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

We highly appreciate the detailed valuable comments of the three 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 in black, our replies in blue, and the changes in revised manuscript in red.

Anonymous Referee #1

The authors have addressed my comments carefully. I therefore recommend publication in ACP after the technical corrections.

[Response] Thank you so much for your approval of our revised work. Please see the detailed response below.

1. Line 53-56: Rephrase this sentence to make it clearly.

[Response] Thanks for your suggestion. The sentence has been revised as follows:

"Early studies found that secondary aerosols dominate the aerosol particles at background sites and regional transport affects the air pollution of the background atmosphere."

2. Line 116: The time of MS and PToF modes are not clear in the text.

[Response] Thanks for your reminding. The time of MS and PToF modes has been added in the revised manuscript:

"Under V-mode operation, the AMS cycle through the mass spectrum (MS) mode and the particle time-of-flight (PToF) mode every 30s, spending 10s and 20s, respectively."

3. It is better to show the correlation coefficients in Figure 4.

[Response] Thanks for your suggestion. The correlation coefficients have been added in Figure 4.

4. Line 356-357: What does it mean “demonstrating the regional characteristics”? The authors should explain it.

[Response] Thanks for your reminding and we have rewritten the sentence as follows:

“In comparison to the diurnal patterns of nitrate, sulfate showed flatter diurnal cycles in each season, identifying the regional characteristics of sulfate formation (Sun et al., 2016).”

5. In Section 3.5: It is interesting that relatively high PM1 concentrations were also observed with air masses form north and west regions of Xinglong in summer. The authors should explain more about the result.

[Response] Thanks for your suggestion and more explanation was added in the revised manuscript:

“The air masses from the southern regions (clusters 2 and 3) were dominant in summer and accounted for 56% of all the air masses in summer, which was obviously higher than the percentage in other seasons (27–38%). Cluster 3 in summer started at Bohai Bay and passed through the Shandong Peninsula and over Bohai Bay. The PM_1 concentrations for clusters 2 (14.7 μg m^{-3}) and 3 (12.2 μg m^{-3}) were both high. These results suggest a dominant role played by southern transport in submicron aerosol concentrations over the NCP in summer. Furthermore, the transport distances of clusters from the north and west regions in summer were shorter than those in other seasons. In general, with a decrease in the transport distance of clusters from the north and west regions, particle concentrations gradually increase (Hu et al., 2017). Although the clusters from these regions in summer only accounted for 15% and 8% of all the air masses, respectively, the PM_1 concentrations for the two clusters (cluster 1: 12.8 μg m^{-3}; cluster 5: 10.2 μg m^{-3}) were both at high levels and similar to those associated with the southern air masses (cluster 2: 14.7 μg m^{-3} and cluster 3: 12.2 μg m^{-3}). All these characteristics suggest that regional transport from Inner Mongolia (west and north regions of Xinglong) also partially contributes to the particle pollution in the background area of the NCP in summer.”

Reference:

Sun, Y., Du, W., Fu, P., Wang, Q., Li, J., Ge, X., Zhang, Q., Zhu, C., Ren, L., and Xu, W.: Primary and secondary aerosols in Beijing in winter: sources, variations and processes, Atmospheric Chemistry and Physics, 16, 8309-8329, 2016.

Anonymous Referee #2

While the dataset is valuable, there are still many mistakes or improper interpretations in this study.

[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.

For example, Line 215: This reviewer does not agree that higher acidity of sulfate is due to the low volatility.

[Response] Thanks for your comments. We now admit that the higher acidity of sulfate was not directly due to the low volatility, but for leading to a much higher concentration of H_{air}^{+} than nitrate. To make it clear, we revised it in the manuscript as follows:

"Previous studies show that sulfate can lead to a much higher concentration of H_{air}^{+} than nitrate due to its low volatility (Ding et al., 2019; Tan et al., 2018; Xu et al., 2019)."

Line 245: The indication is quite confusing as the strong influence of fossil fuel combustion is due to regional transport at this background site.

[Response] Thanks for your reminding. To make it clear, we revised it as follow:

"As shown in Table 2, NOx exhibited its highest concentration in winter and correlated well with chloride (R^2 = 0.6), suggesting a strong influence of fossil fuel combustion, such as coal combustion. The enhancement of emission in winter in the NCP leaded to the increase of NOx in background atmosphere, which was mainly due to the regional transport."

Line 268: The indication here is not correct. How previous studies in urban Beijing indicate the formation mechanisms in background atmosphere?

[Response] Thanks for your reminding. We have revised the sentence as follows:

"Previous studies in urban Beijing show a successive increase in SIA with increased RH (Li et al., 2019;Liu et al., 2016), suggesting that aqueous-phase processing affects nitrate formation in urban atmospheres."

Line 275: The authors should not draw a conclusion that the impact of regional transport weakened as the regional transport can be related to air masses both from clean and polluted areas. With the decrease in wind speed, do the authors see changes in back trajectories?

[Response] Thanks for your reminding. We agree with you that the decrease of surface wind speed does not indicate the weakening of regional transportation. When RH was above 60%, the PM_1 species decreased in spring as RH increased, which was different from other seasons. This behavior may be due to the low overall humidity in spring, the number of samples with humidity above 60% was limited, and the statistical results may be not representative enough. Therefore, the sentence was rewritten as follows:

"When RH was > 60%, the SIA concentrations decreased rapidly, which would be mainly due to insufficient samples (n<30) with humidity above 60%."

Line 277: The authors should conclude the important role of photochemistry in the formation of sulfate and OOA based on the relationship between OA/sulfate and RH.

[Response] Thanks for your suggestion. The sentence was revised as follows:

"Notably, the OA and sulfate concentrations were high even when RH was low (RH < 40) in summer, which was significantly different from what occurred in other seasons, suggesting the important role of photochemistry in the formation of sulfate and OOA."

Line 293: Did the greater level of NPF in winter occur in Xinglong? It's confusing by adding this description.

[Response] Thanks for your reminding. The sentence was deleted in the revised manuscript.

Line 296-321: This part regarding the evaluation of PMF results is partly repetitive of those in the Methods.

[Response] Thanks for your reminding. We have checked carefully and deleted the repetitive part in the revised manuscript.

Line 322-325: The characteristics of f44 and f43 are indicators to name the factors as MO-OOA or LV-OOA. It's not because of these behaviors that MO-OOA has a higher oxidation state than LO-OOA.

[Response] Thanks for your reminding. We agree with you and revised this part as follow:

"The high f44 values were permanent in the mass spectrum 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% and 8.1 to 13.8%, respectively. The f43 values for MO-OOA and LO-OOA ranged from 4.8 to 5.2% and 6.8 to 9.1%, respectively. 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)."

Line 344: The diurnal of nitrate is not only related to the PBL influence but also contributions from lower temperature and higher RH at night.

[Response] Thanks for your reminding. We agree with you and revised this part as follow:

"This behavior was closely related to the variation of the PBL, temperature and RH. The lower concentration of nitrate during the daytime can be attributed to the higher PBL and the evaporation of NH_4NO_3 due to the higher temperatures. In comparison, the higher concentration at night was closely related to lower temperature and higher RH, which favor to nitrate formation through heterogeneous reactions."

Line 347: It's not correct to conclude the importance of N2O5 hydrolysis based on low NO concentration and high O3 concentration. Do they check NO2 concentration?

[Response] Thanks for your reminding and suggestions. We agree with you and revised this part as follow:

"High concentrations of O_3 were observed during nighttime in the four seasons, which were about 45, 70, 35 and 25 ppb, for spring, summer, autumn and winter, respectively. In addition, the NO_2 concentrations ranged from 4 to 14 ppb in all seasons. These results suggested the background atmosphere exhibited high atmospheric oxidation capacity, even at night, especially in summer, which indicated the hydrolysis of dinitrogen pentoxide (N_2O_5) would contribute significantly to nitrate formation at night under high RH conditions."

Line 362: The high sulfate concentration at night can also be contributed by lower PBL.

[Response] Thanks for your reminding. We revised it following your suggestion:

"At night, however, the high sulfate concentration might be attributable to the lower PBL and enhancement of aqueous-phase processing under high temperatures and humidity"

Line 387: The high O/C ratio indicates the high aging of the aerosols.

[Response] Thanks for your reminding. We have changed the sentence in the revised manuscript:

"Mainly due to the weak influence of primary emissions, the O/C ratios in Xinglong in all seasons (0.54–0.75) were slightly higher than those in urban Beijing (0.47-0.53), suggesting the high aging of the aerosols in Xinglong."

Line 439-441: The reviewer cannot understand the meaning or the main point here.

[Response] Thanks for your reminding. We have rewritten this sentence to make it clearly:

"MO-OOA increased significantly with Ox, while it increased slightly with RH (40% < RH < 60%) firstly, and then decreased with RH when RH was above 60%. This characteristic suggested photochemical processing dominated MO-OOA formation, but the role of aqueous-phase processing under moderate RH (40% < RH < 60%) conditions cannot be ruled out in summer in the background atmosphere. In comparison, previous studies found that aqueous-phase processing dominated MO-OOA formation in urban Beijing in summer (Xu et al., 2017;Duan et al., 2020). This kind of difference in MO-OOA formation between urban and background site in the NCP would be mainly due to the higher atmospheric oxidation capability in the background atmosphere. Previous study showed that Ox concentration in the background sites was 30% higher than that in the urban site during summertime (Wang et al., 2013)."

Line 468-473: The authors said that the medium-distance cluster (1, 3, 4, and 5) were dominant in summer but then they focused on cluster 2 and 3 from the southern regions and said southern transport played a dominant role. This is contradictory.

[Response] Sorry for the misleading. Based on the distances over which the air masses were transported, the medium-distance cluster (1, 3, 4, and 5) accounted for 57% in all the clusters and the short-distance cluster (cluster 2) accounted for 43%. Therefore, we said the medium-distance cluster (1, 3, 4, and 5) were dominant in summer in the previous manuscript. However, based on the directions of the air masses, clusters from the southern regions (cluster 2 and cluster 3) accounted for 66%, so that southern transport played a dominant role. To avoid the misleading, we have rewritten this part as follow:

"The air masses from the southern regions (clusters 2 and 3) dominated in summer and accounted for 56% of all the air masses, which was obviously higher than the percentages in other seasons (27–38%). Cluster 3 in summer started at Bohai Bay and passed through the Shandong Peninsula and over Bohai Bay. The PM_1 concentrations for clusters 2 (14.7 μg m^{-3}) and 3 (12.2 μg m^{-3}) were both high. These results suggest a dominant role played by southern transport in submicron aerosol concentrations over the NCP in summer. Furthermore, the transport distances of clusters from the north and west regions in summer were shorter than those in other seasons. In general, with a decrease in the transport distance of clusters from the north and west regions, particle concentrations gradually increase (Hu et al., 2017). Although the clusters from these regions in summer only accounted for 15% and 8% of all the air masses, respectively, the PM_1 concentrations for the two clusters (cluster 1: 12.8 μg m^{-3}; cluster 5: 10.2 μg m^{-3}) were both at high levels and similar to those associated with the southern air masses (cluster 2: 14.7 μg m^{-3} and cluster 3: 12.2 μg m^{-3}). All these characteristics suggest that regional transport from Inner Mongolia (west and north regions of Xinglong) also partially contributes to the particle pollution in the background area of the NCP in summer."

Line 507: The reviewer cannot agree that sulfate dominates submicron particles in southern and western China generally.

[Response] Thanks for your reminding. We admit that this conclusion was obtained from a few previous studies and it is improper to represent for the southern and western China. Therefore, we deleted this sentence revised this part as follows:

"In addition, the high contribution of aerosol nitrate in background NCP highlights how regional reductions in nitrogen oxide emissions are critical for remedying the occurrence of haze events over the NCP."

Reference:

Ding, J., Zhao, P., Su, J., Dong, Q., Du, X., and Zhang, Y.: Aerosol pH and its driving factors in Beijing, Atmospheric Chemistry and Physics, 19, 7939-7954, 10.5194/acp-19-7939-2019, 2019.

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Anonymous Referee #3

This paper presented a four-season measurement of aerosol composition using an AMS at Xinglong site and reported the seasonal variation, diurnal variation, as well as the source appointment. The authors revised the manuscript largely according to the referee's comments, especially add the calculation of aerosol acidity by the thermodynamic model of ISORROPIA II. This is a good improvement. However, similar to the comments of Referee II, this study is to date more like a data report without too much insight to improve the current understanding. I, therefore, recommend a major revision before it can be published in ACP.

[Response] Thank you so much for your approval of our revised work.

Although there are many highly time-resolved aerosols studies in background atmosphere, they mainly conducted in the remote areas, which represent for global background atmosphere rather than regional background atmosphere, while regional background atmosphere is more critical to reflect the general picture of anthropogenic emissions in a hot polluted region, such as the NCP region. However, chemical composition and evolution and formation mechanisms of secondary organic aerosol (SOA) in the background atmosphere in the NCP are still not fully understood. As far as we known, only two studies investigated the aerosol chemical composition and OA sources in the background site in the NCP, using the high-resolution AMS so far (Li et al., 2019;Yan et al., 2020). What's more, both of the two studies were concentrated in wintertime, and didn't investigated the oxidation state of SOA and the evolution and processes of SOA.

In our study, we present the OA variations and SOA formation mechanism in a background site in each season in the NCP. Our results showed that the secondary organic aerosols in the background area in the NCP was highly oxidized in all seasons. This suggests the high atmospheric oxidation capacity in the NCP on a regional scale. What's more, we analyzed the evolution and formation mechanisms of SOA. These results showed a clearer picture on the evolution of PM_1 species and SOA at the background atmosphere, and could improve our understanding on the formation mechanism of SOA under the high atmospheric oxidizing capacity of NCP, which could provide the direct observation results to verify the models. For example, we found the dominant role of aqueous-phase processing on SOA formation in winter, while both of photochemical and aqueous-phase processing contribute to LO-OOA and MO-OOA production in spring and autumn. In summer, the photochemical processing dominant MO-OOA formation, but the role of aqueous-phase processing under moderate RH (40%<RH<60%) condition cannot be ruled out. In comparison, LO-OOA formation was mainly contributed by photochemical processing in summer.

Therefore, we believe that our study provides valuable information to the scientific community on understanding OA sources and SOA formation in the background atmosphere in the NCP.

1. I did suggest to make the analysis more focus on 1-2 scientific question, other than report the data as seasonal variation, diurnal variation et al

[Response] Thanks for your suggestion.

Actually, the compositions of PM_1, the sources and oxidation state of OA and impacts of regional transport on aerosol mass loadings and chemistry changed significantly in different background sites and seasons. However, no systematic measurements with high time resolution of the mass–size distributions of chemical components in fine aerosol particles, covering four seasons, have yet been reported in the background atmosphere in the NCP, which would hinder our understanding on the seasonal variations of evolution and chemical processes of secondary aerosol and regional transport on a regional scale. Therefore, we reported the data as seasonal variation and diurnal variation to fill in the gap of characteristics of background aerosols in the NCP.

We found obvious seasonal variations in chemical species, aerosol acidity and regional transport. Specifically, nitrate dominated PM_1 in spring, autumn and winter, while sulfate dominated PM_1 in summer. Aerosol was acidity in summer and moderate acidity in other three seasons. Backward trajectory analysis showed that higher concentrations of submicron particles were associated with air masses transported short distances from the southern regions in all four seasons, while long-range transport from Inner Mongolia (western and northern regions) also contributed to summertime particulate pollution in the background areas of the NCP.

As we found SOA in the background atmosphere was highly oxidized in the NCP, we focused on the specific question of the formation mechanisms of SOA in the background atmosphere and found that aqueous-phase processing dominated SOA formation in winter, while both of photochemical and aqueous-phase processing contribute to LO-OOA and MO-OOA production in spring and autumn. In summer, the photochemical processing dominant MO-OOA formation, but the role of aqueous-phase processing under moderate RH ($40\%<RH<60\%$) condition cannot be ruled out. In comparison, LO-OOA formation was mainly contributed by photochemical processing in summer. These results showed a clearer picture on the evolution of PM_1 species and SOA at the background atmosphere, and could improve our understanding on the formation mechanism of SOA under the high atmospheric oxidizing capacity of NCP. The related discussion had been

added in the revised manuscript.

2. In section 3.4: the authors drew their conclusions without enough supports. E.g. it's hard to conclude the role of aqueous-phase reactions solely by the observational that MO-OOA increased with RH.

[Response] Thanks for your reminding. We agree with you and have added the discussion about the change of SOA with LWC in the revised manuscript to further prove the conclusion.

"Both LO-OOA and MO-OOA increased significantly as RH increased when RH was < ~90% in autumn and winter. Meanwhile, both of LO-OOA and MO-OOA increased significantly as LWC increased when LWC was below 160 μg m^{-3} and 140 μg m^{-3} in autumn and winter, respectively (Figure S 21). These behaviors suggested aqueous-phase processing had a significant influence on OOA formation in these two seasons. MO-OOA increased more rapidly than those of LO-OOA as RH increased. As a result, the mass fractions of MO-OOA increased by 25% in autumn and by 12% in winter when RH increased from 20 to 90%. Corresponding, the mass fraction of LO-OOA decreased by 15% in autumn. These characteristics indicated that aqueous-phase processing plays more important role in MO-OOA formation than that in LO-OOA in these two seasons. Note that the mass fraction of MO-OOA did not increase as Ox elevated in winter, while it increased ~from 30% to 40% as RH increased from 30 to 90%. This characteristic suggested a more dominant important role of aqueous-phase processing on SOA formation than photochemical processing in this season."

"In spring, LO-OOA and MO-OOA only increased under moderate RH (RH < 70%) as RH increased. Notably, Ox also increased when RH was < 70% as RH increased. The LWC value in spring was far lower than those in other three seasons. LO-OOA and MO-OOA only increased when LWC was below 30 μg m^{-3}, and then decreased when LWC was above 30 μg m^{-3}. This characteristic suggested the weaker effect of aqueous-phase processing on SOA formation in spring than in autumn and winter. Ox concentration in spring was as high as other three seasons. Meanwhile, LO-OOA and MO-OOA increased rapidly at moderate Ox levels when Ox changed from 50 to 70 ppb, and then remained unchanged at high Ox levels. RH maintained at low levels (RH < 40%) as Ox increased, suggesting a more important role of photochemical processing on SOA formation than aqueous-phase processing in spring."

"In summer, both LO-OOA and MO-OOA showed overall increasing trends as Ox increased, while RH showed a corresponding overall decreasing trend. This behavior indicates a strong influence of photochemical processing on both LO-OOA and MO-OOA production. LO-OOA concentration decreased significantly as LWC increased and maintained

low concentration (< 1 μg m^{-3}) when LWC was above 40 μg m^{-3}. Meanwhile, LO-OOA showed a continuously decreasing trend as RH increased in summer, except for a slightly increasing trend when RH increased from 40 to 60%, indicating photochemical processing dominated LO-OOA formation. MO-OOA increased significantly with Ox, while it increased slightly with RH (40% < RH < 60%) firstly, and then decreased with RH when RH was above 60%. This characteristic suggested photochemical processing dominated MO-OOA formation, but the role of aqueous-phase processing under moderate RH (40% < RH < 60%) conditions cannot be ruled out in summer."

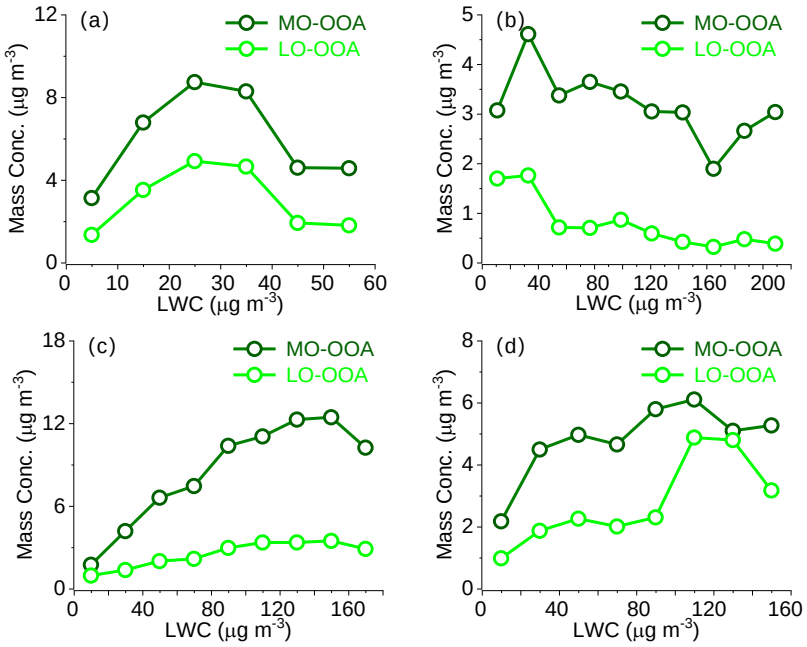

Figure S21. Variations in the mass concentrations of LO-OOA and MO-OOA as a function of LWC in (a) spring, (b) summer, (c) autumn, and (d) winter.

3. Line 194-195: Similarly, it's hard to evaluate the role of aqueous-phase reactions in nitrate formation solely by the observational fact that nitrate proportion increased with PM1.

[Response] Thanks for your comments and we agree with you. We have revised the sentence as follows:

"The proportions of nitrate in PM_1 increased slightly in spring, summer and autumn. In comparison, the proportion of nitrate increased first and then decreased with a high PM_1 concentration (PM_1 > 50 μg m^{-3}) in winter. The difference may be due to the seasonal differences of heterogeneous reactions on nitrate formation."

The role of aqueous-phase reactions in nitrate formation was detailed evaluated in Section 3.1.3.

4. The language writing needs to be polished.

[Response] Thanks for your suggestion and we have polished the language of the revised manuscript.

5. Line 146: there is a lack of evidence to support the identification/definition of FFOA.

[Response] Thanks for your reminding. We have added the evidence to support the identification/definition of FFOA in the revised manuscript.

"The mass spectrum pattern of the POA factor (Fig. S4, factor 3) mainly consisted of hydrocarbon ions ($C_nH_{2n+1}^+$ and $C_nH_{2n-1}^+$), which are commonly related to combustion emissions (Zhang et al., 2015;Sun et al., 2013). The mass profile of the POA factor had some similarity with that of HOA and coal combustion OA (CCOA) (Hu et al., 2017;Elser et al., 2016). The correlation coefficient between POA and NOx was 0.58, and that between POA and chloride was 0.78, in spring, suggesting a significant contribution of coal combustion and traffic-related sources to the POA factor in Xinglong. Moreover, HOA and CCOA show remarkably similar mass spectrum patterns when m/z is below 120 (Sun et al., 2016;Sun et al., 2018), which is sometimes difficult to be separated by PMF analysis, so FFOA can be considered as a combined factor of HOA and CCOA (Sun et al., 2018). In this study, it was difficult to separate CCOA form HOA because of the low percentage of POA in OA. Therefore, the POA factor in spring could also be considered as FFOA, which is a typical profile in Xinglong."

6. Figure 2: Figure numbers did not match with the figure capture

[Response] Thanks for your reminding. The figure numbers have matched the figure capture in the revised manuscript.

7. Correlation analysis for the PMF factors: since a regional background site, the temporal variation of most aerosol compositions can be predicted to be correlated to each other, but cannot say too much on the source appointment. For instance, in Figure 4, the LO-OOA, MO-OOA, nitrate, and sulfate were correlated well to each other.

[Response] Thanks for your comments and we agree with you that the temporal variation of most aerosol compositions can be predicted to be correlated to each other in a regional background site.

Actually, LO-OOA, MO-OOA, nitrate, and sulfate were correlated well to each other in spring, autumn and winter. However, the mass spectrum of LO-OOA and MO-OOA showed different characteristics in these three seasons. For example, in spring, the LO-OOA was characterized by a high 43/44 ratio, and the MO-OOA was defined by having a dominant peak at m/z 44 (Fig. 1). What's more, the O/C ratio of MO-OOA was obviously higher than that of LO-OOA. In addition, both of the mass spectrum and time series of LO-OOA and MO-OOA showed different characteristics in summer (Fig. 2). Therefore, the formation of LO-OOA and MO-OOA may be influenced by different chemical processes.

The similar phenomenon has been found in a background site (1570m a.s.l.) in the western Mediterranean Basin (WMB). The diurnal cycles of OA components were studied as a function of air mass origin (Fig. 3). The SV-OOA was characterized by a high 43/44 ratio, and the LV-OOA was defined by having a dominant peak at m/z 44 (Fig. 4). Both of SV-OOA and LV-OOA showed similar diurnal cycles (Fig. 3).

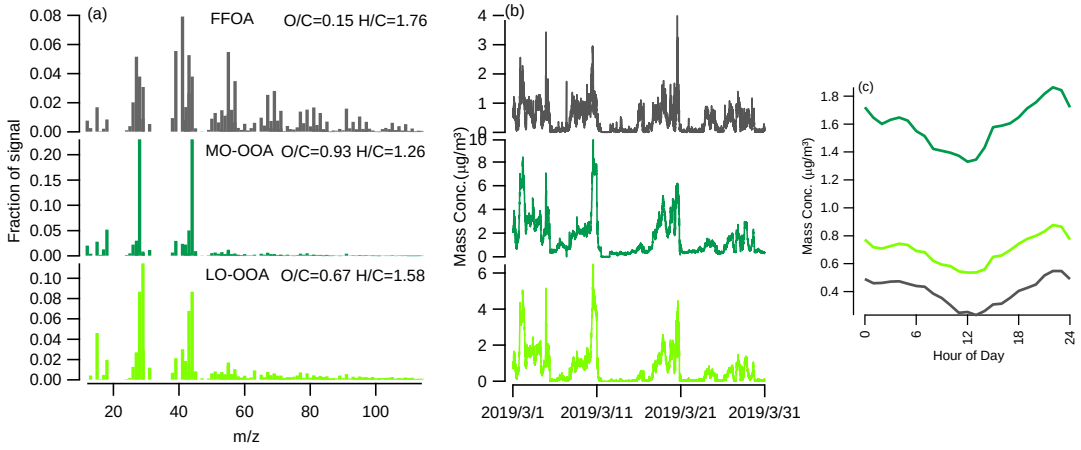

Figure 1. The mass spectra, time series, and diurnal variations of ME-2 analysis for the spring observation

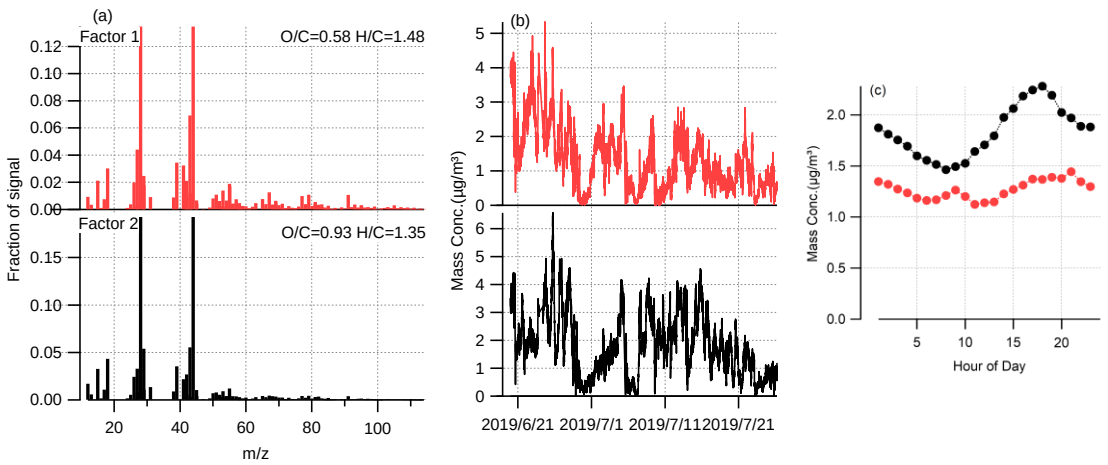

Figure 2. The mass spectra, time series, and diurnal variations of 2-factor solution of PMF analysis for the summer observation.

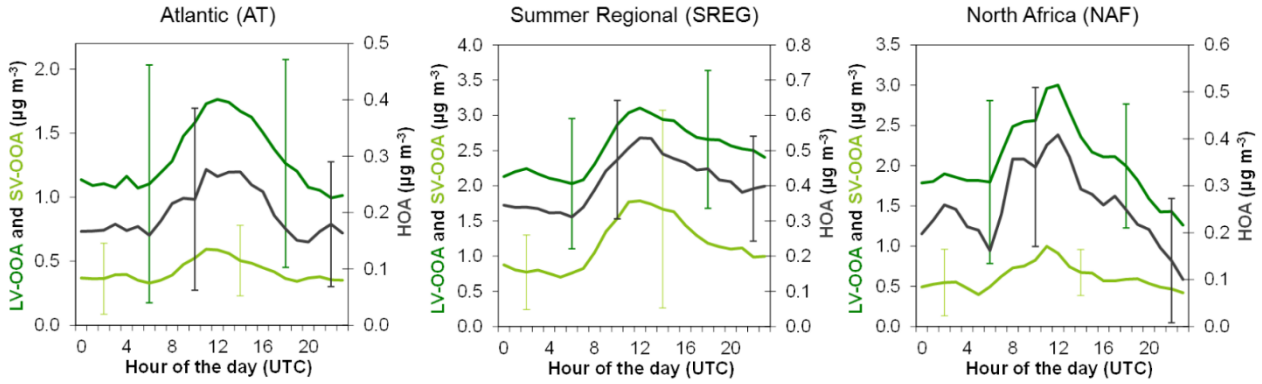

Figure 3. Diurnal cycles of organic species concentrations averaged as a function of atmospheric episode for the summer period. (Ripoll et al., 2015)

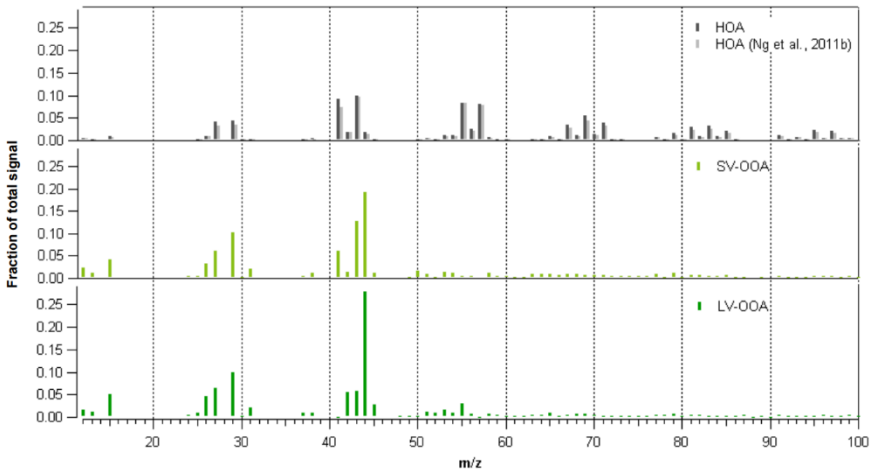

Figure 4. Organic species profiles extracted from the ME-2 analysis for the summer period (14 Jul 11-24 Sep 11). (Ripoll et al., 2015)

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