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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal variations in the high time-resolved aerosol composition, sources, and chemical process of background submicron particles in North China Plain" *by* Jiayun Li et al.

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Response to referees' comments on "Seasonal variations in the highly time-resolved aerosol composition, sources, and chemical process of background submicron particles in North China Plain"

We highly appreciate the detailed valuable comments of the two referees on our manuscript. The suggestions are quite helpful for us to improve the quality of our paper. Please see the detailed point-by-point response below. We list the comments

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Discussion paper

in black, our replies in blue, and the changes in revised MS in red.

Anonymous Referee #1

This study was conducted at a regional background site of NCP for four seasons using a high-resolution aerosol mass spectrometer. Highly time-resolved chemistry and sources of submicron aerosols were investigated. The authors found that nitrate was the most abundant inorganic species and submicron particles were almost neutralized by excess ammonium in all seasons except summer. Source apportionment of organic aerosol (OA) identiïňĄed two oxidized OAs, more-oxidized oxygenated OA (MO-OOA) and less-oxidized oxygenated OA (LO-OOA) in all seasons. SigniïňĄcant contributions of aged secondary organic aerosol in OA were observed in all seasons, especially in summer. The oxidation degree and evolution process of OAs in the four seasons and the comparison with urban studies were further investigated. Overall, the long-term dataset provided in this study is a valuable addition to the literature. I recommend publication after the following issues are addressed.

[Response] Thank you very much for your helpful comments and we have taken all of them into account in the revised version of the manuscript. Please see the detailed response marked blue below and the changes marked red in the revised manuscript.

SpeciïňĄc comments:

1. In the introduction, the importance of background site should be emphasized, which is important for highlighting the signiïňĄcance of this paper. [Response] Thanks for your suggestion. It is suggested that investigate the air quality and the chemical compositions of particles at the background sites could reflect the characteristics of regional air pollution. We have emphasized the importance of background site and highlighted the significance of our study. The revised part of the introduction section are as follows: “The high-resolution time-of-flight aerosol mass spectrometry (HR-ToF-AMS) has been widely used to characterize nonrefractory submicron particles (NR-PM1) at numerous urban sites and a few background sites on the Qinghai–Tibet Plateau (QTP) in west-

ern China, the Lake Hongze site in northern China, the Mount Wuzhi site in southern China, and the Mount Tai and Xinglong station in the NCP (Zhang et al., 2019;Xu et al., 2018;Du et al., 2015;Zhu et al., 2016;Zhang et al., 2020;Li et al., 2019). These previous studies on air quality and aerosol chemical composition in the background area of North China indicated that the aerosol species were well-mixed and highly aged from regional transport. Meanwhile, the background atmosphere had strong atmospheric oxidizing capacity and the organics were highly aged (Wang et al., 2013;Li et al., 2019). However, the evolution and formation mechanisms of SOA in the background area in the NCP are still unclear. Currently, researches focused on the evolution and formation mechanism of SOA is mainly concentrated in urban areas and the results varied in different places and seasons. For example, photochemical processing dominated the oxidized degree of OA in haze events, whereas aqueous-phase processing was the main reason that affected the oxidized degree of OA in foggy events in Hong Kong (Li et al., 2013;Qin et al., 2016). In urban Beijing, Xu et al. (2017a) found that aqueous-phase processing dominated MO-OOA formation in all seasons. While in Li's et al (2020) study, the impact of photochemistry on MO-OOA formation enhanced as the photochemical age increased in early autumn in Beijing. Due to the stronger atmospheric oxidizing capacity and higher oxidized degree of organics in the background atmosphere than in urban atmosphere in the NCP, the evolution and formation mechanisms of SOA must be different from those of urban areas and is of great significant to investigate, but the research in this field in background areas is limited. Also, there are only very limited studies investigating the seasonal difference so far. The formation and evolution of SOA vary greatly in different areas and seasons, mainly due to the complex interaction of local emissions, chemical reactions, and meteorological influences. Therefore, we presented four season measurements and discussed the seasonal difference in aerosol sources and formation processes. In particular, based on robust data analyses, we evaluated the influence of photochemical and aqueous-phase processing on different SOA productions (LO-OOA and MO-OOA) in different seasons in the background atmosphere."

2. Two OOA factors resolved in this study are less-oxidized OOA (LO-OOA) and more-oxidized OOA (MO-OOA), rather than semi-volatile OOA (SV-OOA) and low-volatile OOA (LV-OOA). Please provide more information about the deïňĄnition for LO-OOA and MO-OOA? [Response] Thanks for your comments. OOA is usually identified in two categories, including low-volatility (LV-OOA) and semi-volatile OOA (SV-OOA), based on their correlations with sulfate and nitrate, respectively, and inferred volatilities. Previous studies confirmed the relative volatility characteristics of SV-OOA and LV-OOA by thermodenuder measurements (Huffman et al., 2009;Cappa and Jimenez, 2010). However, the terminology LO-OOA and MO-OOA for less and more oxidized OOA, respectively, is also appropriate, especially for datasets for which volatility dates are not available (Zhang et al., 2011). In this study, the high f44 values were permanent in the MS of both LO-OOA and MO-OOA. The f44 values for MO-OOA and LO-OOA in the four seasons ranged from 16.3 to 23.5%, 8.1 to 13.8%, respectively. The f43 values for MO-OOA and LO-OOA ranged from 4.8 to 5.2%, 6.8 to 9.1%, respectively. These behaviors indicated that MO-OOA had a higher oxidation degree than LO-OOA. The O/C ratios of the MO-OOA factors in the four seasons were 0.93, 0.93, 0.84, and 0.83, respectively, higher than those in the corresponding LO-OOA (0.69, 0.58, 0.67, and 0.49). More detailed information of PMF analysis can be seen in the revised supplementary materials.

3. The names of submicron aerosol species in the ïňĄgures should be consistent, e.g., “ClâËŸ AËĞR” vs Chl, “NO3” vs. “NO3-”. [Response] Thanks for your suggestion and the names of submicron aerosol species have been changed to be consistent in the revised manuscript.

4. Line 77-80: please provide more discussion and reference(s) about the higher atmospheric oxidizing capacity in the background site. [Response] Thanks for your suggestion and the related references have been added in the revised manuscript.

5. Line 153: Please add the standard deviation of the average PM1 concentrations of all seasons here. [Response] Thanks for your suggestion and the standard deviation of

the average PM1 concentrations were added in the revised manuscript in both Section 3.1.1 and conclusion (Section 4)

6. Line 154: The full name is needed when abbreviation is shown at the ïňĄrst place, such as “SIA” here. [Response] Thanks for your reminding. We have checked carefully and defined the abbreviation in the revised manuscript.

7. Line 337-339: “These characteristics were similar to the results found in previous researches conducted in urban Beijing showing that.....” please add references. [Response] Thanks for your reminding. The related references have been added.

8. Line 328: “in the each season” should be “in each season”. Line 309: “high oxidized ability” should be “high atmospheric oxidizing capacity”. Please go through the manuscript for similar typos. [Response] We are so sorry for making the grammatical mistakes to make trouble to your review work. We have carefully checked and corrected the errors sentence by sentence.

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