December 27, 2016

Dear Editor,

We have received the comments from the reviewer #1 of the manuscript. Below are our responses and the revisions that we have made in the manuscript.

Thank you for your efforts on this manuscript. We look forward to hearing from you.

Best Regards,

Guohui Li

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.

The manuscript has been improved from the last version. However, I still think it has not yet met the standard of an ACP paper.

Major comments

1. Comment: My major concern is the writing style of the paper. The authors tend to provide too much information and do not organize it well, which makes a reader difficult to follow.
An example is Line 506-534, where the authors try to explain a lot of things in a very long paragraph without a clear reasoning flow. In addition, Section 3.2.2 is 7 pages long and the topic of the section changes from O_3 to $PM_{2.5}$ and then to aerosol composition without indications of topic changes.

Response: We have divided the paragraph from Line 506-534 into two parts, including the explanations for the contributions of emission interactions to organic and inorganic aerosols, respectively. We have added Section 3.2.3 "*Trans-boundary transport contributions to $PM_{2.5}$ in Beijing*" and Section 3.2.4 *"Trans-boundary transport contributions to aerosol species in Beijing"* in the manuscript, and revised Section 3.2.2 as "*Trans-boundary transport contributions to O_3 in Beijing*".

Comment: Line 408-417 is yet another example. The authors described several previous studies one by one with some unnecessary details. In my opinion, this information should be presented in a more concise way and should be put in the context of the current study.

Response: We have added the sentence in Section 3.2.2 as follows: "*The O_3 contributions in Beijing induced by the trans-boundary transport of non-Beijing emissions is about 31.5% of the O_3 concentration during the study episodes, which is in agreement with previous studies (Streets et al., 2007; Wang et al., 2008), indicating that the trans-boundary transport constitutes the*

main reason for the elevated O_3 level in Beijing after implementation of the APPCAP."

Comment: In addition, some wording may not be right, for example, "pure contribution" (L207), "pure impact" (L211), "outside emissions" (L24, 26, 375, 390, 509, 560), and "only-Beijing emissions" (throughout the text).

Response: We have revised the wording in the manuscript as suggested.

2. Comment: I am also concerned about how a reader would interpret the policy implication of the study because the authors did not thoroughly discuss the matter in the text. I think the authors need some extra work on the discussion, especially on how their work adds to the debate on whether local or non-local emissions play the major contribution to the air pollution in Beijing (Guo et al., 2014; Li et al., 2015; Zhang et al., 2015).

Response: We have added a further discussion in Conclusion to explain the role of non-Beijing emissions as follows: *"However, it is still controversial on whether local or non-local emissions play a dominant role in the air quality in Beijing (Guo et al., 2010, 2014; Li et al., 2015; Zhang et al., 2015). When only considering the local emissions, Beijing only experiences O_3 pollution, and the $PM_{2.5}$ level is low during summertime, which is comparable to the air quality in Mexico City. Mexico City has once been one of the most polluted cities in the world, but the air quality has been greatly improved in recent years after taking emission control strategies (Molina et al., 2002, 2007, 2010). Beijing and Mexico City now have similar emission sources, including transportation and residential living, but Beijing is surrounded by the highly industrialized areas in the south and east. When considering the trans-boundary transport of the pollutants from non-Beijing emissions, the O_3 and $PM_{2.5}$ levels in Beijing are remarkably increased, much higher than those in Mexico City, showing the important role of trans-boundary transport in the air quality in Beijing. Hence, the cooperation with neighboring provinces to decrease pollutant emissions is the optimum approach to mitigate the air pollution in Beijing."*

The paper may lead a reader to conclude that the major culprit for air pollution in Beijing is neighboring regions. However, this interpretation may not be completely right.

(1) Comment: As shown in Figure 2, the emission rate from neighboring regions is not significantly higher than Beijing. Although the total emissions (Table 1) from the neighboring regions are large, the areas of those regions are also very large. From Figure 2, the area average emission rate (numbers in Table 1 divided by areas) of Beijing is probably still highest.

Response: We have clarified in Section 2.2: "*As shown in Figure 2, the total emissions from neighboring regions are much more than those in Beijing, and the emission rates in Tianjin, the south of Hebei and Shandong are also higher than those in Beijing, particularly with regard to SO_2 emissions. Therefore, when the south or east wind is prevailing in NCP, the severe air pollution can be formed in Beijing when precursor emissions in highly industrialized areas chemically react as they are carried toward Beijing, blocked by mountains and further accumulated and interacted with those in Beijing.*"

(2) Comment: Consistent with (1), Figure 9 and SI-Table 1 shows that the flux is almost symmetric around 0, suggesting Beijing is likely to contribute equal amount of pollution to the neighboring regions.

Response: We have clarified in the Section 3.2.1: *"As discussed in Section 3.1.3, the prevailing south wind dominates in 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 O_3, respectively (SI-Table 1), indicating that the pollutants are mainly from the south. It should be noted that the flux of O_3 is mainly focused on the afternoon from 12:00 to 18:00 BJT. The average net horizontal transport fluxes for $PM_{2.5}$ and O_3 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: BTH is a polluted air basin (Zhao et al., 2009; Parrish et al., 2015). Applying FSA to any city in BTH may generate similar results as in this paper.
The paper may also lead a reader to conclude that the most effective way to control air pollution in Beijing is to reduce non-Beijing emissions in BTH. The FSA method is based on simulations completely turn on/off emissions from a certain region. Because a) the method cannot provide information on the local sensitivities of air pollution to emission reduction and b) emission

reduction to zero in a vast region is apparently an infeasible scenario, inference of control strategies from the results is improper. In Line 568-575, the authors briefly mentioned this but this limitation is not explicitly stated.

Response: We have clarified the limitation of FSA method in Conclusion as follows: "*BTH has been considered as a polluted air basin (Zhao et al., 2009; Parrish et al., 2015). However, although Beijing has implemented aggressive emission control strategies, it still experiences* O_3 *and* $PM_{2.5}$ *pollutions during summertime, showing that the effective way to improve air quality in Beijing is to reduce non-Beijing emissions in BTH. The FSA method is based on simulations in which emissions from a certain region are completely turned on/off, which can calculate the individual and synergistic contribution of local Beijing and non-Beijing emissions by including or excluding the local or non-local emissions in this study. However, considering the nonlinear chemistry of* $PM_{2.5}$ *and* O_3*, especially regarding* O_3 *formation, the method might not well provide how the air quality is accurately when taking different emission reduction measures, and also emission reduction to zero in a vast region is apparently an infeasible scenario. This study mainly aims at providing a quantification of the effect of trans-boundary transport on the air quality in Beijing. Therefore, in the future study, sensitivity simulations of different emission reduction measures are needed to design reasonable emission control strategies."*

Minor comments

1 Comment: Line 128 "2.2 Model Configuration"=> 2.2 Pollution episode simulation.

Response: We have revised the section title as "*2.2 Pollution Episode Simulation"*.

2 Comment: Line 219: "2.4 Statistical Methods for Comparisons". => Statistical metrics for observation-model comparisons

Response: We have revised the section title as *"2.4 Statistical Metrics for Observation-Model Comparisons"*.

3 Comment: Line 230: "2.5 Pollutants Measurements" =>2.5 Pollutant Measurements

Response: We have revised the section title as *"2.5 Pollutant Measurements"*.

4 Comment: Line 379: Apparently, …

Response: We have changed the " Therefore" to "Apparently" in Section 3.2.2.

5 Comment: Line 419: inducing the high O_3 concentrations level in Beijing

Response: We have revised the sentence as *"play an important role in inducing the high O_3 concentrations level in Beijing"* in Section 3.2.2.

6 Comment: Line 424-431: "The contribution of background to O_3 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 O_3 (Figure 9). Given NOx lifetime in the summer is short, regional transport of NOx is not important. Furthermore, the emissions of NOx and VOCs 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."

Too much redundant or irrelevant sentences. I would change to "Compared to the direct input of regional O3, the regional transport of NOx is unlikely a significant contributor to high O_3 concentrations in Beijing, partly due to its short lifetime in the summer."

Response: We have changed the sentences as suggested in Section 3.2.2: "*Compared to the direct input of regional O_3, the regional transport of NO_x is unlikely a significant contributor to high O_3 concentrations in Beijing, partly due to its short lifetime in the summer."*

7 Comment: Line 464: e.g., i.e.,

Response: We have revised it in the manuscript.

8 Comment: Line 483: "high atmospheric oxidation capability caused by elevated O_3 concentrations during summertime". High atmospheric oxidation capacity is not directly caused by high O_3 concentrations.

Response: We have revised the sentence as "*due to the increased atmospheric oxidation capability caused by elevated O_3 concentrations during summertime.*"

9 Comment: Line 489-491: Replicate of line 482-483.

Response: We have revised the sentence in Section 3.2.4 as follows: *"Secondary aerosol species dominate the $PM_{2.5}$ mass concentration in Beijing, with a contribution of 77.9%."*

10 Comment: Line 500-501: "…which is caused by the aerosol radiative effect. It is clear that the PBL-pollution interaction plays an important role in the pollutant accumulation in Beijing…" This effect may not be called the aerosol radiative effect. I think PBL-pollution interaction is more proper.

Response: We have revised the "*the aerosol radiative effect*" as "*PBL-pollution interaction*" in Section 3.2.3 and Conclusion.

11 Comment: Section SI-3: My suggestion of a "control" case in last review was to present some results in the clean period, which the authors seemed to misunderstand. I was thinking a comparison between a clean and a polluted episode may bring some insight into the regional contribution. Section SI-3 presents a polluted episode similar to the one in the main text. The results are essentially the same. I would suggest removing this section and relevant sentences in the main text.

Response: We have removed this section in the main text. According to previous studies and the analysis of meteorological conditions, when the north wind is prevailing in BTH, the air quality

in Beijing is good due to the much more clean air transported from the north of China. In addition, we have performed analysis of the weather in Beijing during the summertime of 2015. There are 46 rainy days in Beijing, but the O_3 exceedance days with O_3 concentration more than 200 μg m^{-3} are 53 days. So it is difficult to find a clean episode in Beijing without precipitation.

References

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