



# Supplement of

## Simultaneous aerosol mass spectrometry and chemical ionisation mass spectrometry measurements during a biomass burning event in the UK: insights into nitrate chemistry

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## S1. Bonfire/firework locations during bonfire night 2014.

Locations of nine parks with main bonfire/fireworks during November 5<sup>th</sup> 2014.



Figure S1: Manchester map with locations of parks with bonfires/fireworks displays (red flames) and monitoring site (blue dot) at the University of Manchester. Map produced with Google Maps and location of bonfires was taken from [http://www.pocketmanchester.com/bonfire-night-2014-in-manchester/, accessed 03/05/2017].

#### S2. Literature review on lack of interference of CH<sub>2</sub>O<sup>+</sup> fragment to m/z30.

Table S1.  $CH_2O^+$  signals at m/z 29, 30 and 31 from HR-ToF-AMS data of previous studies. Comparison of m/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 study	4.38	35.00	0.08	0.35	0.01	sPON_ME2
	This study	1.42	8.50	0.06	0.09	0.01	pPON_ME2
	(Aikan at al. 2010)	0.16	0.32	0.05	0.008	0.025	pine burn
	(Alkell 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
ent	(Collier et al. 2016)	0.20	0.60	3	0.6	1	Ground plume
biq	(Collier et al., 2010)	0.23	0.67	3.5	0.8	1.2	aircraft plume
ar		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
	(21100 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
	(He et al. 2010)	0.20	0.56	0.05	0.01	0.018	Willow
	(116 61 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 of al. 2011)	0.25	0.67	4	1	1.5	роа
	(Herniga 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)
pa		0.07	0.10	270	20	200	aged (pine)
-Ż	(Corbin et al. 2015b)	0.20	0.80	4	0.8	1	start
ato	(COIDITI ET al., 2013b)		0.83		0.05	0.06	flaming
po d			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.

 $CH_2O^+$  identification at m/z 30 is accompanied with signals at m/z 29 and m/z 31

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 and 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., 2015a;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  $CH_2O^+$  interference at m/z30 would be unlikely.

#### **S3.** Aethalometer correction.

Aethalometer measurements (absorption coefficients,  $b_{ATN}$ ) need to be corrected from two main effects: filter loading (R) and scattering correction (C) that compensates for the multiple-scattering effects from the matrix. There are different methods to correct aethalometer data (Weingartner et al., 2003;Arnott et al., 2005;Schmid et al., 2006). Coen et al. (2010) proposed a new method through a critical analysis of the effectiveness of the other methods, which involves corrections based on absorption and scattering measurements. In this study, wavelength-dependent scattering measurements were not available (A Photo Acoustic Soot Spectrometer was used to measure aerosol optical absorption coefficients. However, the scattering channels failed to report data during the bonfire event), thus the Weingartner method (Weingartner et al., 2003) was used to do these corrections.

The light attenuation (ATN) is defined by equation S1, where  $I_o$  is the intensity of the incoming light and I is the remaining light after passing through the filter.

$$ATN = -100 * \ln\left(\frac{I_0}{I}\right)$$
(S1)

The attenuation cross section ( $\sigma_{ATN}$  in m<sup>2</sup>.g<sup>-1</sup>) is calculated using the equation S2, where 14625 [m<sup>2</sup>.g<sup>-1</sup>] is the mass specific attenuation cross-section proposed by the manufacturer and  $\lambda$  is the wavelength in nm.

$$\sigma_{\text{ATN}_{\lambda}} = \frac{14625}{\lambda} \tag{S2}$$

The absorption coefficient ( $b_{ATN}$ , Mm<sup>-1</sup>) was calculated using equation S3, where BC is black carbon [µg.m<sup>-3</sup>] measured by the aethalometer.

$$b_{ATN_{\lambda}} = BC_{\lambda} * \sigma_{ATN_{\lambda}}$$
(S3)

 $b_{ATN \lambda}$  values need to be corrected by calculating  $b_{abs}$  (corrected absorption coefficient).

$$b_{abs\_\lambda} = \frac{b_{ATN\_\lambda}}{C^{*R}}$$
(S4)

Where C is a parameter for scattering correction and R, a wavelength dependent parameter, is related to the filter loading effect.

C is calculated as the slope of  $b_{ATN_{630}}$  from aethalometer and  $b_{abs_{630}}$  from MAAP, using the values with ATN<10% (Calculating C with this approach the effects from filter loading are minimized). The Aethalometer does not measure at 630 wavelength thus  $b_{ATN_{630}}$  is calculated using equation S6, where the absorption Ångström exponent ( $\alpha$ ) is calculated using equation S5.

$$\alpha = \frac{\ln\left(\frac{b_{ATN_{470}}}{b_{ATN_{950}}}\right)}{\ln\left(\frac{950}{470}\right)}$$
(S5)

$$b_{ATN_{630}} = b_{ATN_{660}} * \left(\frac{630}{660}\right)^{-\alpha}$$
(S6)

C represents the slope of  $b_{ATN_{630}}$  from aethalometer vs  $b_{abs_{630}}$  from MAAP. Following this method, a value of C = 3.16 was calculated.

The shadowing parameter (*f*) is determined, similar to other studies (Sandradewi et al., 2008;Sciare et al., 2011;Ji et al., 2017) as the average of  $b_{ATN}$  ratios after and before filter changes for the complete dataset in order to minimise the difference before and after filter changes. The f values obtained were  $f_{470} = 1.49$  and  $f_{950} = 1.28$ .

R is calculated with the following equation:

$$R = \left(\frac{1}{f} - 1\right) \frac{\ln(\text{ATN}) - \ln(10\%)}{\ln(50\%) - \ln(10\%)} + 1$$
(S7)

Finally, with C and R being determined, the corrected absorption coefficients  $(b_{abs_{\lambda}})$  are calculated with equation S4.

#### S4. Aethalometer model.



Figure S2: Absorption coefficients (b<sub>abs</sub>) for wood burning and traffic.

#### S5. Back trajectories for the different pollutant episodes.

Hysplit model was used to run back trajectories, with 48 hrs of duration and three different hights (0, 205 and 500 m about ground level), for the episodes with different pollutant concentrations: an event with high secondary pollutants is observed from October  $30^{th}$  – November  $1^{st}$ ; an event with low concentrations from November  $1^{st} - 3^{rd}$ . Bonfire night from November  $5^{th} - 7^{th}$ ; an event with high primary emissions from November  $8^{th}$  - $10^{th}$ .



Figure S3: Back trajectories run for events with high secondary pollutant concentrations (a), low pollutant concentrations (b), bonfire night (c) and winter-like event (d).

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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_organi						
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<b>F</b> 7	18	0.002*frag_016[1			18,-frag_air[18],-f	0.01*frag_air[28]	0.225*frag_orgar						
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### S6. Fragmentation table to add PON to PMF analysis.

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

## S7. OA source apportionment.

PMF and ME-2 source apportionment analysis was performed following the strategy proposed by (Reyes-Villegas et al., 2016). A series of solutions were run under different conditions in order to determine the best way to deconvolve OA factors. PMF was run with f-peaks from -1.0 to 1.0 and steps of 0.1. ME-2 was run using different a-values to partially constrain the solutions (table S1), using mass spectra (BBOA, HOA and COA) from Young et al. (2015a) and Crippa et al. (2013) as target profiles (TP). Figure S5 shows the labelling used to identify the different runs performed with ME-2 and table S2 shows the different a-value combinations used to explore different solutions.



Figure S5: Labelling used to identify runs.

### Table S2: List of ME-2 runs

Run	Run
B5H2C5	H1C3
B3H1C3	H2C5
B3H1C5	B3H1
B5H1C5	B5H2
B5H1C3	

#### S7.1 Strategy to select the solution that best apportions OA sources.

The OA source apportionment was performed using different f-peaks when running PMF and different a-values when running ME-2 (table S2) looking at solutions with 4, 5 and 6 factors,. These solutions were explored comparing their residuals and Q/Qexp for m/z's and time series; Total Q/Qexp and total residuals; diurnal profiles and trilinear regression (Reyes-Villegas et al., 2016). Looking for solutions with low residuals and Q/Qexp values. Trilinear regression is performed between BBOA, HOA and COA and NOx, since these three OA sources and NOx are related to combustion sources. With trilinear regression analysis, partial slopes should be positive as we are working with aerosol concentrations. Moreover, COA partial slope should be close to zero due to its low contribution to NOx. The chi square value from multilinear regression is used as goodness of fit, thus the lowest the value the best correlation between the different sources.

Here the analysis carried out to all the dataset is explained in detail.

Step 1. PMF runs looking at 4-factor solutions with f-peaks from -1.0 to 1.0 and steps of 0.1. One solution is chosen to be compared with ME-2 solutions.

Step 2. ME-2 runs looking at 4-factor solutions with different a-values using TP from Paris and London.

Step 3. Two solutions from step 2 are chosen together with the PMF solution, form step 1, to be the three 4-factor solutions to use in the further comparison.

Step 4. Repeat steps 1 to 3 to look at 5-factor and six-factor solutions to finally compare the 9 solutions.

Step 5. Choose one solution that better separates, according to this analysis, OA sources. Perform this analysis for the four tests mentioned in table S2 in order to have one solution for each test.

These steps were used to explore solutions for the different tests performed, generating more than 60 different plots that were analysed. Here, in order to avoid making an overly massive supplement material, only the final comparison between solutions from the different tests performed is shown (Section S7.2). Table S3 shows the chosen solution for each one of the tests performed.

### S7.2 Chosen solutions for the different tests.

	Ana	lysis	So	lution	Strategy					
ID	а	b	a	b	From solution a to b					
Test 1	° all		pH1C3_5all							
Test 2	* nbf	bfo	pB3H1C3_5nbf	<sup>×</sup> nB3H1C5_6bfo	nbf mass spectra were used as TP to analyse bfo dataset.					
Test 3	+ bfo	all	PMF_6_0.7	<sup>∆</sup> bB5H2C5_5all	bfo mass spectra were used as TP to analyse all dataset.					
Test 4	nbf	all	pB3H1C3_5nbf	×nH2C5_5all	nbf mass spectra were used as TP to analyse all dataset.					
Test 1_ON	° all		wB3H1_ON_5all							
Test 2_ON	* nbf	bfo	pH1C3_ON_5nbf	nB5H1C3_ON_6bfo	nbf mass spectra were used as TP to analyse bfo dataset.					

Table S3: Tests done to determine the solution that better deconvolves OA factors.

<sup>o</sup> all = the whole dataset was analysed: 29/Oct/2014-10/Nov/2014

\* nbf = not bonfire event: from 29/Oct to 05/Nov 15:00 and from 06/Nov 06:35 to 10/Nov/2014

<sup>+</sup> bfo = bonfire only event: 05/Nov 15:00 - 06/Nov 06:35

 $^{\times}$ n = mass spectra from analysis a (nbf) were used as TP in the analysis b.

 $^{\Delta}$ b = means mass spectra from analysis a (bfo) were used as TP in the analysis b for test 3.

ON means the tests were performed after modifying the fragmentation table to determine a PON source.

When doing two analyses with ME-2 (in the case of tests 2, 3 and 4), in analysis "a" mass spectra from London (Young et al., 2015b), labelled as "w" and Paris (Crippa et al., 2013) labelled as "p", were used as target profiles (TP). PMF runs were explored with different fpeak values ranging from -1.0 to 1.0 with steps of 0.1.



Figure S5: Comparison of the chosen solution for the four tests performed. Time series for the complete dataset (a), time series with a close up to y-axis to show low concentrations (b), time series during bonfire night event (c) and mass spectra.

#### S7.3 Comparison of different solution tests.

Here, the different tests performed without modifying the fragmentation table (test 1, test 2, test 3, and test4) and modifying it (test1\_ON and test2\_ON) are compared in order to determine the test that better separates OA factors.

The analysis was carried out by comparing the residuals and Q/Qexp for m/z's and time series for all the dataset (figure S6) and more into detail for the bonfire night (figure S7); trilinear regression between BBOA, HOA and COA with NOx (figure S8) and diurnal profiles (figure S9).

Analysis without modifying the fragmentation table is the first comparison performed, where test 2 resulted to be the test that better deconvolved OA factors with; low residuals (figures S6 and S7) and low chi square, used as a goodness of fit (figure S8). Then test1\_ON and test2\_ON were performed in order to determine the best way to deconvolve OA sources including organic nitrate factors, where test 2 showed to be a better way to deconvolve OA sources compared to test1\_ON.



Figure S6: Comparison of the different solutions for all sampling period.



Figure S7: Comparison of the different solutions for the bfo event.



Figure S8: Trilinear regression between OA sources and NOx.



Figure S9: Diurnal profiles.

#### S8. Primary (pPON) and secondary (sPON) organic nitrate estimation.

The slope from a linear regression between PON, obtained from 46:30 ratios analysis in Section 2.2.2 in manuscript, and BBOA was used to calculate primary and secondary organic nitrate. Blue circles show the period where the slope between PON and BBOA was calculated (Section 4.2 in manuscript).



Figure S10: Time series used to calculate the slope between PON and BBOA



Figure S11: Time series of pPON and sPON for the whole period.



Figure S12: pPON\_ME2 and sPON\_ME2 obtained from ME-2 analysis.

Two methods have been used to determine primary and secondary PON. In the following plots we can see primary PON comparison has a good correlation with a pearson value of 0.7 while secondary PON comparison shows a different behaviour between them.



Figure S13: PON comparison for the two methods used.

## S9. r<sup>2</sup> values between OA sources and CIMS measurements.

Table S4 show the  $r^2$  values between the OA factors and CIMS measurements, for the different analyses; ALL, LC, bfo and WL. Only  $r^2$  higher or equal to 0.4 are displayed.

Table S4: R<sup>2</sup> 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
C7H6O2	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
C6H6O2	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	
C7H8O2	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.

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