Response to Referee #2

We greatly appreciate the helpful comments from the reviewer, which have helped us improve the paper. We have addressed the comments carefully, as detailed below.

The review paper jumps to the hot topics on PM2.5 pollution and aerosol optical property in China. It is well-written and very helpful for understanding the current situations and challenges ahead for alleviating severe PM2.5 pollution in China. This reviewer has a few minor comments for authors considering before publishing in ACP

1. The authors are encouraged to use either OC or OM through the manuscript.

Response: After a careful consideration, we feel both OC and OM are needed in the discussions in various places. For example, OC is measured directly and needed to be discussed in the measurement data as well as in comparing related emission inventories. OM is converted from OC using different conversion factors in different regions and is needed in assessing $PM_{2.5}$ mass distributions among different chemical components.

2. As a rural site downwind of BTH, the study by Feng et al. (JRG, 117, D03302, doi:10.1029/2011JD016400, 2012) is worthy of inclusion for comparison.

Response: We have included this reference and the following discussion in the revised paper: "At an Asian continental outflow site (Penglai in Shandong province), annual average contribution of secondary inorganic aerosols to $PM_{2.5}$ reached to 54% (Feng et al., 2012b), evidently higher than those in urban and inland rural sites in China, while that of carbonaceous aerosols was 31%, close to those in BTH. This finding suggested that intensive emissions of SO_2 and NO_x in China enhanced the downward transport of secondary inorganic aerosols to Pacific Ocean."

3. Lines 191-194, temperature effect should be included. A few very recent studies suggest that extreme weather could also be important factors for heavy $PM_{2.5}$ pollution in winter. The authors may have no time to read, but these studies are really worthy of inclusion for a complete review.

Response: We have added the following discussion in the revised paper**: "**Moreover, extreme weather events such as weakening monsoon circulation, depression of strong cold air activities, strong temperature inversion, and descending air motions in the planetary boundary layer also played important roles in wintertime heavy $PM_{2.5}$ pollution (Niu et al., 2010; Wang et al., 2014c; Zhao et al., 2013). Several extreme wintertime air pollution events in recent years covered vast areas of northern China and were all correlated to some extent with extreme weather conditions (Zou et al., 2017)."

4. Lines 288-295, the reviewer suggest to include these contentious studies for sulfate formation in atmospheric particles published in 2016 and add a few arguments as well. It is helpful for students and young scientists.

Response: We have added the following discussion in the revised paper**:** **"**It is worth to note that several recent studies have highlighted the important role NO_2 might play in sulfate formation in the polluted environment in China (Cheng et al., 2016; Wang et al., 2016c; Xie et al., 2015a). Nevertheless, the aqueous SO_2 + H_2O_2/O_3 oxidation should still be the dominant mechanism in most cases, especially at a background site (Lin et al., 2017). The aqueous SO_2 + oxygen (catalyzed by Fe(III)) reaction can also be important under heavy haze condition in north China (Li et al., 2017b). Extensive measurements of stable oxygen are needed to confirm the relative contributions of different sulfate formation mechanisms."

5. Lines 616-624, relative humidity is also important factor to determine spatial variation of AOD.

Response: We have included meteorological factors (which cover RH) in AOD discussion in the revised paper, which reads: "Satellite retrievals of AOD have been widely applied to estimate surface $PM_{2.5}$ concentrations using statistical models (Liu et al., 2005; Hu et al., 2013; Ma et al., 2014; Wang and Christopher, 2003). Although the correlation between AOD and $PM_{2.5}$ mass concentration depends on many factors, such as aerosol size distribution, refractive index, single-scattering albedo, and meteorological factors (Che et al., 2009; Guo et al., 2009b; Guo et al., 2017), the predicted $PM_{2.5}$ mass from satellite AOD data compared well with ground-level measurements (Ma et al., 2014; Xie et al., 2015b). Moreover, the spatial distributions of AOD measured using sun photometers mostly agreed with those retrieved from satellite data (Che et al., 2014; Che et al., 2015; Liu et al., 2016b; Pan et al., 2010)."

6. In Section "4.3 Aerosol gyroscopic properties", the authors are encouraged to include aerosol particle size information if possible.

Response: We agree that particle size distribution is an important factor affecting *f*(RH) curves. However, size distributions of the dominant chemical components (NH_4^+, SO_4^{2-}, NO_3^-, OC, and EC) were only available in autumn of 2007 in urban Beijing. Thus, it is difficult to investigate the impact of different size distributions on the differences in *f*(RH) curves in different years (autumns of 2007, 2011 and 2014) in urban Beijing. For most of the other cities, only one study was available for *f*(RH) curves and particle size distribution data were also very limited. Thus, we chose not to go to the details on size-distribution related impacts. We, however, simply pointed out the potential influence of size distribution on *f*(RH) curves, which reads: "however, *f*(80%<RH<90%) values in rural Guangzhou were evidently higher than those in urban Guangzhou, likely due to the much higher fraction of secondary inorganic aerosols in fine mode particles in rural Guangzhou than urban Guangzhou in the dry season (Lin et al., 2014; Liu et al., 2008b)."

Additional references added in the revised paper adding the comments from this reviewer are listed below:

Cheng, Y., Zheng, G., Wei, C., Mu, Q., Zheng, B., Wang, Z., Gao, M., Zhang, Q., He, K., Carmichael, G., Pöschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, Science Advances, 2, 10.1126/sciadv.1601530, 2016.

Feng, J. L., Guo, Z. G., Zhang, T. R., Yao, X., Chan, C. K., and Fang, M.: Source and formation of secondary particulate matter in PM2.5 in Asian continental outflow, Journal of Geophysical Research, 117, 2012b.

Li, G., Bei, N., Cao, J., Huang, R., Wu, J., Feng, T., Wang, Y., Liu, S., Zhang, Q., Tie, X., and Molina, L. T.: A possible pathway for rapid growth of sulfate during haze days in China, Atmos. Chem. Phys., 17, 3301-3316, 10.5194/acp-17-3301-2017, 2017b.

Lin, M., Biglari, S., Zhang, Z., Crocker, D., Tao, J., Su, B., Liu, L., and Thiemens, M. H.: Vertically uniform formation pathways of tropospheric sulfate aerosols in East China detected from triple stable oxygen and radiogenic sulfur isotopes, Geophysical Research Letters, 44, doi:10.1002/2017GL073637, 2017.

Niu, F., Li, Z., Li, C., Lee, K., and Wang, M.: Increase of wintertime fog in China: Potential impacts of weakening of the Eastern Asian monsoon circulation and increasing aerosol loading, Journal of Geophysical Research, 115, 2010.

Wang, G., Zhang, R., Gomez, M. E., Yang, L., Zamora, M. L., Hu, M., Lin, Y., Peng, J., Guo, S., and Meng, J.: Persistent sulfate formation from London Fog to Chinese haze, Proceedings of the National Academy of Sciences, 113, 13630-13635, 2016c.

Wang, Y., Yao, L., Wang, L., Liu, Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.: Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China, Science China Earth Sciences, 57, 14-25, 2014c.

Xie, Y., Ding, A., Nie, W., Mao, H., Qi, X., Huang, X., Xu, Z., Kerminen, V. M., Petaja, T., and Chi, X.: Enhanced sulfate formation by nitrogen dioxide: Implications from in situ observations at the SORPES station, Journal of Geophysical Research, 120, 12679-12694, 2015a.

Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W., Dong, F., He, D., and Shi, Q. F.: Analysis of a winter regional haze event and its formation mechanism in the North China Plain, Atmospheric Chemistry and Physics, 13, 5685-5696, 2013.

Zou, Y., Wang, Y., Zhang, Y., and Koo, J.H.: Arctic sea ice, Eurasia snow, and extreme winter haze in China, Science Advances, 3, 10.1126/sciadv.1602751, 2017.