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 authors employed the WRF-CHEM model to assess the contributions of trans- boundary transport to air quality in Beijing during a persistent air pollution episode from 5 to 14 July 2015 in Beijing-Tianjin-Hebei (BTH), China. They showed that the WRF- CHEM model reproduced well the temporal variations of the aerosol species compared to the measurements in Beijing. The authors indicated a large contribution from the regional transport and suggested that a coordinated mitigation for pollutant emissions with neighboring provinces is key to improve air quality in Beijing. This is a good work to investigate the air pollution problem in China and, in particular, provides quantitative insight into the contributions of trans-boundary transport of outside emissions the $PM_{2.5}$ and O_3 levels in Beijing. The paper was in general reasonably well written, although it could be benefitted from additional editing. I recommend publication of this work, after a revision to address the following issues.

1 Comment: Their last statement in the abstract sounded to be out-placed and non-substantiated. It has been commonly well established that the accuracy of simulations by chemical transport models (such as WRF-CHEM) is largely dependent of several factors, including emission inventory, chemistry, and meteorological fields (including the PBL height) (e.g., Zhang et al., Formation of urban fine particulate matter, Chem. Rev. 115, 3803, 2015). What was really missing from this manuscript is a careful account of those various factors that impact their simulations and conclusions.

Response: We have removed the sentence from the abstract and included following discussions in the conclusion: *"The discrepancies between the predictions and observations are possibly caused by the uncertainties in the emission inventory, chemistry, and the meteorological fields simulations (Zhang et al., 2015).*".

2 Comment: For example, the different VOC types exhibit distinct kinetic behaviors, and their contributions to O_3 and SOA formation are also distinct (e.g., Suh et al., Oxidation mechanism of aromatic peroxy and bicyclic radicals from OH-toluene reactions, J. Am. Chem. Soc. 125, 12655,

2003; Fan et al., Atmospheric oxidation mechanism of isoprene, Environ. Chem. 1, 140, 2004). How well was the VOC EI represented in the model, and how did the VOC EI uncertainty impact their simulations?

Response: We have not provided quantitative evaluations of VOCs simulations in the manuscript due to lack of VOC measurements. We have included the following discussions in Section 2.2: *"For example, different VOCs types exhibit distinct kinetic behaviors, and as an important fraction of total VOCs in the urban atmosphere, aromatics are responsible for the photochemical O_3 production and secondary organic aerosol formation (Suh et al., 2003; Fan et al., 2004). In the SAPRC99, aromatics are lumped into ARO1 and ARO2. ARO1 mainly includes toluene, benzene, ethylbenzene, and other aromatics with the reaction rate with OH (kOH) less than 2×10^4 ppm^{-1} min^{-1}. ARO2 includes xylene, trimethylbenzene, and other aromatics with kOH greater than 2×10^4 ppm^{-1} min^{-1}. Additionally, biogenic VOCs also play a considerable role in the O_3 production (Li et al., 2007), and monoterpenes and isoprene are the main biogenic VOCs in the SAPRC99 chemical mechanism."*

3 Comment: The PM problem in Beijing has been well characterized by efficient and rapid secondary formation (Guo et al., Elucidating severe urban haze formation in China, Proc. Natl. Acad. Sci. USA 111, 17373, 2014; Zhang et al., Insufficient evidence for the contribution of regional transport to severe haze formation in Beijing, Proc. Natl. Acad. Sci. USA 112, E2741, doi:10.1073/pnas.1503855112, 2015). How well did their model handle those secondary PM formation processes, including nucleation and growth from the various organic and inorganic species (Fan et al., Contribution of secondary condensable organics to new particle formation: A case study in Houston, Texas, Geophys. Res. Lett. 33, L15802, doi:10.1029/2006GL026295, 2006)

Response: We have included a paragraph about the secondary PM formation process in Section 2.1： *"The aerosol component of the Community Multiscale Air Quality (CMAQ) model is designed to be an efficient and economical depiction of aerosol dynamics in the atmosphere (Binkowski and Roselle, 2003). The particle size distribution in the study is represented as the superposition of three lognormal subdistributions, called modes, which includes the processes of coagulation, particle growth by the addition of mass, and new particle formation. Following the*

work of Kulmala et al. (1998), the new particle production rate presented here is calculated as a parameterized function of temperature, relative humidity, and the vapor-phase H_2SO_4 concentration due to binary nucleation of H_2SO_4 and H_2O vapor, and the new particles are assumed to be 2.0 nm diameter. A number of recent studies have shown that organic compounds can play an important role in the nucleation process (Zhang et al., 2009, 2012, 2015). The contribution from organic acids likely explains the high level of aerosols, especially in polluted urban area, where large amount of organic acids can be emitted directly and produced by photochemical oxidation of hydrocarbons (Fan et al., 2006), which needs to be considered in the further study."

4 Comment: How well did their model handle the particle-phase reactions, including those associated with small di-carbonyls (glyoxal and methyl glyoxal) that could be particularly important for urban PM formation because of their traffic origin (Zhao et al., Heterogeneous reactions of methylglyoxal in acidic media: Implications for secondary organic aerosol formation, Environ. Sci. Technol. 40, 7682, 2006; Gomez et al., Heterogeneous chemistry of glyoxal on acidic solutions – An oligomerization pathway for secondary organic aerosol formation, J. Phys. Chem. 118, 4457, 2015).

Response: In the SOA module, we have included the contribution of glyxal and methylglyoxal to the SOA formation, and we have clarified in Section 2.1: *"Recent studies have shown that small di-carbonyls (glyoxal and methylglyoxal) are important for the aerosol formation due to their traffic origin (Zhao et al., 2006; Gomez et al., 2015). Li et al. (2011a) have indicated that glyoxal and methylglyoxal can contribute about 10% of the SOA in the urban area of Mexico City. The SOA formation from glyoxal and methylglyoxal in this study is parameterized as a first-order irreversible uptake by aerosol particles and cloud droplets, with a reactive uptake coefficient of 3.7×10^{-3} for glyoxal and methylglyoxal (Zhao et al., 2006; Volkamer et al., 2007; Gomez et al., 2015)."*

5 Comment: Another problematic area was related to the MET part, including PBL. In particular, it has become clear that the PBL-pollution interaction plays a key role in pollutant accumulation in Beijing (Wang et al., Light absorbing aerosols and their atmospheric impacts, Atmos. Environ.

81, 713, 2013; Peng et al., Markedly enhanced absorption and direct radiative forcing of black carbon under polluted urban environments, Proc. Natl. Acad. Sci. USA 113, 4266, 2016).

Response: We have considered the role of PBL-interaction in pollutant accumulation in the manuscript. We have clarified in Section 3.2.2: *"It is clear that the PBL-pollution interaction plays an important role in the pollutant accumulation in Beijing (Wang et al., 2013;Peng et al., 2016). Mixing of Beijing local emissions with those outside of Beijing increases the aerosol concentrations in the PBL and decreases the incoming solar radiation down to the surface, cooling the temperature of the low level atmosphere to suppress the development of PBL and hinder the aerosol dispersion in the vertical direction."*

Stylistic/grammatical/typographic errors

(1) We have removed "the" in the title.

(2) We have replaced "emissions outside of Beijing" by "outside emissions" or "non-Beijing" emissions in the manuscript.

(3) The word "pure" in the manuscript has been removed.

(4) We have changed "local emissions with those outside of Beijing" to "local and outside emissions".

(5) We have changed "The pure emissions outside of Beijing" to "The outside emissions", and "pure Beijing local emissions" to "the local emissions".

(7) We have changed "The emissions interactions" to "The interactions between local and outside emissions".

(8) We have changed "need to be performed to improve" to "are needed to improve".