#### Response to comments of Referee 2.

#### Comments from referee are in blue and response from co-authors is in black.

### Major comments.

Quantification of PON from biomass burning using a CToF-AMS: The m/z 46:30 ratio is used for the quantification of PON, which is based on a method done by Farmer et al 2010 that used an HR-AMS and did not include any biomass burning emissions. The large fraction of OA from biomass burning certainly will produce a large CH2O+ contribution and will make the quantification of PON very different compared to the Farmer et al paper. On page 7 line 250 the authors say that the m/z 30 interference is likely small, but this might be very different in biomass burning and needs to be shown or given a reference. In addition it is written on page 6 line 206 that the interference of CH2O+ is discussed in section 3.4.1, but no such section exists in the paper. So it is not clear, how the authors deal with the m/z 30 interference and how large the uncertainty or error estimate on the quantification of PON is. In addition in the later section, where the primary versus secondary PON is discussed, it is certainly possible that the m/z 30 interference is different as the composition changes during the later part of the night, when the authors claim that they observed secondary PON. A section needs to be added, where the interference is clearly explained and the effect needs to be quantified. This should result in an uncertainty range for the m/z 30:46 ratio and error estimate for PON. All of this should be added to the instrumentation section, where there needs to be a description of the AMS added as well that includes a discussion on the calibration for NH4NO3 and, if available, for PON compounds. Here it should be made clear what m/z 30:46 ratio NH4NO3 has in this particular instrument? The resulting error estimate needs to be taken into account for the following discussions.

Section 2.2.2 in methods has been edited as follows:

# 2.2.2 Particulate Organic Oxides of Nitrogen (PON)

Concentrations of PON were calculated following the method proposed by Farmer et al. (2010) and the considerations used by Kiendler-Scharr et al. (2016). This method has been previously used in studies looking at aerosols from biomass burning (Tiitta et al., 2016;Zhu et al., 2016;Florou et al., 2017). Equation 5 calculates the PON fraction ( $X_{PON}$ ), using the signals at m/z 30 and m/z 46 to calculate m/z ratios 46:30 from AMS measurements ( $R_{meas}$ ), from ammonium nitrate calibrations ( $R_{cal}$ ), and from organic nitrogen ( $R_{ON}$ ) to quantify PON concentrations.

$$X_{\text{PON}} = \frac{(R_{\text{meas}} - R_{\text{Cal}})(1 + R_{\text{ON}})}{(R_{\text{ON}} - R_{\text{cal}})(1 + R_{\text{meas}})}$$
(5)

Where ratios from ammonium nitrate calibrations  $R_{cal} = 0.5$ ;  $R_{meas} = m/z$  46:30 ratio from measurements; m/z 46:30 ratio from ON  $R_{ON} = 0.1$ , Following Kostenidou et al. (2015) consideration,  $R_{ON} = 0.1$  was calculated as the minimum m/z 46:30 ratio observed.  $R_{ON}$  value of 0.1 has been used in previous studies (Kiendler-Scharr et al., 2016;Tiitta et al., 2016).

$$PON = X_{PON} * NO_3^-$$
(6)

Finally, equation 6 calculates PON concentrations  $[\mu g.m^{-3}]$  where NO<sub>3</sub><sup>-</sup> is the total nitrate measured by the cToF-AMS. The method proposed by Farmer et al. (2010) is based on HR-ToF-AMS measurements were m/z 30 represents NO<sup>+</sup> ion and m/z 46 NO<sub>2</sub><sup>+</sup> ion while the cToF-AMS gives unit mass resolution mass spectra information, hence, there is the possibility to have interference of CH<sub>2</sub>O<sup>+</sup> ion at m/z 30. However, when analysing mass spectra from previous laboratory and ambient studies using HR-ToF-AMS to investigate biomass burning emissions, we can confirm that the signal of CH<sub>2</sub>O<sup>+</sup> at m/z 30 is low compared to signals at m/z's 29 and 31, while in this study m/z 30 is the main signal (Fig. 5.c). Hence, in this study an interference of CH<sub>2</sub>O<sup>+</sup> at m/z 30 is unlikely and if there were any interference of CH<sub>2</sub>O<sup>+</sup> it would be negligible. Table S1 in

supplement shows m/z 30/29 and 30/31 from previous laboratory and ambient studies investigating biomass burning emissions.

Another possible interference would be the presence of mineral nitrates at m/z 30 (e.g. KNO<sub>3</sub> and NaNO<sub>3</sub>). However, mineral nitrate salts tend to be large particles (Allan et al., 2006;Chakraborty et al., 2016) and also have low vaporisation efficiency (Drewnick et al., 2015), which makes it unlikely to be measured by the AMS in large quantities.

Table S1. CH <sub>2</sub> O <sup>+</sup> signals at m/z 29, 30 and 31 from HR-ToF-AMS data of previous studies. Comparison of n	n/z
ratios 30/29 and 30/31 with values found in this study.	

	Reference	30/29	30/31	m/z 29	m/z 30	m/z 31	Notes
	This should	4.38	35.00	0.08	0.35	0.01	sPON_ME2
	i his study	1.42	8.50	0.06	0.09	0.01	pPON_ME2
	(Ailian at al. 2010)	0.16	0.32	0.05	0.008	0.025	pine burn
	(Alken et al., 2010)	0.20	0.45	0.045	0.009	0.02	BBOA Mex
		0.25	0.56	4	1	1.8	Ground plume
ut i	(Calling and 2010)	0.20	0.60	3	0.6	1	Ground plume
bie	(Collier et al., 2016)	0.23	0.67	3.5	0.8	1.2	aircraft plume
am		0.25	1.25	4	1	0.8	aircraft plume
	(7hou at al. 2017)	0.18	0.88	8	1.4	1.6	no bb
	(Zhou et al., 2017)	0.32	0.95	6	1.9	2	bb inf
		0.30	0.90	6	1.8	2	bb plm
		0.25	0.75	0.06	0.015	0.02	Fir (diluted/cooled)
		0.21	0.68	0.07	0.015	0.022	pine burn
	$(U_{0} \text{ at al} 2010)$	0.20	0.56	0.05	0.01	0.018	Willow
	(He et al., 2010)	0.30	0.90	0.06	0.018	0.02	Wattle
		0.30	0.90	0.06	0.018	0.02	SugaCaneLeave
		0.30	0.08	0.05	0.015	0.2	Rice Straw
	(Horings at al. 2011)	0.25	0.67	4	1	1.5	роа
	(Herifiga et al., 2011)	0.25	0.50	4	1	2	5h aging
		0.15	0.50	13	2	4	start (oak)
	(Ortega et al., 2013)	0.20	0.50	50	10	20	aged (oak)
sed		0.04	0.05	250	10	220	start (pine)
-ba:		0.07	0.10	270	20	200	aged (pine)
-Ż	(Corbin at al. 2015b)	0.20	0.80	4	0.8	1	start
ato	(COIDITI Et al., 2013b)		0.83		0.05	0.06	flaming
poq			0.50		0.01	0.02	Filtered and Oxid
La	(Corbin et al., 2015a)		0.50		0.01	0.02	Oxidized
		0.25	0.50	0.04	0.01	0.02	Primary
		0.43	6.00	0.07	0.03	0.005	OH and UV exp.
		0.34	1.00	0.065	0.022	0.022	OH and UV exp.
		0.40	1.00	0.045	0.018	0.018	OH and UV exp.
	(Bruns et al., 2015)	0.34	1.00	0.065	0.022	0.022	OH and UV exp.
		0.40	1.00	0.045	0.018	0.018	OH and UV exp.
		0.23	1.00	0.048	0.011	0.011	OH and UV exp.
		0.20	1.00	0.04	0.008	0.008	OH and UV exp.
		0.25	1.00	0.048	0.012	0.012	OH and UV exp.

These paragraphs have been added to describe Table S1 in the supplement.

Table S1 shows  $CH_2O^+$  signals at m/z's 29, 30, and 31 from HR-ToF-AMS studies. It is possible to observe the low  $CH_2O^+$  contribution to m/z 30 with 30/29 ratios between 0.01-0.40. The high values of 0.4 – 6 were observed when exposing aerosols to OH and UV. We can also see that 30/31 30/29 ratios do not show variations during and after biomass burning events or during fresh and aged emissions (Ortega et al., 2013;Corbin et al., 2015b), suggesting there is not substantial  $CH_2O^+$  variability over the biomass burning process.

In this study, a large contribution of m/z 30 signal to the mass spectra was observed with both sPON and pPON with 30/29 ratios (4.38 and 1.42 respectively) and 30/31 ratios (35.0 and 8.5 respectively) higher than unity. Showing that a CH<sub>2</sub>O<sup>+</sup> interference at m/z30 would be unlikely.

For biomass burning the fragmentation tables need to be modified, which the authors do later in the paper. It does not make sense to me to run PMF on not-corrected data as was done in Section 3.3, when you know you are using incorrect data.

We agree on showing both solutions in the main manuscript is confusing. The PMF solution without modifying the fragmentation table has been moved to the appendix. Part of this paper is to explore different ways to run ME-2 and evaluate its performance under different conditions, hence we consider important to show this solution on the appendix.

The section on the FragPanel modification is very specific to AMS users and all the three bullet points cannot be understood by anybody else without explaining all the abbreviations and acronyms. Especially sentences like on page 7 line 238: time series of PON:mz30 were calculated with the equation PON:mz30=PON/mz30, where PON=m/z 46:30. This makes mathematically no sense. Again all of this need to be included in the instrument section together with the quantification of PON and clearly will increase the already large uncertainty in the PON quantification during the biomass burning event.

The following paragraphs have been added to the end of section 2.2.3 Multilinear engine 2 (ME-2):

PON may exhibit covariance with other types of OA, thus their inclusion in the source apportionment analysis may give a more complete factorisation and highlight their co-emission with other OA types. Therefore, a different experiment was designed by modifying the fragmentation table, through the AMS analysis toolkit 1.56, in order to identify a PON source. The fragmentation table contains the different chemical species measured by the AMS, with each row representing m/z for specific species and where the user can define peaks that exist in each species' partial mass spectrum and their dependency on other peaks (Allan et al., 2004). The following steps were performed to modify the fragmentation table:

• Time series of a new ratio named RON\_30 is calculated by RON\_30 = PON/mz30, were PON is the time series calculated in section 2.2.2 and mz30 is the time series of the signal at m/z=30 measured by the AMS.

• Using the AMS analysis toolkit; the fragmentation table is modified, in the column frag\_Organic at the m/z 30, by multiplying RON\_30\*30. See figure S4 in supplement for a screenshot of the fragmentation table.

• PMF inputs are generated to be used in SoFi software.

Figure S4 has been added to the supplement to show how fragmentation table has been modified.

I FragPanel													
Use the radio buttons to add or remove a wave to the table	font size	e <u>12</u>	old	Export frac Compare fra	) waves in table as .itx aq waves in two folder:	Simple recursion results in	<ul> <li>Define Org to include PAHs (typical)</li> <li>Define Org and PAH separately (rare)</li> </ul>						
		R29	<u> </u>	RON_30*3		ె							
CO2	mz	frag_air	fra	ag_CO2	frag_016	frag_water	frag_RH	frag_organic	frag_PAH	fra			
<b>V</b> 016	15	0.00368*frag_air						15,-frag_NH4[15]		frag	1 ^		
V water	16	frag_016[16],frag			0.39*frag_air[14]	0.04*frag_water[1	0.04*frag_RH[18]	0.04*frag_organiı					
RH	17	0.000391*frag_0				0.25*frag_water[1	0.25*frag_RH[18]	0.25*frag_organii					
	18	0.002*frag_016[1				18,-frag_air[18],-f	0.01*frag_air[28]	0.225*frag_organ					
V organic	19	frag_RH[19]				0.000691*frag_w	0.000691*frag_R	0.000691*frag_or					
delta02	20	20,-frag_organic[:				0.002*frag_water	0.002*frag_RH[1)	0.002*frag_organ					
	21												
sulphate	22												
✓ 503 ✓ H2SO4	23												
	24							24,-frag_sulphate			_		
	25							25					
	26							26					
	27							27		frag	1		
✓ NH4 ✓ NH4 17	28	28						frag_organic[44]					
	29	0.0064*frag_air[2						29,-frag_air[29]		frag	1		
	30	0.0000136*frag_;						RON_30*30			_		
I K	31							31,-frag_nitrate[3			_		
Use the listbox & buttons	32	32,-frag_sulphate									_		
to add or remove a wave	33	0.000763*frag_ai									- 1		
to the table. Update list	34	0.00402*frag_air									-		
frag_orgLessPAH	35										-		
-	36	0.00338*frag_air(									-		
Add/Remove Free ) ( such	37							37,-frag_chloride			Ŧ		
Add/Hemove Frag Wave										-			

Figure S4: Modifying fragmentation table to add PON to PMF analysis.

The description of PMF and the ability of PMF to resolve the biomass burning event:

In my opinion, PMF is pushed way too far in this manuscript given the quality of the data. It is well known that PMF has difficulties to resolve large individual peaks and this becomes very clear in this paper as well.

As mentioned to Referee 1, we agree that, during bonfire night, LV-OOA, COA and HOA may be mixed with BBOA concentrations. We mentioned in section 4.1 that this would be the case. Conclusions will be modified mentioning that even when using the methodology of first analysing the period before and after bonfire night and then analyse the bonfire night period, it was not possible to completely separate the OA sources. However, we consider that, while BBOA concentrations would be mixed with other OA sources implying a decrease on BBOA concentrations, the actual BBOA trend remains and BBOA factor is considered to be representative of biomass burning OA emissions. This is supported by the good correlation BBOA shows with  $b_{abs_470wb}$  r<sup>2</sup>=0.880, a marker of emissions from biomass burning. Thus we consider BBOA time series to be accurate enough to determine primary and secondary PON concentrations.

The following paragraph has been added to the end of section 4.1 OA source apportionment:

Here is shown the importance of performing OA source apportionment using different approaches in order to identify the best way to deconvolve OA sources. PMF and ME-2 source apportionment tools could not completely deconvolve OA sources during the bfo event. However, due to the high correlation between for  $b_{abs\_470wb}$  and BBOA\_2 ( $r^2 = 0.880$ ) we consider that while BBOA\_2 might not represent the total OA concentrations from the bonfire night event, it does represent the trend of OA emmitted from the biomass burning.

First of all this paper is clearly tailored to the AMS community, but it should at least be somewhat understandable to anybody else, especially when some of the main findings are related to the absorption of PON. None of the factors that are used such as HOA, COA or LVOOA are explained anywhere or even a reference given. What are those factors, how are they characterized, how do they relate to any of the other measured tracers and how do the ones determined in this manuscript compare to the AMS data base?

We have added a more understandable description of the OA sources with their respective references, The following paragraph has been edited in the section 3.3 OA source apportionment:

Two steps were involved in Test2\_ON: in step a, PMF/ME-2 were run for the event before and after the bonfire night (named as not bonfire event, nbf). In Step b, mass spectra from the solution identified in step a were used as TP, to analyse the bonfire-only (bfo) event. Finally, both solutions (nbf and bfo) were merged for further analysis. Different OA sources were identified in Test2\_ON (Fig. 5), five sources were identified during nbf event: biomass burning OA (BBOA), hydrocarbon-like OA (HOA), cooking OA (COA), secondary particulate organic oxides of nitrogen (sPON\_ME2) and low volatility OA (LVOOA). These sources are identified by characteristic peaks at their mass spectrum; BBOA, which is generated during the combustion of biomass, has a peak at m/z 60, related to levoglucosan (Alfarra et al., 2007); HOA, related to traffic emissions, presents high signals at m/z 55 and m/z 57 typical of aliphatic hydrocarbons (Canagaratna et al., 2004); COA, emitted from food cooking activities, is similar to HOA with a higher m/z 55 and lower m/z 57 (Allan et al., 2010;Slowik et al., 2010; Mohr et al., 2012); LVOOA, identified as a secondary organic aerosol, has a high signal at m/z 44 dominated by the  $CO_2^+$  ion (Ng et al., 2010); sPON ME2 has a strong signal at m/z 30 and it has been identified to be secondary as it follows the same trend as LVOOA (Figure 5.a). In the case of the bfo event six different sources were identified: BBOA, HOA, COA, LVOOA and two factors with peaks at m/z30, which is related to PON (Sun et al., 2012). These two PON factors may have different sources; one may be secondary (sPON\_ME2) and the other primary (pPON\_ME2) which has similar trend as BBOA (Fig. 5.b). Further details about pPON ME2 and sPON ME2 nature will be explored in section 4.2.

The next issue is that PMF was done in many different ways in this manuscript, many tests were performed and a lot of these tests were subsequently discarded. So why do you describe all the tests that have not worked. As it is written, this is very confusing and really hard to follow. Furthermore, two different PMF tests were actually used in the manuscript and they are very clearly different and the results that fit the story the best were used for no apparent reason. Even the number of BBOA factors changes between the two tests.

One of the objectives of this paper is to explore different ways to perform source apportionment, hence we consider important to mention the different tests perform. However, we agreed that mentioning both solutions in the main text is confusing. Hence, we have modified the manuscript by only explaining in the main text the optimal solution (Test2\_ON) and moving the other solution to the appendix.

It is very clear that PMF, even in the way as done here with separating the biomass burning event from the rest of the time series, cannot resolve the single event. In Figure 6 the combined increase of LVOOA, COA, and HOA is about twice as large as the combined BBOA\_2, sPON\_ME and pPON\_ME signals. One might argue that during bonfire night activities such as cooking and traffic might increased as well, but certainly not to such large extents. Especially the LVOOA signal, which is larger than the BBOOA signal, has to be from biomass burning as well. So it seems that the interferences in the biomass burning event are larger than the BBOA signal itself.

Given this large uncertainty and interferences on the PMF results, it seems clearly a step too far to try to separate sPON from pPON with two different methods that have large uncertainties and are not even fully consistent with each other. The I-CIMS measures some primary and secondary ON tracers, some of which are specific to biomass burning such as nitroaromatics, why have those measurements not been used to correlate with the sPON signal? It is not clear from the text on page 9, what compounds from the I-CIMS have actually been used, except some of the inorganic tracers.

This is the correct table, which is in the supplement: Table S4:  $R^2$  values between OA factors and CIMS measurements.

				BBOA			COA				sPON				LVOOA				pPON
Formula	Name	ALL	HSC	LC	bfo	WL	ALL	LC	bfo	WL	ALL	LC	bfo	WL	ALL	LC	bfo	WL	bfo
C4H6O2	methacrylic acid	0.89			0.92	0.53	0.64		0.77	0.48					0.78		0.82		0.52
C3H4O2	Acid_Acrylic	0.85			0.90	0.65	0.62		0.70	0.43				0.48	0.79		0.88		
H2COH2O	methylhydroperoxide	0.78			0.90		0.54		0.69						0.66		0.85		
C6H6O	Phenol	0.89			0.89		0.59		0.73						0.75		0.73		0.57
С7Н6О2	Benzoic acid	0.89		0.57	0.89	0.86	0.65		0.83	0.45		0.71		0.73	0.67	0.72	0.64	0.58	0.57
C2H5NO	Methylformamide	0.88			0.89	0.47	0.61		0.79						0.65		0.67	0.56	0.65
C2H3NO	Methyl isocyanate	0.89	0.49	0.44	0.89	0.71	0.55		0.66					0.50	0.85		0.88		
C5H10O2	Pentanoic acid	0.77			0.87		0.60		0.76						0.54		0.66		
HNO2	nitrous acid	0.81			0.86	0.66	0.59		0.84					0.57	0.61		0.66		0.70
CH2O2	formic acid	0.52			0.86				0.62						0.58		0.88		
C3H7NO	Dimethylformamide	0.80			0.85		0.59		0.76						0.56		0.63	0.60	0.63
C3H6O2	propionic acid	0.87		0.67	0.85	0.72	0.53	0.45	0.62			0.41		0.67	0.78		0.78	0.63	
C2H5N3O2	C2H5N3O2				0.83				0.77								0.59		
CHNO	Isocyanic acid	0.86		0.64	0.83		0.56		0.68			0.81			0.84	0.80	0.86		0.47
C4H6O4	succinic acid				0.83				0.71								0.60		
C6H6O3	trihydroxybenzene	0.83	0.48	0.72	0.82	0.85	0.62		0.79	0.42		0.75		0.71	0.59	0.82	0.54	0.59	0.49
C4H8O2	butyric acid				0.80				0.58								0.76		
C2H2NO3	C2H2NO3	0.61			0.79		0.48		0.56			0.49			0.63		0.90		
H02H2O	H02H2O	0.53			0.77				0.63								0.70		
CHN	Hydrogen cyanide	0.80		0.66	0.76	0.84	0.57	0.36	0.70			0.60		0.74	0.62	0.69	0.61	0.54	0.77
С6Н6О2	Catechol	0.73			0.73		0.44		0.56						0.63		0.62		
С7Н8О	Cresol	0.79			0.72		0.50		0.59						0.59		0.51		0.65
C3H4O4	Malonic acid				0.69				0.50								0.52	0.54	
С7Н8О2	guaiacol	0.63			0.62	0.78			0.45	0.43				0.62	0.58		0.57		
C2H4O3	Glycolic Acid				0.62		0.42		0.63										
CNO	anion isocyanate	0.66		0.61	0.61		0.48		0.50			0.81			0.74	0.76	0.74		
C3H7NO2	L-Alanine				0.54				0.64										0.65
* NO		0.40			0.63				0.59								0.58		0.46
* NO2					0.45	0.51			0.41				0.50				0.54		
* Nox					0.60	0.47			0.57								0.59		
* CO		0.79	0.55		0.81	0.67	0.64		0.80	0.42				0.48	0.58		0.56		0.78
* SO2					0.63				0.57								0.52		0.72
CINO3	Chlorine nitrate			0.45							0.45	0.69	0.53			0.66			
CINO2	nitryl chloride			0.47								0.74	0.52			0.67			
CI2	Chlorine												0.51						0.44
C6H5NO3	nitrophenol		0.41															0.55	

ALL = all dataset, LC = low concentrations, bfo = bonfire night, WL = winter-like.

The following paragraph has been added at the end of the section 4.3 OA factors and CIMS correlations, which supports the primary and secondary nature of PON:

during the bfo event, pPON\_ME2 showed high  $r^2$  values with carbon monoxide (0.78) as well as hydrogen cyanide (0.77), Methylformamide (0.65) and Dimethylformamide (0.63) which are typical primary pollutants related to combustion processes [(Borduas et al., 2015) and references therein]. sPON\_ME2 showed low correlations with ClNO<sub>2</sub> (0.52) and ClNO<sub>3</sub> (0.53). High  $r^2$  values were also observed during LC episode between ClNO<sub>2</sub> - ClNO<sub>3</sub> and LVOOA (0.67 - 0.66) and sPON (0.74 - 0.69) proving their secondary origin. Cl<sub>2</sub>, which has been previously identified to be related to both primary and secondary sources (Faxon et al., 2015), shows low correlations with pPON\_ME2 (0.44) during bfo event and sPON\_ME2 (0.55) during LC event.

# Minor Comments:

There are a large number of minor issues mostly about missing Tables, wrong numbering of Sections and typos in axis labels and such, so I only point out the two most obvious ones.

We are sorry for not updating the numbers and labelling in the last version of the manuscript before submitting it to ACPD. A full update of numbering to tables and figures has been performed.

- I mentioned that before, but all the references to other sections are wrong in the manuscript. Most importantly there are references to sections that don't even exist such as Section 3.4.1, in which supposedly the interference of m/z 30 is discussed. –

The interference of m/z 30 is being discussed in section 2.2.2

- Another glaring omission is Table 1 mentioned on page 9 line 325. This Table could be the most important evidence to support the separation of sPON from pPON, but is unfortunately missing.

This table is in supplement as Table S4.

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