Response to the comments of referees

We would like to thank the referees for their constructive comments and suggestions in improving the scientific content of our manuscript. We have revised the manuscript accordingly. Listed below are our point-by-point responses.

Referee #2:

This paper describes chemical composition and sources of fine aerosol (PM_1) measured with an Aerosol Chemical Speciation Monitor (ACSM) during two different harvest seasons in China. Organic aerosol (OA) was the most abundant PM_1 component and it was apportioned by means of Positive Matrix Factorization (PMF) (supported by correlation with ancillary measurements) in 3 different classes: hydrocarbon-like OA (HOA), oxygenated OA (OOA) and biomass burning OA (BBOA). Much of the focus is on quantifying BBOA and discussing the importance of biomass burning as main contributor to the increase of PM_1 loadings during harvest periods because of agriculture fires. Importantly authors elaborated an equation for on-line estimating of the BBOA mass concentrations starting from the simple measure of the *m/z* 60 fragment in the ACSM mass spectrum. This is a well-written paper that clearly describes measurements and analyses, but maybe quite poor of incisive new findings. Nevertheless, it is a good manuscript suitable for publication after a few minor changes which are detailed below.

Response: We thank the referee's constructive comments and suggestions to improve the scientific content of our manuscript. We have revised the manuscript accordingly. The detailed responses to the comments and suggestions are shown below.

Section 3.2: Not enough information is presented regarding the organic source apportionment by PMF analysis. The authors don't state their reasons for not using the solution sets with 4+ factors in section 3.2 and these results are not shown graphically, which would be very useful to see. These could very easily be included as supplementary material. In particular the choice of keeping only one factor that encompasses HOA and COA should be better explained by adding (for example, as I already said) details on the solutions with more than 3 factors.

Response: We thank the referee's comments and suggestions. The ACSM organic mass spectra in this study were analyzed using the PMF model. We have reevaluated the PMF results and selected a better PMF-solution with more than 3 factors, i.e., 4-factor solution

1

including a new factor (oxidized biomass burning-influenced OA, OOA-BB) for the summer and autumn harvest, respectively. Furthermore, some details in the 4-factor solution in this study have been further discussed in section 3.2, and more accessorial information was added into the supplementary. The details in the revised manuscript can be found in lines 180 - 202, page 8 - 9.

Section 3.4: The estimation of BBOA directly from a tracer mass (m/z 60) is a very useful task to address and is well attempted, but is here a very specific result: the proposed equations relate to two specific campaigns in a specific region. It would be necessary to better comment this point. Moreover it would be very useful to try to extrapolate one general equation valid at least for all the harvest seasons in the region (not one for each measurement campaign). However this work is a first effort to achieve this task (as far as I know) that can be used as a comparison in other future studies and could be definitely appreciate.

Response: We thank referee's comments and suggestions. The BBOA concentrations have been successfully estimated for both the summer and autumn harvest via a unified equation. And we have added some information for explaining the different estimated equations for BBOA loadings in two specific campaigns. The details in the revised manuscript can be found in lines 519 - 542, page 22 - 23.

PG9111, L17: "dominate" should be replaced with "dominant".

Response: We thank referee's suggestions. This sentence has been changed. The details in the revised manuscript can be found in lines 31 - 35, page 2.

PG9111, L27: This concluding sentence seems to be a little pointless. The heavy pollution events result associated both with transport and local sources giving misleading information to the reader: specify better the sentence or remove it.

Response: This concluding sentence was revised as "Analysis of air masses back-trajectory indicates that the high BB pollutants are linked to the air masses from the western (summer harvest) and southern (autumn harvest) areas."

PG9117, L4: add a link to the organic source apportionment section (3.2) in order to facilitate the reading.

Response: The link has been added. The details in the revised manuscript can be found in lines 201 - 202, page 9.

PG9121, L9: why PMF doesn't isolate the COA factor? Specify better this point, even just showing the 4+ factors solution.

Response: We thank the referee's comments and suggestions. We have presented the 4+ factors solution in the supplementary information. As response above, the HOA factor mixed with COA and traffic HOA cannot separate a COA factor even the PMF factors go up more than 3 or 4 factors in this study. Similar to previous studies, it was also not able to distinguish the HOA and COA factors from PMF analysis of unit mass resolution organic spectra obtained by Aerodyne Quadrupole AMS(Q-AMS)/ACSM in Beijing, China (Sun et al., 2010; Sun et al., 2012). One of the reasons is due to the much similar spectra of HOA and COA. However, PMF analysis of high resolution mass spectra of OA was able to distinguish the COA from the traffic HOA and cooking-related OA (e.g. Huang et al., 2010; Crippa et al., 2013). The details in the revised manuscript can be found in lines 180 - 202, page 8 - 9.

PG9121, L23&27: check the consistency of r^2 values with figures.

Response: The r^2 values have been checked and modified.

PG9122, L28: a little pointless and misleading sentence: it seems that BBOA (a primary component) was enhanced during the nighttime for the same reasons of OOA (aqueous-phase oxidation etc.) that is quite contradictory. Specify better or remove the sentence.

Response: This sentence has been removed.

Referee #3:

In their manuscript "Insights into characteristics, sources and evolution of submicron aerosols during harvest seasons in Yangtze River Delta (YRD) region, China" Zhang and coworkers present results from two two-week measurement periods in summer and fall 2013 using an Aerodyne ACSM for non-refractory sub-micrometer aerosol components together with a MARGA to measure potassium ions in the aerosol and additional instruments to measure PM_1 and black carbon in $PM_{2.5}$. Furthermore gas analyzers to measure CO, NO_2 and O_3 were also deployed as well as a met station. The measurements were performed on top of a not further specified 6-story building in urban Nanjing (more than 8 Million inhabitants) during the main periods of wheat harvest (summer) and rice harvest (autumn).

Time series of PM_1 components as well as meteorological factors were presented and average values for the two intervals were determined. Diurnal variations of these data were presented and partially discussed. Using PMF three different types of organic aerosol, i.e. HOA, OOA, and BBOA, were extracted from the ACSM data and their time series were compared to other species time series and discussed. The contribution especially of the BBOA organics fraction to total PM_1 with a special focus on very polluted days is discussed in order to determine the biomass burning contribution to air quality. This analysis was also used to determine a simplified method to determine BBOA concentrations from the marker peak *m/z* 60 and total organics signal. Briefly, information on oxidation processes is attempted to be extracted from the data and finally with the help of HYSPLIT back trajectories the contributions from different regions around the sampling site are investigated.

Generally the content of the paper is suited for ACP and the paper is well written with few linguistic weaknesses. As detailed below there are a few passages that are hard to understand and should be reworded. For several of the Figures it is hard to see the information, e.g. because different shapes of symbols cannot be distinguished or because information is printed on top of each other. The two major weaknesses of the manuscript, however, are the missing focus of the work and many conclusions that are either not sufficiently supported by the data or drawn from pretended behavior of the data that is not in agreement with the data presented in the figures (see detailed comments below). Data from a set of aerosol and trace gas measurements operated over two relatively short time intervals are presented together with a very broad analysis of these data. For a real characterization of the aerosol or of the sources or of the transformation both, the data and the analysis are not sufficiently deep. I have the impression that everything that can be done with this data set has been done and is presented in this paper without a real focus of the work. As a result the

paper is rather long with many figures (13 figures in the main text and additional 5 figures in the supplement that are also referenced in the text; many of the figures with two or multiple panels) and at the same time only gives a rather limited depth of information. Here I agree with the first referee that not much new information is provided by this manuscript.

I also agree with the first referee that the "poor man's PMF" method, the method to derive information on different types of organics (e.g. on BBOA) without the need to perform a full PMF analysis, is a very interesting approach. However, this approach is not new. Weimer et al. submitted a comparable approach to ES&T in 2008 (Dissertation S. Weimer, ETH Zürich, 2008). In 2009 Aiken et al. (ACP) presented also such a method to estimate BBOA and other organic particle types concentrations which was also used in Ng et al. 2011 (ES&T) or Lee at al., 2010 (AS&T). While this approach allows a good approximation of BBOA concentrations without the need to perform PMF it has a major limitation: For each data set the factors used to convert the m/z 60 signal into a BBOA concentration are different. Since these factors are determined using the PMF results this method cannot be used to determine BBOA concentrations on-line as stated by the authors here. Since there are already several attempts to determine BBOA from the marker peak at m/z 60 I think a more thorough analysis and discussion of the variability of the factors used for this calculation would be appropriate in order to develop this approach into the direction of a real on-line BBOA estimate.

Taking this and the many other comments below into account I cannot suggest publication of this manuscript in ACP before major revisions that result in a more focused and deeper analysis and presentation of the interesting data. For example focusing on the identification of the contribution of the harvest-related emissions to local air quality with more in-depth apportionment to the sources together with a discussion of the limitations of this apportionment would provide valuable information on this special type of aerosol and its impact on air quality in this area. For this purpose also external information on the sources should be included and presented. Also information on the typical aerosol during the times between the harvest periods could be used to identify and distinguish the harvest contribution. At the same time the manuscript would gain if the authors avoid conclusions that are not properly supported by the data. The detailed comments below are partially intended to make this clearer.

==Response:== We thank the reviewer so much for the careful reviewing and do the best to integrate his suggestions to improve the scientific content of our manuscript. We fully agree with that. Also, we have revised the manuscript accordingly.

P9111L16-17: BBOA mass concentrations increase with increasing PM_1 for large mass loadings. However, this does not imply that BBOA plays a "dominate" (better: "dominant") role in high PM pollution. According to the data presented in this paper the BBOA contribution is in the order of only 20-25% even for these large PM concentrations. This is not a dominant contribution.

Response: We thank the referee's comment. This sentence has been re-worded. with "PM_1 components were shown to be dominated by organic fraction (OA, 39% and 41%) and nitrate (23% and 20%) during the harvest seasons (summer and autumn respectively)."

P9111L18-20: The good correlation between the K^+ ion and BBOA concentrations can make K^+ a good tracer for wheat and rice straw burning. However, before you can claim that it "is" a good tracer you need to show that there are no other relevant sources for this ion.

Response: We agree with the referee's comments. This sentence has been removed.

P9111L2527: This sentence is not specific at all and does not really contain any relevant information.

Response: This sentence has been re-worded with "Analysis of air masses back-trajectory indicates that the high BB pollutants are linked to the air masses from the western (summer harvest) and southern (autumn harvest) areas."

P9112L11-14: This sentence is hard to understand – please reword.

Response: This sentence has been re-worded. The details in the revised manuscript can be found in lines 55 - 56, page 3.

P9112L22-24: PMF is used frequently in the AMS/ACSM community. Other groups also use other types of source apportionment approaches.

Response: We agree with the referee's comments. This sentence has been re-worded. The details in the revised manuscript can be found in lines 65 - 70, page 3.

P9112L25: To make clearer what you want to say I suggest changing "…can only be employed to analyze OA datasets (…), but cannot be easily utilized in real-time …" into "…can only be employed to analyze OA datasets a posteriori (…), but cannot be easily utilized in real-time …"

This sentence has been changed.

P9113L7: Replace "appointments" by "apportionment"

Response: "appointments" has been replaced by "apportionment".

P9113L11: Replace "predict" by "determine"

Response: This sentence has been re-worded, combining with the next comment. The details in the revised manuscript can be found in lines 84 - 92, page 4.

P9113L14-24: This text is rather unclear. The information is partially hard or not to understand. Please reword.

Response: This text has been re-worded. The details in the revised manuscript can be found in lines 84 - 92, page 4.

P9113L25-26: What do you want to say with this sentence?

Response: We have been removed this text, according to the comments above.

P9114L11-13: This is not true. The BBOA mass concentrations have not been determined on-line based on the m/z 60 mass concentration. Before this calculation can be done first PMF has to be performed in order to obtain the conversion factors for this calculation.

Response: We agree with the referee that the estimated BBOA loadings have to be based on the results from the PMF analysis firstly. Thus, this sentence has been removed. In fact, we want to develop this on-line technology for estimating BBOA in further work when improving the standard ACSM analysis software with some codes, similar to the on-line technology for estimating HOA and OOA loadings in a previous study (Ng et al., 2011a).

P9112-P9114: In the introduction some detailed information should be given on the sources that are the focus of this investigation. What is typically burned during harvest season? Who burns it? How is it burned? Is there a typical time when material is burned? How dry is the material typically?

Response: We accepted the referee's comments and revised the text accordingly. We also have added some detailed information in the introduction. The details in the revised manuscript can be found in lines 59 - 61, page 3.

   In recent years, as the decreasing use of agricultural residues as a renewable fuel in the

rural YRD region, a lot of famers harvest the agricultural crops during the daytime, and then directly burn its residues during the nighttime and/or evening in their filed. As we know, both the burning conditions, i.e., flaming and smoldering phases likely play an important role in the field biomass burning events, because the agricultural residues could generally be mixed with more and/or less dry material. Figure R1 shows the typically agricultural residues burning events in China.

Figure R1. Pictures for the typical agricultural residues burning events in the field of China.

P9114L19-26: Unfortunately this section does not provide a good idea of the measurement site. According to the text it is located in a city of more than 8 million which is almost a megacity. Therefore, enormous influence from traffic, residential or industrial activities on air quality can be expected. What is the distance to such sources? What is the influence by such sources? How far are biomass burning sources away? How large is the expected transport time from such sources to the measurement site? If there are many restaurants in the vicinity of the sampling site as stated I wonder why not cooking aerosol is found and identified in PMF. During other field campaigns in cities it has been identified as a major fraction (>20%) of the organic aerosol.

<mark>Response:</mark> We agree with the referee's comments and suggests. We have revised the text accordingly. The sampling site was located on the roof of a six-story building approximately 18 m above ground level and ~ 15 m from the nearest heavy-traffic road), and ~ 50 m from the nearest restaurants and residents in this study. Therefore, the air quality should be affected by the local traffic and cooking-emission sources as we expected in this study. In addition, there are many fire locations around Nanjing (Figure S1), particular distributing in the western areas (Anhui province) of Nanjing. Logically, the biomass burning emissions should also be identified during the two harvest seasons. As shown in Figure S1, we could find that the biomass burning sources present the characteristics area pollution, impaling that the air quality of Nanjing might be potentially affected by the regional biomass burning sources, when air masses and/or wind direction are from there. It is approximately dozens of

8

kilometers and/or hundreds of kilometers from the main fire locations areas to urban Nanjing (Figure S1), as we expected. This meant the urban Nanjing site is significantly influenced by the biomass burning plumes originated the rural areas and undergone the aging in BB plumes measured dozens or hundreds of kilometers away from the fire locations, but little/negligible local biomass burning influence. Therefore, there should be more aged/secondary biomass burning-related OA than the fresh biomass burning-related OA in the biomass burning plumes, as we expected. The details in the revised manuscript can be found in lines 103 - 119, page 5.

We also agree well with the referee that cooking-emission sources play a significant role in aerosol pollution, particular for urban areas with many restaurants and resident areas around sampling sites. COA should also be identified in this study, in the absence of potential cooking-emission sources near the sampling site. As a matter of fact, we also found the cooking-emission sources could contribute to the aerosol concentrations. However, it has been mixed with the traffic HOA in this study, since the mass spectra of COA and traffic HOA are highly similar and they can be difficult to distinguish via PMF analysis with the Q-AMS or ACSM OA mass spectrum (e.g. Sun et al., 2010; Sun et al., 2012). As previous studies, the fraction of the organic mass fragments signal at m/z 55 and m/z 57 in total OA signal (i.e. f_{55} and f_{57}) can be used to differentiate COA from traffic HOA (e.g. Sun et al., 2011, 2012). Furthermore, Mohr et al. (2012) suggested a V-shape plot between f_{55} and f_{57} after subtracting the contributions from oxygenated OA factors (OOAsub) to distinguish the COA and traffic HOA in ambient datasets using an Aerodyne High Resolution Time-of-Flight AMS (HR-ToF-AMS). Their data points fall into a V-shape in a scatter plot, with strongly influenced HOA data aligned to the right arm and strongly influenced COA data points aligned to the left arm (Mohr et al., 2012). In a recent study, Xu et al. (2014) also found a similar result using HR-ToF-AMS. The same method has been used in this study for distinguishing the COA and traffic HOA (Figure R2). The relationships between f_{55} and f_{57} in this study, however, have no obvious variations at all, as the increasing of f_{55} and/or f_{57} during the summer and autumn harvest, which differs from the previous studies using HR-ToF-AMS (Mohr et al., 2012; Xu et al., 2014). Therefore, this is likely to a potential reason why it is hard to distinguish the COA and traffic HOA using the ASCM in this study.

In addition, it has been tried to disentangle the traffic and cooking contributions exploring the solution space using the mulitlinear engine (ME-2, Paatero 1999) with the newly developed source finder (SoFi, Canonaco et al., 2013) for the harvests data. Several techniques have been tested, e.g. constraining a traffic, a cooking and a biomass burning factor profiles from other PMF studies (Crippa et al., 2013, Ng et al., 2011b), increasing the

number of factors to possibly extract cleaner sources or reweighting the *m/z* 57 variable (mostly due to the fact that traffic is difficult to be extracted). In addition, unconstrained PMF runs have been performed on morning data (6 − 8 am) or midday data (11 am − 13 pm) in order to identify a traffic and cooking factor profile directly from the data and to employ them as constraints in subsequent runs over the full period. None of these strategies really led to a satisfactory result, since to some extend a mixing behavior was always notable, especially for the diurnal cycle. Based on these additional tests, it can be concluded that although some indications for a possible separation of HOA and COA for the harvest seasons is indicated from the f_{55}/f_{57} plot, it was not possible to achieve a reasonable and well-separated representation of these two factors. Therefore, the authors decided to keep these two sources in one single factor and label it mixed HOA and COA factor.

The HOA + COA also shows the high concentrations during the cooking time (noon and early night, see Fig. 3). Apart from the BB-related sources influence, the HOA + COA can contribute a major fraction to OA concentrations (> 40% for the summer and > 50% for the autumn) in the nighttime during the summer and autumn harvest (Figure R3), similar to the contribution of POA (= traffic HOA + COA) in Beijing (Sun et al., 2012) and in New York City (Sun et al., 2011).
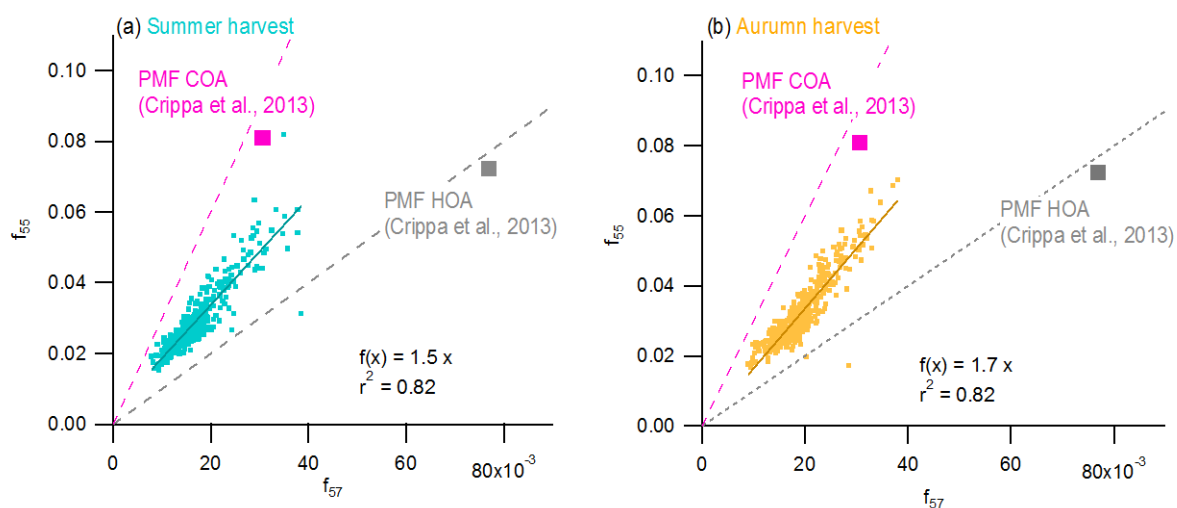

Figure R2. Scatter plots of f_{55} vs. f_{57}. The results between f_{55} and f_{57} during the two harvests highly differ from previous reports in an Aerodyne High Resolution Time-of-Flight AMS (HR-ToF-MAS) studies (Mohr et al., 2012; Xu et al., 2014).
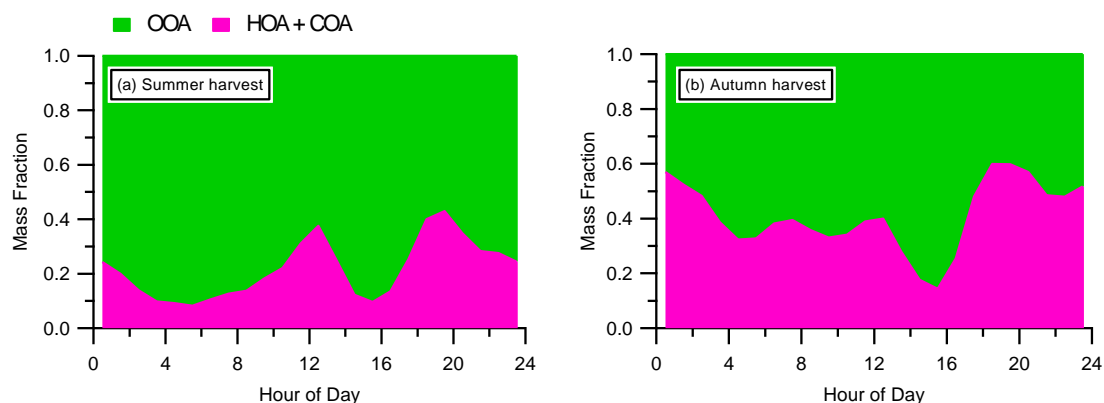
Figure R3. Diurnal variations of the mass fractional contributions of HOA + COA and OOA, excepting the biomass burning related influence, during the summer and autumn harvest respectively.

P9115L14, 17: It is unfortunate that the MARGA and the aethalometer were used with a PM$_{2.5}$ cyclone. Therefore, it would be desirable to get an idea on the fraction of K$^+$ and BC that is expected in the PM$_1$-PM$_{2.5}$ size range.

Response: We thank the referee's suggestions. We also agree with referee. It is unfortunate that the K$^+$ and BC were measured for PM$_{2.5}$. As a previous report, the mass of BC was mainly distributed below 1 μm at urban area in South China (Huang et al., 2012a). This means that the PM$_{2.5}$ BC could be approximately represented the PM$_1$ BC in the atmosphere at least in South China, which also agrees well with a report by Huang et al. (2012b) in YRD region. Therefore, the BC mass in atmosphere is mainly dominated by the PM$_1$ BC mass. The K$^+$ concentration was just used for as a tracer species, i.e., biomass burning emissions, aiming to further support the reasonable PMF BBOA in this study.

P9115L24ff: What exactly are the "fire products" used here? What are the limitations of these products – e.g. minimum size of fires, time resolution?

Response: The fire locations at a time resolution of ~ 24 h used in this study were available from MODIS (Moderate-resolution Imaging Spectroradiometer) mounted on NASA's Terra and Aqua satellites, NASA's Earth Observing System (EOS). MODIS can present fire distributions in details at 1 km resolution through Fire Information for Resource Management System (FIRMS) on global scale.

P9116ff: Relative ionization efficiencies (RIEs) and collection efficiencies (CEs) are discussed here. Can you also provide the RIE for organics? Can you provide some

information on ionization efficiency (IE) calibrations? How often, how, where have they been performed? How reproducible were the results?

We thank the referee's comments and suggestions. The default RIE value of 1.4 is used for organics. In general, the IE_{NO3} and RIE_{NH4} can be directly calibrated during the NH_4NO_3 calibrations in the standard AMS systems, because the high time resolution detection electronics allow for precise measurements of single ions and single particles, and thus $I_{s,i}$ and IE_s (Ng et al., 2011b), where $I_{s,i}$ is in units of ions/species (s), and IE_s is in units of ions/molecule. However, the ACSM cannot directly measure the single particles due to the slower detection electronics, i.e., no capability to time resolve single ions (Ng et al., 2011b). Thus, in practice, calibration of the ACSM is based on determining an instrument response factor (RF), using NH_4NO_3 calibration aerosol. The ACSM calibration is based on a combination of a Differential Mobility Analyzer (DMA) and Condensation Particle Counter (CPC), as well as the sum of the ion signals representing the NO_3 moiety is recorded with the ACSM for the IE and RIEs in this study. The mono-dispersed, size-selected 300 nm pure NH_4NO_3 particles within a range of concentrations were sampled into both the ACSM and a condensation particle counter (CPC). RF, is measured in units of amps of signal per µg m^{-3} of sampled aerosol. When normalized to the calibration volumetric sample flow rate Q_{cal} (in units of cm^3 s^{-1}) and multiplier gain G_{cal} (~20,000), RF is proportional to the ionization efficiency of s (in units of ions/molecule), following the procedures detailed in Ng et al. (2011b). IE was then determined by comparing the RF of ACSM to the mass calculated with the known particle size and the number concentrations from CPC. Once the IE is determined, the changes of air ions, e.g., m/z 28 (N_2^+) can be used to account for the degradation of detector (Ng et al., 2011b; Sun et al., 2012). As shown in Figure R4, the results show that the RF_{NO3} is 3.96×10^{-11} in this study. In addition, our ACSM was mainly used for the long-term and real-time online measuring the PM_1 species, because this Aerodyne ACSM can be operated unattended and continuously for a long observation. Nevertheless, we calibrate it around every half year at the superstation in this sampling site.
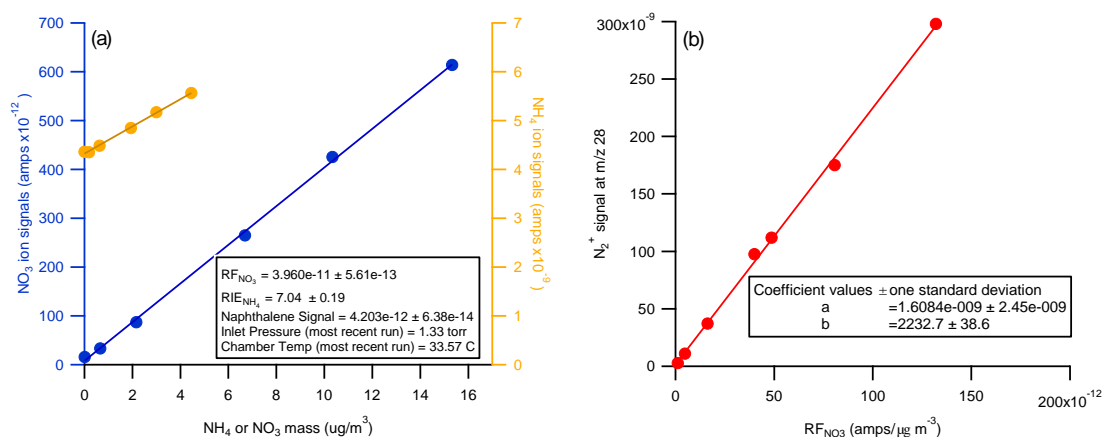
Figure R4. The results of (a) the RF_{NO3} and RIE_{NH4} calibration and (b) the relationship between N_2^+ signal at m/z 28 and RF_{NO3} in this study.

P9117L15ff: In Figure 1 time series of many meteorological factors and PM_1 components are shown. Most of the time series are not discussed. No information is taken from the temporal evolution of all these data, e.g. determination of meteorological situations, typical temporal trends of variables, etc. What is the point of showing the time series if the data are not used to extract information from them?

Response: We thank the referee's comments and suggestions. We also agree with the referee that the typical temporal trends of variables are very important and interesting. Thus, we expanded the discussions in the revised manuscript. The details in the revised manuscript can be found in lines 245 - 267, page 11 - 12.

P9118L12-14: What are the reasons for these sharp peaks of very high mass concentrations? There are in total 3 such peaks during the 4 weeks of measurement. Can they be associated with certain sources in close proximity? How long do they last? Should they be used to calculate correlations or do they represent "contaminated" data?

Response: We thank the referee's comments and suggestions. The 3 peaks in this study are reasonable. In addition, more details for those peaks have been explained in text (see section 3.1.1). For example, the first peak in the summer harvest was mainly affected by the biomass burning source emissions, with the highest concentrations of chloride (12.6 µg m^{-3}), BC (19.2 µg m^{-3}), K^+ (16.5 µg m^{-3}), BBOA (25.9 µg m^{-3}), and OOA-BB (101.6 µg m^{-3}), etc., while with low concentration of HOA + COA (2.1 µg m^{-3}).The secondary peak was mainly dominated by the urban pollution plumes, with high concentrations of HOA + COA (79.9 µg m^{-3}) and BC (16.9 µg m^{-3}), but with the relatively low concentrations of chloride (0.5 µg m^{-3}), K^+ (1.2 µg

m^{-3}), and BBOA (4.1 µg m^{-3}). The third peak, however, was likely affected by the local cooking-emissions plume during the nighttime, with high concentration of HOA + COA (58.0 µg m^{-3}) and the relative low concentrations of chloride (0.4 µg m^{-3}), BC (7.2 µg m^{-3}), K$^+$ (1.5 µg m^{-3}), BBOA (3.8 µg m^{-3}). Therefore, the three peaks can all be reasonably explained. Despite this, the 3 peaks were removed from curve fitting and/or correlating to avoid biasing the real correlations. The details in the revised manuscript can be found in lines 245 - 267, page 12.

P9118L22ff: How are average wind directions calculated? According to Figure S2 in autumn the majority of wind directions were from north (345 °15). This is not at all reflected in the average diurnal wind direction patterns in Figure 2. According to this the typical wind was from south (180), which according to Figure S2 almost never occurred. Also the statement that the diurnal variations of wind directions are similar for the two periods seems therefore not correct. When looking at Figure S2 one sees clearly that the wind patterns are very different during the two periods. For other variables there are also clear differences in the diurnal variations. Even though the general temporal trends are similar partially strong differences in the heights of peaks during certain times of the day occur.

Response: We agree with the referee that the average value of wind direction could not well reflect the diurnal wind direction pattern. Therefore, this text has been removed accordingly.

P9119L6-8: Are the three peaks in the SO$_4$ diurnal pattern significant? With only 2 weeks of data I would expect such small peaks to be generated by fluctuations of concentrations. Is it realistic to assume that the morning peak is generated by sulfur containing fuel? What is the fuel sulfur content in that region?

Response: We also agree with the referee that the three peaks of the sulfate in the diurnal pattern are not significant, while relatively flat. This reflects that sulfate shows the non-volatile character and it is a more regional pollutant in the YRD region during the summer and autumn harvest. The details in the revised manuscript can be found in lines 279 - 283, page 12.

P9119L15: The "two distinct peaks" in the NO$_3$ diurnal pattern looks more like one peak during night. The "minor peak" mentioned in line 23 is not visible to me – is it significant?

Response: We thank the referee's comments and suggestions. We also agree with the referee that the highest and important peak occurred in night, while the "minor peak" is not important.

The details in the revised manuscript can be found in lines 283 - 292, page 12.

P9118L20-P9120L4: To explain the diurnal patterns of the different PM components only potential sources are discussed. The effect of boundary layer height is completely ignored. Please include this into the discussion.

<mark>Response:</mark> We thank the referee's comments and suggestions. The variations of boundary layer height play a significant role in the pollutants dilution, particular for the diurnal variations of pollutants. Therefore, we have added some information of boundary layer height to the text.

P9120L7: PMF was used to separate three different types of organic aerosol: HOA, BBOA and OOA. This is a bit disappointing, especially since the measurements were performed in an urban area with multiple restaurants around the measurement site. It would be nice if HOA could be separated into a traffic-related HOA and a cooking related COA.

<mark>Response:</mark> We thank the referee's comments and suggestions, and also agree with referee. Both traffic-related HOA and COA should be distinguished in urban areas with some traffic and cooking sources around the sampling site. However, because of their similar mass spectra, they cannot be resolved from PMF analysis of unit mass resolution of organic spectra. The similar issues that COA cannot be separated from HOA have been reported previously by Sun et al. (2010) and Sun et al. (2012) in urban areas. In addition, it has been tried to disentangle the traffic and cooking contributions exploring the solution space using the mulitlinear engine (ME-2, Paatero 1999) with the newly developed source finder (SoFi, Canonaco et al., 2013) for the harvests data. Several techniques have been tested, e.g. constraining a traffic, a cooking and a biomass burning factor profiles from other PMF studies (Crippa et al., 2013, Ng et al., 2011b), increasing the number of factors to possibly extract cleaner sources or reweighting the m/z 57 variable (mostly due to the fact that traffic is difficult to be extracted). In addition, unconstrained PMF runs have been performed on morning data ($6 - 8$ am) or midday data (11 am $- 13$ pm) in order to identify a traffic and cooking factor profile directly from the data and to employ them as constraints in subsequent runs over the full period. None of these strategies really led to a satisfactory result, since to some extend a mixing behavior was always notable, especially for the diurnal cycle. Based on these additional tests, it can be concluded that although some indications for a possible separation of HOA and COA for the harvest seasons is indicated from the f_{55}/f_{57} plot, it was not possible to achieve a reasonable and well-separated representation of these two factors. Therefore, the authors decided to keep

these two sources in one single factor and label it mixed HOA and COA factor.

P9120L11-12: The average absolute BBOA contribution to the PM_1 aerosol is 50% larger in autumn compared to summer. I would not call this "similar" – especially if much smaller differences are used to draw conclusions further down the text.

Response: We thank the referee's comments and suggestions. The text has been re-worded. The details in the revised manuscript can be found in lines 310 - 314, page 13.

P9120L24: Add "used" between "can be" and "as a diagnostics"

Response: "used" has been added between "can be" and "as a diagnostics".

P9121L22: Should "consistent" not be "correlated"?

Response: "correlated" was changed to "consistent".

P9121L23: According to Figure 4 r^2 is 0.93, not 0.96.

Response: We thank the referee for carefully reviewing. The r^2 value has been changed accordingly.

P9122L5: What are the "BB activities" that are mentioned here? Can you specify this further?

Response: We thank the referee's comments and suggestions. "BB activities" has been changed to "BB emissions".

P9121-9122: It would be interesting to see whether there is a wind direction dependence of the concentrations of the different OA types.

Response: We agree with the referee. It is very interesting to analyze the wind direction dependence of OA components. We have added some information in the revised manuscript accordingly. For example, it is seen that the high HOA + COA concentration (> 6 µg m^{-3}) occurred when WD was from southeast during the summer harvest (Fig. S4). During the autumn harvest, the high concentration of HOA + COA was associated the northerly and easterly wind. This result is well consistent with the areas of local cooking and traffic sources emissions around sampling site. In addition, the uniform distribution of its concentrations is almost in association with all kinds of WD during the summer and autumn harvest respectively (Fig. S4). This is a good evidence for explaining the regional pollution of OOA in the YRD region during the harvest seasons. Some information about that has been added

(see section 3.2).

P9122L14-15: "OOA …is one of the most important secondary sources of …". OOA is not a "source", it is a type of organic aerosol.

<mark>Response:</mark> We agree with the referee. This sentence has been removed.

P9123L2-4: Since in Figure 5 only averages for the time intervals are shown I would not call this "dynamic variations". This term would be more appropriate for time series of data. I would also not call these differences "strong". For the visibility no significant difference is observed (within the error ranges) and for the other variables no error bars are presented. I would expect these differences also not to be significant. For SO_4, NH_3 and Chl no difference can be seen in the data. There seems to be a NO_3 and NH_4 increase in the BB-events compared to the non-BB times as well as an increase in HOA and OOA. Can you comment on this? Do you have an explanation for this?

<mark>Response:</mark> We thank the referee's comments. The original Fig. 5 have been changed with new one (Fig. 7), in order to understand the contribution of the harvest-related emissions to local air quality during the different biomass burning periods and compare with other sites. The increasing of NO_3, NH_4 and Chl, as well as an increase in HOA and COA during the different biomass burning periods, because the highest peaks of HOA occur at night and biomass burning emissions can also contribute oxygenated OA during those periods. The details in the revised manuscript can be found in lines 465 - 496, page 20 - 21.

P9123L15: The "value of 11.9 km" does not agree with the bar in Figure 5a.

<mark>Response:</mark> The related text has been removed.

P9123L22-25: Why do the secondary species concentrations increase linearly with increasing PM_1 concentrations? From my point of view if this relationship is significant this would mean that there are either no local sources (in an almost-megacity) or the concentration of species from local sources increases exactly as the concentrations of secondary species.

<mark>Response:</mark> We thank the referee's comments and suggestions. The secondary species (including nitrate, sulfate, ammonium and OOA) increase linearly with the increase of the PM_1 concentrations in this study. As show in Fig. 1d, the nitrate, sulfate, ammonium and total OA contribute the largest fraction to the total PM_1 loadings during the whole study. Also, the secondary OA accounts the largest fraction to the total OA concentrations (Fig. 7). This

means that the secondary species dominate the total PM_1 loadings for the entire study. Nevertheless, we have revised this text accordingly. The details in the revised manuscript can be found in lines 597 - 513, page 21.

P9123L27-28: The authors state that BC does not show a clear relationship with PM_1. According to Figure 6 BC concentrations increase in summer about as linearly as the secondary species with increasing PM_1.

Response: We thank the referee's comments and suggestions. We have compared the relationship between the fractions of all PM_1 species and the total PM_1 loadings during the summer and autumn harvest again, which were also presented in two new plots (Fig. 8a - b). The details in the revised manuscript can be found in lines 510 - 513, page 21.

P9123L20-P9124L27: There is a long discussion on the variations of individual species concentrations as a function of total PM_1 concentration. All kinds of features in the graphs in Figure 6 are interpreted. Taking into account that all the graphs in Figure 6 are strongly influenced by the data points at large concentrations of PM_1 (e.g. PM_1 above 50 $\mu g/m^3$) and that this PM_1 concentration range is often discussed in this paragraph one has to recognize that there are only 3 very short time intervals during the 2×2 weeks of measurement (see Figure 1c) where such concentrations are reached. Therefore all these discussions are based on only a handful of data points and are very likely not significant at all.

Response: We thank the referee for carefully reviewing. We also agree with the referee that a lot of discussions are focusing on only a handful of data points and are very likely not significant. Thus, we have removed some information for that and with new one. The details in the revised manuscript can be found in lines 497 - 513, page 21.

P9124L15-17: I do not agree with the statement "… the results suggest that BBOA plays a key role in causing the highest PM pollution during the harvest seasons, whereas the role of secondary species is less significant." For the largest PM_1 concentrations the fraction of BBOA is only about 20%. So biomass burning is only one out of several sources causing this high PM concentrations. In addition the inorganic secondary species NO_3, SO_4 and NH_4 together have a contribution of 40 - 45% to total PM_1. This is definitely not "less significant" than BBOA!

Response: We thank the referee's comments and suggestions. We also agree with the referee that the secondary species also play a significant role in total PM_1 including the high biomass

burning periods. This text has been revised, as response above. The details in the revised manuscript can be found in lines 497 - 513, page 21.

P9124L20-22: The authors state: "… when the mass fraction of OA is >70%, the POA mass concentration reaches ~100 μg/m^3, while SOA is generally below ~10 μg/m^3." How is this possible? According to Figure 1 there are only 3 very short events with PM>100 μg/m3. According to Figure 6 the POA mass fraction for PM > 100 μg/m^3 is about 25%. With maximum concentrations of about 145 μg/m^3 OA this is about 35 μg/m^3 (and not about 100μg/m^3)! In addition according to Figure 6 for PM>100 μg/m^3 SOA (OOA) is about 20-25 μg/m^3 (and not below 10 μg/m^3). The statement is probably only correct for individual single data points and therefore probably not really significant.

Response: We agree with referee that more discusses focusing on only individual single/several data is not really significant. Therefore, several high peaks have been presented in Table S1. The details in the revised manuscript can be found in lines 497 - 513, page 21.

P9124L28 and others: POA should not be identified with HOA, COA and BBOA; and SOA should not be identified with OOA. Likely most of the PMF-generated organic aerosol types are associated with either POA or SOA, however identifying them with primary or secondary aerosol is not sound since this cannot be proved with AMS data. There has been a long and intense discussion in the early days of AMS organics analysis on this issue, which needs no repetition.

Response: We thank the referee's comments and suggestions. We agree with the referee. BBOA often contains some secondary BBOA (S-BBOA) and primary BBOA (P-BBOA) in the BB plumes, although the fraction of P-BBOA vs. S-BBOA is difficult to estimate (DeCarlo et al., 2010), which was also reported by a recent field study (Young et al., 2014). However, OOA is a surrogate of SOA in AMS studies, which has been reported by previous studies (Zhang et al., 2005). Nevertheless, we have removed the related text.

P9125L1-4: During autumn harvest POA loadings are about 2 times higher than during summer harvest. The authors explain this with the amount of fire sites around urban Nanjing. This explanation is hard to believe when looking at Figure S1. During autumn the number of fires shown in the Figure and also the number of fires in the Nanjing area is much smaller than during summer. Is there a potential influence by lower boundary layer height in autumn?

Response: We thank the referee's comments and suggestions. It is very interesting to add the

boundary layer height for explaining its potential influence on higher POA loadings in the autumn harvest. This is likely a fact, although we have no the boundary layer height data. Therefore, we have revised that by adding some information of boundary layer height.

P9125L5-17: During times of high BBOA concentrations also SOA and SPM is increased. What is the reason for this? Are there different sources that cause high concentrations by coincidence? Is this a wind direction effect? Is the SOA from precursors that are co-emitted by the fires?

<mark>Response:</mark> The referee is right. Some precursors of SOA might be from the biomass burning emissions, and/or SOA might be in the biomass burning plumes with its aging. As a matter of fact, the two kinds of BBOA, i.e., a typical BBOA and a biomass burning-influenced OOA (OOA-BB), have been resolved with the PMF analysis in the revised manuscript, which has already been presented in section 3.2.2. Here we thank the referee's comments and suggestions.

P9125L20-21: Is f_{60} (= m/z 60 / total organics) or m/z 60 (= signal at m/z 60) a marker for BBOA? In the formulae used here and in other studies m/z 60 is used to calculate the BBOA concentration. In the next sentence here f_{60} is used. This should be handled more consistently or at least the difference should be made clear.

<mark>Response:</mark> We thank the referee's comments and suggestions. The f_{60} (the ratio of the integrated signal at m/z 60 to the total signal in the organic component mass spectrum) level can be as a marker for biomass burning emission and BBOA (Aiken et al., 2009; Cubison et al., 2011). The m/z 60 loadings can also be as a good marker for BBOA, when removing the influence of non-biomass burning m/z 60 (~0.3% of total OA signal, in general). "m/z 60" was changed to "Δ m/z 60" in the title.

P9125L22-23: The background level of f_{60} was determined for non-BB periods. What are "non-BB periods"? From a later section of the manuscript it becomes apparent that these are periods between the two 2-week measurement periods. However, this is rather unclear. Explain and introduce these measurements in an earlier section of the manuscript. Additionally: how do you know that during these periods there was no BB-related aerosol in the air?

<mark>Response:</mark> We thank the referee's comments and suggestions. The background level of f_{60} (~ 0.26% of total OA signal) was determined for little or negligible biomass burning influence

(non-BB periods, in July 1 to 8), which is consistent with the result (~0.3% of total OA signal, in general) for background level of biomass burning emission in previous studies (Aiken et al., 2009; Cubison et al., 2011). The details in the revised manuscript can be found in lines 515 - 542, page 22.

P9125L27: There are different slopes for the correlations between Delta-60 and BBOA. What does this mean? Can you discuss the possible reason for this and its implications. If this slope is not known a priori, a real on-line calculation of BBOA seems not feasible.

Response: We thank the referee's comments and suggestions. We also agree well with the referee that the results of BBOA loadings based on on-line calculation of BBOA are dominated the results from PMF BBOA before (with a slope). The details in the revised manuscript can be found in lines 524 - 532, page 22.

P9126L8-11: How is the information in Figure 10 different from that in Figure 9?

Response: We thank the referee's comments. We have rearranged the figures with new discusses in the revised manuscript. Figure 10 (before) shows results of the "slopes" for estimating BBOA only. However, we also want to see the typical temporal trends of variables between the estimated BBOA and PMF BBOA during the two harvests.

P9126L16: I suggest replacing "… overall mass concentration and fraction of OOA and ..." by "… overall mass concentration of OOA and its fraction of OA and …"

Response: This sentence has been changed.

P9126L20-22: Please reword. This sentence is hard to understand.

Response: This sentence has been re-worded. The details in the revised manuscript can be found in lines 545 - 547, page 23.

P9126L22-24: When looking at Figure 11b, I do not think this "linear relationship" is significant.

Response: We thank the referee's comments and suggestions. The related text has been removed.

P9127L4-5: Also the "small peak" at 30 μg/m^3 – 80 μg/m^3 is not really significant.

Response: We also agree with the referee. The related text has been removed.

P9127L8-12: This conclusion might be right, but how do the data suggest this process?

Response: We thank the referee's comments and suggestions which are available for leading to our revision. In the revised manuscript, moreover, we have removed this sentence and made a major revision for explain the mixing and/or aging processes. The details in the revised manuscript can be found in lines 545 - 563, page 23.

P9127L13-24: This paragraph is hard to understand. Please reword.

Response: This paragraph has been re-worded.

P9127L15-16: The degree of OA aging might be affected by atmospheric photochemical activity. Does this make sense? While O_x describes the actual level of photochemical activity the f_{44}/f_{43} ratio describes the oxidation level of the aerosol which is the result of the oxidation during the history of the particles.

Response: We thank the referee's comments and suggestions. We also agree with the referee. As discussed in Aiken et al. (2008), and Jimenez et al. (2009), the f_{44} can be considered as indicator of atmospheric aging due to photochemical aging processes leading to the increase of f_{44} in the atmosphere. However, it does not need to further emphasize and/or explain using the O_x loadings. Thus, we have removed the related information about O_x in this study accordingly.

P9127L25-P9128L2: What are the "BB campaigns"? What is the "photochemical activity campaign"? The fraction of BBOA in total OA decreases with increasing oxidation degree of OA. Is this a result of a transformation process as suggested here – or does this just reflect the different mixing of OOA and BBOA in different air masses? The same comment is also true for the rest of this paragraph.

Response: We thank the referee's comments and suggestions. The "BB campaigns" was changed to "BB emissions", and "photochemical activity campaign" was removed. We also agree with the referee that the BBOA can be mixed with OOA and/or other pollutants in the different air masses and/or BB-related plumes. As previous reports (DeCarlo et al., 2010; Bougiatioti et al., 2014), the OA / ΔCO ratio is related to the emission ratios of the main sources of OA and CO, their relative strengths, and SOA formation, while in the absence of biomass burning plumes, the dominant processes affecting OA / ΔCO are mixing of the source region plume with clean regional air and any additional evolution of OA (additional

SOA formation, oxidation, volatilization, etc.). Therefore, we have explained this evolution process using the relationship between (BBOA + OOA-BB) / ΔCO and f_{44} for the two campaigns. Figure 11 shows the BB-related OA (i.e. BBOA + OOA-BB) to ΔCO ratio as a function of the f_{44} during the summer and autumn harvest respectively, to investigate further the probable importance of the aging processes of BB plumes. The CO background is determined as 14.9 µg m^{-3} for summer harvest and 17.9 µg m^{-3} for autumn harvest, based on an average of the lowest 5% CO during two plumes (Takegawa et al., 2006). The ratio of BBOA + OOA-BB to ΔCO can remove the effect of dilution in the regional air (DeCarlo et al., 2008). As discussed in de Gouw et al. (2005), Aiken et al. (2008), Jimenez et al. (2009), and Ng et al. (2010), the f_{44} can be considered as indicator of atmospheric aging due to photochemical aging processes leading to the increasing of f_{44} in the atmosphere. Overall, the (BBOA + OOA-BB) / ΔCO ratio shows an obvious reduction with increasing of f_{44} values during the summer and autumn harvest respectively, apart from the influence of traffic-like plumes. This is likely due to a combination of rapid SOA formation from BB emissions and mixing with urban air and with higher CO content. Similar results have also been found by DeCarlo et al. (2010), from aircraft measurements during MILAGRO in Mexico City and the Central Mexican Plateau. It is interesting that the BB plumes in the summer harvest show a higher oxidation level ($\Delta f_{44} = 0.04$) than that in the autumn harvest (Fig. 11a-b). This might be a potential factor leading to a higher oxidation level in the mass spectra of OOA-BB in summer harvest, compared to that in the autumn harvest (Fig. 3c).

P9128L10-11: I do not see how this is the consequence of the previous sentence. The logical cause-effect relationship is missing.
<mark>Response:</mark> We thank the referee's comments and suggestions. We have replaced the original plots with two new plots for presenting the logical cause-effect relationship (see Fig. 12c-d).

P9128L23-26: According to the text there are high BBOA contributions in the air masses of group 3 during summer and autumn. From Figure 13 I can see the large contributions for BBOA from air mass group 3 in autumn, but not in summer. In addition the authors state that this is "consistent with the fire location distributions around this sampling site". For the summer field campaign back trajectory group 3 is from north to north-west. This is indeed in agreement with a large number and density of fires according to Figure S1a. However, for this trajectory group no increased BBOA fraction is observed (Figure 13a). For the autumn field campaign trajectory group 3 is from SE. According to Figure S1b there is no large

number or concentration of fires in this direction during this time. The same is true for the direction of group 4 trajectories, for which also an increased BBOA fraction is observed according to Figure 13b.

Response: We thank the referee for carefully reviewing. We have made major revisions for this section. The 48-h back trajectories (BTs) staring at 500 m above the ground level of Nanjing were recalculated every 2 h in the campaigns, and then clustered according to their similarity in spatial distribution using the HYSPLIT4 software. The corresponding BTs can be broadly classified into four principal clusters of air masses based on the change in total spatial variance during the summer and autumn harvests, respectively, i.e., northeasterly (NE) BTs, easterly marine (EM) BTs, southeasterly marine (SEM) BTs, and southwesterly continental (SWC) for the summer harvest; northerly continental (NC) BTs, northeasterly marine (NEM) BTs, easterly marine (EM) BTs and southerly continental (SC) for the autumn harvest.

P9128L28-P9129L5: In the plots of Figure S5 I do not see an increase of BBOA and Chl for greater wind speeds. In addition, what do you mean with "suggesting a vital role of the BB campaign around urban Nanjing in the BB pollution."? Finally, how do high BBOA and Chl concentrations associated with wind speeds of ~ 2 m/s denote the dominant role of local burning events around Nanjing? I do not see how this provides a strong basis for this conclusion. In addition I do not see this association of high BBOA and Chl concentrations with wind speeds of ~ 2 m/s.

Response: We thank the referee for carefully reviewing. We have removed Figure S5 and added some new plots for explaining the relationship between OA components and wind direction (see Figure S4). In addition, we also revised the text (see section 3.6).

P9129L6-8: Again I cannot agree with the statement and the conclusion. According to Figure 13 HOA and BC are not larger for group 2 and 4 compared to the other groups in summer. In autumn HOA and may be BC is larger for group 2 but not for group 4. In addition according to Figure 13 both, group 2 and group 4 back trajectories arrive at the measurement site from east to northeast during both measurement periods and not from north as stated by the authors.

Response: We thank the referee's comments and suggestions. We have revised the text, as response above.

P9129L14-16: Again it is hard to follow the text and to agree with the statements. This sentence is hard to understand. In addition I do not see that in group 2 in summer and in group 1 in autumn the OOA fractions are significantly larger than in the other groups of the respective measurement periods – they all seem quite similar. In addition: What do the authors mean with "longest trajectory"? Indeed, for the simulated time interval (48 h) the group 2 and group 1 trajectories are the longest ones. This means that during the last 48 h before arrival the aerosol has traveled the longest distance for these groups. However, for OOA generation it is probably rather a question of how large the emissions of precursors are along the track of the air masses during the last couple of days than the velocity of the transport. For example group 2 trajectories from the summer field campaign arrived from the sea with probably little OOA precursor emissions. Thus it would be reasonable to assume lower OOA concentrations for this group compared to the other groups where the air spent several days over the continent before the measurement.

Response: We thank the referee's comments and suggestions. The related text has been reworded. The air masses originated from marine/sea with less OOA and higher HOA and COA fractions to OA were presented in this study, compared with the air masses from continental areas (Fig. 13a-b).

P9129L20: I also do not see larger SO_4 fractions for the groups that are associated with air masses from the northwest and northeast.

Response: This sentence has been removed.

P9131L4: Jiangsu province was not mentioned before. Please introduce first in the main text.

Response: This sentence has been removed.

Figure 2: It would be easier to identify times of the day if the scale would be 0/6/12/18/24 instead of 0/10/20. (Also Figure 7b, d)

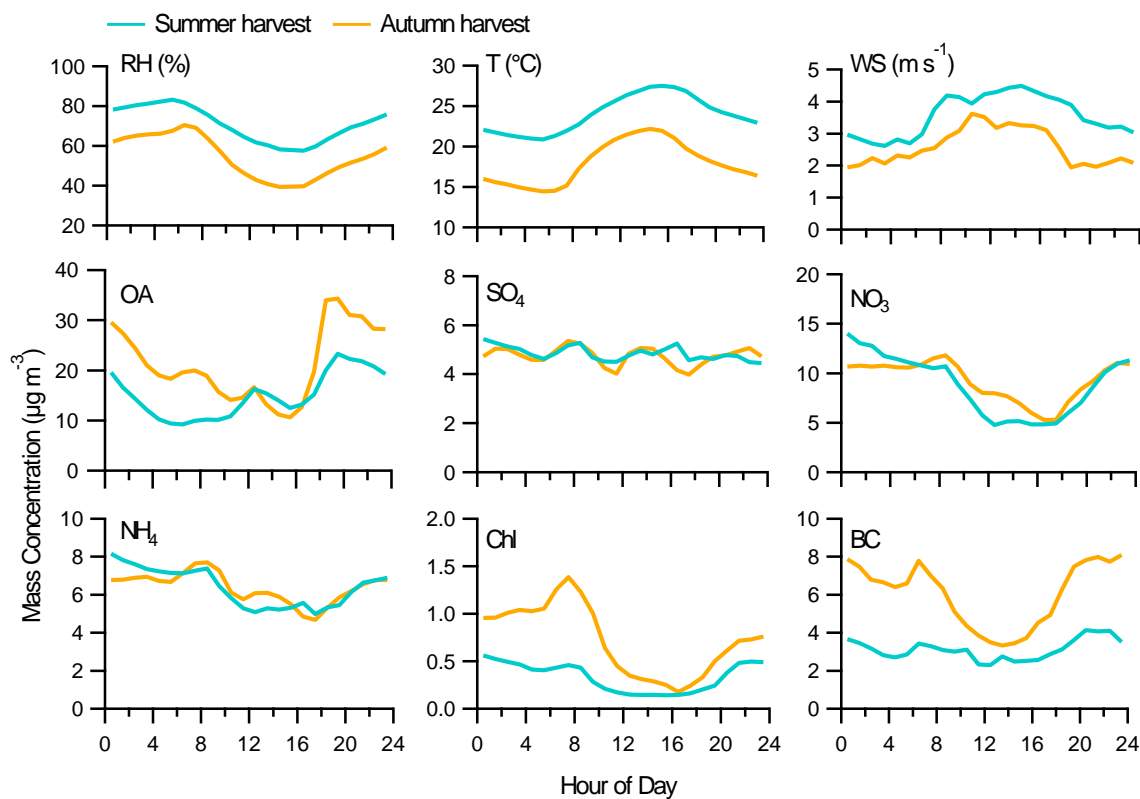Response: The time scale has been changed, i.e., 0/4/8/12/16/20/24 instead of 0/10/20.

Fig. 2. Diurnal variation patterns of meteorological factors (i.e. RH, T, and WS), PM_1 species including organic aerosol (OA), nitrate (NO_3), sulfate (SO_4), ammonium (NH_4), chloride (Chl), and black carbon (BC) during the harvest seasons.

Figure 3: The summer and autumn mass spectra are hard if not impossible to distinguish. I suggest shifting one of the spectra by 0.5 amu.

Response: We thank the referee's comments and suggestions. Figure 3 was revised with different colors and line thickness.
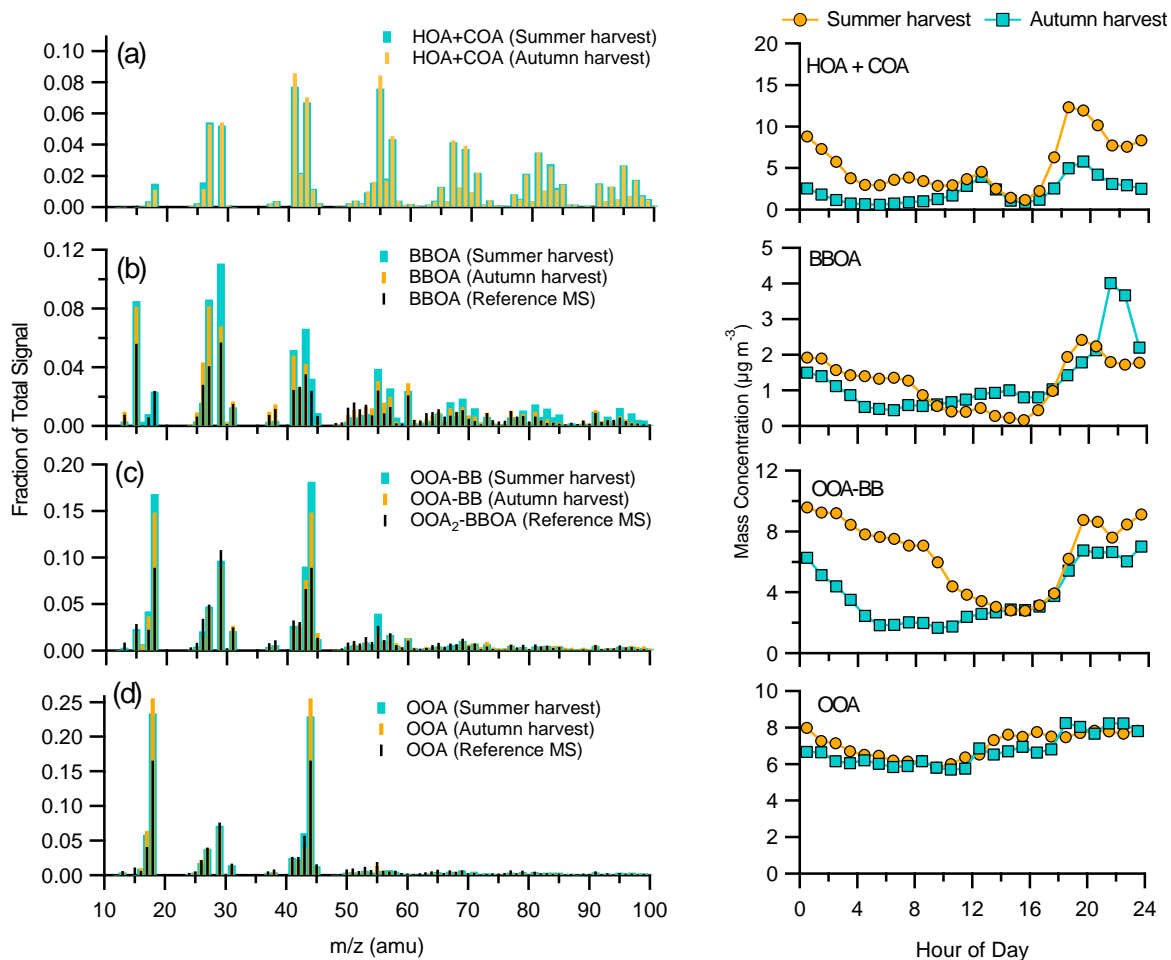

Fig. 3. Mass spectra profiles (left) and diurnal variations (right) of four OA components, i.e., hydrocarbon-like and cooking-emission related OA (HOA + COA), fresh biomass burning (BB) OA (BBOA), oxygenated BB-influenced OA (OOA-BB), and oxygenated OA (OOA). Note that the reference mass spectra (MS) data is from the results by Crippa et al. (2013).

Figure 10: Are the fits forced through zero? Can you give the fit equation in a more meaningful form, e.g. PMF BBOA = 1.06 × estimated BBOA?

<mark>Response:</mark> Yes, the fits are forced through zero. Following the reviewer's suggestion, we added "PMF BBOA = 1.0 × estimated BBOA" in the plot.
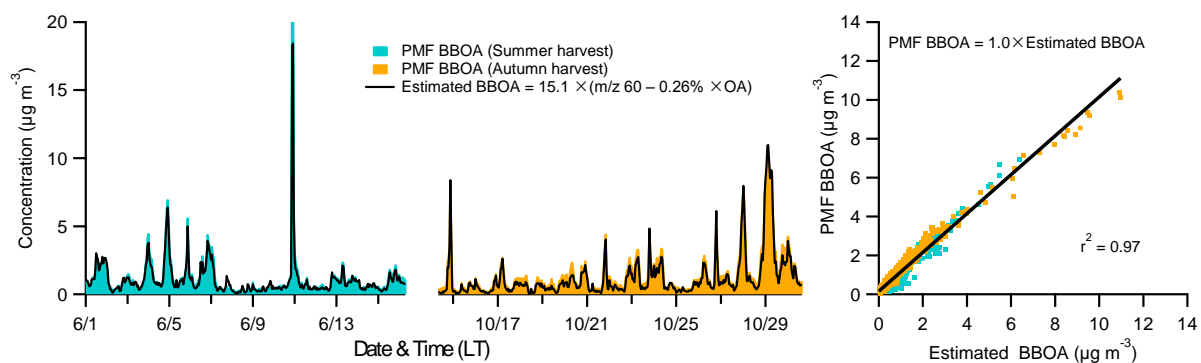

Fig. 10. Time series of BBOA identified by PMF (PMF BBOA) and estimated BBOA during the harvest seasons, as well as correlation plot of estimated BBOA vs. PMF BBOA. Note that the highest values for case 1 (Fig. 1c) during the summer harvest have been removed for fitting.

Figure 11: It is impossible to distinguish the two types of symbols in the graphs.

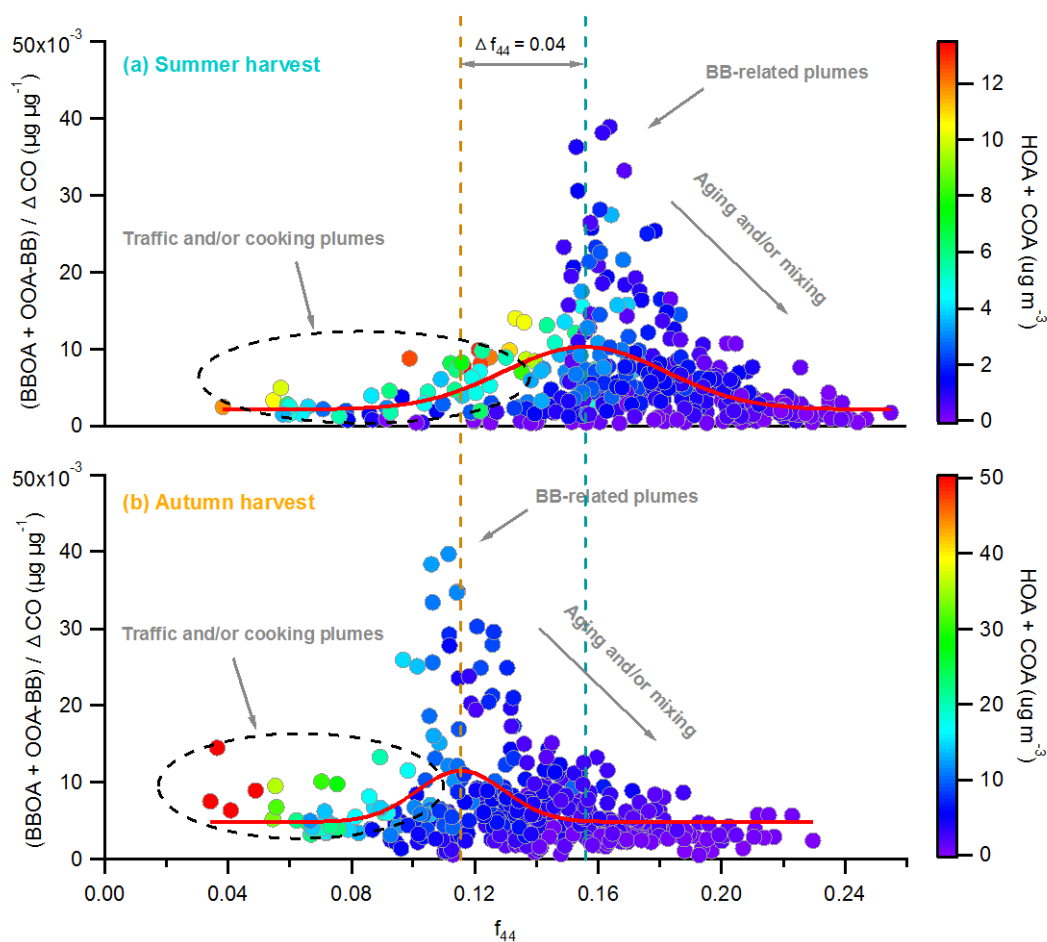We agree with the referee. The original Figure 11" has been changed to a "new Figure 11" in the revised manuscript.

Fig. 11. The (BBOA + OOA-BB)/ΔCO ratio as a function of f_{44} during the summer and autumn harvest, respectively. Colored by the HOA + COA mass concentrations, and the red curve lines are the Gaussian curve fitting for the summer and autumn harvest respectively.

Figure 12a: What are the blue dots? What are the green and red symbols?
<mark>Response:</mark> The blue dots are the data points for the plot of f_{44} vs. f_{43}, and the green and red symbols have also been further explained in the figure caption.
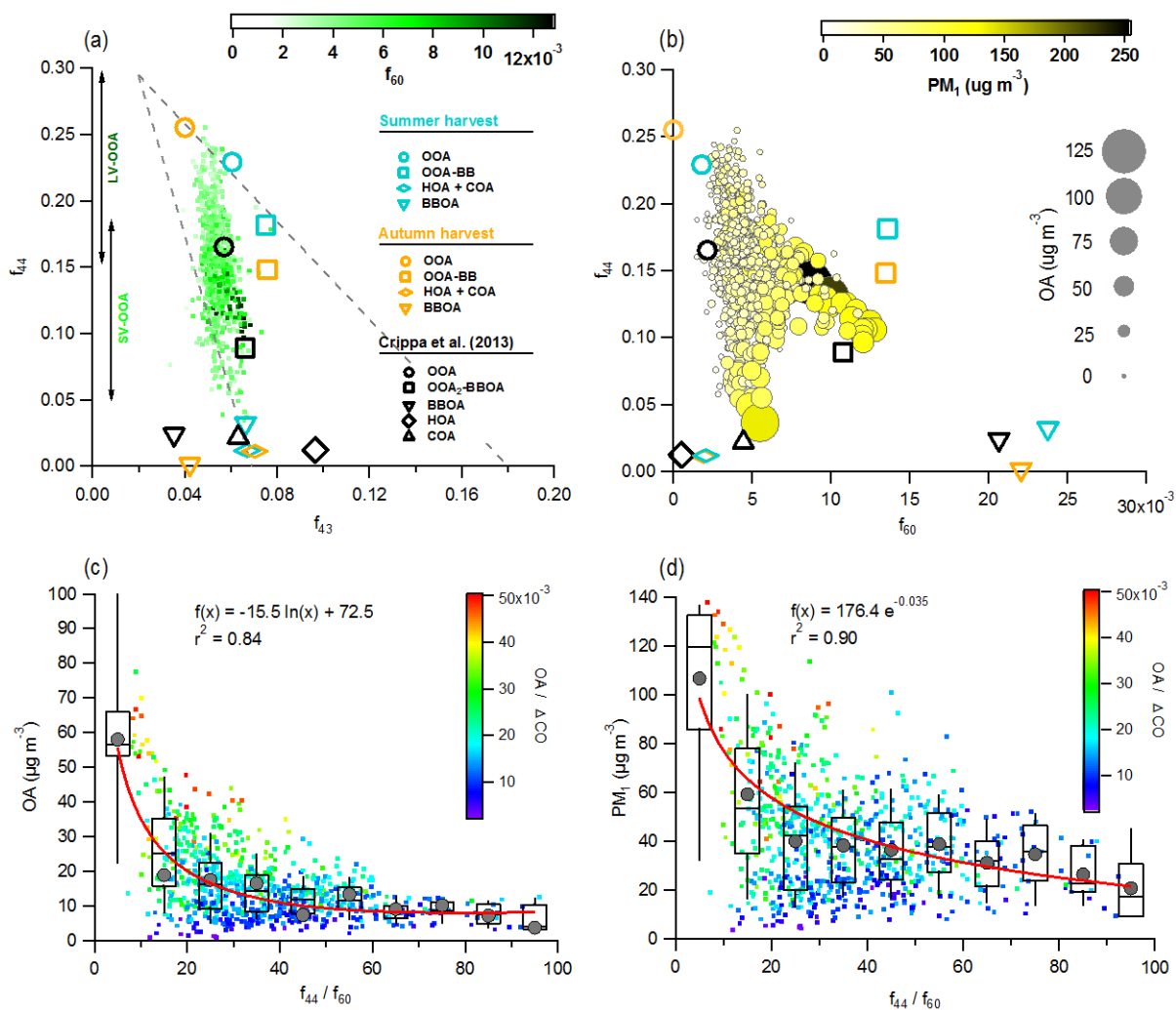

Fig. 12. Summary plots showing **(a)** triangle plot (f_{44} vs. f_{43}), SV-OOA and LV-OOA are represented for semi-volatile OOA and low-volatility OOA respectively. The dots are colored by f_{60} as a biomass burning marker; **(b)** f_{44} as a function of f_{60} (f_{44} vs. f_{60}), colored by the PM_1 mass concentration and sized by the OA loadings; **(c-d)** the total OA and PM_1 mass concentration as a function of the ratio f_{44}/f_{60}, colored by the OA / Δ CO ratio, respectively. Note that using the mean values (gray points) for fitting.

References

Aiken, A. C., DeCarlo, P. F., Kroll, J. H., Worsnop, D. R., Alex, H. J., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y. L., Zhang, Q., Trimborn. A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, R., Prévôt, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., Jimenez, J. L.: O/C and OM/OC Ratios of Primary, Secondary, and Ambient Organic Aerosols with High-Resolution Time-of-Flight Aerosol Mass Spectrometry, Environ. Sci Technol, 42 (12): 4478-4485, 2008.

Aiken, A. C., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D., Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner, E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L.: Mexico City aerosol analysis during MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) – Part 1: Fine particle composition and organic source apportionment, Atmos. Chem. Phys., 9, 6633-6653, doi:10.5194/acp-9-6633-2009, 2009.

Bougiatioti, A., Stavroulas, I., Kostenidou, E., Zarmpas, P., Theodosi, C., Kouvarakis, G., Canonaco, F., Prévôt, A. S. H., Nenes, A., Pandis, S. N., and Mihalopoulos, N.: Processing of biomass-burning aerosol in the eastern Mediterranean during summertime, Atmos. Chem. Phys., 14, 4793-4807, doi:10.5194/acp-14-4793-2014, 2014.

Crippa, M., DeCarlo, P. F., Slowik, J. G., Mohr, C., Heringa, M. F., Chirico, R., Poulain, L., Freutel, F., Sciare, J., Cozic, J., Di Marco, C. F., Elsasser, M., Nicolas, J. B., Marchand, N., Abidi, E., Wiedensohler, A., Drewnick, F., Schneider, J., Borrmann, S., Nemitz, E., Zimmermann, R., Jaffrezo, J.-L., Prévôt, A. S. H., and Baltensperger, U.: Wintertime aerosol chemical composition and source apportionment of the organic fraction in the metropolitan area of Paris, Atmos. Chem. Phys., 13, 961-981, doi:10.5194/acp-13-961-2013, 2013.

DeCarlo, P. F., Ulbrich, I. M., Crounse, J., de Foy, B., Dunlea, E. J., Aiken, A. C., Knapp, D., Weinheimer, A. J., Campos, T., Wennberg, P. O., and Jimenez, J. L.: Investigation of the sources and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements during MILAGRO, Atmos. Chem. Phys., 10, 5257-5280, doi:10.5194/acp-10-5257-2010, 2010.

de Gouw, J. A., Middlebrook, A. M., Warneke, C., Goldan, P. D., Kuster, W. C., Roberts, J. M., Fehsenfeld, F. C., Worsnop, D. R., Canagaratna, M. R., Pszenny, A. A. P., Keene, W. C., Marchewka, M., Bertman, S. B., and Bates, T. S.: Budget of organic carbon in a polluted atmosphere: Results from the New England Air Quality Study in 2002, J. Geophys. Res. Atmos., 110, D16305, 2005.

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prévôt, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., E, Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S.,

Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of organic aerosols in the atmosphere, Science, 326, 1525-1529, 2009.

Huang, X.-F., He, L.-Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L., Zeng, L.-W., Liu, X.-G., Zhang, Y.-H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.: Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer, Atmos. Chem. Phys., 10, 8933-8945, doi:10.5194/acp-10-8933-2010, 2010.

Huang, X.-F., Sun, T.-L., Zeng, L.-W., Yu, G.-H., and Luan, S.-J.: Black carbon aerosol characterization in a coastal city in South China using a single particle soot photometer, Atmos. Environ., 51, 21-28, http://dx.doi.org/10.1016/j.atmosenv.2012.01.056, 2012a.

Huang, X.-F., He, L.-Y., Xue, L., Sun, T.-L., Zeng, L.-W., Gong, Z.-H., Hu, M., and Zhu, T.: Highly time-resolved chemical characterization of atmospheric fine particles during 2010 Shanghai World Expo, Atmos. Chem. Phys., 12, 4897-4907, doi:10.5194/acp-12-4897-2012, 2012b.

Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U., and Prévôt, A. S. H.: Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data, Atmos. Chem. Phys., 12, 1649-1665, doi:10.5194/acp-12-1649-2012, 2012.

Ng, N. L., Canagaratna, M. R., Jimenez, J. L., Zhang, Q., Ulbrich, I. M., and Worsnop, D. R.: Real-Time Methods for Estimating Organic Component Mass Concentrations from Aerosol Mass Spectrometer Data, Environ. Sci. Technol., 45, 910-916, 2011a.

Ng, N. L., Herndon, S. C., Trimborn, A., Canagaratna, M. R., Croteau, P. L., Onasch, T. B., Sueper, D.,Worsnop, D. R., Zhang, Q., Sun, Y. L., and Jayne, J. T.: An Aerosol Chemical Speciation Monitor (ACSM) for Routine Monitoring of the Composition and Mass Concentrations of Ambient Aerosol, Aerosol Sci. Tech., 45, 770–784, 2011b.

Sun, J. Y., Zhang, Q., Canagaratna, M. R., Zhang, Y. M., Ng, N. L., Sun, Y. L., Jayne, J. T., Zhang, X. C., Zhang, X. Y., and Worsnop, D. R.: Highly time-and size-resolved characterization of submicron aerosol particles in Beijing using an Aerodyne Aerosol Mass Spectrometer, Atmos. Environ., 44, 131-140, 2010.

Sun, Y.-L., Zhang, Q., Schwab, J. J., Demerjian, K. L., Chen, W.-N., Bae, M.-S., Hung, H.-M., Hogrefe, O., Frank, B., Rattigan, O. V., and Lin, Y.-C.: Characterization of the sources and processes of organic and inorganic aerosols in New York city with a high-resolution time-of-flight aerosol mass apectrometer, Atmos. Chem. Phys., 11, 1581-1602, doi:10.5194/acp-11-1581-2011, 2011.

Sun, Y. L., Wang, Z. F., Dong, H. B., Yang, T., Li, J., Pan, X. L., Chen, P., and Jayne, J. T.: Characterization of summer organic and inorganic aerosols in Beijing, China with an Aerosol

Chemical Speciation Monitor, Atmos. Environ., 51, 250-259, 2012.

Weimer, S., Alfarra, M. R., Schreiber, D., Mohr, M., Prévôt, A. S. H., and Baltensperger, U.: Organic aerosol mass spectral signatures from wood-burning emissions: Influence of burning conditions and wood type, J. Geophys. Res., 113, D10304, doi:10.1029/2007JD009309, 2008.

Xu, J., Zhang, Q., Chen, M., Ge, X., Ren, J., and Qin, D.: Chemical composition, sources, and processes of urban aerosols during summertime in Northwest China: insights from High Resolution Aerosol Mass Spectrometry, Atmos. Chem. Phys. Discuss., 14, 16187-16242, doi:10.5194/acpd-14-16187-2014, 2014.

Young, D. E., Allan, J. D., Williams, P. I., Green, D. C., Harrison, R. M., Yin, J., Flynn, M. J., Gallagher, M. W., and Coe, H.: Investigating the two-component model of solid fuel organic aerosol in London: processes, PM_1 contributions, and seasonality, Atmos. Chem. Phys. Discuss., 14, 20845-20882, doi:10.5194/acpd-14-20845-2014, 2014.

Zhang, Q., Worsnop, D. R., Canagaratna, M. R., and Jimenez, J. L.: Hydrocarbon-like and oxygenated organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols, Atmos. Chem. Phys., 5, 3289-3311, doi:10.5194/acp-5-3289-2005, 2005.