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Dear Dr. Maenhaut,

Thank you very much for your and reviewers' useful comments and suggestions on our manuscript! We have revised the manuscript and response letter accordingly. The detailed responses to the comments and suggestions are shown below point by point.

Thank you very much for your considerations!

Sincerely yours,
Dr. Lili Tang

Response to the Editor's comments and suggestions

General comments:

The language and grammar of the authors' replies to the referee comments are rather weak. Therefore, the replies are somewhat difficult to follow. Anyway, referee #3 had very major comments for the ACPD version and I feel that he should be invited to have a look at the authors' replies and at the revised version to decide whether his comments were properly addressed and to assess to which extent the revised manuscript needs further revision. Also referee #2 of the ACPD version and new referees are invited to review the revised version. Furthermore, I have several comments and technical corrections myself, so that further revision is definitely needed.

Response: Thanks for the comments and suggestions! We have modified the response letter accordingly!

Specific comments:

The language and grammar should certainly be improved.

Lines 38-39: It is not clear what BBOA represents here. Is it only "fresh biomass burning OA" or the sum of "fresh biomass burning OA" and "biomass burning-influenced OA"?

Line 49: "Ge et al., 2011" is not in the reference list.

Line 54: “Bougiatioti et al., 2013” is not in the reference list.

Line 66: Replace “identify the sources apportionment” by “identify and apportion the sources”.

Line 75: Replace “were developed” by “was developed”.

Line 95: Replace “is resolved” by “are analyzed”.

Line 103: Replace “of about” by “of”.

Line 110: Replace “In this study, the” by “The”.

Line 116: Replace “negligible” by “ negligibly”.

Line 123: Acronyms and abbreviations (here “NR”) should be defined (written full-out) when first used.

Line 135: Replace “detection limits” by “detection limit”.

Line 157: Replace “And the pure” by “Pure”.

Line 162: Replace “a RIE” by “the RIE”.

Line 175: Replace “was also applied” by “was applied”.

Line 178: Replace “combing” by “combining”.

Line 199: “Young et al., 2014” is not in the reference list.

Line 263: Replace “shown in” by “seen in”.

Line 263: Replace “presents the” by “presents”.

Lines 280-281: “This means the non-volatile and character” should be rephrased.

Line 376: Replace “were characterized” by “ was characterized”.

Line 379: Replace “mass spectrums” by “mass spectra”.

Line 388: “Zhang et al., 2005c” is not in the reference list.

Line 393: Replace “thorough the” by “through the”.

Line 393: Replace “slightly increase” by “slight increase”.

Line 404: Replace “mass spectrums” by “mass spectra”.

Line 431: It is unclear what is meant by “the increase of temperature condition”.

Line 440: Replace “show a similar trend with” by “shows a similar trend as”.

Line 442: Replace “recent filed” by “recent field”.

Line 444: “Robinson et al., 2007” is not in the reference list.

Line 457: Replace “Pear River Delata” by “Pearl River Delta”.

Line 475: “to some discusses in” should be reworded.

Line 476: Replace “than that in” by “than in”.

Line 480: “Lathem et al., 2013” is not in the reference list.

Line 485: Replace “than that in” by “than in”.

Line 492: Replace “are much” by “is a much”.

Line 493: Replace “have also been” by “have been”.

Line 499: Replace “maintained at a” by “remained at a”.

Line 530: “for explaining the OOA-BB loadings depended on” should be rephrased.

Line 540: Replace “equations of” by “equation of”.

Line 557: Replace “have also been” by “have been”.

Line 594: Acronyms and abbreviations (here “BTs”) should be defined (written full-out) when first used.

Line 611: Replace “associated” by “is associated”.

Line 618: Replace “that a” by “a”.

Line 621: Replace “sources emission play” by “sources play”.

Line 651: Replace “than that” by “than”.

Line 662: Replace “to the sulfate” by “to that of sulfate”.

Line 667: Replace “equations of” by “equation of”.

Line 676: Replace “are mainly” by “is mainly”.

Lines 691-692: “Andreae and Merlet, 2001” should come after “Allan et al., 2010”.

Lines 693-695: “Alfarra et al., 2007” should come after “Aiken et al., 2010”.

Lines 875-877, lines 881-882, lines 897-899, lines 918-920, lines 926-927, lines 969-971, and lines 993-994: The title of journal articles should be in lower case instead of in title case.

Lines 878-880: “Lipsky et al., 2007” should come after “Li et al., 2006”.

Line 1048: Replace “Averagely relative” by “ Average relative”.

Line 1050: Replace “Pear River” by “Pearl River”.

Line 1060: Replace “(orange) harvests” by “harvest (orange)”.

Line 1076: Replace “are represented for” by “indicate”.

Table S4, second line of heading: Replace “showed in” by “showed” and replace “will be” by “are”.

Table S5, second line of heading: Replace “showed in” by “showed” and replace “will be” by “are”.

Figure S1, third line of caption: Replace “1km” by “1 km”.

Figure S4, first line of caption: Replace “plots show the” by “plots of the”

Response: Thanks for the suggestions! All the suggestions have been adopted.

Response to the Reviewers' comments and suggestions

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: Thanks for the constructive comments and suggestions! We have revised the manuscript accordingly. Specific corrections are listed as follows point by point.

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: Thanks for the comments and suggestions! In this study, the ACSM organic mass spectra were analyzed using the PMF model. In the revised manuscript, the PMF results were re-evaluated, and a better PMF-solution with more than 3 factors, i.e., a 4-factor solution including a new factor (oxidized biomass burning-influenced OA, OOA-BB), was selected. The reason why the 4-factor solution was selected is because that the four factors can be well resolved. The details in the revised manuscript can be found in lines 180-202, pages 8-9, and Supplementary Information.

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: Thanks for the comments and suggestions! The BBOA concentrations for the summer and autumn harvest were estimated via a unified equation, BBOA = 15.1 $\times$ (m/z 60 – 0.26% $\times$ OA). The details in the revised manuscript can be found in lines 520-539, page 22.

PG9111, L17: “dominate” should be replaced with “dominant”.

Response: Thanks for the suggestion! This sentence has been reworded. The details in the revised manuscript can be found in lines 29-31, 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: Thanks for the suggestion! This concluding sentence has been reworded. The details in the revised manuscript can be found in lines 41-43, page 2.

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

Response: Thanks for the suggestion! 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: Thanks for the comments and suggestions! As mentioned above, the HOA factor that is composed of COA and traffic HOA cannot be separated by a COA factor even if the PMF factors go up more than 3 or 4 factors under this study. Previous studies also concluded that it was very difficult to distinguish HOA and COA factors from the PMF analysis of unit mass resolution organic spectra obtained by Aerodyne Quadrupole AMS(Q-AMS)/ACSM, which is due to the fact that HOA and COA show rather similar spectra (Sun et al., 2010; Sun et al., 2012). However, the PMF analysis of high resolution mass spectra of OA is able to distinguish COA from traffic HOA and cooking-related OA (Huang et al., 2010; Crippa et al., 2013). Unfortunately, we currently don't have such high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) for measuring together. Nevertheless, we have re-evaluated the PMF results together with the multilinear engine (ME-2, Paatero 1999) analysis of the ACSM OA data. The details in the revised manuscript can be found in lines 180-202, pages 8-9, and Supplementary Information.

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

Response: Thanks for the suggestion! The r^2 values have been checked. The details in the revised manuscript can be found in line 364, page 15.

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: Thanks for the suggestion! The 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: Thanks for the constructive comments and suggestions! All the suggestions have been adopted. We also have revised the manuscript accordingly. Firstly, we are sorry for missing the focus of the work and many conclusions. We have re-analyzed our data and reworded the text according to the comments and suggestions. In the revised manuscript, we also did our best to analyze the data in order to explore new findings and improve the quality of the manuscript. Moreover, we have re-plotted some figures to better distinguish valuable information. The details in the revised manuscript can be found in lines 29-43, page 2; lines 279-306, pages 12-13; lines 498-510, pages 20-12; lines 592-634, pages 25-26. Secondly, we agree that the estimated BBOA loadings have to be calculated based on the results from the PMF analysis. Accordingly, this text has been reworded. The details in the revised manuscript can be found in lines 516-539, pages 20-21. Additionally, we agree that the presented data show a relatively weak analysis in the ACPD version. Thus, we made a major revision for the manuscript. For example, the contribution of the harvest-related emissions (i.e., biomass burning emissions) to local air quality was investigated. The details in the revised manuscript can be found in lines 453-493, pages 19-20; The PMF results were re-analyzed and a better PMF-solution with a 4-factor solution including a new factor (oxidized biomass burning-influenced OA, OOA-BB) was selected. The details in the revised manuscript can be found in lines 401-450, pages 18-19. Based on the comments and suggestions, more details in our revision are shown below point by point, please check them!

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: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in lines 29-31, page 2.

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: Thanks for the comment! The sentence has been removed.

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

Response: Thanks for the comment! The sentence has been reworded. The details in the revised manuscript can be found in lines 41-43, page 2.

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

Response: Thanks for the comment! This sentence has been reworded. 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: Thanks for the suggestion! This sentence has been reworded. 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 …"

Response: Thanks for the suggestions! This sentence has been reworded. The details in the revised manuscript can be found in lines 70-73, pages 3-4.

P9113L7: Replace "appointments" by "apportionment"

Response: Thanks for the suggestions! The term "appointments" has been replaced by "apportionment". The details in the revised manuscript can be found in line 81, page 4.

P9113L11: Replace "predict" by "determine"

Response: Thanks for the suggestions! This sentence has been removed, according to the next comment. The details in the revised manuscript can be found in lines 82-93, page 4.

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

Response: Sorry for the confusion! This text has been reworded. The details in the revised manuscript can be found in lines 82-93, page 4.

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

Response: Sorry for the confusion! This sentence has been removed.

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: Thanks for the comments! We agree with the referee's viewpoint. This sentence has been removed.

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: Sorry for the confusion! We have added some detailed information into the introduction according to the suggestions. The details in the revised manuscript can be found in lines 59-61, page 3.

In recent years, with the decreasing use of agricultural residues as a renewable fuel in the rural YRD region, lots of farmers in China harvest the agricultural crops during the daytime and then directly burn the agricultural residues during the nighttime and/or evening in the fields. As we know, the burning conditions, namely flaming phase and smoldering phase, can play an important role in the field biomass burning events. This is because that the agricultural residues generally are mixed with more or less dry materials. Typical burning events of agricultural residues in the field of China are shown in Figure R1.

Figure R1. Pictures for typical burning events of agricultural residues 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.

Response: Thanks for the comments and suggestions! In this study, 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. Therefore, the air quality could be affected by the local traffic and cooking-emission sources. In addition, there are many fire locations around Nanjing (Figure S1), in particular for the western areas of Nanjing. As shown in Figure S1, we also found that the biomass burning sources present characteristic area pollution, implying that the air quality of Nanjing might be affected by the regional biomass burning sources when air masses and/or wind direction are from the main fire locations. It is approximately dozens of kilometers and/or hundreds of kilometers from the main fire locations areas to urban Nanjing (Figure S1), implying that the urban Nanjing site is significantly influenced by the biomass burning plumes originated from the rural areas and undergone the aging in BB plumes measured dozens or hundreds of kilometers away from the fire locations, but little/negligibly influenced by the local biomass burning. Therefore, there could be more aged/secondary biomass burning-related OA than the fresh biomass burning-related OA in the biomass burning plumes. The details in the revised manuscript can be found in lines 103-119, page 5.

We agree that cooking-related sources can play a significant role in aerosol pollution, in particular for urban areas with many restaurants and resident areas around sampling sites. As a matter of fact, we also realized that COA should be identified because of the presence of potential cooking related sources near the sampling site. In this study, however, the traffic HOA was mixed. Therefore, it is difficult to distinguish the COA and traffic HOA via PMF analysis with the ACSM OA mass spectrum, which is due to the fact that the mass spectra of COA and traffic HOA are highly similar. Previous studies by Sun et al. (2010) and Sun et al. (2012) also found that COA cannot be separated from traffic HOA in urban areas via PMF analysis with the Q-AMS or ACSM OA mass spectrum.

Some studies addressed that 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 (Mohr et al., 2012; Sun et al., 2011, 2012). In this study, however, it was observed that f_{55} is not related with increased f_{57} for both the summer and autumn harvest (Figure R2). We also tried to use the multilinear engine (ME-2, Paatero 1999) with the newly developed source finder (SoFi, Canonaco et al., 2013) to distinguish the contributions of traffic and cooking sources. However, the COA and traffic HOA were not distinguished as well. Moreover, other related techniques, including constraining a traffic, a cooking and a biomass burning factor profiles from other PMF studies (Crippa et al., 2013, Ng et al., 2011), increasing the number of factors, and reweighting the *m/z* 57 variable (mostly due to the fact that traffic is difficult to be extracted), have also been performed to extract cleaner source. 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 the traffic and cooking factor profiles directly from the data and to employ them as constraints in subsequent runs through the period. However, the results are not satisfactory because a mixing behavior was always observed, in particular for the diurnal cycle. All in all, to our knowledge it is not possible to obtain a reasonable and well-separated representation of HOA and COA under this study. Therefore, we decided to keep the two sources with a single factor, and then labeled it as a mixed factor of HOA and COA, i.e., HOA + COA. The details in the revised manuscript can be found in lines 181-202, pages 8-9.

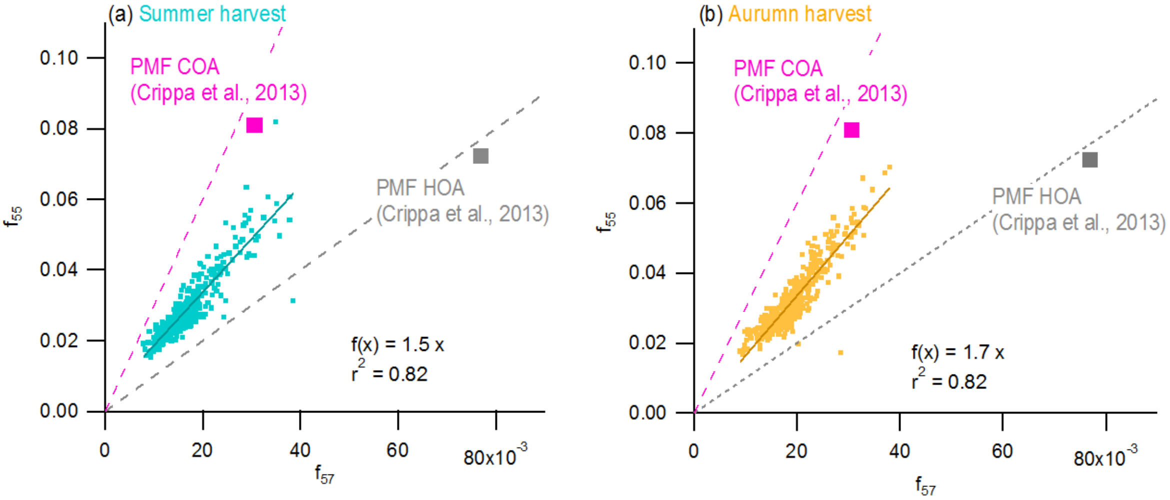

Figure R2. Scatter plots of f_{55} vs. f_{57}. The results between f_{55} and f_{57} during the two harvests highly differ from previous reports in an Aerodyne High Resolution Time-of-Flight AMS (HR-ToF-MAS) studies (Mohr et al., 2012; Xu et al., 2014).

HOA + COA also shows high concentrations during the cooking time (noon and early night, please see Figure 3 in the revised manuscript). In addition to the BB-related source influence, the HOA + COA might contribute a major fraction to OA concentrations (> 40% for the summer harvest and > 50% for the autumn harvest) in the nighttime (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). The details in the revised manuscript can be found in lines 310-314, page 13; lines 463-470, pages 19-20.

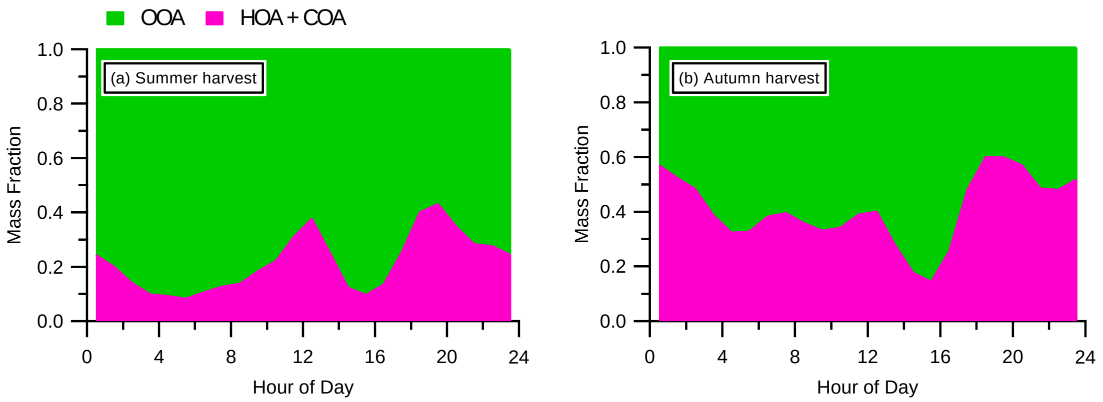

Figure R3. Diurnal variations of the mass fractional contributions of HOA + COA and OOA, excepting the biomass burning related influence, during the summer and autumn harvest respectively.

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: Thanks for the comments! In this study, the measurements of K^+ and BC were performed for $PM_{2.5}$. A previous study also found that 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. (2012) in South China. Therefore, the BC mass in the 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. The details in the revised manuscript can be found in lines 431-437, page 18.

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: Sorry for the confusion! 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). The MODIS can present fire distributions in details at 1 km resolution through Fire Information for Resource Management System (FIRMS) on global scale. The details in the revised manuscript can be found in lines 142-144, page 6.

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?

Response: Thanks for the 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. This is because that 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., 2011), where $I_{s,i}$ is in units of ions/species (s), and IE_s is in units of ions/molecule. However, the ACSM cannot be directly used to measure the single particles due to the slower detection electronics, i.e., no capability to time resolve single ions (Ng et al., 2011). Thus, the calibration of the ACSM is based on the determination of an instrument response factor (RF), using NH_4NO_3 calibration aerosol. In this study, 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. 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. (2011). 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., 2011; Sun et al., 2012). As shown in Figure R4, the results show that the RF_{NO3} is 3.96×10^{-11}. In addition, our ACSM was mainly used for the long-term and real-time online measuring the PM_1 species. This is because that the Aerodyne ACSM can be unattended and continuously for a long observation. Nevertheless, we calibrated the used ACSM around every half year at the superstation in this sampling site. The details in the revised manuscript can be found in lines 154-164, page 7.

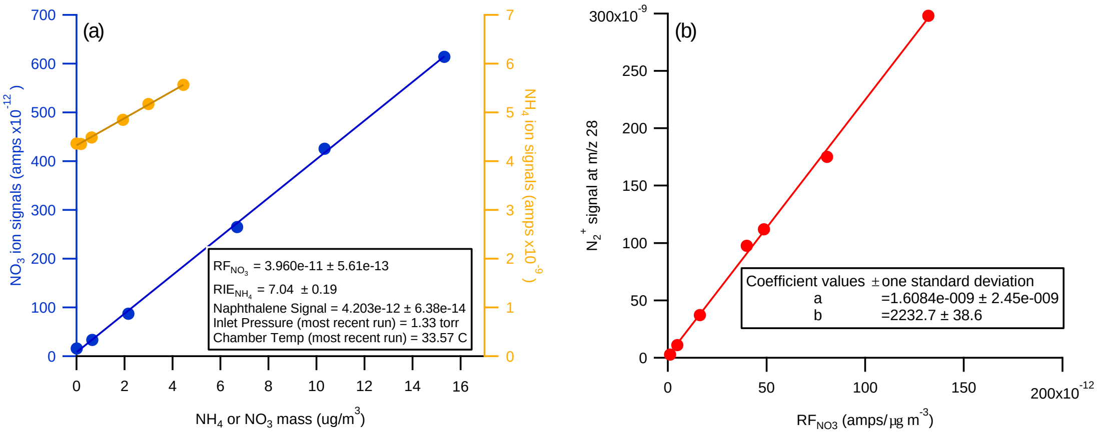

Figure R4. The results of (a) the RF_{NO3} and RIE_{NH4} calibration and (b) the relationship between N_2^+ signal at *m/z* 28 and RF_{NO3} in this study.

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: Thanks for the comments and suggestions! Indeed, the typical temporal trends of variables are very important and interesting. Thus, we have added some relevant discussions into the revised manuscript. The details in the revised manuscript can be found in lines 245-267, page 11.

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: Thanks for the comments and suggestions! The 3 peaks in this study are reasonable. More details for those peaks have been explained in text (please see section 3.1.1 in the revised manuscript). 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, all the three peaks can be reasonably explained. Despite this, the 3 peaks have been 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 11.

P9118L22ff: How are average wind directions calculated? According to Figure S2 in autumn the majority of wind directions were from north (345º-15º). 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 (180º), 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: Thanks for the comments and suggestions! This text has been removed.

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: Sorry for confusion! We also agree 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. We have revised the text. 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: Sorry for confusion! We also agree 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.

Response: Thanks for the comments and suggestions! The variations of boundary layer height play a significant role in the dilution of pollutants, particular for the diurnal variations of pollutants. Therefore, we have added some information about boundary layer height into the text. The details in the revised manuscript can be found in lines 292-293, page 12; lines 305-306, page 13; lines 429-430, page 18; lines 464-470, pages 19-20.

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.

Response: Thanks for the comments and suggestions! As a matter of fact, we also realized that COA should be identified because of the presence of potential cooking related sources near the sampling site. In this study, however, the traffic HOA was mixed. Therefore, it is difficult to distinguish the COA and traffic HOA via PMF analysis with the ACSM OA mass spectrum, which is due to the fact that the mass spectra of COA and traffic HOA are highly similar. Previous studies by Sun et al. (2010) and Sun et al. (2012) also found that COA cannot be separated from traffic HOA in urban areas via PMF analysis with the Q-AMS or ACSM OA mass spectrum.

Some studies addressed that 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 (Mohr et al., 2012; Sun et al., 2011, 2012). In this study, however, it was observed that f_{55} is not related with increased f_{57} for both the summer and autumn harvest (Figure R2). We also tried to use the multilinear engine (ME-2, Paatero 1999) with the newly developed source finder (SoFi, Canonaco et al., 2013) to distinguish the contributions of traffic and cooking sources. However, the COA and traffic HOA were not distinguished as well. Moreover, other related techniques, including constraining a traffic, a cooking and a biomass burning factor profiles from other PMF studies (Crippa et al., 2013, Ng et al., 2011), increasing the number of factors, and reweighting the *m/z* 57 variable (mostly due to the fact that traffic is difficult to be extracted), have also been performed to extract cleaner source. 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 the traffic and cooking factor profiles directly from the data and to employ them as constraints in subsequent runs through the period. However, the results are not satisfactory because a mixing behavior was always observed, in particular for the diurnal cycle. All in all, to our knowledge it is not possible to obtain a reasonable and well-separated representation of HOA and COA under this study. Therefore, we decided to keep the two sources with a single factor, and then labeled it as a mixed factor of HOA and COA, i.e., HOA + COA. The details in the revised manuscript can be found in lines 181-202, pages 8-9.

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: Thanks for the comments and suggestions! The text has been reworded. The details in the revised manuscript can be found in lines 309-314, page 13.

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

Response: Thanks for the comments and suggestions! The term "used" has been added between "can be" and "as a diagnostics". The details in the revised manuscript can be found in line 327, page 14.

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

Response: Thanks for the comments and suggestions! The term "correlated" has been changed to "consistent". The details in the revised manuscript can be found in line 364, page 15.

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

Response: Thanks for the suggestion! The r^2 value has been changed accordingly. The details in the revised manuscript can be found in line 364, page 15.

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

Response: Thanks for the comments and suggestions! "BB activities" has been changed to "BB emissions". The details in the revised manuscript can be found in line 367, page 15.

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

Response: Thanks for the comments and suggestions! Accordingly, we have determined whether there is a wind direction dependence of the concentrations of the different OA types in the revised manuscript. For example, it has been observed that the high HOA + COA concentration (> 6 μg m^{-3}) occurred when WD was from southeast during the summer harvest (Figure 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 (Figure S4). This is a good evidence for explaining the regional pollution of OOA in the YRD region during the harvest seasons. The details in the revised manuscript can be found in lines 335-339, page 14; lines 446-450, page 19.

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

Response: Thanks for the comments and suggestions! 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?

Response: Thanks for the comments and suggestions! 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 original Figure 5 have been replaced by a new one, please see Figure 7 in the revised manuscript. In addition, the average mass concentrations of PM_1 species and OA components were also presented in Table S2 (Please see supplementary information) during the different BB periods, i.e., the low BB (L-BB), medium BB (M-BB), and high BB (H-BB). The secondary inorganic aerosols (including SO_4, NO_3, and NH_4) can be seen in lower fraction in the H-BB period than in the L-BB period. However, the mass concentrations of SO_4, NO_3, and NH_4 are higher in the H-BB period than in the L-BB period (Table S2). These findings indicate that BB contributes more to organics than that to the secondary inorganic aerosols in transported pollution air masses. The peaks of HOA+COA that are consistent with the peaks of BBOA at the same scale were observed at night. This means that both local primary sources (including traffic and cooking emissions) and regional biomass burning plumes contribute to the total OA mass during the nighttime. Thus, HOA and COA show an increase during the biomass burning periods. In addition, OOA also showed an increase during the biomass burning periods, which is due to the fact that additional OOA component can be formed via the oxidation of BBOA precursors in the biomass burning plumes (Aiken et al., 2009; DeCarlo et al., 2010). The details in the revised manuscript can be found in lines 463-493, pages 19-20.

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

Response: Thanks for the comments and suggestions! 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.

Response: Thanks for the comments and suggestions! In this study, the secondary species (including nitrate, sulfate, ammonium and OOA) increase linearly with the increase of the PM_1 concentrations. As shown in Figure 1d, the nitrate, sulfate, ammonium and total OA contribute the largest fraction to the total PM_1 loadings during the whole study period. Moreover, the secondary OA accounts the largest fraction to the total OA concentrations (Figure 7). This means that the secondary species dominate the total PM_1 loadings during the whole study period. We have revised this text accordingly. The details in the revised manuscript can be found in lines 494-510, 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: Thanks for the 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 graphs (Figure 8a - b). The details in the revised manuscript can be found in lines 494-510, 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 μg/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: Thanks for the comments! We agree with the referee. The related text has been removed and reworded accordingly. The original Figure 6 has also been replaced by a new one, please see Figure 8 of the revised manuscript. The details in the revised manuscript can be found in lines 494-510, 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: Thanks for the comments and suggestions! We also agree that the secondary species also play a significant role in total PM_1 including the high biomass burning periods. This text has been revised. The details in the revised manuscript can be found in lines 494-510, 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: Thanks for the comments and suggestions! Accordingly, several high peaks have been presented in Table S1. The details in the revised manuscript can be found in lines 494-510, 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: Thanks for the comments and suggestions! We agree with the referee. The related text has been removed.

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: Thanks for the comments and suggestions! Different boundary layer height can lead to the seasonal variations of POA loadings (Sun et al., 2012, 2013). Generally, the boundary layer height is lower in autumn than that in summer, implying that air pollutants are more difficult to be diluted in autumn, compared with that in summer (Schneider et al., 2002). Therefore, the POA loadings in this study showed higher during the autumn harvest than that during the summer harvest. The related text has been revised with some information of boundary layer height. The details in the revised manuscript can be found in lines 468-470, pages 19-20.

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?

Response: Thanks for the comments and suggestions! 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. The details in the revised manuscript can be found in lines 395-450, pages 17-19.

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.

Response: Thanks for the 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 be also 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. The details in the revised manuscript can be found in line 512, page 21; lines 516-539, pages 21-22.

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?

Response: Thanks for the 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 513-539, pages 21-22.

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: Thanks for the comments and suggestions! We agree that the results of on-line BBOA loadings are dominated by the results from PMF BBOA before (with a slope). In the revised manuscript, the two kinds of BB-related OA (i.e. fresh BBOA and OOA-BB) were identified. As shown in Figure 5b, the strong correlations (r^2 = 0.95, r^2 = 0.98, and r^2 = 0.97) between the fresh BBOA and Δ *m/z* 60 with the similar slopes, i.e., 16.3 for summer, 14.6 for autumn, and 15.1 for the total harvest seasons, were observed. The OOA-BB mass loadings also show the high correlations with Δ *m/z* 60 (r^2 = 0.95 and r^2 = 0.97), but with very different slopes (74.8 and 64.4) during the summer and autumn harvest, respectively (Figure 5b). This is likely due to the fact that the OOA-BB loadings depended on the aging processes of BB pollutants, its mathematically mixing of sources in the BB plumes, and burning conditions (Weimer et al., 2008; DeCarlo et al., 2010; Cubison et al., 2011; Crippa et al., 2013; Bougiatioti et al., 2014). Therefore, the fresh BBOA concentrations for the harvests are estimated as BBOA = 15.1 × (*m/z* 60 – 0.26% × OA). The details in the revised manuscript can be found in lines 524-529, page 22.

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

Response: Thanks for the comments and suggestions! The figures have been rearranged with discussion in the revised manuscript. In order to present the typical temporal trends, the time series of the estimated BBOA and PMF BBOA were also shown in the revised manuscript. The details in the revised manuscript can be found in lines 516-539, pages 21-22.

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: Thanks for the suggestion! This sentence has been changed. The details in the revised manuscript can be found in lines 542-544, pages 22-23.

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

Response: Thanks for the comment! This sentence has been reworded. The details in the revised manuscript can be found in lines 542-544, pages 22-23.

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

Response: Sorry for the confusion! 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: Sorry for the confusion! The related text has been removed.

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

Response: Thanks for the comments and suggestions! This sentence has been removed. An explanation for the mixing and/or aging processes has also been added into the revised manuscript. The details in the revised manuscript can be found in lines 542-560, pages 22-23.

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

Response: Thanks for the comments and suggestions! The paragraph has been reworded. The details in the revised manuscript can be found in lines 561-574, pages 23-24.

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: Thanks for the suggestion! We agree with the referee. The related information on O_x has been removed in this study. The details in the revised manuscript can be found in lines 561-574, pages 23-24.

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: Thanks for the constructive comments and suggestions! The "BB campaigns" has been changed to "BB emissions"; "photochemical activity campaign" has been removed for clarity.

As shown in Figure 11, the (BBOA + OOA-BB) / ΔCO ratios showed an obvious decrease with the increasing of f_{44} values during the two harvests in the absence of traffic-like plumes, implying that BBOA might be mixed with OOA and/or other pollutants in the different air masses and/or BB-related plumes. This is likely due to a synergistic effect of the rapid formation of OOA from BB plumes and the mixing of BBOA with regional OOA and/or CO. The details in the revised manuscript can be found in lines 542-569, pages 22-23; lines 575-582, page 24.

P9128L10-11: I do not see how this is the consequence of the previous sentence. The logical cause-effect relationship is missing.

Response: Thanks for the comments and suggestions! The original plots have been replaced by two new plots which present the logical cause-effect relationships. The details in the revised manuscript can be found in Figure 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: Sorry for the confusion! 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 changes in total spatial variance during the summer and autumn harvest, 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. The details in the revised manuscript can be found in lines 592-611, pages 24-25.

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: Sorry for the confusion! The original Figure S5 has been replaced by a new Figure S4, which presents a relationship between the OA components and wind direction. In addition, we have also revised the text. The details in the revised manuscript can be found in lines 370-373, page 14; lines 622-625, page 26.

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: Thanks for comments! We have revised the statement and conclusion. The details in the revised manuscript can be found in lines 612-620, pages 25-26.

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: Sorry for the confusion! We agree the referee. The related text has been reworded for clarify. The details in the revised manuscript can be found in lines 620-634, page 26.

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: Thanks for the suggestion! This sentence has been removed.

P9131L4: Jiangsu province was not mentioned before. Please introduce first in the main text.
Response: Thanks for the suggestion! 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: Thanks for the suggestion! The time scale has been changed. Please see the Figure 2 and Figure 3 in the revised manuscript, lines 1028-1039, pages 45-46.

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: Sorry for the confusion! We have revised the Figure 3 accordingly. Please see the Figure 3 in the revised manuscript, lines 1034-1039, page 46.

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?

Response: Sorry for the confusion! Yes, the fits are forced through zero. According to the suggestion, we have added "PMF BBOA = 1.0 × estimated BBOA" into the graph. Please see the Figure 10 in the revised manuscript, lines 1076-1079, page 53.

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

Response: Thanks for suggestion! We have revised the graphs accordingly. Please see the Figure 11 in the revised manuscript, lines 1081-1084, page 54.

Figure 12a: What are the blue dots? What are the green and red symbols?

Response: Sorry for the confusion! The blue dots colored by the O_x loadings were data points for the plot of f_{44} vs. f_{43}, and the green and red symbols were the f_{44} vs. f_{43} of the PMF-resolved factor (OOA and HOA, respectively) mass spectra in the original Figure 12a. The details can be found in the revised figure caption of Figure 12, please see lines 1087-1091, page 55.

References

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.

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., He, L.-Y., Hu, M., Canagaratna, M. R., Kroll, J. H., Ng, N. L., Zhang, Y.-H., Lin, Y., Xue, L., Sun, T.-L., Liu, X.-G., Shao, M., Jayne, J. T., and Worsnop, D. R.: Characterization of submicron aerosols at a rural site in Pearl River Delta of China using an Aerodyne High-Resolution Aerosol Mass Spectrometer, Atmos. Chem. Phys., 11, 1865-1877, doi:10.5194/acp-11-1865-2011, 2011.

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., 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, 2011.

Schneider, J. and Eixmann, R.: Three years of routine Raman lidar measurements of tropospheric aerosols: Backscattering, extinction, and residual layer height, Atmos. Chem. Phys., 2, 313-323, doi:10.5194/acp-2-313-2002, 2002.

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.

Sun, Y. L., Wang, Z. F., Fu, P. Q., Yang, T., Jiang, Q., Dong, H. B., Li, J., and Jia, J. J.: Aerosol composition, sources and processes during wintertime in Beijing, China, Atmos. Chem. Phys., 13, 4577-4592, doi:10.5194/acp-13-4577-2013, 2013.

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.