot of the daily retrieved and simulated CF averaged in the NCP from 05 December 2015 to 31 December 2015. Generally, the simulated daily average CF correlates well with that retrieved, with a correlation coefficient of 0.69. The simulated average CF over the NCP during the episode is*

52.8%, lower than the MODIS retrieved 78.4%. Numerical models still have difficulties in representing accurately clouds in terms of microphysical processes, cloud morphologies, occurrence and dissipation. In addition, many uncertainties also significantly impact CF retrievals, such as the satellite's view zenith angle, cloud microphysics assumptions, namely cloud phase, particle size and shape, et al. (An and Wang, 2015; Platnick et al., 2017; Zeng et al., 2012; Li et al., 2014). Therefore, it is still difficult to validate cloud simulations using the satellite cloud products. ".

7 Comment: L232-233: Why do you choose to focus on the NCNST site?

Response: The hourly submicron sulfate, nitrate, ammonium, and organic aerosols are measured by the Aerodyne Aerosol Chemical Speciation Monitor (ACSM) at NCNST, which are used to validate the model performance.

Grammatical/wording recommendations: Please see the attached PDF.

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2020-597/acp-2020-597-RC2- supplement.pdf.

Response: Thanks. We have revised the manuscript according to the attached comment