

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.

The authors used the WRF-CHEM model to investigate the contributions of the non-local emissions to the summertime air pollution episode in Beijing. By turning on/off the local and/or non-local emissions and using the factor separation method, the authors found that during the episode (July 5 to 14, 2015), non-Beijing emissions contributed dominantly over local emissions to the PM_{2.5} and ozone concentrations in Beijing. Although previous studies have discussed the topic of local vs. non-local contributions in Beijing, this paper provided updated information on the topic by focusing on the summer of 2015, after Beijing has taken drastic emission control measures in recent years. My major criticism, however, is that the paper lacks sufficient quantitative analysis and scientific discussion as a research paper. Therefore, I recommend a major revision before publication.

Major comments

Note that I make a bunch of suggestions in the major comments. I do not think it is necessary for the authors to follow these suggestions completely but I do think the major questions or concerns should be addressed in the revision.

1 Comment: The author gave a lengthy description of model evolution of PM_{2.5} and ozone in the BTH region during the episode (Line 257-301, Fig. 5-9). However, these descriptions are mostly qualitative and are not scientifically insightful. The multi-panel figures (Fig. 5-9) are too complex to help a reader understand what the authors say in the text. I would suggest the author to rewrite the section and keep it succinct. It may also be a good idea to move some of the content to a supplement.

Response: We have moved Figure 6, Figure 7, and the corresponding analysis to Supplementary Information Section SI-1.

2 Comment: In addition, Fig. 5-8 shows that $PM_{2.5}$ and O_3 in BTH are being transported by wind. However, this is not equivalent to the contribution of non-local emissions. First, $PM_{2.5}$ and O_3 in Beijing can also be contributed by local productions from precursors emitted outside Beijing. Second, it is also possible that Beijing emissions can contribute to the production of $PM_{2.5}$ and O_3 outside Beijing, and then these $PM_{2.5}$ and O_3 are transported back to the city. Since the authors did not provide quantitative analysis to rule out these possibilities, Fig. 5-8 cannot support the author's conclusion very well. Instead of describing the 12-panel figures, I would suggest the author to do more quantitative analysis (for example, diagnosis of the flux of $PM_{2.5}$ and O_3 across the city boundary) and discuss whether precursor transport is important.

Response: We have added a figure (Figure 9) and a table (SI-Table 1) to provide the quantitative analysis of the flux of $PM_{2.5}$, O_3 , and NO_2 across the city boundary to highlight the importance of trans-boundary transport of pollutants. We have added a section 3.2.1 to clarify the horizontal transport:

“3.2.1 Analysis of Horizontal Transport of O_3 and $PM_{2.5}$

The analysis in Section 3.1.3 has shown the strong correlation between the airflow and the high level of pollutants in Beijing during the study episode. It is essential to confirm whether the continuous air pollutions in Beijing are directly related to the airflow transport from outside of Beijing (An et al., 2007; Yang et al., 2010). In the present study, the horizontal transport flux intensity is defined as the horizontal wind speed on the grid border multiplied by the pollutants concentration of the corresponding grid from which the airflows comes (Jiang et al., 2008). Considering that trans-boundary transport mainly occurs within the PBL, the study also focuses on the contribution of trans-boundary transport of pollutants within PBL over Beijing and its surrounding areas. Previous studies have shown that the average mixing layer height is approximately between 600 - 800 m during summertime, with the maximum during noontime higher than 1000 m (Wang et al., 2015; Tang et al., 2016). Figure 9 shows the temporal variations of net horizontal transport flux of $PM_{2.5}$, O_3 and NO_2 through Beijing boundary and the pollutants contributions from non-Beijing emissions to the air quality in Beijing city. The hourly $PM_{2.5}$, O_3 and NO_2 contributions of non-Beijing

emissions generally have the same variation trend as the horizontal transport flux, indicating that the contribution of surrounding sources plays an important role in high pollutants concentrations in Beijing during the study episode. For example, the O₃ net flux also has the similar peak in the afternoon as the O₃ contribution from the non-Beijing emissions. As discussed in Section 3.1.3, the prevailing south wind dominates BTH, so the largest flux intensity are from the south, with the average of 103.3 g s⁻¹ and 244.5 g s⁻¹ for PM_{2.5} and O₃, respectively (SI-Table 1), indicating that the pollutants are mainly from south. The average horizontal transport fluxes for PM_{2.5} and O₃ during the episode are 68.2 g s⁻¹ and 68.5 g s⁻¹, respectively, showing important contributions of non-Beijing emissions to the air quality in Beijing. ”

3 Comment: For the purpose of this paper, the accuracy of the emission inventory is very important. However, the description of the emission inventory in the manuscript is too brief. What year does the emission inventory based upon? Is it for the year 2006 as in Zhang et al. (2009) or is it updated? What are emissions from Beijing compared with those from the surrounding regions? How uncertain is the emission inventory? This information is essential for a reader to assess the significance of the paper’s conclusion.

Response: We have added a Figure (Figure 2) to show the spatial distribution of anthropogenic NO_x, VOCs, OC, and SO₂ emission rates, and a table (Table 1) to compare the emissions of Beijing with its surrounding areas. We have clarified in section 2.2: “*The anthropogenic emissions are developed by Zhang et al. (2009), which is based on the 2013 emission inventory, including contributions from agriculture, industry, power generation, residential, and transportation sources. The SO₂, NO_x, and CO emissions have been adjusted according to their observed trends from 2013 to 2015 in the present study, but the VOCs emissions are not changed considering that the VOCs emissions are still not fully considered in the current air pollutant control strategy. The major pollutants emissions used in the model simulation for Beijing, Tianjin, and the neighboring provinces (Hebei, Shanxi, and Shandong) are summarized in Table 1. Obviously, high anthropogenic emissions are distributed outside of Beijing, especially in Hebei and Shandong provinces. Figure 2 presents distributions of the emission rates of VOCs, NO_x, OC, and SO₂ in the simulation domain,*

showing that the anthropogenic emissions are generally concentrated in urban areas. It is worth noting that uncertainties of the emission inventory used in the study are still large taking into consideration the rapid changes in anthropogenic emissions that are not fully reflected in the current emission inventories, particularly since implementation of the APPCAP, and the complexity of pollutants precursors.”

4 Comment: In addition, as the authors pointed out, the emissions have been greatly reduced in Beijing in recent years. Therefore, if emission inventory for multiple years were available, it would be very interesting to conduct additional simulations and calculations and see how the implementation of APPCAP affects the contributions of trans-boundary transport.

Response: We have conducted sensitivity simulations during the period from 5 to 14 July 2015 using the 2013 emission inventory. The detailed discussion has been included in the Supplementary Information Section SI-2:

“Section SI-2: Contributions of Trans-boundary Transport to PM_{2.5} and O₃ Concentrations in Beijing from 5 to 14 July 2015 Based on 2013 EI

In order to investigate the effect of the APPCAP on the contribution of trans-boundary transport, sensitivity simulations for the air pollution event from 5 to 14 July 2015 have been performed using the 2013 emission inventory. SI-Table 3 shows the average PM_{2.5} contribution in Beijing from only-Beijing emissions, non-Beijing emissions, emission interactions, and background. During the study episodes, the average PM_{2.5} contribution in Beijing from only-Beijing emissions from 5 to 14 July 2015 is 13.2%, which is lower than that using 2015 emission inventory. The non-Beijing emissions and the emission interactions contribute 65.2% and 6.5% to the PM_{2.5} level in Beijing, respectively. Therefore, the PM_{2.5} contribution caused by the trans-boundary transport is about 71.7% of PM_{2.5} concentration in Beijing, which is higher than that with 2015 emission inventory. The background PM_{2.5} contribution to Beijing is 15.0%. SI-Table 4 gives the average O₃ contributions from 12:00 to 18:00 BJT in Beijing from only-Beijing emissions, non-Beijing emissions, emission interactions, and background. The only-Beijing emissions contribute about 23.6% on average to the afternoon O₃ level in Beijing, varying from 16.4% to 29.7%, slightly higher than that with 2015 emission inventory. The outside emissions contribute more than local sources, with

an average of 38.0%, and the emissions interactions also decrease the O₃ level by 5.5% on average. The background also plays an important role in the O₃ level in the afternoon when using the 2013 emission inventory, with an average contribution of 44.0%. The O₃ contribution caused by the trans-boundary transport of outside emissions is approximately 32.5% of the O₃ concentration, higher than that with 2015 emission inventory. Hence, in general, the O₃ and PM_{2.5} contribution of outside emissions has gradually decreased since implementation of the APPCAP.”

5 Comment: The authors used the FSA method to separate the impact of local and non-local emissions. I have several questions about the results that the authors present.

(a) In Table 2, the background contribution f_0 varies from 32.6 ppbv to 62.9 ppbv. Why does background vary so much? In addition, f_0 and f'_S (surrounding) anti-correlates very well ($R^2=0.89$ based on my calculation). In Table 3, f_0 and f'_S also anti-correlates really well ($R^2=0.92$). Why is that? The author should give more discussion to provide insight into these interesting results.

Response: We have clarified in Section 3.2.2: *“The background O₃ contribution varies from 32.6% to 62.9% during the episode, which is primarily determined by the prevailing wind direction. When the northerly wind is prevalent, the clean airflow from the north affects Beijing, enhancing the background O₃ contribution, such as on 5, 13, and 14 July 2015. However, when the polluted airflow from the south impacts Beijing, the background O₃ contribution is decreased.”*

In general, the airflow influencing Beijing includes the O₃ from outside emissions and background, so when the background O₃ or PM_{2.5} contribution is low, the O₃ or PM_{2.5} contribution of outside emissions is high, Therefore, f_0 and f'_S (surrounding) anti-correlates very well.

(b) In Table 2 and Figure 10(a). The authors show that local contribution to O₃ is much less than non-local contributions. Beijing, with so much traffic, should have large amount of NO_x emissions. Given NO_x lifetime in the summer should be on the order of several hours,

regional transport of NO_x should not be very significant. So the question follows, why is regional contribution so larger? Is the input of regional O_3 or input of precursors? If it's the input of O_3 , why do regions surrounding Beijing have such high O_3 production? Do they emit a lot of NO_x and VOC_s . The authors should discuss the matter by referring to emission inventory (See major comment 2) and flux diagnosis (See major comment 1).

Response: We have added a Table in the supplement (SI-Table 2), and also explained the role of the O_3 precursors by referring to the emission inventory and flux diagnosis according to the referee's suggestion. We have added a paragraph in section 3.2.2 to provide a detailed description of the role of O_3 precursors, and the reason why the regional transport is dominant: *“Previous studies have proposed that the regional transport of O_3 precursors can play an important role in inducing the high O_3 level in Beijing (Wang et al., 2009; Zhang et al., 2014). SI-Table 2 provides the average NO_2 contributions in Beijing from only-Beijing emissions, non-Beijing emissions, emission interactions, and background. Different from O_3 , the local emissions dominate the level of NO_2 in Beijing area, with an average contribution of 70.3% during the study episode. The average contribution of non-Beijing emissions, emission interactions and background are 24.8%, 0.9% and 4.0%, respectively. The contribution of background to O_3 concentrations is obvious, which is much more different from that for NO_2 . In addition, the trans-boundary transport flux of NO_2 is much lower than that of O_3 (Figure 9). Given NO_x lifetime in the summer is short, regional transport of NO_x is not important. Furthermore, the emissions of NO_x and VOC_s around Beijing are much more than those in Beijing, especially in Hebei and Shandong provinces, which is subject to contribute more O_3 production (Table 1). Compared to the direct input of regional O_3 , the transport of O_3 precursors probably does not play an important role in the high O_3 level in Beijing.”*

(c) In Table 3 and Figure 10(b), the authors show somewhat counter-intuitive results that non-local emissions almost always contribute more than 50% of Beijing and f'_S follows the $\text{PM}_{2.5}$ concentration perfectly. In my opinion, the authors should show additional results using emission inventory, flux diagnosis, etc. to convince a reader that their calculation is right and consistent. Or, it may be a good idea for the authors to show simulation and FSA

results outside an episode in the same summer to give readers a sense of how the “control” case looks like.

Response: We have conducted another simulation from 22 to 28 May 2015 to clarify the significant contribution of trans-boundary transport to the air quality in Beijing. The detailed discussion can be seen in the Supplementary Information Section SI-3:

“Section SI-3: Contribution of Trans-boundary Transport to $PM_{2.5}$ and O_3 Concentrations in Beijing from 22 to 28 July 2015 based on 2015 EI

The model simulations and FSA results from 5 to 14 July 2015 show that the non-Beijing emissions play an important role in the air quality in Beijing. In order to further confirm the important role of trans-boundary transport, a severe air pollution episode from 22 to 28 May 2015 in NCP is simulated using the WRF-CHEM model. SI-Table 5 shows the average $PM_{2.5}$ contribution in Beijing from only-Beijing emissions, non-Beijing emissions, emission interactions, and background. The average contribution from only-Beijing emissions is 11.5%, while the average contribution from non-Beijing emissions is 62.4%, varying from 35.4% to 73.4%, which is much higher than the contribution from local source. The emission interactions also increase the $PM_{2.5}$ level in Beijing, with the average contribution of 6.3%, varying from 2.4% to 9.4%. The background contributes 17.6% to the $PM_{2.5}$ concentration in Beijing on average. Therefore, the $PM_{2.5}$ contribution induced by trans-boundary transport is about 68.7% of $PM_{2.5}$ level in Beijing during the episode from 22 to 28 May 2015, indicating the substantial impact of trans-boundary transport on the $PM_{2.5}$ concentration in Beijing. SI-Table 6 presents the average O_3 contributions from 12:00 to 18:00 BJT in Beijing from only-Beijing emissions, non-Beijing emissions, emission interactions, and background. The local sources contribute about 17.6% on average in the afternoon to the O_3 level in Beijing. The non-Beijing emissions contribute more than local emissions, with the average contribution of 42.4%, ranging from 16.9% to 51.0%. The emission interaction between only-Beijing emissions and non-Beijing emissions increase or decrease the O_3 level in Beijing in different time due to the nonlinear process of O_3 production, with contributions ranging from -2.5% to 2.1%, but the emission interactions in Beijing decrease the O_3 concentration by 0.4% on average. The background plays also an

important role in the afternoon O₃ concentration, with average contribution of 40.4%. The contribution from trans-boundary transport of the outside emissions is about 42.0% during the period from 22 to 28 May 2015, further indicating that the important role of trans-boundary transport in the O₃ level in Beijing.”

(d) More fundamentally, I am concerned that the FSA results in the paper are somewhat misleading to readers because in the four simulations to derive FSA, the author turn on/off the local or non-local emissions completely. As a result of the nonlinear chemistry, these simulations cannot give accurate information about the local sensitivity of air quality to emission reduction. Theoretically, it is possible that reducing local emissions may still be more effective than reducing non-local emissions. I suggest the authors to make clear in the text about the limitation of their method.

Response: We have clarified in Conclusion: *“The FSA method can calculate the individual and synergistic contribution of only-Beijing and non-Beijing emissions by including or excluding the local or non-local emissions. However, considering the nonlinear chemistry of PM_{2.5} and O₃, especially regarding O₃ formation, the method might not well provide how the air quality is accurately when taking different emission reduction measures. Therefore, in the future study, sensitivity simulations of different emission reduction measures are needed to design reasonable emission control strategies.”*

(e) The paper also lacks sufficient discussion of the results in the context of previous studies. The authors mentioned several previous studies on local vs. non-local emissions. The result of this paper stands out as reporting most significant non-local contributions. The paper will be much better if the authors can discuss their paper in context of these studies (in terms of method, results, discrepancies, or agreement, etc.).

Response: We have included discussions of the results in context of previous studies in Section 3.2.2 as follows:

“Using the ozone source apportionment technique, Wang et al., (2009) have emphasized that local emissions are the most important contributor to high O₃ levels from June to July in

2000 in Beijing urban area because the emissions rates there are significantly higher than the average level in the surrounding areas. Based on CMAQ simulations of Beijing, Streets et al., (2007) have estimated that 35%–60% of the high O_3 concentration at the Olympic Stadium site could be attributed to non-Beijing emissions, with Hebei Province contributing 20%–30% of Beijing's ozone concentrations during the prevailing south wind. Wang et al., (2008) have found that the average contribution from non-Beijing emissions to the O_3 levels in Beijing is 30.0% and the maximum of daily contributions is as high as 56.5% in August 2006.”

“Previous studies have also demonstrated the dominant role of non-Beijing emission in the $PM_{2.5}$ level in Beijing. Based on CMAQ model, Streets et al., (2007) have reported that average contribution of regional transport to $PM_{2.5}$ at the Olympic Stadium can be 34%, up to 50%–70% under prevailing south winds. Guo et al. (2010) have provided a rough estimation that the regional transport can contribute 69% of the PM_{10} and 87% of the $PM_{1.8}$ in Beijing local area using the short and low time resolution data in the summer. Combining the $PM_{2.5}$ observations and MM5-CMAQ model results, regional transport is estimated to contribute 54.6% of the $PM_{2.5}$ concentration during the polluted period, with an annual average $PM_{2.5}$ contribution of 42.4% (Lang et al., 2013). Using the long-term measurements of $PM_{2.5}$ mass concentrations from 2005 to 2010 at urban Beijing, and trajectory cluster and receptor models, the average contribution of long-distance transport to Beijing's $PM_{2.5}$ level can be approximately 75.2% in the summer (Wang et al., 2015).”

Minor comments

(1) Abstract. The words “pure local emissions” and “pure emissions outside Beijing” are confusing. Are there impure emissions? I suggest using “local emissions”, “non-local (or non-Beijing) emissions”, and “interactions between local and non-local emissions”.

Response: We have changed “pure local emissions” and “pure emissions outside of Beijing” to “only-Beijing emissions” and “non-Beijing emissions”, as the referee suggested.

(2) Line 121. Is NCEP reanalysis only used for boundary and initial conditions? Is WRF configured to nudge the meteorology fields towards reanalysis?

Response: We have clarified in Section 2.2: *“The NCEP 1° × 1° reanalysis data are used to obtain the meteorological initial and boundary conditions, and the meteorological simulations are not nudged in the study.”*

(3) Section 3.1. This section is only a summary of changes in air quality of Beijing during recent years, which are background information rather than research results. Therefore this section should not be within “results and discussion”.

Response: The Section 3.1 has been moved into the Section 2.5 as the referee suggested.

(4) Line 217-219, 243-238, 290. The author attributes the model errors to inability of WRF-CHEM to resolve convection in several occasions. However, these statements are not backed by any data (e.g., observations of convective clouds etc.). Given there are so many plausible error sources in a 3-D chemical transport model, I would suggest not to make guesses on what leads to the model errors (emission errors, meteorology errors, etc.), unless supported by observation data or model sensitivity test.

Response: We have removed the sentences as the referee suggested. *“The underestimation of PM_{2.5} concentrations on July 8 and overestimation on July 11 and 14 are still rather large, perhaps caused by the simulated wind field uncertainties that influence the pollutants transports from outside of Beijing or lack of resolving convective clouds due to the 6 km horizontal resolution. ”* in the line 217-219, *“There are two possible reasons for the biases in sulfate simulations. Firstly, the model is not able to resolve well convective clouds due to the 6 km horizontal resolution used in simulations, reducing the sulfate production from cloud processes. Secondly, a large amount of SO₂ is released from point sources, such as power plants or agglomerated industrial zones, and the transport of SO₂ from point sources is much sensitive to wind field simulations. ”* in the line 243-248, and the sentence *“possibly caused by the active convections in the afternoon, which cannot be well resolved in WRF-CHEM model. ”* in the line 290.

(5) Line 250. The title number should be 3.2.3.

Response: We have changed the title number.