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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.

Anonymous Referee #2

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This paper reports the seasonal variations of submicron particles and its chemical composition at a background station using HR-ToF-AMS. Using ME-2 analysis, the authors identified different sources of organic aerosol and explored the oxidation degree and evolution process of different OA in four seasons. Backward trajectory analysis was conducted to see the influence of air mass transport. The seasonal dataset of HR-ToF-AMS measurements at a background station in north China is valuable and suitable for a measurement report. But to publish as a scientific article in ACP, this reviewer did not find good novelty or significance of this study compared to previous findings.

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Discussion paper

Also, many conclusions drawn in this study are not well supported by the observations or interpretations. Overall, this reviewer do not think the scientific significance of the paper meet the scope of ACP scientific articles and cannot recommend the publication of the paper.

Major concerns:

1. With similar data analysis and similar results from this study compared to so many published AMS papers, it is difficult to see the novelty and significance of this paper. Regarding the background atmosphere in NCP, there have also been many studies focusing on air quality and particle chemical composition, including AMS studies. The authors should make it clear what is the specific values of this study. It should not be because you did measurements in a different location, or your measurement period is longer. Instead, the authors should state clearly the scientific questions or valuable findings from these measurements that can improve current understanding of aerosol chemistry.

2. The reviewer cannot be convinced by the PMF analysis and result evaluation in this study, and details are lacking. (1) Regarding the first PMF run with PET, the authors showed in Fig. S2 that OOA was over split in a 5-factor solution without only showing the similar mass profiles. But how about the time series, diurnal variations and O/C ratios of different OOA factors? The authors should not conclude this by only checking the mass profiles. Are they representative of other OOA rather than LO-OOA and MO-OOA? For example, as the authors have been emphasizing the significance of aqueous-phase processing, is there any single OOA factor related to aqueous phase chemistry? Please provide these details either in the manuscript or SI. (2) To perform ME-2 analysis, the authors constrain the FFOA profiles with the POA factor resolved in the five-factor solution of PMF analysis in spring and apply it to all seasons. One concern here is that how do the authors believe the POA factor from the five-factor solution is good enough to represent the primary sources? How does the profile look like when performing PMF analysis to six or seven factor? Does the POA factor in this

study comparable to those resolved from AMS studies previously, especially those in NCP area? Another big concern is that how robust it is to apply this factor from spring to all the other seasons. To check this, the author should perform PET PMF analysis for the other season and see if a POA factor will appear when go to more factors, and then check if the mass profile of POA are comparable in all seasons. As shown in Fig. 5, the mass contribution of FFOA in total OA is 5%, which is in the uncertainty range of PMF analysis. The authors should prove if it is reasonable to manually constrain the ME-2 to separate a single FFOA factor in summer. (3) Details about the evaluation of PMF and ME-2 results are lacking. To interpret PMF/ME-2 results, the authors should carefully follow the procedures proposed by Ulbrich et al. (2009) and Zhang et al. (2011). For example, evaluation of Q/Qexp as a function of factor number and Q/Qexp or scaled residual as a function of m/z should be provided in the manuscript or SI.

3. It is questionable that the authors solely based on the ratio of measured NH4+ to predicted NH4+ to evaluate aerosol acidity. While this ratio has been used as an indicator of aerosol acidity by some previous AMS studies, it has been proved recently that this ion balance method is not reliable to evaluate particle acidity (Guo et al., 2015; Song et al., 2018). Thermodynamic models, i.e., E-AIM and ISORROPIA, should be used. With thermodynamic models, Guo et al. (2017) found that aerosol is always acidic in NPC region, which is contradictory to results from this study.

4. Many interpretations or conclusions from this study cannot be well supported by the observations. For example, the authors conclude in the end of the abstract that the neutralized state of submicron particles highlight the significance of NOx and ammonia reduction (also in conclusion part, Line 486 and Line 510). But these two do not have causal relationship. Line 215, the authors should not make a conclusion that the high NOx concentration is solely due to strong influence of traffic. Line 280, the authors conclude that POA factor was closely related to traffic emission without evaluating the characteristic of POA. While POA showed a good correlation with NOx, does its diurnal profile show morning and evening peaks, which is a typical feature of traffic-related

pollutant? It is not correct to conclude that the high concentration of FFOA at night indicate the high primary emissions. The variations in boundary layer height play a significant role. Similar issues happened again in Sect. 3.3. The authors draw conclusions regarding species formation mechanisms during daytime and nighttime based on the variations in their concentrations without considering the major influence of boundary layer height. Line 323, why the authors think the increase in nitrate concentration is caused by regional transport? The observations cannot support this. In the end of this paragrath, the authors emphasize the strong effects of local chemical production and regional transport on nitrate diurnal pattern. But the reviewer did not find any related discussion about the relative contribution of local production and regional transport. Again, Line 336, why the increased concentration from noon to evening indicate the regional characteristics of MO-OOA? Overall, these conclusions without detailed interpretation and well supported observations would confuse the audience a lot. Line 426, from the observations, the authors can only conclude that aqueous-phase and photochemical processing both play roles. How do they evaluate which is more important? Line 432, how do the authors conclude that photochemical processing enhanced during regional transport without any observation or interpretation regarding this (Also in the conclusion part, Line 499)? Line 434, the authors said that the impact of photochemical processing on MO-OOA production was limited in summer. Then what is the major formation mechanisms of MO-OOA in summer? Do they provide any evidence regarding different formation mechanism of LO-OOA and MO-OOA from this study? Line 505, the longer transport distance of air masses in summer does not mean that the influence of regional transport is strongest in summer. Not proper quantification.

Specific comments:

(1) In the introduction, Line 42, it's not proper to summarize that sulfate dominated in the south of NPC and nitrate dominate in the north of NCP. It is not determined by the location, but more by the emission characteristics.

(2) Line 55, the authors said that previous studies at background NCP site are lim-

ited by the low resolution. Any new findings do they draw from their high-resolution measurements?

(3) Line 105, to quantify aerosol concentrations, what are the RIE values of sulfate and ammonium according to the standard calibration?

(4) Since the measurement station is 960 meters above sea level, why do the authors choose the height of 500 m to permafrost back trajectories? How does it compare to 1000m or 1500m?

(5) Line 114, the AMS does not measure chlorine but chloride. Please revise the whole manuscript accordingly.

(6) Line 162, are high RH and high PM1 concentration correspond to air masses from the south?

(7) Line 170, it should be clearly noted that the frequency distribution of PM1 is shown in Fig. 1 using the white curve.

(8) Line 221, how do the authors define the wind dilution ratio? Please make it clear.

(9) In Fig. 3, how do the authors average WD? Do they follow the vector average wind direction?

(10) Line 280, please provide the number of correlation coefficient of POA vs. NOx and POA vs. chloride.

(11) Line 300, how do their correlations look like? According to previous studies, LO-OOA correlates better with nitrate, while MO-OOA better with sulfate.

(12) In Fig. 5, the nitrate time series is missing in Fig. 5b.

(13) Line 360, the higher O/C ratio in Xinglong should be due to the weak influence of primary emissions.

(14) Line 390, please define the Ox.

(15) Figure 9, the plots as a function of Ox not RH.

(16) Please define what is “dva” in the manuscript.

(17) Many grammatical mistakes. The reviewer recommends to do editing service. For example, in the title, it should be “highly time-resolved”. Line 309, should be “be attributed to”. Line 83, should be “a HR-ToF-AMS was deployed. . . . . with collocated measurements of meteorological parameters and gaseous species. ” Line 89, should change “of” to “on”.

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