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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 accordingly. The detailed responses to the comments and suggestions are shown below point by point, and a marked-up manuscript version in blue is provided.

Thank you very much for your considerations all the time!

Sincerely yours,
Dr. Lili Tang

Response to the Editor's comments and suggestions

Comments to the Author:
The comments of the three referees should be taken into consideration for preparing a revised version. Besides, I have a number of comments and corrections myself, which have to be addressed.

Main text:
Line 41: Replace "masses back-trajectory" by "mass back-trajectories".
Line 47: "Ramanathan et al., 2001" is not in the Reference list.
Lines 89-91: This sentence is unclear; it needs to be rephrased.
Lines 113-114: There is something wrong with this sentence; it needs to be rephrased.
Line 189: Replace "is difficult to distinguish the" by "has difficulties distinguishing the".
Line 198: Replace "found the highly" by " found highly".
Line 222: Replace "were shown" by "are shown".
Line 300: Replace "As previous studies" by "As in previous studies".
Line 311: Replace "were presented" by "are presented".

Line 318: Replace “from mass spectrum were characterized as the components” by “ from the mass spectrum were characterized as components”.

Line 368: POA has not been defined; acronyms and abbreviations should be defined (written full-out) when first used.

Line 384: Replace “As shown in Figure 4d, the” by “In Figure 4d the”.

Line 428: Replace “as it will be” by “as they will be”.

Line 430: Replace “shows the relatively” by “ shows relatively”.

Line 442: Replace “filed” by “field”.

Line 444: “Lipsky et al., 2006” is not in the Reference list; there is “Lipsky and Robinson, 2006” in that list, to which no reference is made within the text.

Line 456: Replace “Shanghai), suburban/remote” by “Shanghai) and suburban/remote”.

Lines 506-509: This sentence is unclear; it needs to be rephrased.

Line 623: Replace “account the” by “provide”.

Line 629: It is unclear what “the others” denote.

Line 671: Replace “Air masses” by “Air mass”.

Line 793: Titles of journal articles should be in lower case and not in title case.

Lines 955-959: There is not referred to this reference within the text.

Supplementary Information:

Figure S6a: Replace in the abscissa “Number of factor” by “Number of factors”.

Figure S11a: Replace in the abscissa “Number of factor” by “Number of factors”.

Figure S11, second line of caption: Replace “summer harvest” by “autumn harvest”.

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

Response to the Reviewers' comments and suggestions

Referee #2

-P8, lines192-195: this sentence is completely pointless. Rephrase or delete.

Response: Thanks for the comments and suggestion! This sentence has been deleted.

-P9, lines 197-199: sentence definitely not clear. Rephrase or delete.

Response: Thanks for the comments and suggestion! This sentence has been deleted.

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 autumn 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 $PM_{2.5}$.

The authors discuss characteristics of the ambient aerosol measured during the two periods with a focus on the biomass burning contribution to ambient PM pollution. Diurnal patterns, individual pollution events and contributions from different source regions are discussed. PMF is used to separate four different types of organic aerosol: oxygenated organic aerosol (aged secondary organic aerosol), biomass-burning oxygenated organic aerosol (likely secondary organic aerosol from biomass burning-related precursors), biomass burning organic aerosol (primary organic aerosol from biomass burning), and hydrocarbon-like organic aerosol from traffic and cooking activities. Also for these organic aerosol types diurnal patterns and source regions are discussed to obtain a more complete picture of the harvest-related biomass burning contribution to ambient PM pollution during the harvest times.

This is the second version of the manuscript that was submitted to ACPD which includes major changes compared to the first version. The authors improved the manuscript significantly regarding the two major points in my review for the first version: The current version of the manuscript is much more focused compared to the first version and many conclusions that were not based on the presented data have been changed or removed. Even though the authors focused their work much more than in the previous version, this manuscript is still quite long, especially with a very

large number of figures. In my first review I wrote that the manuscript has 13 Figures with additional 5 Figures in the supplementary information which were also referenced in the text. Now the manuscript has 13 Figures with 15 (!) additional Figures in the supplement.

Most of the comments in the first review have been answered and treaded satisfactorily. Nevertheless, there are still many shortcomings of the manuscript in many locations of the text. However, these issues are much less serious compared to the previous version of the manuscript. Since the manuscript is now much better focused and since generally the data are well analyzed and the results are well founded on the data, I think that it can be published in ACP with only minor (although not really few) revisions after the detailed comments below are taken into account. Generally, I suggest that the authors ask a native English speaker to proofread the text. There are still many sections in the text that are confusing.

Response: Thanks for the constructive comments and suggestions! All the suggestions have been adopted. We also have revised the manuscript accordingly.

Detailed comments:

P1L4-5: While the content of the manuscript did not change a lot and mainly PMF has been re-done, ending up with a different selection of factors, the authors list grew by two persons from PSI, Switzerland. Such a contribution to one detail of the content of a manuscript could also be honored in the acknowledgements.

Response: Thanks for the comments! The two new authors not only re-evaluated the PMF results, but also provided many constructive suggestions in improving the contents and help us modify the manuscript during the revision stage. Thus, we invited the two authors as co-authors. They all agree on this.

P2L29: According to the authors Nanjing has 8 million inhabitants. This is below the threshold of 10 million above which a city is called “megacity”. (also: P5L103)

Response: Thanks for the comments! The “megacity” has been removed.

P2L35: OOA-BB does not dominate 80% of total BBOA but contributes 80% to total BBOA.

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in line 35, page 2.

P2L37-38: High RH and low *T* do not facilitate the formation of semi-volatile species (this is a chemical process) but supports the partitioning of such species into the particle phase.

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in lines 37-38, page 2.

P2L40-41: "The OA mass decreases with the aging of BB plumes, indicating that the fresh BB plumes contribute to the OA burden significantly." How is this conclusion an inevitable result of these findings?

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in lines 40-43, page 2.

P2L43: Change "… high BB pollutants are linked …" into: "… high BB pollutant concentrations are linked …" Not only BB pollutants are linked to these source areas, also other pollutant concentrations are increased for air masses from these continental source areas (as expected) compared to marine source areas. This is not necessarily a result of biomass burning.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 44, page 2.

P3L58: Change "… a large number of agricultural crop residue …" into "… a large amount of agricultural crop residue …"

Response: Thanks for the suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 60, page 3.

P3L61: Change "… which may result in BB emission." into "… which results in BB emissions."

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 63, page 3.

P3L61-63: "The understanding of the compositions, sources and processes of atmospheric aerosol particles during harvest seasons is urgently needed to design measures to improve the quality of air in China." It probably would be much more efficient to simply reduce biomass burning by the farmers on the fields in order to

improve air quality.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 65, page 3.

P4L80-81: "However, limited information is available for estimating the source apportionment of BBOA." What is the information of this sentence?

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in lines 82-83, page 4.

P5L98-99: "source" and "origin" is twice almost the same.

Response: Thanks for the comments! The term of "source" has been removed.

P5L112ff: These sentences need some major rewording. In addition: There are no agricultural fields in the urban Nanjing area. This means that all BB emissions are transported "dozens to hundreds of kilometers" before reaching the measurement site. With the average wind speed of 2.5 m/s this means that the transport time is in the order of 11 hours (for one hundred km). So a significant delay in the maxima of pollutants from BB burning would be expected compared to the times when the fires are burning (during night). This is not observed in the diurnal patterns.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in lines 115-117, page 5.

P6L127: Use "l min-1" instead of "L min-1" (also line 135) and use "cm3" instead of "cc".

Response: Thanks for the suggestion! The related text has been revised. The details in the revised manuscript can be found in lines 130 and 138, page 6.

P6L129: Is the mass spectrometer really scanned with a rate of 500 ms amu^{-1}? This means that a single scan takes 70 seconds.

Response: Thanks for the comments! Yes, the mass spectrometer really scanned with a rate of 500 ms amu^{-1} during the whole experiment. Actually, it needs 12 scans (~15 min) in every measurement, i.e., 6 times for the closed model and open model, respectively.

P6L135-138: Be consistent in how to name instruments. In line 135 you write "The MET ONE BAM-1020" while in line 138 you write "… a gas analyzer (Thermo Scientific, Model 48i)."

Response: Sorry for the confusion! The MET ONE BAM-1020 was employed to measure PM_1. CO was measured using a gas analyzer (Thermo Scientific, Model 48i).

P6L140: What is a ground meteorology station?

Response: Sorry for the confusion! The "ground meteorology station" is the term of the place in which we obtained meteorology data including temperature, relative humidity, wind speed, and wind direction. The details in the revised manuscript can be found in line 143, page 6.

P7L157-158: I assume NH_4NO_3 particles are not used for "quantitative measurements" but for the calibration of the instrument.

Response: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in line 161, page 7.

P7L162: Explain what "response factor" is.

Response: Sorry for the confusion! Response factor (RF) measured as a unit of amps of signal per μg m^{-3} of sampled aerosol (NH_4NO_3) was applied to calibrate the ACSM under this study. 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 described by Ng et al. (2011).

P8L177-178: What do you mean with the sentence "Furthermore, the OA …"?

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

P8L180: PMF was not restricted to *m/z* 120 but "up to *m/z* 120" or "to *m/z* 10 – 120".

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in line 182, page 8.

P8L182-183: Fpeaks were not varied in steps of 0.1 during the summer and autumn harvest but fpeaks were varied in steps of 0.1 for the data of the summer and autumn

harvest.

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in line 185, page 8.

P9L197: How can the 4-factor solution be "valid"? PMF cannot separate different types of organic aerosol that in reality exist. PMF can only be used to describe the organic aerosol using different factors, i.e. types of organic aerosols. These factors are never "valid", they can just describe the total aerosol better or worse. Therefore the PMF solution of the previous version of the manuscript was not "wrong" or "invalid" and this solution is now not "valid", but this solution describes the total OA better than the previous one and provides more information.

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in line 197, page 9.

P9L208: Why were back trajectories calculated for 500 m arrival height while the measurements were performed at 18 m above ground level? Are there significant differences in back trajectories calculated for ground level and for 500 m?

Response: Thanks for the comments! We also calculated back trajectories for 18 m. Results show that there are not significant differences in back trajectories calculated for ground level and for 500 m. This arrival height (i.e. 500 m) was also used in many previous studies (e.g. Sun et al., 2010; Huang et al., 2012; Sun et al., 2012).

P10L227-230: Why may PM_1 be overestimated due to the uncertainties in the conversion of the measured signals into mass concentrations? It also could be underestimated.

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

P10L233-236 and L238-240: The first of these two sections is in contradiction to the second one. According to the second of these sections OA is similar during both harvest seasons while for the other species except nitrate during summer the fractions are smaller. According to the first of these sections all species are similar for both harvest seasons with the exception of BC, for which during autumn harvest a larger fraction was found.

Response: Thanks for the comments and suggestion! The sentence has been reworded.

The details in the revised manuscript can be found in line 237, page 10.

P10L240f: What is consistent with the findings in these other papers? The dominance of OA in ambient PM_1 is consistent all over the world (e.g. Jimenez et al., Science, 2009).

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

P11L246ff: Wind direction is also an important meteorological factor affecting PM. This should be named not only in line 246 but also in line 249 as an important factor that can cause dramatic changes in PM load. What is "dilution of the atmosphere" (line 249)?

Response: Sorry for the confusion! We agree with the referee. The sentence has been reworded. The details in the revised manuscript can be found in line 242, page 10.

P11L256-262: The explanation provided for case 1 provides little evidence for such a sharp, short and intense peak as observed in the data.

Response: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in lines 252-255, page 11.

P11L262-265: Again this explanation fails in explaining what causes these sharp and intense peaks.

Response: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in lines 255-257, page 11.

P11L265-267: How is this a conclusion from the previous sentences? There is no reasonable explanation for the sharp and intense peaks. There is also no reasonable explanation what biomass burning has to do with these peaks.

Response: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in lines 252-259, page 11.

P12L275: Check the times of the peaks in the diurnal patterns (also P13L299-300). How can the OA peak around 19:00-22:00 be "in agreement with the emission behaviors of pollution sources, i.e. (…) BB emissions"? BB emissions are during the night. Transport times for the average wind speed and the typical distance of the

sources is in the order of 10 hours or more. So they should arrive not before the early morning.

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in line 269, page 11.

P12L279 and L282: Sulfate does not show any significant diurnal trend and not a "weaker diurnal variation" (L279). Therefore a "similar diurnal trend" (L282) does not make sense.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 272, page 12.

P12L282-285: The lower nitrate concentrations during the day and higher values during the night is not necessarily a harvest-related feature. This is probably due to the partitioning of nitrate between gas- and particle phase and is related to RH and T.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in lines 280-281, page 12.

P13L298-306: For the identification of the BB contribution to the ambient PM burden and to separate it from the contribution from other sources it would be very helpful to have diurnal patterns for the non-harvest times between the two harvest seasons. As it is right now the discussion on the BB contribution is to a large degree speculation – especially if one takes into account the large travel times (and related unknown delays in arrival time) BB emissions need to reach the sampling site.

Response: Thanks for the comments and suggestion! Unfortunately, we currently don't have black carbon data for the non-harvest period (July 1 to 8). In addition, it is difficult to accurately assess the travel times of BB plumes from fire locations to the sampling site. This is mainly due to agricultural fires distributed at many locations (Figure S1). The related text has been modified. The details in the revised manuscript can be found in lines 126-127, page 6.

P14L335-339: This is a strange result. Does this mean that the location of the main traffic and cooking sources changed from summer until autumn?

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

P14L343: How do you know that *m/z* 60 is almost all $C_2H_4O_2^+$ without any high resolution mass spectra?

Response: Thanks for the comments! The related text, i.e., (almost all $C_2H_4O_2^+$), has been removed.

P15L345-356: Check the language of this paragraph.

Response: Thanks for the suggestion! The language of this paragraph has been checked.

P15L359: Change "… this factor might be fresh/primary BBOA …" into "… this factor might be associated with fresh/primary BBOA …". You cannot identify a PMF factor with a certain type of organic aerosol, it can only be a reasonable representation of OA.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 346, page 14.

P15L364: Do you have an explanation why the correlation between BBOA and K^+ is so different during the two harvest seasons?

Response: Thanks for the comments and suggestion! As shown in Fig. 5, two relatively high peaks (i.e. at ~19:00 on 23 and 26 October) of BBOA in the autumn may affect the correlation between BBOA and K^+. The correlation shows better after the two peaks of BBOA are removed (Fig. 6). The two high peaks of BBOA may be affected by local plumes with very high pollutants sourced from cooking activities. He et al. (2010) also found that very strong cooking emission (e .g. ~100 μg m^{-3}) can contribute to *m/z* 60 in AMS measurement. Therefore, several BBOA peaks have been removed for fitting. The details in the revised manuscript can be found in line 350, page 15.

P15L365: Add "(Fig. 3)" between "… at nighttime" and "which is …".

Response: Thanks for the suggestion! The related text has been added. The details in the revised manuscript can be found in line 352, page 15.

P16L372: "means" seems too strong. I suggest replacing with "suggests".

Response: Thanks for the comments and suggestion! The related text has been

replaced. The details in the revised manuscript can be found in line 358, page 15.

P16L376: Replace "Fig. 3d" with "Fig. 3c and d"

Response: Thanks for the comments and suggestion! The related text has been replaced. The details in the revised manuscript can be found in line 363, page 15.

P16L379: The statement on m44 is a repetition of a similar statement two lines above.

Response: Thanks for the comments and suggestion! A similar statement has been removed.

P16L384: Replace "As shown in Figure 4d …" with "In Figure 4d …"

Response: Thanks for the comments and suggestion! The related text has been replaced. The details in the revised manuscript can be found in line 371, page 15.

P16L386: Do you think that R^2 of 0.46 describes still a "good correlation"?

Response: Thanks for the comments and suggestion! The term "good" has been removed. The details in the revised manuscript can be found in line 372, page 16.

P16L390&393: What is a "stable variation"?

Response: Sorry for the confusion! The "stable variation" has been replaced by "stable trend". The details in the revised manuscript can be found in line 377, page 16.

P17L395: OOA is not formed by photochemical processing during the daytime only during the harvest seasons but also during other seasons.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in lines 381-382, page 16.

P17L404: Add "and identified" between "… has been resolved" and "as oxidized …"

Response: Thanks for the comments and suggestion! The related text has been added. The details in the revised manuscript can be found in line 390, page 16.

P17L405: What are "oxidized signals"?

Response: Sorry for the confusion! The "oxidized signals" has been replaced by "oxidized ions". The details in the revised manuscript can be found in line 391, page

16.

P17L406-407: Replace "… which correlates well with BB-emission …" with "… which correlate well with those of BB-emission …". Otherwise you state that the m60 correlates with OOA2-BBOA.

Response: Thanks for the comments and suggestions! The related text has been replaced. The details in the revised manuscript can be found in line 393, page 16.

P17L417: Replace "burn phase" with "burning conditions"

Response: Thanks for the comments and suggestions! This sentence has been removed.

P18L419-P19L451: This paragraph is not really well structured and leaves the reader with quite a bit of confusion. Here comparisons of the OOA-BB factor mass spectrum with several other mass spectra from the literature are presented. After reading this text the reader is likely confused about the actual meaning of the OOA-BB factor: It could be BBOA mixed with OOA, it could be aged BBOA, it could be secondary OA from gaseous biomass burning-related precursors, it could be BBOA from smoldering. Either this paragraph is better structured to extract solid information on the (likely) nature of OOA-BB or it is clearly stated that the nature of OOA-BB is unknown.

In addition, there are again several statements within this paragraph which are not well based on the data and are more or less speculation or assumptions. This should be marked clearly. If ozonolysis and NO_3 reactions are assumed to be the processes that generate OOA-BB from precursors it would be helpful if O_3 and NO_3 concentrations or relationships between their concentrations and OOA-BB formation would be presented.

Response: Sorry for the confusion! We agree with the referee. The related text has been revised and/or removed. The details in the revised manuscript can be found in lines 401-424, pages 17-18.

P18L432: The OOA-BB time series strongly correlates with "the time series of "K^+ and Delta *m/z* 60 … (insert "the time series"). Please provide R^2 for these correlations to support the "strong".

Response: Thanks for the comments and suggestions! The R^2 value has been added.

P19L449-450: Change "… high concentration from southeasterly wind … and from northerly wind …" into "… high concentration during times with southeasterly wind … and with northerly wind …" or something like this.

Response: Thanks for the suggestions! The sentence has been removed.

P19L459-460: What were the criteria for the separation? What do the percentages mean?

Response: Thanks for the comments and suggestions! Using the relative contribution of the sum of BBOA and OOA-BB to OA, the harvest season was separated into 3 time periods, i.e., low BB (L-BB, 28% and 29%) period, medium BB (M-BB, 49% and 38%) period, and high BB (H-BB, 93% and 50%) period, during the summer and autumn harvest respectively.

P20L490-493: I suggest changing "contribution" to "fraction".

Response: Thanks for the suggestions! The related text has been changed. The details in the revised manuscript can be found in lines 444, 446, and 447, pages 18-19.

P21L499: I do not agree with this statement. According to Figure 8 the total OA fraction does NOT remain at a stable level: It increases from about 15% to 40% and from 30% to 45% during summer and autumn, respectively.

Response: We agree with referee. The related text has been changed. The details in the revised manuscript can be found in lines 476-477, page 20.

P21L500ff: How many percent is the "significant" increase of OOA-BB and BBOA? Here the impression is given that only an increase of the fraction of a certain particle component with increasing total PM concentration shows a contribution to increased PM. Also components that have constant fractions for all total PM levels contribute to the increase of total PM.

Response: Thanks for the comments and suggestions! The related text has been added. The details in the revised manuscript can be found in lines 479-481, page 20.

P21L515: "The BBOA mass loadings during the harvest season were estimated using a simple method." I suggest inserting "a posteriori" between "estimated" and "using"

to make clear that this was a calculation that was done after the data analysis was finished. However, it would be more honest not to write that BBOA mass loadings were "estimated", but to write that Delta *m/z* 60 concentrations correlate with PMF BBOA. Here this correlation is shown once again, BBOA is not estimated.

Response: Thanks for the comments and suggestions! The related text has been changed. The details in the revised manuscript can be found in lines 495, page 20.

P22L519: Change "background level" into "background fraction" (also L521 and L523: "value"). What is "little/negligible" BB-influence – how is it defined?

Response: Thanks for the comments and suggestions! The little/negligible BB-influence represents the period without significant agricultural burning impacts in July 1 to 8 in this study. The "little/negligible" BB-influence has been defined. The details in the revised manuscript can be found in lines 126-127, page 6.

P22L531: What is "mathematically mixing"? The whole sentence is hard to understand.

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

P22L535-539: If Delta *m/z* 60 correlates with BBOA (as was shown in the text above) it is not surprising that Delta *m/z* 60 multiplied by the ratio of BBOA/Delta *m/z* 60 agrees well with BBOA. This is a trivial result and no Figure is needed to present this. Therefore I suggest removing Figure 10.

Response: Thanks for the comments and suggestions! The Figure 10 has been removed from the manuscript.

P23L545: Explain what Delta CO is and how it was calculated.

Response: Thanks for the comments! The CO background is determined as 14.9 μg m^{-3} for summer harvest and 17.9 μg m^{-3} for autumn harvest, respectively, based on an average of the lowest 5% CO during two plumes (Takegawa et al., 2006).

P23L549f: This ratio can only remove the effect of dilution of the plume in regional air if there is no other CO source and the dilution air is CO-free. Is this the case?

Response: Thanks for the comments! Yes, that is. The sentence has been reworded. The details in the revised manuscript can be found in lines 526-527, page 22.

P23L560: Explain what Delta f_{44} is.

Response: Sorry for the confusion! The related text about Delta f_{44} has been removed.

P23L559-562: Here in two consecutive sentences the same information is given twice. The second sentence is not a consequence of the first one it just repeats the information.

Response: Thanks for the comments! The related text has been removed.

P23L543-562: It is not really clear to me what the reader learns from Figure 11.

Response: Sorry for the confusion! The (BBOA+OOA-BB/CO) decreases as a function of f_{44}, which might indicate that BBOA was oxidized to less volatile OOA, e.g., LV-OOA during the aging process. The details in the revised manuscript can be found in lines 521-536, page 22.

P23L566: What is the "result observed by Crippa et al. (2013)" ?

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

P23L567: HOW does this "further demonstrate(s) that BBOA represents mainly the primary BBOA"? How is this conclusion based on the previous text?

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

P24L571-576: Where is the evidence in the data that BBOA wanders up the triangle when aging?

Response: Sorry for the confusion! With the aging process in the atmosphere, OA clusters within a well-defined triangular region (Fig. 11a). This trend is consistent with some previous studies (e.g. Sun et al., 2012b; Ng et al., 2010, 2011). Moreover, f_{60} value, a marker for BBOA (Cubison et al., 2011), shows a decrease with the aging (the f_{60} color-scale in Fig. 11a).

P24L578-580 and P24L587: While in the first of the two locations in the text it is clearly stated (and I agree with this) that in Figure 12b no evidence for a dominance of either aging or dilution is presented, in the second location (line 587) only "aging" is named as the underlying process. This is not what the data show!

Response: Thanks for the comments and suggestion! The related text has been removed.

P23L543-P24L591: All the results presented here show no clear evidence for aging processes of the BB plume! All the effects observed here can also be explained by dilution of the plume only. This should clearly be stated.

Response: Thanks for the comments and suggestion! The related text has been removed.

P25L594-595: Not back trajectories for every 2 hours are presented in Figure 13 as stated here, but only the cluster back trajectories.

Response: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in line 554, page 23.

P25L606: Are "the total PM_1 loadings" the average PM_1 concentrations for the whole cluster-related time intervals?

Response: Thanks for the comments！ Yes, the average PM_1 concentrations is for the whole cluster-related time intervals.

P25L611: Which "continental-related cluster"? State clearly.

Response: Sorry for the confusion! Actually, the "continental-related cluster" is the continental-related air masses. However, this term of "continental-related cluster" is not clear. The "continental-related cluster" has been changed by "continental-related air masses". The details in the revised manuscript can be found in lines 570-571, page 23.

P25L606-613: This finding is similar to what is found almost everywhere on the world. Ambient measurements are typically impacted by the source region of the air that is measured. How are the large concentrations measured for some of the trajectory clusters related to the fire locations?

Response: Thanks for the comments and suggestion! The WC BTs and SC BTs are mainly related to the fire locations during the summer and autumn harvest, respectively. The average PM_1 loadings are the highest (71.3 μg m^{-3}) for the WC BTs,

which is almost twice higher than that of the lowest (24.4 μg m^{-3}) for the EM BTs during the summer harvest. This suggests that the long-range transported pollutants from southwestern areas can cause the high PM pollution in the YRD region during the summer harvest. Similarly, the highest average concentration of PM_1 (80.9 μg m^{-3}) is associated with the continental-related air masses during the autumn harvest. Therefore, source regions related to the fire locations (Figure S1) are of utmost importance to the high air pollution in the YRD region during the harvest seasons.

P25L616-617: I do not completely agree with this statement: According to Figure 13 BC fractions are not larger in WC compared to the other clusters, OOA-BB and HOA/COA are similar in WC and SEM, HOA/COA is less in WC than in EM.

Response: Thanks for the comments and suggestion! The related text has been removed.

P26L621f: The statement "local sources play an important role in the relatively low PM pollution" sounds quite strange.

Response: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in line 578, page 24.

P26L624-627: I do not understand what is meant here in this paragraph. In addition: did you confuse "chloride" with "nitrate" in the list of aerosol components in parenthesis?

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in line 586, page 24.

P26L630-631: I do not understand this sentence.

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

P26L631-636: The result that marine air masses (EM, NEM) have higher PM concentrations than continental air masses (NC) during autumn is surprising and different to the summer result. This should be discussed.

Response: Thanks for the comments and suggestion! This point has been discussed in the manuscript. The details in the revised manuscript can be found in lines 581-585, page 2.

P27L647-649: The similar diurnal patterns of the secondary species during summer and autumn are probably due to similar chemical processing as stated, but also due to physical processes like gas-particle partitioning which is similar independent of season. I do not think that source emission patterns need to be similar since for this type of aerosol the sources are largely de-coupled from the aerosol measured days later.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 605, page 25.

P27L659-661: I cannot follow the argument here completely. Do you suggest that OOA-BB is oxidized precursor material that is condensed onto primary BB particles? This is quite unclear here. It could also be oxidized primary particles.

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in lines 616, page 25.

P27L662: Change "reflecting their regional pollution" into "reflecting their regional origin"

Response: Thanks for the suggestion! The related text has been changed. The details in the revised manuscript can be found in lines 618 page 25.

P28L667-668: This simple estimate of BBOA using *m/z* 60 was already presented several times. It would be interesting how well this method used here would work in other environments.

Response: We agree with the referee that the simpler method to estimate fresh BBOA loadings would be interesting and how well this method is used here would work in other environments.

P28L671-674: Low pollution levels were associated with marine air masses only during the summer harvest.

Response: Thanks for the comments! The related text has been removed.

Table 1: In this table not the mass concentration of PM_1 is shown but the concentrations of the species and OA types within PM_1.

Response: Thanks for the comments and suggestion! The caption of Table 1 has been changed.

Figure 1: The letters ("a)", "b)", ..) are missing in the Figure. I have the impression the unit for precipitation is not correct. Is it really "mm", not "mm/h"?

Response: Thanks for the comments and suggestion! The related letters has been added in Figure 1. And the "mm/h" has been replaced by "mm".

Figure 3: How well do the mass spectra correlate? R^2-values would be interesting. Are the reference spectra from ACSM or from AMS? Do you have an explanation why the spectra from this work show so much more m44 and m28 compared to the reference spectra? Especially for the spectra in b) the reference spectrum is systematically lower than the measured spectra. How can this be if both spectra are normalized to total signal?

Response: Thanks for the comments and suggestion! Firstly, the PMF-solution, i.e., four factors (HOA+COA, BBOA, OOA-BB, and OOA), is similar to the results at an urban site by Crippa et al. (2013). Particularly, two BB-related components, i.e., BBOA and OOA-BB, were identified, which is also similar to the finding by Crippa et al. (2013). In order to compare the two BB-related components with previous studies, we selected a study by Crippa et al. (2013). R^2-values have been added into the Figure 3. The reference spectra was from High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS). The average f_{44} in this study shows higher value than the results measured HR-ToF-AMS (Crippa et al., 2013). The high *m/z* 44 fraction in ACSM measurements is reasonable, due to the following reasons: (1) the ACSM f_{44} value was only calculated as the fraction of *m/z* 44 in total OA signal (*m/z* 12-120). The contributions of *m/z*'s > 120 were not included because the *m/z*'s > 120 measured using the ACSM have larger uncertainties (Sun et al., 2012). (2) Some ions in the *m/z*'s < 120, such as *m/z* 39 ($C_3H_3^+$) and 14 (CH_2^+), cannot be measured by ACSM (Ng et al., 2011). Similarly, higher f_{44} values were also measured based on ACSM in previous studies (e.g. Canonaco et al., 2013; Bougiatioti et al., 2014; Sun et al., 2014).

Figure 7: All the information on composition in other cities is not discussed in the text. Either discuss this information or remove it from the graph.

Response: Thanks for the comments and suggestion! We have discussed this

information in the manuscript. The details in the revised manuscript can be found in lines 436-442, page 18.

Figure 11: While the fits are probably the best approximations of the data points using this fit function, they do not represent the data and the behavior of the data at all.

Response: Thanks for the comments and suggestion! The fits have been removed.

Figure 12: The yellow scale in panel b) is not very useful since almost all data points have the same color.

Response: Thanks for the comments and suggestion! We have adjusted the color scale in Figure 12b.

Figure 13: The Figure only shows the average (is this correct?) cluster back trajectory. There is no information on the individual trajectories that contribute to the averages. How well do the averages represent all these trajectories within the clusters? Is it possible to visualize this somehow? E.g. instead of showing a single trajectory showing the range of trajectories that is spanned by the individual trajectories within a cluster?

Response: Thanks for the comments and suggestion! The corresponding back trajectory clusters can be broadly classified into principal clusters of air masses based on the spatial distributions (Draxler and Rolph, 2003). The four-cluster solution was found to be the optimum solution according to the change in total spatial variance, with the mean BT of each cluster. In order to visualize, we were using the results of cluster analysis based on the HYSPLIT back-trajectories (BTs). This is because the potential source regions are mainly investigated but no the individual air masses. Therefore, the cluster analysis was performed in this study. This analysis method is consistent with many previous studies (e.g. Wehner et al., 2008; Huang et al., 2012, 2010; Sun et al., 2011; Zhang et al., 2014).

Referee #4

General Comments

1. Overall, the language and grammar could be improved, as I found a number of instances were I could not quite understand the sentence. I have highlighted a number of these below however the authors are advised to thoroughly check the whole manuscript.

Response: Thanks for the comments! The language and grammar for the whole has been improved.

2. My main comment is regarding the manuscript is that the authors appear to not consider local or other sources of biomass burning apart from agricultural residue burning. While the authors state that there is negligible local biomass burning, based on their findings I am not sure that the authors can discount local source influences. I realize that the measurements were taken during summer and autumn and so the contribution from domestic heating would be minimal, however this does not exclude local emissions. The authors identify with PMF a fresh and an oxidized BBOA source factor, and while I am convinced that are two distinct BBOA sources, I am not sure about attributing the source of both of these factors to the agricultural residue fires.

How far away are the harvest fires from the sampling site, as they appear to be a considerable distance from Nanjing in Figure S1? As the authors mention in the manuscript, primary BBOA emissions will rapidly oxidize in the atmosphere and if the fires are a fair distance, would not the BBOA have been mostly oxidized by the time they reach the receptor site? Or to put it another way, how can you 'see' fresh biomass burning aerosols from these agricultural residue fires? Furthermore, as mentioned by the authors in the manuscript, differing fuel, burn conditions and efficiency will affect the chemical composition of primary BBOA. In previous work with aerosol mass spectrometry, the differing fuels and burning conditions was put forward as the reason for finding two biomass burning source factors (e.g. Young et al. 2014). Therefore rather than fresh and aged BBOA from the same source (agricultural fires), could not the BBOA factor be more from local emissions of a different source (with differing fuel, etc.) and the OOA-BB more related to the agricultural fires?

Response: Thanks for the comments and suggestion! As mentioned above, 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 in the fields. Therefore, heavy pollution events frequently occurred in the east China during the harvest seasons, particular in YRD region (Wang et al., 2009; Ding et al., 2013). This area is also one of the most important agricultural bases in China, and the agricultural activities, like intensive seasonally burning activities of agricultural straw in rural areas (Ding et al., 2013). However, there was little agricultural activity and/or biomass burning campaign in urban Nanjing (Ding et al., 2013). In addition, there is no domestic heating in Nanjing. As a matter of fact, it doesn't need domestic heating in summer and/or autumn with relatively high air temperature (24.1 ℃ and 18.1 ℃ for the summer and autumn harvest, respectively). Urban biomass burning emission, however, plays an insignificant role in the contribution to BB-related pollutants in this study. Local emissions contribute to BBOA less in this study. In addition, the background value of f_{60} value was 0.26 % on average in July, implying that agricultural burning showed insignificant impact (Fig. 9).

This further supports that there is no local biomass burning source. The agricultural residue fire is a major biomass burning source affecting air quality in harvest seasons, which has been reported by previous studies (e.g. Li et al., 2007; Wang et al., 2009; Li et al., 2010; Ding et al., 2013; Huang et al., 2013; Cheng et al., 2014). Therefore, those evidences support that both of fresh BBOA and OOA-BB are mainly attributing to the agricultural residue fires in this study. The details in the revised manuscript can be found in lines 115-117, page 5.

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 thousands of meters 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, but little/negligibly influenced by the local biomass burning emissions. The sentence has been reworded. The details in the revised manuscript can be found in lines 115-117, page 5.

As mentioned above, the agricultural residue fire is a major biomass burning source affecting air quality. In addition, there was little biomass burning events in urban areas.

As shown in Figure 3b, the mass spectrum of BBOA extracted in this study shows a prominent peak of *m/z* 60 which is a well-known tracer ion for BB emissions. In addition, the BBOA is also characterized by higher peaks at masses *m/z* 27, 29, 41, 43, 55, 57, 77 and 91 that are indicative of freshly emitted organic aerosol, because fresh *m/z* 43….. *m/z* 57 can be also from BB-related emissions (Aiken et al., 2009; Heringa et al., 2011; Bougiatioti et al., 2014). For example, primary BBOA (P-BBOA) has a significant contribution from a non-oxygenated ion $C_3H_7^+$ at *m/z* 43, but not from an oxygenated ion $C_2H_3O^+$ (*m/z* 43) in smog chamber experiments by Heringa et al. (2011). The BBOA spectrum profiles with the lack of *m/z* 44 signal (CO_2^+) during the summer and autumn harvest show high correlation (r^2 = 0.82 and r^2 = 0.87) with a result in Paris (Crippa et al., 2013). Moreover, the spectrum of BBOA in this study is qualitatively similar to published BB spectra from the fresh BB smoke in a smog chamber (Grieshop et al., 2009). These findings suggest that this factor can be related to BBOA with low atmospheric oxidants, and thus this factor might this factor might be associated with fresh/primary BBOA during the harvests. Furthermore, fresh BBOA shows a similar trend (Fig. 4) and wind rose patterns (Figure S4) as OOA-BB during the whole study, suggesting that fresh BBOA and OOA-BB might be from same plumes.

3. I think that the title should be changed slightly to reflect that the focus of the work is on biomass burning emissions of agricultural waste during the harvest season.

Response: Thanks for the comments and suggestion! 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 afternoon, nighttime and/or evening in the fields. And air pollution events frequently occurred in harvest seasons in China. Thus, the harvest season can indirectly reflect the biomass burning influence in China (Ding et al., 2013; Cheng et al., 2014). We have modified the related text in the introduction. The details in the revised manuscript can be found in lines 63-65, page 3.

Specific comments

1. Page 3, line 61. The authors state that farmers burning agricultural residues may

result in BB emissions, surely this WILL result in biomass burning emissions?

Response: Thanks for the comments! As mentioned above, the agricultural residue fire is a major biomass burning source affecting air quality during harvest seasons in the YRD region.

2. Page 4, line 82-83. Suggest rewording this sentence as it is difficult to follow.

Response: Thanks for the comments! The sentence has been reworded. The details in the revised manuscript can be found in lines 85-86, page 4.

3. Page 9, line 218-220. Perhaps the meteorological values (e.g. WS, WD, etc) could go in Table 1, for clarity.

Response: Thanks for the suggestion! The meteorological factors have been added into Table 1.

4. Page 10 line 230. "An over estimation was previously suggested by Huang et al. (2011)". What was the extent of this over estimation found by Huang et al.? The authors may wish to comment on how much this could have affected the observed correlation between measured and reconstructed PM_1 based on Huang et al. findings.

Response: Thanks for the comments and suggestion! 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.

5. Page 10, line 242. "Overall, those species also show a similar contribution between summer and autumn harvest". To which species are you referring to here? It is not clear in the text, please clarify.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 237, page 10.

6. Page 11, line 260-1. "This speculation is consistent with the highest loadings of K^+, BBOA, OOA-BB, chloride and BC during the summer harvest". When were these highest loadings observed? During case 1? Please clarify.

Response: Thanks for the comments and suggestion! The case 1, at 21:00 – 22:00 on 10 June, with the highest PM_1 mass (253.1 μg m^{-3}) during the summer harvest is characterized by high loadings of K^+, BBOA, OOA-BB, chloride, and BC, indicating the significant impacts of agricultural burning from the northwest of Nanjing (Fig. 1). The sentence has been reworded. The details in the revised manuscript can be found in lines 252-255, page 11.

7. Page 11, line 263. From figure 1, case 2 and 3 appear to be at times when the wind came from different directions, from the NW and E, respectively. Earlier in the paragraph it is stated that the agricultural fires are located to the NW of Nanjing, so case 3 also in the direction of agricultural fires? The authors should perhaps clarify this in the text.

Response: Sorry for the confusion! The wind direction in the cases should be from north to northeast (Fig. 1 and Table S1). The sentence has been reworded. The details in the revised manuscript can be found in lines 255-257, page 11 and Table S1.

8. Page 12, line 280. "This means the non-volatile character of sulfate and its more regional pollution in the YRD region during the summer and autumn harvest." Please re-word.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 274, page 12.

9. Page 15, line 366. The peak of the diurnal profile for BBOA is around 8 pm, which is usually characteristic of domestic burning, so as mentioned in an earlier comment could this not indicate local emissions?

Response: Thanks for the comments and suggestion! As mentioned above, local emissions contribute to BBOA less in this study. In addition, the background value of f_{60} value was0.26 % on average in July, implying that agricultural burning showed insignificant impact (Fig. 9).

10. Page 19, line 447-451. "As shown in Figure S4, for the BB-emissions related OA (including BBOA and OOA-BB), they show a very similar wind rose pattern with high concentration from southeasterly wind during the summer harvest, and from northerly wind during the autumn harvest. This further supports that the production of

OOA-BB is related to the BB plumes." From Figure S1, the main concentration of agricultural fires appear to be to the NW of Nanjing during the summer, how does corroborate with highest concentrations coming from the SE, particularly for BBOA? A similar trend is also observed in the autumn, with the highest BBOA and OOA-BB concentrations coming from SE whereas the fires mainly appear to be concentrated in the west (Fig S1). How a more useful analysis may be to also include wind speed, such as a polar plot (where concentrations are plotted as a function of both wind direction and speed), which could indicate the whether local or regional emissions were contributing and their direction.

Response: Sorry for the confusion! In order to investigate the relationship between wind direction and the OA components loadings, we didn't include information on wind speed in the polar plots (Figure S4). The related text has been removed.

11. Page 20, line 475. "This is also corresponding to some contention discussed in section3.2.3." Please re-word, to what contention are you referring?

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

12. Page 20, line 489. "Therefore, these findings indicate that BB contributes more fractions on organics than that on the secondary inorganic aerosols in the transported pollution air masses." Please re-word, not quite sure what you are trying to say.

Response: Sorry for the confusion! The sentence has been reworded. The details in the revised manuscript can be found in lines 465-466, page 19.

13. Page 21, line 496. I am not sure what Fig 8 adds to the analysis based on the discussion that it not already shown in Fig 7 and so Fig 8 could perhaps be removed but will leave that for the authors to decide.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in lines 476-483, page 20.

14. Page 23, line 5559-562. How is a higher Δf_{44} a factor for the observed higher oxidation levels during summer? Is not the f_{44} a measure of OA oxidation and so how can the f_{44} be a factor for the observed higher oxidation levels during summer?

Response: Thanks for the comments and suggestion! The related text has been removed.

15. Page 25, line 616, “The contributions of BC, HOA + COA, and BB related OA (BBOA and OOA-BB) to PM_1 are rather high in the WC BTs”. However, my understanding of Fig 13 is that the % contributions for these species did not change that much during the summer harvest and perhaps the authors may wish to clarify this statement. Overall, I found the discussion in Section 3.6 a little hard to follow and would recommend that the authors review this section.

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

16. Page 27, line 659-661. “This suggests that OOA-BB may be quickly oxidized a bit and condensed on the particle phase during the nighttime with the high RH and low *T* conditions.” Please re-word.

Response: Thanks for the comments and suggestion! The sentence has been reworded. The details in the revised manuscript can be found in line 616, page 25.

17. Figure 3. Why have you used results by Crippa at al. (2013) from Paris as reference mass spectra and not those obtained locally such as He et al. (2010) or Huang et al. (2013)?

Response: Thanks for the comments and suggestion! Firstly, the PMF-solution, i.e., four factors (HOA+COA, BBOA, OOA-BB, and OOA), is similar to the results at an urban site by Crippa et al. (2013). Particularly, two BB-related components, i.e., BBOA and OOA-BB, were identified, which is also similar to the finding by Crippa et al. (2013). In order to compare the two BB-related components with previous studies, we selected a study by Crippa et al. (2013).

18. Included in Fig 9 are measurements taken during July 2013, as they are considered to have no biomass burning influence. However, I found no mention of these measurements in the Methods. Were these taken at the same location and with the same instrument? Please include a brief description in the methods. Furthermore, to give some context to the measured PM masses during the harvest season in Nanjing, it may be worth including a comparison of the measured PM_1 masses during the July measurements.

Response: Thanks for the comments and suggestion! The related text has been added. The details in the revised manuscript can be found in line 126-127, page 6.

References

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.

Canonaco, F., Crippa, M., Slowik, J. G., Baltensperger, U., and Prévôt, A. S. H.: SoFi, an IGOR-based interface for the efficient use of the generalized multilinear engine (ME-2) for the source apportionment: ME-2 application to aerosol mass spectrometer data, Atmos. Meas. Tech., 6, 3649-3661, doi:10.5194/amt-6-3649-2013, 2013.

Cheng, Z., Wang, S., Fu, X., Watson, J. G., Jiang, J., Fu, Q., Chen, C., Xu, B., Yu, J., Chow, J. C., and Hao, J.: Impact of biomass burning on haze pollution in the Yangtze River delta, China: a case study in summer 2011, Atmos. Chem. Phys., 14, 4573-4585, doi:10.5194/acp-14-4573-2014, 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.

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(D16), D16305, doi:10.1029/2004JD005623, 2005.

Ding, A. J., Fu, C. B., Yang, X. Q., Sun, J. N., Petäjä, T., Kerminen, V.-M., Wang, T., Xie, Y., Herrmann, E., Zheng, L. F., Nie, W., Liu, Q., Wei, X. L., and Kulmala, M.: Intense atmospheric pollution modifies weather: a case of mixed biomass burning with fossil fuel combustion pollution in eastern China, Atmos. Chem. Phys., 13, 10545-10554, doi:10.5194/acp-13-10545-2013, 2013.

Li, H.Y., Han, Z.W., and Cheng, T. T.: Agricultural fire impacts on the air quality of Shanghai during summer harvest time, Aerosol Air Qual. Res. 10, 95-101, 2010.

Li, X. H., Wang, S. X., Duan, L., Hao, J. M., Li, Y. S., and Yang, L.: Particulate and trace gas emissions from open burning of wheat straw and corn stover in China, Environ. Sci. Technol., 41 (17): 6052-6058, 2007.

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.

He, L.-Y., Lin,Y., Huang,X.-F., Guo,S., Xue,L., Su,Q., Hu,M., Luan,S.-J., and Zhang,Y.-H.: Characterization of high-resolution aerosol mass spectra of primary organic aerosol emissions from Chinese cooking and biomass burning, Atmos. Chem. Phys., 10, 11535-11543, doi:10.5194/acp-10-11535-2010, 2010.

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., Xue,L., Tian, D.-X., Shao, W.-W.,Sun,T.-L., Gong, Z.-H., Ju, W.-W., Jiang, B., Hu, M., and He,L.-Y.: Highly time-resolved carbonaceous aerosol characterization in Yangtze River Delta of China: Composition, mixing state and secondary formation, Atmos. Environ., 64, 200-207, 2013.

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, 2012.

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, 2012.

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., Q. Jiang, Wang, Z., Fu, P., J. Li, J., Yang, T., and Y. Yin, Y.: Investigation of the sources and evolution processes of severe haze pollution in Beijing in January 2013, J. Geophys. Res. Atmos., 119, 4380–4398, 2014.

Wang, G. H., Kawamura, K., Xie, M. J., Hu, S. Y., Cao, J. J., An, Z. H., Waston J. G., and Chow, J. C.: Organic molecular compositions and size distributions of Chinese summer and autumn aerosols from Nanjing: characteristic haze event caused by wheat straw burning, Environ. Sci. Technol., 43 (17): 6493-6499, 2009.

Wehner, B., Birmili, W., Ditas, F., Wu, Z., Hu, M., Liu, X., Mao, J., Sugimoto, N., and Wiedensohler, A.: Relationships between submicrometer particulate air pollution and air mass history in Beijing, China, 2004–2006, Atmos. Chem. Phys., 8, 6155-6168, doi:10.5194/acp-8-6155-2008, 2008.

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, PM1contributions, and seasonality, Atmos. Chem. Phys. Discuss., 14, 20845-20882, doi:10.5194/acpd-14-20845-2014, 2014.

Zhang, J. K., Sun, Y., Liu, Z. R., Ji, D. S., Hu, B., Liu, Q., and Wang, Y. S.: Characterization of submicron aerosols during a month of serious pollution in Beijing, 2013, Atmos. Chem. Phys., 14, 2887-2903, doi:10.5194/acp-14-2887-2014, 2014.