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Interactive comment on "Aging of aerosols emitted from biomass burning in northern Australia" *by* Andelija Milic et al.

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Author response to Referee #2

The authors appreciate the reviewer's comments that helped us to improve the manuscript. We have addressed each of comments as outlined below. All page and line numbers refer to the revised manuscript (Revised_manuscript_trackChanges.docx) where all changes (track changes) are available (revised supplementary material is included in the revised manuscript pdf file). The link for the revised manuscript is given at the end of this document. If the text is significantly changed only the number of section is given (e.g. "Biomass burning events", Sect. 3.2.4).

Comment: This paper discussed the aging of biomass burning (BB) aerosols from

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Northern territory of Australia based on aerosol mass spectrometer (AMS) at a ground observation site. The entire field study (29 May to 20 June, 2014) was divided to be “close BB” and “distant BB” periods. The BB aerosols were investigated by comparing aerosol chemical composition, aging process, ozone formation, size distribution and SOA formation between those two periods. PMF analysis was performed on the background OA period. Ambient Isoprene Epoxydiols-Derived SOA (IEPOX-SOA) factor from three PMF-factors solution was discussed and showed similar performances to the same factor reported in the previous studies. This paper showed a good dataset for investigating the biomass burning influences on aerosols. However, the analysis in this paper is not clear and is not presented in a quite logic way. Thus I recommend a major revision for this paper before considering its publication in ACP.

Response: Number of changes is made throughout the manuscript in order to present data in more clear way. The following has been modified or added to the manuscript: The title of the manuscript is modified. Separation on “close BB” and “distant BB” periods is clarified. More analysis and discussion are added for BB events, including backtrajectories and Sentinel hot spot locations. The suggestions regarding isoprene-derived OA are incorporated in the manuscript. Discussion regarding the sea salt-chlorides and f44-resolved size distribution is modified and clarified.

Comment 1: The authors analysed the data in section 3.2 based on dividing the entire study to be “distant BB” and “close BB” periods in section 3.1. However, it is not clear that how the “close BB” and “distant BB” periods were defined. Was it based on distance or the emission intensity, or the correlation between organic, CO and particle number concentration? The authors only labelled “close BB” period in Figs.1-2, “distant BB” period and background OA period are unknown.

Response 1: The authors agree that more specific definitions should be given for “close” and “distant” BB periods and clear presentation of data separation should be given.

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The measurement period was separated into "close BB" and "distant BB" periods based on organics, carbon monoxide and particle number concentration and their correlation, as well based on the distance of known fires (Sentinel hot spots) from the ATARS.

Firstly, we were looking at the time series of organics, carbon monoxide and particle number concentration (Figure 2, Page 25) in order to identify the periods of significant BB influences, e.g. their high signals (e.g. on 9th and 26th of June) gave us a guide on which days intense/close BB took place. The increases in BB markers (levoglucosan, soluble non-sea salt potassium and Org 60) also showed the same trend (Figure 3, Page 26). After identifying these periods, the next step was to relate these signals to the possible BB events. Therefore, we have mapped all fires (Sentinel hot spots) that have happened during these periods and have confidence level of 50% and more. To make sure that everything is included (e.g. close fires with low intensity) we have gone through every measurement day and mapped all the events that have occurred.

Hot spots detected within 20 km occurred on 30th of May, between 7th and 11th and 25th and 26th of June (Figure 2, Page 25). Taken the distance of 20 km and increased concentrations of organics, carbon monoxide and particle number, these days were taken as "close BB" periods. Knowing that ATARS was constantly affected by the BB air masses the remaining measurement days were taken as "distant BB" periods. In summary, "close BB" periods refer to periods where close fires (within 20 km) have taken the place and correlated increases in carbon monoxide, organics and particle number concentration were observed (Fig. 2), while the rest of the measurement is defined as "distant BB" periods.

In order to clarify the close and distant BB periods the paragraph at Page 8, line 3- Page 8 Line 16 is modified: "The sampling site was constantly impacted by fire emissions with periods of heavy BBs characterized by high aerosol and gas phase concentrations, for instance CO reaching up to ~104 ppb and organics up to 350 μg m-3 (Fig. 2). The most intense BB episodes were on the 30th of May, between the 7th and 11th of June,

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and on the 25th and 26th of June (Fig. 1c) (Mallet et al., 2016). During intense fire periods, organics, CO and particle number concentration showed correlated increases (Fig. 2). Moreover, looking at Sentinel Hot spots during these periods, hot spots were detected within 20 km from the ATARS. Based on this, the dataset was separated into periods of “close BB” (corresponding to high organics, CO and particle number concentration signals and close events (< 20 km)) and “distant BB” (corresponding to less intense organics, CO and particle number concentration signals and distant events (> 20 km)). It is important to emphasize that all periods during the measurement that have not been included in close BB periods have been assigned to distant BB periods, as ATARS was constantly influenced by BBs. The selection does not mean that emissions from distant fires were not present during the close BB periods but that the influence of fires near the measurement station during these periods was dominant. Nine intense BB events were identified from close BB periods as single source emissions (Desservettaz et al., 2016). Five of the nine BB events (Fig. 2) were analysed here (Sect. 3.2.4), due to the instrument not sampling during the remaining events. Most of the events occurred in the afternoon/night time (Table 1)”.

In order to indicate distant BB periods the following is added to Fig. 2 caption at Page 25: “The distant BB periods cover all days of the measurement other than days included in close BB periods”.

Background period is related just to the PMF analysis and therefore it was not introduced in the 3.1 chapter, but it is clearly defined at Page 15, lines 1 – 4 and illustrated in Figures S13 and S14. Comment 2: Suggest revising the structure of the results and discussion part. After all comparisons between the “close BB” and “distant BB” being discussed in section 3.2.1-3.2.3, the paper started to describe the details about the “close BB” period in section 3.2.4, which is very confusing. I do not quite understand what the relationship between the 5 BB events in section 3.2.4 with the “close BB” period analysed in section 3.2.1-3.2.3 is. I suggest the authors introduce the details on “close BB” and “distant BB” period first, and then discuss the general comparison results. In this way, the readers can get a rough idea on what “close BB” and “distant BB” are then understand later comparison better. I also suggest the authors add a separate figure for readers to see the details of those “close BB” and “distant BB” periods. In this way, the good correlation addressed in page 7 Line 30 can also be understood.

Response 2: As suggested (previous comment) the “close BB” and “distant BB” periods were clearly defined and introduced before the comparison of the results. Moreover the following is added to the manuscript at Page 8, lines 14-16: “Nine intense BB events were identified from close BB periods as single source emissions (Desservettaz et al., 2016). Five of the nine events (Fig. 2) were analysed here (Sect. 3.2.4), due to the instrument not sampling during the remaining events”. This is repeated at the beginning of the “Biomass burning events” section (Sect.3.2.4) at Page 12, lines 10-12: “Five single source BB events were analysed here (Fig. 2). These episodes were within previously defined close BB periods“. This clearly explains that nine events were identified as a single source events (coming just from the one fire source) and that these events are within the close BB periods. The AMS was not sampling during all events, therefore, five events labelled on Fig. 2 are addressed in this paper. As discussed in the previous comment, distant BB periods were clarified in the main text and in the figure caption (Fig. 2, Page 25).

Comment 3: Page 8, line 10-20: 1) The authors mentioned there were high chloride (KCl or NH4Cl) mass concentrations from the biomass burning, then said a significant portion of chlorides from BAM measurement came from sea salt. Those two statements are conflictive.

2) If the signals are strong, KCl+ and NaCl+ ions can be resolved in the OA spectra in the HR fitting of PIKA.

3) The chloride with high concentrations in Fig.1 was not shown in the comparison plot of Fig. S2. Were some periods missed in Fig. S2? The authors should point out which periods were used for comparison in Fig.S2.

4) Line 10, “This can be explained by the depletion of chloride with transport and aging of BB plumes (Li et al., 2003; Li et al., 2010;Liu et al., 2000).” There is no proof for this sentence. Since the author has the K+ and Na+ measurement, the authors can calculate the chloride depletion fraction to show the evidence for this sentence. See equation (A1) and Fig. (A2) in (Hayes et al., 2013) for reference.

Response 3: 1) High AMS-resolved chloride concentrations during the close BB periods can be clearly seen from the AMS (Fig.1(c)) and BAM (Fig. S3a) time series and also high correlation of AMS-resolved chlorides and organics during the close BB periods indicates BB-related origin. The high contribution of chlorides originating from fires has been also emphasised in the campaign overview publication (Mallet et al., 2016). However, apart from the fire-related chloride signals, BAM filters collected the sea salt chlorides during the periods of oceanic influence (period between 3rd and 6th and 19th and 22nd of June, as it can be seen in the Fig S3a. Moreover, the Mg2+/Na+ ratio (ions measured by BAM) for the filters collected during the oceanic-influenced periods was close to the ratio for the sea salt (Fig. S3b). Therefore we concluded that significant portion of chlorides from BAM filters were from the sea salt. For more details please see the response to the minor comment number 2, from Referee 1.

To clarify this in the manuscript the following is modified to the Page 9, line 3- Page 9, line 8: “Mg2+/Na+ ratio values for the filters collected during the high oceanic influence (between 3rd and 6th and 19th and 22nd of June) were close to the sea salt Mg2+/Na+ ratio of 0.12 (Fig. S3b). At the same time low terrestrial fetch (low radon concentration) was observed (Mallet et al., 2016). Moreover, the chlorides concentration collected on filters was prominent during the period of oceanic influence (Fig. S3a). This suggests that a significant portion of chlorides detected on the BAM filters was of a sea salt origin, which is refractory and therefore not well measured by the AMS, thereby explaining the poor chloride correlation”. Moreover, the sentence was added at Page 8, line 27: “Increased chloride concentration during the close BB periods was also indicated by the BAM data (Fig S3a)”.

In order to present the BAM chloride time series a new Figure, Fig S3a is added to the supplementary material and Fig. 3 is now to Fig. 3b. Figure 3b is also modified: z-axis (date) was added to the graph.

2) After HR analysis of KCl+ and NaCl+ ions, further analysis couldn't be done as resolving the ions was not possible due to their low contribution (see in Figure below).

3) All AMS data were averaged to BAM 12h data, therefore the AMS concentrations at Fig.1 and Fig. S2 are not the same. To emphasise this, the following sentence is added to the Fig. S2 caption: "The AMS data were averaged to BAM 12h data".

4) Unfortunately, we were not able to track the plumes and it was a challenge to separate the plumes and investigate their aging and thus chlorides depletion. Therefore, further analysis could not be performed.

Comment 4: Page 9, line 7: "The variability observed in f44 vs. f60 for close BB events (Fig. 4a) probably reflect BB plumes generated during different burning conditions rather than different atmospheric processing of BB masses." There is no evidence to support this sentence. In Fig. 5a, higher $\Delta O3/\Delta CO$ was observed for the high f44. The authors explained this figure as (page 9 line 31): "On average, the $\Delta O3/\Delta CO$ ratio increases with f44 and decreases with f60, indicating increased photochemical processing of OA in plumes with atmospheric aging and ozone production". Two statements are contradicting with each other.

Response 4: The authors have changed the sentences at Page 9, lines 25-27 as following: "The variability observed in f44 vs. f60 for close BB events (Fig. 4a) can reflect BB plumes generated during different burning conditions but also different atmospheric processing of BB masses" Ozone enrichments were observed in close as well as in distant plumes, therefore this statement is kept as it is and in addition sentence is added at Page 10, lines 28 and 29: "The $\Delta O3/\Delta CO$ enrichments for close BB period indicate that aging of close emissions cannot be excluded".

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Comment 5: Page 9, line 14: “This confirms that levoglucosan-like species carried by the BB plume did not degrade to background levels even as oxidized species were formed. Thus, f60 is a reasonable marker of distant BB in this study.” This expression is not quite true; lots of points in Fig 4b are around background level when the f44 is very high. Mixing BB plumes with other aged plumes also can degrade the f60.

Response 5: Most of the points in Fig. 4b are higher than the f60 background level when f44 is high (from 0.2 to 0.25). Mixing of BB plumes can result in lower f60, but even then most of the points are higher than the background. So the overall conclusion is that levoglucosan can be used as a reliable marker for distant BBs in this study.

Comment 6: Page 9 line 23 “The wide range in f44 can be attributed to difference in burning conditions for close BB”. What the author missing in this entire section is dilution or mixing BB plumes with other plumes. Not only different burning condition or aging, but also the dilution and mixing will also lead the similar evolution trend of f44 vs f60 in the triangle plot (Fig. 4).

Response 6: Dilution effect: The authors agree that dilution effect should be included in the text, as a factor influencing f44 vs f60 trend. This is added to the manuscript at Page 10, lines 11 and 12: “One more factor that can influence f44 vs f60 trend for both close and distant fire is dilution effect”.

Mixing between plumes: The backtrajectories clearly show that during the close BB periods, apart from the close fires all other hot spots are located on 60km or more from the ATARS (Fig S7 in the supplementary material). The authors thus suggest that the mixing of fresh plumes with the plumes coming from the distant fires in most of the cases won’t influence the f44/f60 changes due to the fact that both m/z 44 (60) and total organics for distant diluted masses are likely much smaller than m/z 44 (60) and total organics for close plume masses. This is simplified through the equation below.

f44=(Org 44 (fresh plume masses)+Org 44 (diluted distant plume masses))/(Org (fresh plume masses)+Org (diluted distant plume masses))

As same implies for f44 and f60, it is assumed that the mixing of fresh plumes with the distant diluted plumes coming from the distant fires won't affect the f44/f60 trend.

As not the same can be concluded in case for the distant BB periods the following is added to the manuscript at Page 10, line 7 and 8: "The observed evolution trend in f44 vs f43 for distant fires can be also influenced by mixing between the plumes" and sentence at Page 14, lines 26-28 is modified: "The thousands of fires that occurred during the SAFIRED campaign contributed to a wide range of OA composition that reflect different burning materials, conditions, mixed fresh and aged emissions and processing in the atmosphere".

Comment 7: Page 10, line 2: Suggest the authors use Ox (=NO_2+O_3) instead of O_3, or use both, then the O_3 loss by NO can be accounted for (Herndon et al., 2008).

Response 7: This comment unfortunately cannot be addressed due to the unreliable NO_x data.

Comment 8: Page 10, lines 6 and 7: How did the authors calculate the average $\Delta O_3/\Delta CO$ from different fires? How did the authors obtain $\Delta O_3/\Delta CO$ from "distant BB" fire plumes since the plume spikes are not obvious as those in "close BB" period?

Response 8: The average $\Delta O_3/\Delta CO$ ratio for particular BB event (5 events that were singled out) was calculated by averaging all values for the time of the fire. The average $\Delta O_3/\Delta CO$ ratio for distant BB periods is the average of all $\Delta O_3/\Delta CO$ values for the whole distant BB periods, not for the fires in periods separately.

Comment 9: Page 10, line 14: The authors should calculate $\Delta OA/\Delta CO$ vs Δf44 (not f44), because it is not clear if high f44 can really reflect the SOA aging since different BB plumes may have different f44 due to burning condition and mixing or dilution. I suggest the authors check the photochemical age calculated from VOCs (de Gouw et al., 2005), which can be another parameter for indicating oxidation/aging process of plumes. And also the authors have a great dataset to see emission ratio of BB emitted aerosols (e.g. ΔSO4/ΔCO, ΔNH4/ΔCO and ΔCl/ΔCO).

Response 9: The authors think that considering Δf44 (the change in f44 for different burning conditions) would only be beneficial for individual plumes, but not for the whole data set as we have presented here. Furthermore, for individual plumes, this is only possible if the plume can be tracked and the initial value of f44 is known. That is not the case for this study (stationary measurement site).

Unfortunately, due to the numerous fires which occurred across the area and the limitations of satellite (passing the region once per day) and ground-based measurements, we were not able to single out individual plumes that could be used for the suggested analysis.

Comment 10: Page 10, 3.2.3 section: Suggest adding size distributions of m/z 44 and m/z 60, which can be used to compare with the size distributions from SMPS. This comparison can help to interpret the comparison between “low f44” and “high f44” periods. In addition, the comparison of size distributions between “close BB” and “distant BB” can be added. This can help to characterize those two periods and see if the “distant BB” are really more aged or mixed with aged aerosols.

Response 10: In order to further analyse changes in size distribution with aging, size distributions for organics (sampled by the AMS) were examined for both close and distant BB periods. Data were categorised based on different f44 ranges, same as for the SMPS data (Fig 6).

More conclusions were made and paragraph for the Sect. 3.2.3 at Page 11 is modified: “Atmospheric aging of plume particles increases particle diameter due to gas to particle transfer of organic and inorganic gaseous species (Martins et al., 1998). In order to estimate whether the aging of BB masses during SAFIRED influenced particle size, average SMPS size distributions and AMS size distributions for organics, both categorised based on different f44 ranges, were examined (Fig. 6). It is important to emphasise that SMPS uses electrical mobility diameter, while AMS uses vacuum aerodynamic di-

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ameter. The close and distant BB periods were analysed separately. The f44 values were classified into four groups that represent different aging stages ($0.05<f44<0.1$, $0.1<f44<0.15$, $0.15<f44<0.2$, $0.2<f44<0.25$). The first f44 bin ($0.05<f44<0.1$) was not considered in case of the distant BB periods, as only a few data points were in this range (Fig. 4). The same was done for the highest f44 bin ($0.2<f44<0.25$) for close BB periods. According to SMPS data, the average particle mode varied between 101 - 113 nm and 104 - 106 nm for close and distant BB periods, respectively. The average mode for organics showed larger sizes and varied between 259 - 293 nm and 293 - 305 nm for close and distant BB periods, respectively. Increased f44 was accompanied by a reduction in SMPS particle size for close plumes, going from 113 nm ($0.05<f44<0.1$) to 101 nm ($0.1<f44<0.15$). The same trend was observed for the organic aerosols. Considering both AMS and SMPS data for distant fires, there was no considerable change in diameter with aging from less aged BB plumes to more aged BB air masses. The particle modes show only slight differences between different f44 bins. This is not consistent with the observed increase in OA for distant fires. Changes in size distribution will be discussed further in the Sect. 3.2.4 where results related to specific BB events are presented".

Please note that after further analyses we realised that only a few points were in the f44 range 0.05-0.1 for distant fires, therefore this was removed from the manuscript and new graph was added (Fig. 6).

Comment 11: Page 14, lines 1-2: What is the monoterpene concentration in this study? Monoterpene-derived SOA also can influence the background level of $C_5H_6O^+$ (Hu et al., 2015). Isoprene concentration was not reported in this study, which is important for discussing IEPOX-SOA.

Response 11: The average monoterpenes concentration was found to be 0.22 ± 0.41 ppb. The average isoprene concentration was 0.49 ± 0.78 ppb. The following is added to the manuscript at Page 16, line 2-5: "The average isoprene concentration measured during the campaign was 0.49 ± 0.78 ppb. However, it should be emphasised that monoterpene-derived OA can also influence the background level of C5H6O+ (Hu et al., 2015). The average monoterpenes concentration for this study was found to be 0.22 ± 0.41 ppb".

Comment 12: Page 14, lines 8-9: Low correlation between the IEPOX-SOA time series with both OOA and BBOA was observed (Fig. S15) suggesting that during BB influenced periods either higher NOX concentrations suppressed IEPOX and consequently IEPOX-SOA generation or the dominant BB aerosol during these periods inhibited measurement of isoprene oxidation products." This sentence does not have evidences to support. In theory, the high OM concentration from BBOA can facilitate the oxidation product to partition into the aerosol phase (Kroll and Seinfeld, 2008). To investigate the BB influence to IEPOX-SOA: 1) suggest the authors start to see if the BBOA plumes can increase the acidity of aerosol, which can promote the IEPOX uptake (Gaston et al., 2014). 2) the NOx concentration can influence the oxidation product of isoprene (Surratt et al., 2010; Bates et al., 2014). At the absent of IEPOX measurement, the authors can investigate the IEPOX-SOA concentration level or the fC5H6O abundance based on different categories of NOx concentrations by checking if there is systematic differences of IEPOX- SOA mass concentrations at high NOx and low NOx. This analysis presumably assumed the isoprene emissions in the large areas were relatively constant within the field study period.

Response 12: Due to the lack of supporting evidence for either of proposed reasons, this sentence is deleted from the manuscript (Page 17, lines 11-14). 1) According to calculations for the composition depended collection efficiency, 22% of the aerosol were acidic. Furthermore, the correlation of IEPOX-SOA and sulfate is found to be weak (R of 0.3) (Fig. S18). However, two periods (period before 5th of June and after 15th of June) could be clearly separated on the plot. While there is no correlation between sulfate and isoprene-derived OA factor for the first period, when plotting only data collected from the 15th of June, correlation is found to be slightly higher (R of 0.4). The following is added to the manuscript at Page 16 line 6-14: "The main path for isoprene-derived OA formation is proposed to be acid-catalysed IEPOX uptake (Lambe et al., 2015; Lin et al., 2011). According to calculations for the composition depended collection efficiency, 22% of the aerosol were acidic. In order to estimate whether the acidity of the particles had an influence on isoprene-derived OA generation in ATARS, the correlation between sulfate (taken as proxy of aerosol acidity) and the isoprene-derived OA factor was examined. The correlation between the factor and sulfate can be considered as weak (R of 0.3) (Fig S18). However, two periods can be clearly distinguished from the graph: the period before 5th of June and the period after 15th of June. While there is no correlation between sulfate and the isoprene-derived OA factor for the first period, when plotting only data collected from 15th of June, correlation is found to be slightly higher than the correlation for all background data (R of 0.4)".

2) The comment unfortunately cannot be addressed due to the unreliable NOx data.

Minor comments

Comment 1: Page 4 lines 1-2: Please give the standard deviations of those reported average values.

Response 1: The standard deviation values for all reported average values are given in the revised manuscript. Sentence at Page 4, lines 1-3 has been modified: " The early dry season was characterized by dry weather conditions (average relative humidity of 67 ± 23 %) and warm days with an average daily and nightly temperatures of 27 ± 5 °C (up to 34 °C) and 19 ± 4 °C (with a minimum of 10 °C), respectively".

Comment 2: Page 4, line 27: How can the authors separate aged POA and SOA? To what extent, the authors will define the aerosol is aged POA since SOA can be from aged POA (Robinson et al., 2007).

Response 2: The authors agree that aged POA and SOA cannot be separated using the fragment analysis that was stated by this sentence. Therefore, "aged POA" was excluded from the sentence (Page 4, line 30): "Organic aerosols measured by the AMS encompassed aerosols that were processed in the atmosphere for different periods of time and included both POA and SOA. As such, a tool was needed to distinguish BB aerosol from other sources and fresh from processed BB aerosol".

Comment 3: Page 5, line 20: Have the authors offset the background of BAM measurement e.g. the background concentration can refer to the mass concentrations of different species on blank filters?

Response 3: Yes, all of the species measured on the BAM PM1 filters were blank corrected. This is added to the manuscript at Page 5, lines 26 and 27: "All of the species measured on BAM filters were blank corrected".

Comment 4: Page 5 line 21: Suggest changing "PM1" to be "PM1", which is more commonly used.

Response 4: "PM1" has been changed in "PM1" throughout the manuscript.

Comment 5: Page 6, line 17: Is the PTR-MS data UMR or HR? How did the authors separate the contributions of isoprene and furan to the m/z 69.

Response 5: An unit mass resolution PTR-MS (quadrupole mass spectrometer) has been used and this is added to the manuscript at Page 6, lines 22-24: "A high sensitivity Proton Transfer Reaction-Mass Spectrometer (PTR-MS, Ionicon Analytik) with a quadrupole mass spectrometer and an H3O+ ion source was employed to measure non-methane organic compounds (NMOCs) that include non-methane hydrocarbons and oxygenated volatile organic compounds".

The authors used the PTR-MS-resolved m/z 69 as a signal related to isoprene. Furan concentrations were significant only at the time of close BB periods (as it can be seen from Fig S17). These have been removed from the data for the PMF (background data) where isoprene-derived OA were examined.

The following is added to the manuscript at Page 7, lines 1 and 2: "In order to distinguish furan and isoprene contribution over the sampling period, a gas chromatography-mass spectrometry method was used" and at Page16, lines 15-18: "The isoprene/furan concentrations at m/z 69 (PTR-MS) were treated as an isoprene contribution due to the dominance of isoprene signal compared to furan, according to the samples analysed by gas chromatography-mass spectrometry (Fig. S17). The furan contribution was more significant in BB plumes, during the close BB period, as suggested previously (Warneke et al., 2011)".

Figure (Fig. S17) illustrating the furan and isoprene data analysed by the gas chromatography-mass spectrometry is presented in the supplementary material.

Comment 6: Page 7, line 14: Suggest adding the OM/OC values.

Response 6: The average OM/OC value of 1.4 is stated at Page 7, line 23.

Comment 7: Page 7, line 20: Please change "organic mass" to be OM. Since the authors start to use OM as an abbreviation, it should be consistent in the following text.

Response 7: "OM" has been used as an abbreviation through the manuscript (Page 7, line 23; Page 7, line 24). "Organic mass" is used just at the beginning of the sentence (Page 7, line 27).

Comment 8: Page 7, line 25: Please give the standard deviations to those reported average concentrations.

Response 8: The standard deviation values for all reported average values are given in the new manuscript version at Page 7, line 29 – Page 8, line 2: "In this study, the remaining submicron non-refractory mass was made up of inorganics including sulfates (4.2 %), ammonium (2.8 %), nitrates (1.5 %) and chlorides (1.3 %), with average concentrations of 0.51 $\pm$ 0.32 μg m-3, 0.35 $\pm$ 0.68 μg m-3, 0.19 $\pm$ 0.45 μg m-3 and 0.17 $\pm$ 1.28 μg m-3, respectively."

Comment 9: Page 8 line 25: It is not clear that Org 60 is UMR or HR. The authors already use m/z 60 refers to UMR and C2H4O2+ refers to HR. Please be consistent. F60 (HR or UMR) has a similar problem e.g., in page 9 line 1.

Response 9: HR peak fitting data (PIKA) were used through the analysis as stated in Page 4, line 27: “Therefore, HR peak fitting data (PIKA) were chosen for further analysis”. Since these are UMR data fitted in PIKA, Org60 and f60 are used in the manuscript instead of C2H4O2+ and fC2H4O2+. The same is done with other fragments.

To make it clear the following is modified to Page 5 lines 1 and 2: “An extensively employed fragments analysis using the AMS-extracted parameters f43 (ions C3H7+ and C2H3O+), f44 (CO2+) and f60 (C2H4O2+) was applied here’ and at Page 4 lines 7-9: “The AMS parameter f60 and accompanying f73 (C3H5O2+) are widely used as BB emission signatures as they are directly related to levoglucosan-like species, which are a substantial fraction of organics emitted in pyrolysis of cellulose (Alfarra et al., 2007; Simoneit et al., 1999)”.

Comment 10: Page 9, line 23: What does “atmospheric processing” refer to?

Response 10: Atmospheric processing refer to aging process which is defined at Page 2, lines 21 and 22 as: fresh particle or gaseous species transformed in the atmosphere through photochemical processing.

Comment 11: Page 11, line 4: Repeated sentence, which was already said in page 7 line 29.

Response 11: This sentence (Page 12, lines 10 and 11) is changed for : “Five single source BB events were analysed here (Fig.2)”.

Comment 12: Page 11, line 27: OOA should be SOA, OOA PMF factor has not been introduced here yet.

Response 12: OOA was used here as an oxygenated fraction of OA, as defined in Page 2, line 28 and not as a PMF-resolved factor. Therefore OOA is kept as it is.

Comment 13: Page 11, line 8: “As these plumes were mostly related to sources close to the sampling site (within 2 to 10 km), differences in m/z 44 and 43 can be mostly related to different burning conditions." No evidence was shown to support this sentence.

Response 13: The sentence was removed from the revised manuscript at Page 12, lines 14-16.

For figures

Comment 1: Figure 1. Suggest adding EC fraction in Fig. 1(a) to the total AMS fraction measured in Fig. 1(b), then a full chemical composition of PM1 can be obtained.

Response 1: Mass fractions for inorganic species collected by BAM are added to Figure 1 (a) bar so full chemical composition of PM1 is obtained. Manuscript text is modified, Page 7, lines 19 and 20: "Organic carbon made up 72 % and EC 15 % of the measured PM1 on the BAM filters (Fig. 1a)." Fig. 1 at Page 25 and its caption are also updated.

Comment 2: Figure 3. Has the background of org 60 been subtracted (0.003). If the answer is negative, please offset the background.

Response 2: The background value of 0.003 is related to f60, not to Org 60. The authors did not offset the background value for Org 60. This is not done in other studies as well.

Comment 3: Figure 4. Empirical triangle area was reported in Cubison et al. (2011). Suggest adding right guide line here as well, which can help to compare this study with other studies.

Response 3: We agree that adding the empirical BBOA-related triangle plot in f44 vs. f60 space, introduced by Cubison et al. (2011) will facilitate comparison with other BB studies. The triangle plot is added to plots in Figure 4 but also to Figure 5 and Figure 6 plots, for the same reason. As Figure 4, 5 and 6 are combined in one panel figure (please see the next comment), the new Figure 4 caption is modified: "f44 vs. f60 coloured by date for (a) close and (b) distant BB periods, by ΔO3/ΔCO ratio for (c) close and (d) distant BB periods, and by ΔOA/ΔCO ratio for (e) close and (f) distant

BB periods. Vertical black lines refer to the f60 background level of 0.003. Red dashed lines refer to the ambient BBOA-related data introduced by Cubison et al. (2011). Note: ozone data from 29th of May until the 1st of June were not available".

Comment 4: Figure 5-6. They are all identical plots. It will be easier to see by combining them together as one panel figure.

Response 4: We agree that combining all f44 vs. f60 plots into the one panel figure gives better insight into the plots. Figure 4 (Page 27) now represents combined figures (previously Figure 4, Figure 5 and Figure 6).

Comment 5: Figure 9 (b): What is the correlation between BBOA with levoglucosan and K+? what is the correlation between IEPOX-SOA with sulfate and isoprene? Does any compound correlate with OOA? Those external tracers should be added on the diurnal variation plot as well. Figure 9 (c): It is better to add the exact fraction values on the bar.

Response 5: The authors have already examined all correlations that have been suggested in this comment. In overall, the correlation of PMF factors was difficult to access due to the "chopped" background dataset that resulted in limited dot points for correlation. The BBOA had a weak correlation with the levoglucosan and nnsK. Moreover, levoglucosan and nssK data are given every 12h, which is the reason why diurnal trend couldn't give useful information. The correlation of isoprene-derived OA and sulfate has found to be weak (R of 0.3) (Please see an answer on the major comment number 12 from Referee 1). There is no correlation between isoprene and isoprene-derived OA, as it was stated and presented in Figure S14. None of parameters have strong trend with the OOA. The exact fraction values for PMF factor contribution are added to the Figure 7 (c).

Comment 6: Figure 10. There are more f44 vs f82 points from IEPOX-SOA factors in Hu et al. (Hu et al., 2015). The authors should add the range of those data, which is more meaningful than showing only two points.

Response 6: The two points present the position of two IEPOX-SOA profiles from two specific studies, Budisulistiorini et al. (2013) and Robinson et al. (2011) in f44 vs. f82 plot.

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Please also note the supplement to this comment:
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