

Author's response:

We thank the Referees for the careful revision and comments which helped improving the overall quality of the manuscript.

A point-by-point answer (in regular typeset) to the referees' remarks (in the *italic typeset*) follows, while changes to the manuscript are indicated in **blue font**.

In the following page and lines references refer to the manuscript version reviewed by the anonymous referees.

Anonymous Referee #4

Received and published: 8th March 2017

General Comments:

This paper presents results from an offline analysis of aerosol samples collected in Marseille France using HR-AMS for the organic and other offline techniques for the inorganic, elemental carbon, and metals analyses. Using PMF, source apportionment was carried out and the results from the offline study were compared to PMF analysis of a smaller sample set of on-line AMS data. While the two time periods did not overlap between the offline and online data sets, good comparisons were observed for the different organic factors between offline and online AMS. The authors found increased BBOA in the winter and that OOA was dominant in the summer. The authors also observed changes in the levoglucosan:BBOC ratio over the course of the winter-spring months that was opposite what would be expected if it was driven only by changes in photochemical processing from the winter to the early spring. They attribute the observed change in the BBOC to changing sources with more domestic wood burning in the cold winter months and more agricultural biomass burning in the early spring. Overall, the analysis is thorough and the conclusions are explained well and match previous studies in similar areas. I recommend publishing the manuscript after some minor revisions.

We thank anonymous referee #4 for the review and the comments.

Specific Comments:

- 1. A blank was run between each sample with ultra-pure water to monitor memory effects. However, the minimum concentration needed to make aerosols that will dry down to a size observable in the AMS is relatively large. Thus, some material may be staying behind in the atomizer that would not be observed. Were any blanks with a clean salt solution ran to test for this carry-over between runs? Were the samples analyzed in any particular order or randomly?*

Filters were measured without following a specific order. As anonymous reviewer #4 suggested, nebulizing solutions of inorganic salts would provide a more representative blank. However, we have previously showed that the organic blank measurements collected by ultrapure water nebulization provide a comparable blank estimate to the organic blanks determined from the nebulization of $(\text{NH}_4)_2\text{SO}_4$ (Bozzetti et al., 2017).

2. Page 16, line 18 “The AMS-PAHs:HOA ratio was : : :” the text gets a bit confusing here. I think the same point is being made and supported with different pieces of data but I recommend breaking the text up into a few paragraphs to make the discussion clearer.

According to anonymous reviewer #4 we subdivided the section into paragraphs as follows. The AMS-PAHs:HOA ratio was 0.0020, while the AMS-PAHs:BBOA was 0.0028.

In general, industrial emissions can be an important source of PAHs at this location as discussed in El Haddad et al. (2013). In presence of an industrial contribution, the BBOA vs. AMS-PAHs correlation would decrease. In this work the correlation between AMS-PAHs and the $C_2H_4O_2^+$ fragment, typically related to levoglucosan fragmentation (Alfarra et al., 2007), was high ($R=0.87$) and no AMS-PAHs spike was observed without a simultaneous increase of $C_2H_4O_2^+$ (Fig. S14). Moreover the industrial-related OA factor resolved by El Haddad et al. (2013) was clearly associated to wind directions from W/SW (225° - 270°), while in this work wind directions were oriented from W/SW only for 7% of the monitoring time, furthermore without being associated to any significant increase in the AMS-PAHs concentration (Fig. S15), indicating the absence of clear industrial episodes.

The BBOA diurnal cycle, similarly to AMS-PAHs, showed higher values at night than during the day (Fig. 4). In addition, the BBOA highest concentrations were detected at night and associated to slow wind speeds from the E/NE which is consistent with the night land breeze direction. Moreover, strong enhancements of the BBOA factor concentrations were perceived when the wind direction shifted to the E/NE (typically around 18 o'clock during the monitoring period) suggesting that BBOA could be transported from the valleys nearby Marseille (Fig. S17).

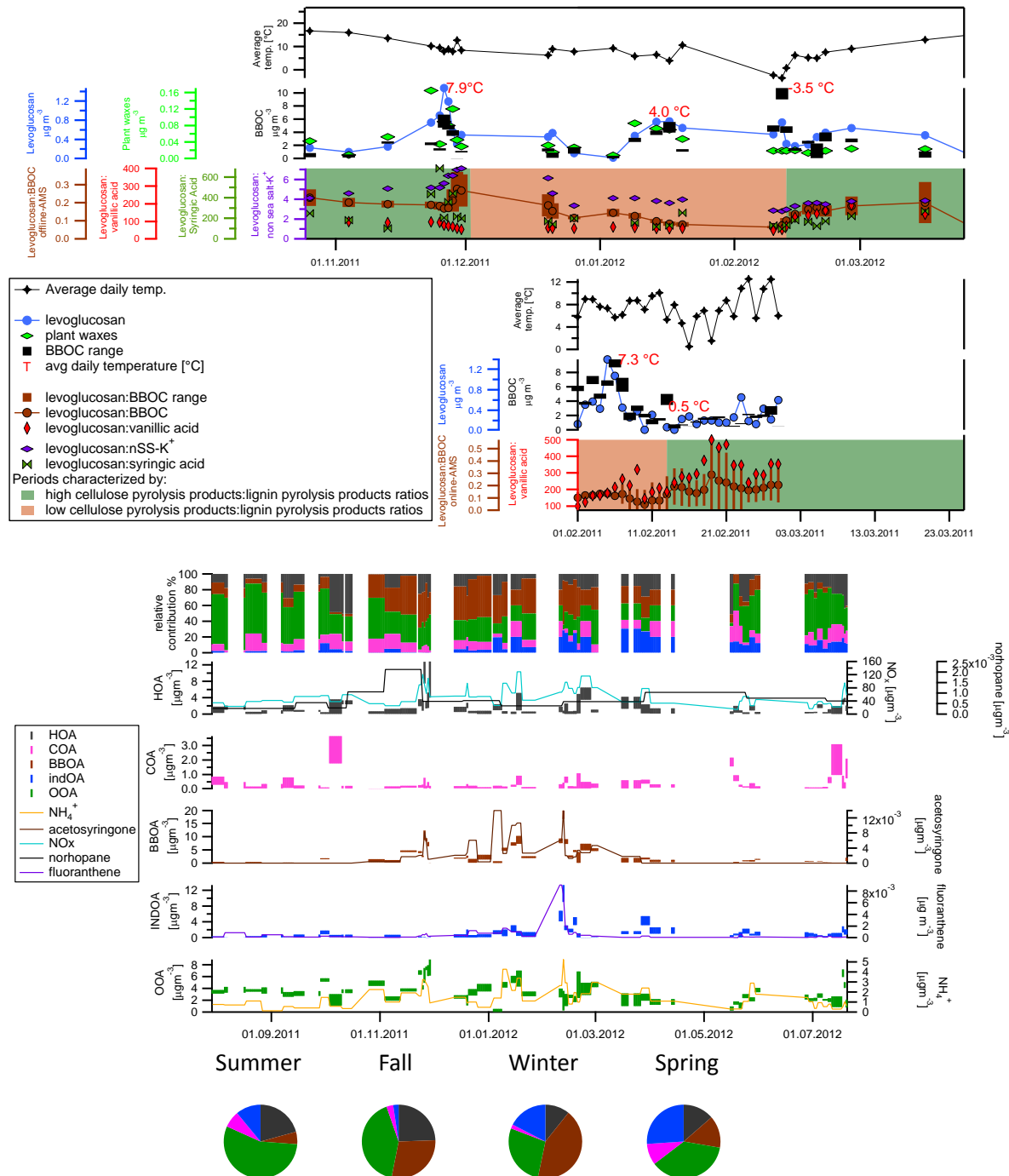
We calculated the BBOC time series by dividing the BBOA concentrations by the OM:OC_{BBOA} ratio calculated from the average BBOA HR spectrum (1.60). The average BBOC:levoglucosan ratio [$\mu\text{g m}^{-3}/\mu\text{g m}^{-3}$] was 0.15, comparable to other European studies (Zotter et al., 2014; Herich et al., 2014; Minguillón et al., 2011).

3. The analysis presented here used IC to measure the salts in the samples but the AMS also quantifies some of those ions. How do the measurements between the two techniques compare?

Since the same question was raised by anonymous reviewer #1, we kindly refer anonymous reviewer #4 to the answer to the major comment I) by anonymous reviewer #1.

4. Figure 11 is very hard to read, both the colors used and the sizes of the markers should be adjusted.

Following the suggestion of anonymous reviewer #4 we changed the background colors and the marker sizes.



5. Figure 6, change the black to grey in the pie chart or clarify in the caption.

Following anonymous reviewer #4 suggestion, we changed the color of HOA in the pie plots to dark grey.

6. Figure 6, the time series for BBOA is shown with acetosyringone but no mention of this is made in the text.

Acetosyringone is listed in Table S1. We added the acetosyringone correlation with BBOA at P18 L6-7.

7. Figure 10 shows dates on the x-axis for 2012 but 2011 online AMS data is overlaid using the same axis. I suggest changing the axis labels to 10.02.20xx or just the month and day to highlight this difference in collection times.

Fig. 10 was modified according to the suggestions of anonymous reviewer #2. The two traces were displayed on two different axes and labelled with different colors.

