## Major Comments

1) Both referees suggested that further citations of previous work are required. Thank you for the incorporating several of the suggested references. However, I agree with referee 2 that this discussion should be included in the introduction, rather than scattered through the paper. Please add this discussion of previous SP-AMS measurements that integrate BC signals into the PMF in the introduction and highlight the differences of your approach. An additional study that you should consider including is Rivellini et al., (2020).

I have added the further citations of previous studies detail in the introduction section.

2) Re: Referee 2 comment 11: Same as the referee, I am unable to follow the argument that supports the use of the term HULIS to describe this factor and I also wonder if MO-OOA would be a better description. While I understand that HULIS has been identified in atmospheric aerosol, in my understanding it is essentially an operational definition (similar to various measurements of BC and MO-OOA) that varies depending on the analytical instrumentation used. Moreover, HULIS generally refers to higher molecular weight species. As AMS measurements provide extremely limited information on molecular weight, I tend to agree with the referee that a terminology such as MO-OOA that has been defined based on AMS measurements is more appropriate. As stated by the referee, identification of this factor as HULIS requires "…other co-located measurements in this work can provide evidence that the HULIS factor has some specific chemical features that cannot be described as MO-OOA."

Thank you so much for your suggestion. I have replaced HULIS with MO-OOA factor. Please see the main manuscript.

3) It strikes me that the mass spectra of the fullerene region differ significantly between the different factors. Does this provide any additional insight/information particularly when compared with the Onasch et al., (2015) results?

Yes, the mass spectra of HOA+Fullerene region have different signal concentrations between different factors. But the fifth factor is named 'HOA+fullerene' because it is heavily weighted by HOA and Fullerene showing a peak at m/z 720 ($C_{60}^{+}$), implying polycyclic aromatic hydrocarbons can transform into soot containing HOA+Fullerene during combustion (Wang et al., 2015, Wang et al., 2016; Reilly et al., 2000). The mass spectra of HOA+Fullerene region have different signal concentrations between different factors (3a). But this factor is considered as HOA+Fullerene due to its high signal strength and most prominent distributions i.e., $60\text{x}10^{-6}$ as compared to other factors (hydrocarbon-Like OA = $2.5\text{x}10^{-6}$, domestic burning = $2.5\text{x}10^{-6}$, BBOA = $8\text{x}10^{-6}$ and BC and MO-OOA = $3.0\text{x}10^{-6}$). Depending on the different phase of combustion, HOA+Fullerene was typically not associated from the traffic source (diesel), Although the mass spectral pattern varies, the signals of most factors are too low to get any meaningful information with any confidence. However, in Onasch et al., 2015, the variations within mass spectral region of fullerene signals between different factors is negligible, but in our case all the factors except 'HOA+Fullerene' factor have very low signals concentration, so this factorisation technique provide obvious and separate HOA+Fullerene factor. The time series of HOA+Fullerene also show the very high concentration of 8000 Counts/s only during the bonfire event, that is why it is the bonfire source (3b).

4) Re: Referee 1 comment 3: I agree with the referee that further information regarding the impacts of the uncertain RIE is required. How was it determined that the RIE did not affect the PMF analysis? Please describe this in the manuscript.

The RIE, as defined by Allan et al. (2004, doi: 10.1016/j.jaerosci.2004.02.007), is a constant factor applied to the signals as part of the conversion from a signal in the mass spectrometer to an ambient mass concentration. Because this is a purely multiplicative operation, this will affect all data and associated errors equally and therefore the factors derived in the PMF model described by Ulbrich et al. (2009) will simply by multiplied by the exact same amount. So, when PMF factors are derived using data that has not had an RIE applied (and corollary to this, other multiplicative factors such as ionisation efficiency, collection efficiency and inlet flow rate), the relative contributions of the different factors as a function of time will be exactly the same as if data that had had this applied. The only difference is that the absolute units of the factors are as an arbitrary mass spectral response (in s-1) rather than an ambient mass concentration (in µg m-3). The only potential difference is in the relative signal strengths of organic and elemental carbon, which will likely have different RIE values (Onasch et al., 2012). This is explored in more detail in section 4.2 where the relative contributions of the different factors to the HR-SP-AMS signal and the BC mass fraction is explained.

## Minor Comments

5) Re: Referee 1 comment 4: Please include the time resolution of the measurements (e.g., 10 s, 1 min, etc.). Additionally, from my understanding, the instrument was operated in different configurations (V mode, W mode, extended m/z range V mode as well as with and without the catalytic stripper). It would be useful to explicitly state the time coverage of the measurements (for example every 3rd minute for half hour of every hour).

I have included this in Instrumentation section from lines 311-320.

6) Re: Referee 1 comment 5: Please include this in the main manuscript.

Thank you so much for your comment. The reason for 1000 m/z range of this PMF analysis is that above m/z 1000, the signals were too small.

7) Re: Referee 2 comment 7: Please include the both the mass spectra (Fig. 4) and the concentration time series (Fig. 3) in the same plot. Line 222 states that Fig. 4 shows both of these, however, only the mass spectra are shown.

Figures updated as follows:

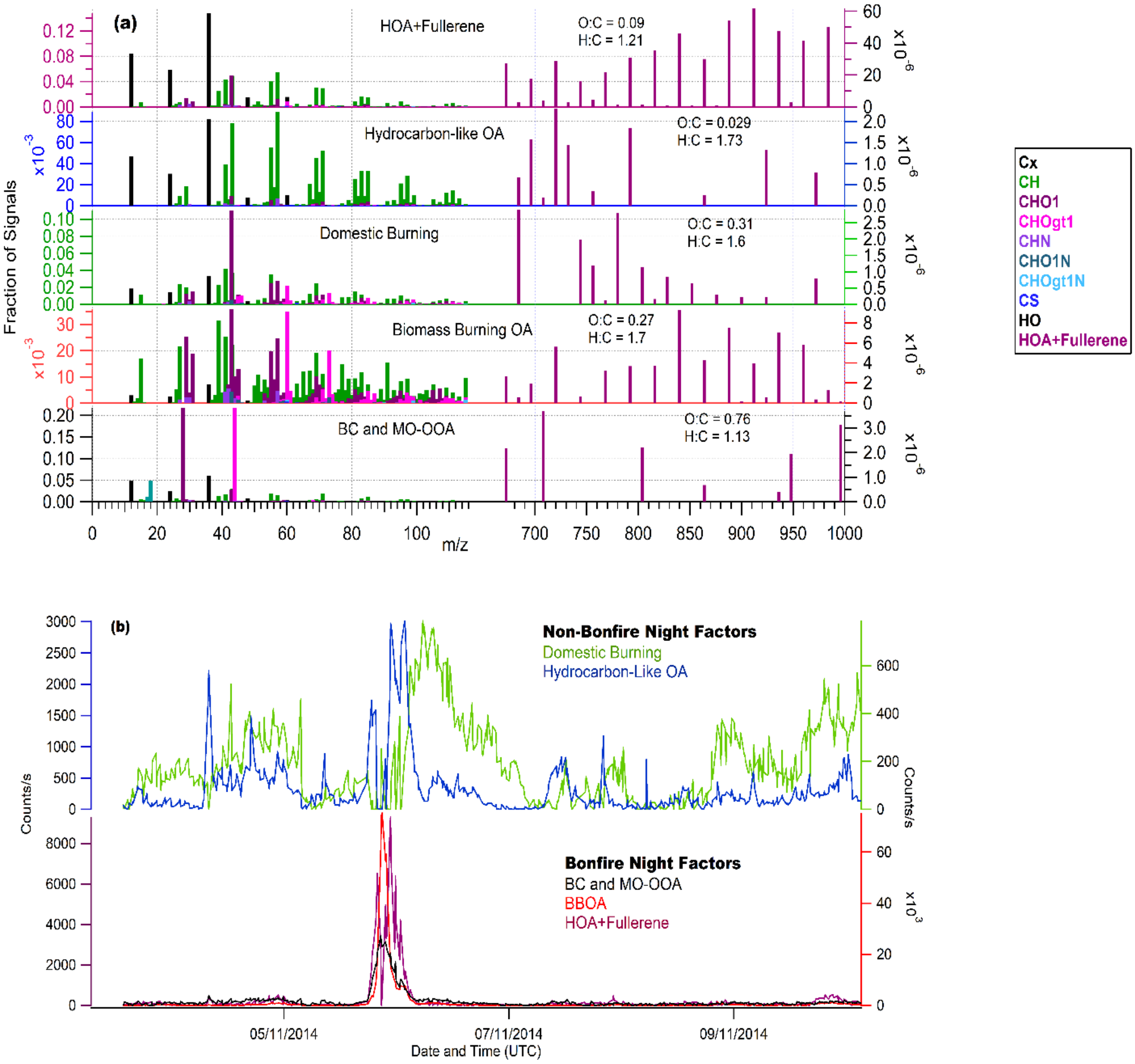


Figure 3. PMF solution (a) Five factors source profile (BC and MO-OOA, BBOA, Domestic burning OA, Hydrocarbon-Like OA and HOA+Fullerene) (b). The time series of non-bonfire and bonfire night factors obtained.

8) Re: Referee 2 minor comments: Please define eBCwb, eBCtr when they are first used (line 206).

Applied changes. eBCwb (Black carbon emissions from wood-burning) and eBCtr (eBCtr (Black Carbon emission from traffic).

9) Lines 22-24 "Positive matrix ….able to do." This sentence is very difficult to read. Please rewrite to improve clarity.

Positive matrix factorisation was applied to positively discriminate between different wood-burning and bonfire sources for the first time, which no existing black carbon source apportionment technique is currently able to do.

10) Lines 66- 69: "In this study … metal nanoparticles." This should be in the experimental section, not the introduction.

Done added in experimental section.

11) Sects 2.1 and 2.2: I suggest renaming to better reflect the contents of the section. 2.1 is focused on both the site and the supporting measurements, while 2.2 is only focused on the SP-AMS rather than on all the instruments.

Renamed.

12) Sect 2.2: The text of this section needs to be revised to more clearly describe the instrument as there are several confusing/incorrect statements. For instance (not the only case), line 118 "…the new vaporiser is designed, to detect the vaporized species through electron ionization for the generation of the chemical ion…" is problematic as it implies that the vaporizer detects the species and the term chemical ion is poorly defined.

Revised.

13) Line 131: I don't understand why the term isotopes is in parentheses or why it is included in this section. Please explain or revise. The fragments should be identified as ions.

Revised and corrected.

14) Line 140: All the signal is integrated in UMR, not "can." Also "robust" in what way"

Corrected.

15) Lines 151-153: Please spend a few sentences describing the Corbin et al approach and how this approach is different. The short comment by Corbin may be of help here.

Revised text as follows:

As with all PMF analysis, error estimates have to be provided but because of the lower signals and the combination of different data retrieval method used for the fullerene signals (UMR rather than HR), greater emphasis had to be placed on these signals. Corbin et al. (2015) presented a very detailed error model for HR data employing a Monte Carlo method to explore multiple sources of error. But because UMR was used in this instance, we were unable to apply this method, so we took an empirical approach. This was done by applying an additional 'model error' to the error matrix, i.e., an error term proportional to the signal intensity in addition to its square root, as per the standard AMS error model (Ulbrich et al., 2009; Comero et al, 2009). The model error value was increased from 0 to 0.10 to down weight the larger signals and place more of an emphasis on the fullerene signals. According to Corbin et al., 2015, the peak width 'w' is predicted during the peak fit integration from an empirical fit to the data. This 'w' prediction has a linked proportional uncertainty σw = w. In that data set, σw = w was 2.5%, which was independently treated as 2 or 3% uncertainty in the isolated peaks heights, so these two can be combined in quadrature. And for the isolated peaks the value for the total percentage uncertainty is about 5% which is conceptually equivalent to 0.05 model error. This is comparable to the 0.1 model error u σw = w sed here. Along with placing greater emphasis on the smaller fullerene signals, the application of this model error also increased the number of "weak" variables, defined as having SNR below 2 (Paatero and Hopke, 2003; Ulbrich et al., 2009), which were down weighted by a factor of two. No variables were "bad" in the sense of having SNR < 0.2 (Paatero and Hopke, 2003).

16) Line 156: Please describe more completely how this reflects that the "data and uncertainties have a lognormal distribution." I did not find this information in the supplement.

Revised text in the main manuscript;

17) Figure 1: Why is Sr included in this time-series? It is not referenced in the text.

I have added the details about Sr peak in the text.

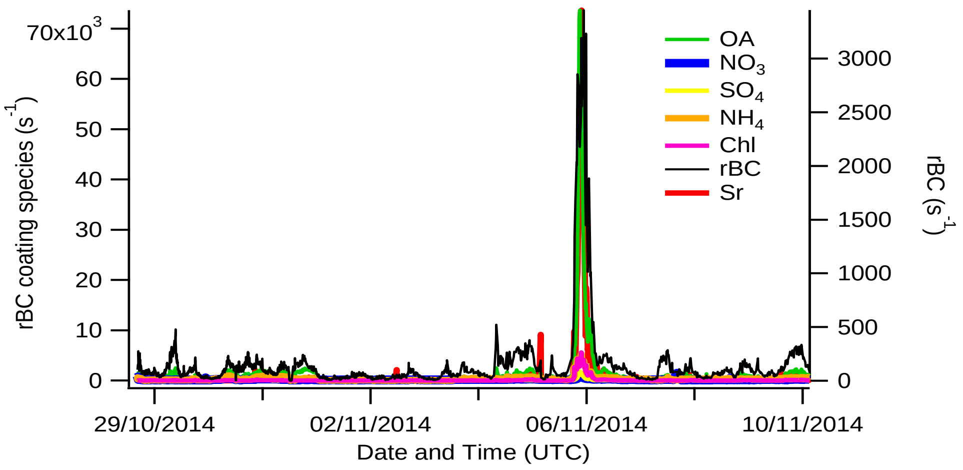
"During the periodic stagnation weather phenomenon, the very high concentrations of BC and Sr was also observed with the signal of 3400 s-1 and 53 s-1 respectively, during the bonfire event at 10:20 pm and 9:50 pm, as compared to (BC concentrations of 100 s-1 – 500 s-1 before Bonfire and 250 s-1 - 300 s-1 after bonfire night) and (Sr concentrations of 5 s-1 and 1 s-1 before and after bonfire night)".

18) Line 199: From the figure (2b) it appears that rBC starts increasing first. Do you mean to say that the peak signals are at different times? Please clarify.

The peak signals for rBC and organic aerosols were at different times such as the concentrations of rBC started increasing first at 07:54 pm followed by organic aerosol concentrations increasing at 8:30 pm to 9:00 pm (highest).

19) Figure 2: Please standardize the x-axes on the panels to the same range and format (date or date +time). Figure 2b: Please use standard AMS colors and remove the negative charge from the legends.

Done.



20) Line 226: Please describe what you mean by "mixing" so readers who are not familiar with PMF analysis more clearly understand the results.

In that case, only two unambiguously bonfire night sources of BC were identified, with a degree of 'mixing' between the bonfire night factor and traffic noted in the HOA factor, which means the factors share similar common features in their time series or profiles.

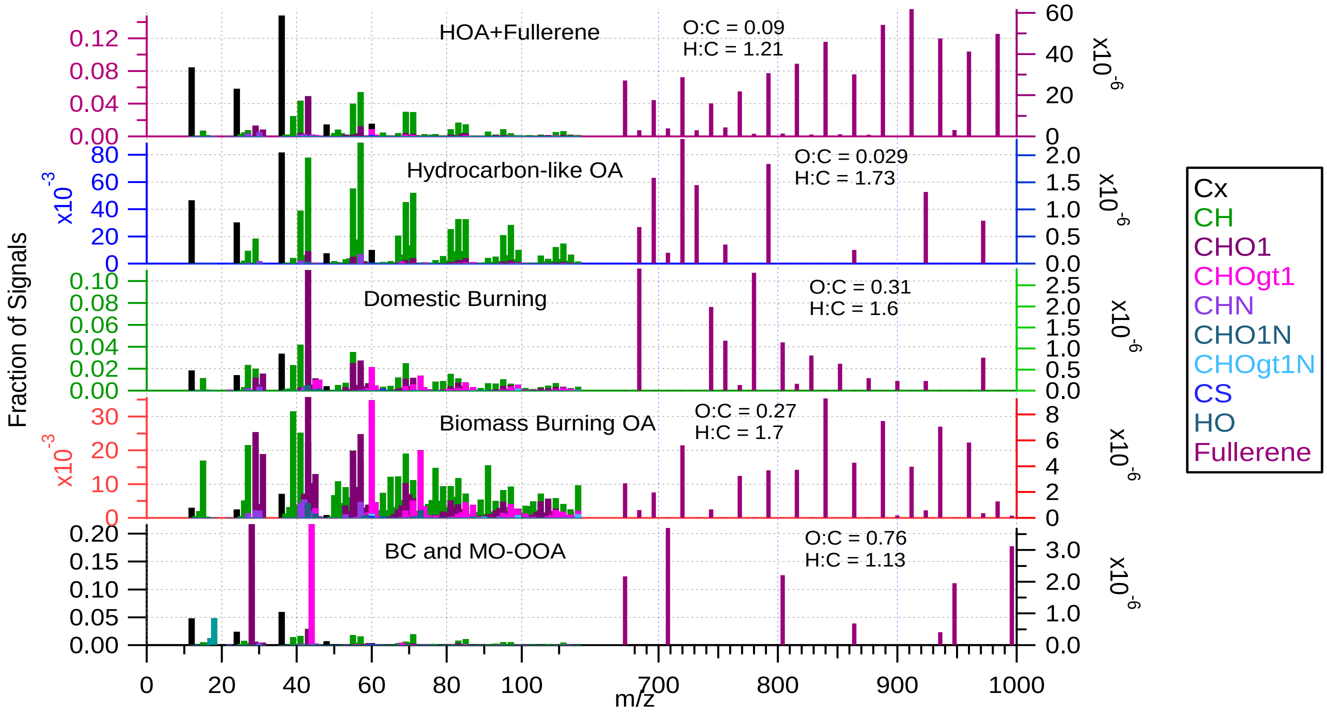
21) For the PMF results without fullerenes included, were the same optimization steps taken as for the PMF performed on the data with the fullerenes (lines 232-236)? Was a 6 factor solution explored and did that improve the results?

Firstly, the factorisation was performed without the inclusion of fullerene signals in the data matrix by choosing the f-peak between -2.0 and +2.0 with fpeak interval of 0.2 and model error of 0.10.

Yes, the six-factor solution was also explored and explained in supplementary materials. The 6-factor solution has two 'split' factors representing the same emissions. These are factor 2 and factor 4 in figure S7b and represent domestic wood burning sources because their peaks were evident before and after the bonfire night event (fig. S7a, S7b).

22) Figure 4: Please fix the y-axes so that the tick marks do not overlap and that the values can be clearly read. Please increase the resolution of the legend and replace "gt1" with ">1." Please color HO and Cx differently. In the legend, point out that the magnitude of the fullerene signal varies widely between the factors.

Done. Please see below;



23) Line 345: I think you mean Figure 6 here, not 7.

Corrected.

24) Line 381-382: This sentence confuses me as it seems to imply that this work and Reyes et al suggest that fireworks are a source of soot. Perhaps you mean "consistent with the results reported here and in Reyes at all that fireworks are not soot sources."

Corrected, please see below:

The firework tracer Sr has shown some correlation with pPON and BBOA, but their peaks occurred at slightly different times. So, in spite, of the high correlation, this implies that they are not identical, and 'Sr' is behaving as a separate factor when subject to PMF analysis. It could be that if the firework display occurred at the beginning of the bonfire event their emissions maybe coincident with the pyrolysis emissions on bonfire begin to be lit, as distinct from the smouldering emissions later (Haslett et al., 2017). But without specific knowledge of the timings of the events that contribute to these emissions, it is difficult to reach firm conclusions. This, coupled with the fact that Sr could not be associated with any of the factors in this study, would be consistent with Reyes et al. (2018) in concluding that fireworks are not a significant source of the material observed.

25) Sect. 4.2: I think your figure numbering in the text versus the figures themselves is off. Figure 8 in the text seems to be what is actually labeled figure 7 and likewise for 8/9.

Corrected.

26) Supplement: Please use the HR coloring on the mass spectra as was done in the main text.

Corrected.

References

Onasch, T. B., Fortner, E. C., Trimborn, A. M., Lambe, A. T., Tiwari, A. J., Marr, L. C., Corbin, J. C., Mensah, A. A., Williams, L. R., Davidovits, P., and Worsnop, D. R.: Investigations of SP-AMS Carbon Ion Distributions as a Function of Refractory Black Carbon Particle Type, Aerosol Sci. Technol., 49, 409–422, https://doi.org/10.1080/02786826.2015.1039959, 2015.

Rivellini, L.-H., Adam, M. G., Kasthuriarachchi, N., and Lee, A. K. Y.: Characterization of carbonaceous aerosols in Singapore: insight from black carbon fragments and trace metal ions detected by a soot particle aerosol mass spectrometer, Atmospheric Chem. Phys., 20, 5977–5993, https://doi.org/10.5194/acp-20-5977-2020, 2020.