Response to Anonymous Referee #3:

In this manuscript, the authors report on measurements of various chemical species in size-segregated particle samples that were collected over one-year period in the urban area of Beijing. Their results showed that the abundance and chemical compositions of particles varied temporally and spatially, with the major contributions from organics and secondary inorganics. They also showed that the mass fraction of organics decreased but the total mass fraction of sulfate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+) increased from non-haze to haze days. The authors further speculated that heterogeneous reactions and meteorological factors, such as relative humidity and wind speed, might be responsible for haze pollution in Beijing. They concluded that future air pollution control strategies should consider wind patterns and the detailed size-resolved information is needed to characterize the sources of particulate matter (PM) in urban regions. This work represents few studies to investigate the size-resolved source apportionment of PM in Beijing, and their results may have important implications on regulatory control for PM in that country. The article was reasonably written, although the paper would certainly be benefited from additional editing for English usage. I recommend that paper can be published in ACP, provided the following issues have been adequately addressed.

Reply: Thank you for the positive comments above and insightful suggestions below, which have been included in the revised version to strengthen the quality of our manuscript. In addition, the revised manuscript was edited by several native English speakers at AJE to improve the language.

Their results showed that OM dominated in fine particles and decreased from 37.9% on non-haze days to 31.2% on haze days. Such an observation may reflect two distinct processes during haze formation in Beijing. For example, new particle formation has been found to be prevalent in Beijing during clean period (Guo et al., 2014). There is large evidence showing that organics play a key role in new particle formation, both

to enhance aerosol nucleation and growth of freshly nucleated particles (Science 304, 1487, 2004; Chem. Rev. 112, 1957, 2012). Such an aspect should be clearly discussed.

Reply: Thank you for your advice. We have added the following discussions in Sect. 4.2.1.

OM dominated in fine particles and decreased from 37.9% on non-haze days to 31.2% on haze days. Such an observation may reflect two distinct processes during haze formation in Beijing. New particle formation has been found to be prevalent in Beijing during clean period (Guo et al., 2014), and the nucleation mode particles contained mainly secondary organics. Nucleation consistently precedes a polluted period, producing a high number concentration of nano-sized particles under clean conditions and the growth process competes with capture/removal of nano-particles by coagulation with preexisting aerosols.

In addition, there is also large evidence showing that organics play a key role in new particle formation, both to enhance aerosol nucleation and growth of freshly nucleated particles. For example, the interaction between organic and sulfuric acids promotes efficient formation of organic and sulfate aerosols in the polluted atmosphere(Zhang et al., 2004; Zhang et al., 2011).

In contrast, their results showed that the contribution of SNA to the PM2.1 mass increased from 19.1% on non-haze days to 33.9% on haze days, indicating that SNA played a key role in haze formation. Although the process related to sulfate formation is still inadequately understood at the present time, but the recent literature on such a topic should be clearly discussed. Also, the issues regarding heterogeneous reactions and the effects of RH on aerosol growth need to be further addressed. They may refer the relevant topics discussed in a recent review article (Chem. Rev. 115, 3803, 2015).

Reply: Thank you for your advice. The issues regarding heterogeneous reactions and the effects of RH on aerosol growth have been further addressed in Sect. 4.2.1. SO_4^{2-}, NO_3^- and NH_4^+ primarily originated from secondary pollution particles that were produced by the transformation of their SO_2, NO_2 and NH_3

precursors (Liu et al., 2016). The pronounced increase of SNA from clear to haze days was primarily due to the faster heterogeneous formation of secondary aerosols under high RH and abundant precursors (Tan et al., 2009; Zhang et al., 2013). For haze pollution that is associated with high RH, the aqueous phase on the aerosol surface provides a means for the rapid heterogeneous gas-liquid conversion of gaseous precursors to produce secondary inorganic aerosols (Wang et al., 2012; Zhang et al., 2015). However, SOA primarily originated from the transformation of VOCs during the optical oxidation process. A high concentration of particulate matter on haze days weakens the incident solar radiation; therefore, the atmospheric oxidization capability of organic matter is considerably weakened.

Furthermore, the size-resolved variations in particle composition, hygroscopicity, and density (Figs. 3 and 4 in that paper) during haze formation in Beijing have already reported in Guo et al. (2014). Comparisons between that work and the present study should be clearly made.

Reply: Thanks for the suggestion. We have added the following discussions in the revised paper:

In our study, peak shifts from 0.43-0.65 μm on non-haze days to 0.65-1.1 μm on haze days were observed when considering the annual average size distributions of SO_4^{2-}, OC, NO_3^-, NH_4^+, Cl^-, K^+ and Cd. Meanwhile, in a previous haze study in Beijing which was mainly focus on more smaller size fractions, a continuous growth from the nucleation mode particles is also clearly depicted by the evolution in the mean particle size, which increases from about 40 nm when the $PM_{2.5}$ level is less than 50 $\mu g \cdot m^{-3}$ to about 190 nm when the $PM_{2.5}$ concentration exceeds 300 $\mu g \cdot m^{-3}$ over the course of 3 d (Guo et al., 2014).

In our study, the relative contributions of OM and CM to the particle mass decreased from non-haze to haze days, and the relative contribution of SNA increased from non-haze days to haze days. Similar trends had been observed in Guo et al (2014), in which the organic mass fraction dominates in the clean period

(74–77%) and decreases slightly during the transition (48–49%) and polluted (35–42%) periods. The contributions of sulfate and nitrate to the particle mass concentration increase throughout the pollution period, with mass fractions of 8–9% and 6–12% for the clean period to 23–26% and 12–27% for the polluted period, respectively.

Suggestions for English improvements

The abstract was too long and should be written in a more concise manner. Perhaps most of the numbers provided could be omitted.

Reply: As suggested, we have revised the abstract in a more concise manner and reduce the words from 504 to 472.

The usage of tenses needs to be consistent. One such an example is given here. "Although OM dominates in fine particles, it decreased from 37.9% on non-haze days to 31.2% on haze days." (lines 818-819)

Reply: As suggested, this sentence was changed to "Although OM dominated in fine particles, it decreased from 37.9% on non-haze days to 31.2% on haze days". In addition, we have also checked the manuscript and made all the usage of tenses be consistent.

There are quite a few acronyms for chemicals throughout the manuscript. Please ensure that those were defined only once at their first usage. For example, OC was defined at least twice (lines 90 and 220).

Reply: Thank you for your advice. We defined OC in Line 90 and OM in Line 220 in the manuscript, respectively. Also, we have also checked the manuscript and made all other terms be defined only once at the first time.

Reference

Liu, Z., Hu, B., Zhang, J., Yu, Y., and Wang, Y.: Characteristics of aerosol size

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Zhang, R., Suh, I., Zhao, J., Zhang, D., Fortner, E. C., Tie, X., Molina, L. T., and Molina, M. J.: Atmospheric new particle formation enhanced by organic acids, Science, 304, 1487-1490, 2004.
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