Response to the comments of co-editor

We thank the referee for the valuable comments which have greatly helped us improve the manuscript. Please find below our responses (in black) after the referee comments. The changes in the revised manuscript are written in *italic*.

Page 3: I agree with one of the reviewers that papers in preparation should not be cited (to be consistent with the accepted practices). Please remove reference to Lu et al.

Done. We remove the reference. (P3, L33)

Page 16, Line 33: missing parenthesis and comma

Done. We revised it (P16, L33-34). "Here we investigate the apportionment of eight ions associated with compounds of interest: C6H10O5 (approximately assigned to levoglucosan), C7H7NO4 (methyl-nitrocatechol), C9H10O5 (syringic acid), C8H8O4 (vanillic acid), C8H6O4 (phthalic acid), C5H6O4 (glutaconic acid), C7H8O4 (tetrahydroxy toluene) and C7H10O5 (pentahydroxy toluene)."

Figure 1: amu is an outdated unit for molecular weight; it is not used for the mass-to-charge ratio in mass spectrometry. The quantity m/z is typically viewed as dimensionless so I would change the X-axis label accordingly, as you did in other mass spectra figures. While you are changing this figure, the yellow font used for the COA factor label is very hard to see, a different color is recommended. Also this figure appears hazy on my screen (the rest of the figures do not have this problem). Perhaps there is a way to export it at a higher resolution.

Done. We changed the label of X-axis to m/z, color of the COA factor, and the resolution of the figures (Fig.1 and Fig. S1).

Figure 3: The figure legend implies that this plot uses H:C ratios for the ions as opposed to neutral species. I think VK plots for the neutrals instead of ions are easier to interpret. Perhaps it is not even an issue given how few H+ adducts (just one?) you observe. Since you are providing formulas of the neutral compounds in the text, why not calculate the H:C for the neutral species? The same comment applies to the rest of the VK plots.

Done. We agree with the comment that the neutrals are easier to interpret and are the whole story. We change the relevant texts and the captions of the figures (Fig. 3, 4, 5, and 9).

Figure 10: the labels is this figure are very small relative to the figure size, I am worried they will be invisible in the final print. IS there any way to increase them? (In general, labels in many of the figures in this paper are disproportionally small, but in this figure it is extreme).

Done. We replot the figure with increasing the size of the labels.

Figure 13 and Figure 14: I would fix capitalization in the Y-axis title (flux -> Flux)

Done. We revised the Y-axis to "Mass Flux" in Fig. 2, 3, 6, 13, 14 and S6. We noted the inconsistent in terms of ag/s versus ag s⁻¹. Then, we use ag s⁻¹ everywhere to be consistent with μ g m⁻³. In this case, the Fig. 2, 3, 5, 6, 10, 13, 14, S6, S8, and S10 are revised.

Bozzetti et al. (2016) reference is missing page numbers

Done. We revised it.

Veres et al. (2010) reference is missing page numbers

Done. We revised it.

Zhang et al. (2017b) reference is missing page numbers

Done. We revised it.

The use of DOI numbers is not very consistent – some have Doi in front of them, and some have nothing.

Done. We checked all the reference to make them consistent.

Figure S1, S3, S5: dat -> date

Done. We revised it in Fig. S1, S3 and S5.

Figure S6: Hourly -> Hour of the Day

Done. We revised it in Fig. 3 and Fig. S6.

Table S2: I think the right column title should be "Molecular Weight of the Neutral (Da)". I actually do not see how the calculated molecular weight is useful (because you can get it from the formula) – if you list values you should list the observed m/z values for the Na+ and H+ adducts. This way you could also include ions that you observed but could not assign.

Done. We agree with the comment to use the observed m/z values in the right column and change the title to "Measured m/z". These ions are from the cluster analysis not the whole fitting ions, so the unknown ions are not included.