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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Burning of olive tree branches: a major organic aerosol source in the Mediterranean" *by* E. Kostenidou et al.

Anonymous Referee #2

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This paper investigated the composition of organic aerosols in the area of ICE-HT institute. By performing PMF analysis on the organics data collected by HR-ToF-AMS, it is found that olive tree burning contributed a substantial fraction (33.7%) to the total organic aerosols loading. The otBB-OA factor determined has a lower m/z 60 than other BBOA factors previously reported. Based on these results, the authors suggested that olive tree burning is one of the most important aerosol sources in the Mediterranean countries.

Overall, the paper is well written and clear. I found the results from this study very interesting. However, I have some concerns regarding the results from the PMF analysis. The authors presented a 3-factor solution (otBB-OA, HOA, OOA) with a fpeak=-0.2. The fpeak value of -0.2 was chosen to obtain the smallest angle between the otBB-OA

factor and the chamber otBB spectrum. While one would expect the ambient otBB-OA factor and the chamber otBB spectrum to be strongly correlated, I do not think that it is appropriate to use this as a criterion for choosing the fpeak value, especially the chamber otBB spectra have some variations among themselves (Table S1). The authors should first decide on the appropriate number of factors before they explore the rotational freedom of their solution (Ulbrich et al., 2009). Based on all the information provided in the manuscript, it seems that the COA factor should be included in the solution and that a fpeak value of zero would be appropriate (please see detailed comments below). This is simply based on my interpretation of the analysis and supporting information provided in the manuscript. I suggest the authors to re-evaluate their PMF solutions again and carefully evaluate the 4 factor solution with fpeak =0 in the revised manuscript. While this could be a major revision, I do not see this posing a large amount of extra work since the authors have explored the different solutions in great details. The issue is in choosing the most appropriate solution for their data.

Specific comments:

1. Page 7227, line 14. It is not clear how the "30g of fine PM per year" is relevant to the context when the authors are discussing PM concentrations in "tons per year".

2. Page 7231, line 15. Is Nafion dryer used at the AMS inlet? The authors cannot simply apply CE from chambers to their ambient data. A CE of $\sim$ 0.5 is recommended if a dryer is used. Otherwise a higher CE might be more appropriate (depending on ambient RH and aerosol composition). Please clarify this.

3. Page 7232, line 19 onwards. It would be clearer if the authors could indicate the positions of the different functional groups in the spectrum shown in Figure 1.

4. Page 7232, lines 25-26. The sentence "the differences in the alkane group region are expected given that the biomass precursors in Mexico and Texas likely do not include olive trees" is not clear. How would biomass precursors affect alkane contribution?

5. Page 7235, line 2. At a first read it seems that the authors injected extra toluene into the chamber as a tracer to estimate OH concentration. Later in page 7236 the authors discussed that toluene is actually present in olive tree burning. It would be clearer if the authors mention this on page 7235 when they discussed how they estimated OH concentration.

6. Page 7235, line 5. The author should include the corresponding O/C calculated from HR analysis in Figure 5.

7. Page 7235, line 11. “through out” should be “throughout”.

8. Page 7235. Are there any aerosol formed in these chamber experiments? If so, how much? The authors should include this info in the revised manuscript.

9. Page 7235, lines 19-23. What about carbonyl functional groups? Did they show a corresponding decrease in intensity?

10. Page 7237, PMF analysis: a. Figure 9. Since HR PMF is performed, the authors should show the HR spectrum instead of UMR spectrum. This would clearly indicate the contributions of different families (CH, CHO, CHN, etc) at each m/z.

b. Page 7238, last two paragraphs. I think some of the info should be included in the supplementary info instead of the main text. In PMF analysis, various m/z with similar temporal variations are grouped into one factor. One would certainly expect the time series of the individual m/z in each factor to correlate well with the time series of that particular factor. Hence, while it is extremely useful to examine the correlations of factors with external tracers such as sulfate, levoglucosan etc, it is not as informative to report R2 values of individual m/z and the factor itself. This information should be moved to the supplementary info and preferably with a table format.

c. Page 7263. There are three large spikes in HOA on Nov 26, Dec 12 and 14. What are the sources? The authors should at least comment on this.

d. I do not agree with the authors reasoning in choosing a fpeak=-0.2. In PMF analysis,

it is advised that one should explore the rotational freedom of the solution (using fpeak parameter) after the approximate best number of factors is determined (Ulbrich et al., 2009). However, in this case, the authors actually used fpeak to determine the number of factors to start with. Their reason for choosing fpeak=-0.2 was solely based on minimizing the angle between otBB-OA chamber spectrum and their otBB-OA factor from ambient data. Given that there is also variation in the chamber otBB-OA spectrum (Table S1, largest angle = 13.2) and that the angle between the otBB-OA chamber spectrum and otBB-OA factor does not vary that much for fpeak values between -2 and 0, I do not see any strong reason why they needed to use a fpeak value other than zero.

e. Based on the discussion provided in the supplementary information, I think that a 4 factor solution would be more appropriate (i.e., there is a COA factor). From Figure S5, it is obvious that an additional factor is needed to describe their data. The authors cannot simply pick and choose to discard certain part of their data (Fat Thursday) and perform PMF analysis on the remaining data. In Figure S8, the COA spectrum is distinctively different from other factors. The authors argued that by creating a COA factor it “destroys” the otBB-OA, I do not agree with this. The authors stated that with the COA factor, the correlation between the chamber otBB-OA spectrum and otBB-OA factor got worse. However, from Figure S6 the angle between these two is only between 15-20 for fpeak =0, which is not too off from the variation in chamber otBB-OA itself (Table S1, largest angle = 13.2). Further, the authors suggested that the correlation of otBB-OA factor and the fourth factor (OOA) increases with increasing fpeak from -0.4 to 2. Looking at Figure S9, even with a fpeak=0, the R2 between the otBB-OA factor and OOA factor is < 0.2. Together with the fact that the otBB-OA spectrum and OOA spectrum are distinctly different, I think it’s suffice to say that the COA factor is a real factor and should be included in the solution.

f. Overall, I would suggest the authors to re-evaluate their PMF solutions. Based on all the info provided in the manuscript, I would guess that a 4 factor solution with fpeak=0

would be appropriate. The authors of course should evaluate this carefully and provide justifications in the revised manuscript.

g. Figures S5 and S6. It is nice that the authors showed these residues in details. I suggest the authors to also include a plot of Q/Qexp in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 7223, 2013.