Referee #1

(1) The paper titled "Sources and chemical characterization of organic aerosol during the summer in the eastern Mediterranean" by Kostenidou et al. deals with the chemical characterization and organic sources identification in Athens and Patras, highlighting the importance of regional sources in that area. Positive matrix factorization has been performed on high resolution time of flight AMS data identifying both primary and secondary organic aerosol sources. This is a relevant topic in the atmospheric science field and therefore it is suitable for ACP. The overall quality of this work is good and the manuscript is quite well-written. I recommend publishing this work after the authors respond to the following comments.
We appreciate the positive assessment of our work

General comments

(2) Despite most of the references cited in the introduction are pertinent, this section should be reorganized to give the clear context for this paper. The authors list many topics, like source apportionment techniques, organic sources, studies in the Mediterranean area etc., but this introduction appears to be independent from the topic treated in this paper. I would recommend to explicitly mention how your study is connected to this overview (you reported just one sentence at the end of the introduction).
We have added in each subsection of the introduction information about the links of the present work with the topic discussed.

(3) Did the authors expect to find marine related OA sources at their measurement sites? Being the two sites influenced by marine air masses, the S:C ratio should probably be higher than in continental urban sites. Can the authors further elaborate this concept?
Given the location of the two sites, we would expect to find a marine OA (MOA) factor. The S:C ratio estimated by the AMS is often underestimated in the presence of organosulfates (Farmer et al., 2010; Docherty et al., 2011). To avoid such artifacts we investigated the contribution of MOA applying a constrained solution in the ME-2 using the MOA mass spectrum of Crippa et al. (2013) with a=0.1. For Patras the average MOA concentration for 4, 5, and 6 factors was around 0.04 μg m^{-3} corresponding to 1% of the OA mass. For Athens the MOA concentration for the 3, 4 and 5 factor solutions was approximately 0.25 μg m^{-3} (3.7% of the OA mass). So if MOA was indeed present its contribution to OA was very low. This information has been added to the revised manuscript.

(4) The conclusion section is quite poor. The authors should highlight the importance of their work also at the end of the paper.
We have rewritten the conclusion section.

Specific and technical comments
(5) Page 3456, line 15: replace "HR-AMS" with "HR-ToF-AMS" (everywhere in the manuscript)
Done.

(6) Page 3457, line 4: replace "contributes" with constitutes.
Done.

(7) Page 3457, line 10: add the reference to Zhang et al., 2011.
Done.

(8) Page 3457, line 12: add the reference to Canonaco et al., 2013.
Done.

(9) Page 3462, line 9: I would mention that CE is usually around 0.5.
Done.

(10) Page 3464, line 18: reformulate "are not that useful".
We replaced it with: "could result in erroneous conclusions"

(11) Page 3466, line 3: replace "types" with "sources".
Done.

(12) Page 3467: it would be interesting to show some of the PTRMS tracers correlations with the AMS SOA factors. I would also move Table S1 from the supplementary material to the main text.
We added 3 figures in the SI showing the time series of the Patras M-OOA, Patras b-OOA and Athens V-OOA compared to PTR-MS tracers (2 tracers for each factor). We moved Table S1 to the main manuscript.

(13) Page 3472, line 11: which m/z where so different from the HOA from Crippa et al. 2013b?
The major differences were in the *m/z's* 44 and 28 which were higher than in Paris, *m/z's* 29 and 43 which were lower compared to Paris and *m/z* 39 which was absent in Paris probably due to the unit mass resolution spectra used as input for the Crippa et al. PMF analysis. Please note that there was a typo and the correct angle thetas between the HOA Paris (SIRTA) winter (of Crippa et al. 2013b) and HOA Patras and HOA Athens are 25^o and 24^o correspondingly (not 33^o and 31^o). We modified the corresponding text in the manuscript to explain the above points.

(14) Figure 6 is expected to be used for SOA components. I would put it in the supplementary material.
We moved Figure 6 to the SI.

(15) Figures S3 and S6: include fitting parameters (slope, intercept).
Done.

(16) Figure S14b: I would also highlight the extremely high contribution of mz44 for HOA-1.
Done.

(17) Figure S16b: I would also mention the lower contribution at mz44 for HOA-1
In Figure S16a HOA-1 is actually HOA-2 and vice versa (there was a typo in this graph, which is now corrected). So, there is almost no difference between the *m/z* 44 for HOA-1 in Figure S16a and the *m/z* 44 for HOA-1 in Figure S16b.

(18) A general recommendation for the SI: you should report at least few sentences describing the shown graphs to guide the reader, although a more complete description has been already reported in the main text.
We have added 1-2 sentences in the SI providing additional information about the corresponding graphs.

References

Canonaco, F., Crippa, M., Slowik, J. G., Baltensperger, U., and Prévôt, A. S. H.: SoFi, an Igor based interface for the efficient use of the generalized multilinear engine (ME- 2) for source apportionment: application to aerosol mass spectrometer data, Atmos. Meas. Tech., 6, 3649–3661, 2013.

Crippa, M., El Haddad, I., Slowik, J., G., DeCarlo, P. F., Mohr, C., Heringa, M., F, Chirico, R., Marchand, N., Sciare, J., Urs, B., and Prévôt, A. S. H.: Identification of marine and continental aerosol sources in Paris using high resolution aerosol mass spectrometry, J. Geophys. Res, 118, 1950–1963, doi:10.1002/jgrd.50151, 2013.

Docherty, K. S., Aiken, A. C., Huffman, J. A., Ulbrich, I. M., DeCarlo, P. F., Sueper, D., Worsnop, D. R., Snyder, D. C., Peltier, R. E., Weber, R. J., Grover, B. D., Eatough, D. J., Williams, B. J., Goldstein, A. H., Ziemann, P. J., and Jimenez, J. L.: The 2005 Study of Organic Aerosols at Riverside (SOAR-1): instrumental intercomparisons and fine particle composition, Atmos. Chem. Phys., 11, 12387–12420, doi:10.5194/acp-11-12387-2011, 2011.

Farmer, D. K., Matsunaga, A., Docherty, K. S., Surratt, J. D., Seinfeld, J. H., Ziemann, P. J., and Jimenez, J. L.: Response of an aerosol mass spectrometer to organonitrates and organosulfates and implications for atmospheric chemistry *PNAS,* 107, 6670-6675, doi:10.1073/pnas.0912340107, 2010.

Zhang, Q., Jimenez, J., Canagaratna, M., Ulbrich, I., Ng, N., Worsnop, D., and Sun, Y.: Understanding atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, Anal. Bioanal.Chem., 401, 3045-3067, 2011.

Referee #2

The manuscript by Kostenidou et al. presents chemical composition and sources of fine particulate matter in the Eastern Mediterranean based on AMS measurements in the suburbs of Patras and Athens and subsequent PMF analysis. There are not too many studies on air quality in the Eastern Mediterranean, which makes the present analysis an important addition. I recommend publication in ACP after the following comments have been addressed:

General comments:

(1) Please add more info and clarifications to the site descriptions. Especially for Patras it is not clear to me at which of the two sites mentioned the instruments described in section 2.3 were measuring. Also add a map that shows the locations of the 2 sampling sites in Patras, and their position relative to the sampling site in Athens.
All of the instrumentation described in Section 2.3 was located at the ICE-HT institute. We added a sentence at the end of this section 2.3 clarifying this point. In addition we added a sentence at the end of section 2.2 highlighting that this paper focuses only in the ICE-HT measurements. We also added two maps in the SI illustrating the locations of the two sampling sites in Patras and the positions of the Patras and Athens sites.

(2) The measurements in Patras and Athens could not be done at the same time due to experimental limitations, I can see that. However, to still be able to make some comparisons between the two sites you could compare parameters that were measured simultaneously (meteorology? SMPS? Etc.). This would give an idea of how different the meteorological and pollution situation was for the 2 measurement periods, and how that potentially influences AMS mass loadings.
A detailed description and intercomparison of the various measurements (including filter samples) performed in Athens and Patras is given in the companion paper by Tsiflikiotou et al. (in preparation). The present work focuses on the OA sources and AMS data. We added a brief summary of the conclusions of Tsiflikiotou et al. about the two periods and the OA levels in the two cities to provide additional context for the present work.

(3) There are quite a few comparisons in the manuscript between PM1 and PM2.5 data, e.g. sulfate for the CE, and (section 3.1) OM and Org. This can be done, but discrepancies should specifically be discussed for the size difference. Also, what conclusions are drawn based on the comparison in section 3.1 (p. 3463, l. 23 – 26)? Section 3.2 (p. 3465, l. 8 – 12): How can AMS PM1 be higher than PM2.5? This needs to be elaborated.
We have made sure that the different size ranges (PM_1 for the AMS and $PM_{2.5}$ for the filter samples) are taken into account during the various comparisons. A number of useful conclusions are also drawn by the agreement or lack thereof between the two techniques. For example the good agreement between the AMS sulphate and the $PM_{2.5}$ sulphate from the filter samples probably suggests that almost all of the $PM_{2.5}$ sulphate was in the submicrometer size range during these campaigns. The scatter observed when comparing the corresponding measurements is due to a number of factors including the positive and

negative artifacts of filter measurements, the uncertainties of the corresponding measurements (e.g., due to AMS collection efficiency), and the different size ranges. The negative artifacts of the filter measurements due to evaporation of the collected OA can explain the occurrence of some measurements in which the $PM_{2.5}$ filter-based OA is less than the PM_1 AMS OA. These issues are now discussed in the revised paper.

(4) What is the reasoning behind calling the 2 non-biogenic OOA factors V- and M-OOA, and not LV-, and SV-OOA, respectively? Are they significantly different from literature LV- and SV-OOA factors? If yes, this should be mentioned, otherwise I suggest renaming the factors.
The names of the 2 non-biogenic OOA factors (very oxygenated OA and moderately oxygenated OA) were chosen based on their oxygenation degree. The PMF analysis separates these factors based mainly on their oxygen content and not on their volatility. Unfortunately, no volatility measurements were made during these campaigns. Recent (not yet published) work by our group suggests that the V-OOA factor also contains some relatively volatile components, and the M-OOA contains low volatility compounds. We understand that the LV-OOA and SV-OOA terms have been used in several past studies and we do explain the correspondence and compare our factors to the corresponding LV-OOA and SV-OOA ones (see for example Table 1). V-OOA was close to the literature LV-OOA spectra, but M-OOA had a moderate correlation with most of the SV-OOA spectra. We have added discussion of these topics in the revised paper but we do prefer to keep the existing names of these factors.

(5) I am a bit confused by the interpretation of the two factors called HOA-1 and HOA-2. P. The factor profile of HOA-1 is similar to HOA factor profiles found in literature, however its factor time series does not correlate with traffic marker time series. The HOA-2 factor profile resembles COA, the diurnal pattern of the time series indicates contributions from cooking emissions, however its time series correlates also with BC. When you did ME-2, was HOA fixed? I wonder whether the different PMF solutions and their rotations could specifically be explored for a distinction of the different primary organic sources.
In a series of sensitivity tests, we tried rotations in the f_{peak} range from -1 to 1. In all of these tests the correlations between our factors HOA-1 and HOA-2 and traffic markers such as BC and toluene practically did not change. We also used a fixed HOA spectrum based on the Aiken et al. (2009) as well as the HOA mass spectrum found in Athens during winter (unpublished results). In these tests too the correlation between HOA-1 and BC did not improve. All these suggest that are conclusions are robust to the details of the PMF process. We have added a summary of the results of these sensitivity tests to the revised manuscript. Our explanation is that the correlations are affected also by the locations of the various sources around the receptor site and the corresponding wind directions. The fact that these area sources (gasoline cars, diesel cars, cooking activities) did not have a homogeneous spatial distribution can lead to these rather unexpected results.

Specific comments:

(6) P. 3456, l. 15 – 16, and throughout manuscript: P. 3459, section 2.1.: Is there a reference about the larger study you could add? If not consider adding a few sentences on this study here.
We have added the corresponding reference for the summer study (Tsiflikiotou et al., in preparation).

(7) P. 3462, l. 1 -4: Add reference to the supplementary information. Also, add (here or in the supplement) information on the ME-2 settings you used – which factor spectra were fixed, how fixed were they, etc. You also mention in the SI several times that you chose fpeak=0 because for fpeak=0 Q/Qexp was minimized. Fpeak=0 will always produce Q/Qexp = min, you specifically use fpeak6=0 to distort the solution (compare Ulbrich et al., ACP, 2009).
We added the Lanz et al. (2008) reference to the Supplementary Information. For the solutions presented in this paper we did not use constrained solutions in ME-2. For this reason we did not include such details. We tried constraining the HOA spectrum in one of the sensitivity tests (see our response to comment 5 above) and we have included the corresponding information about the spectra used as well as the corresponding parameter values. The f_{peak} selection was not based solely on the minimization of $Q/Q_{expected}$, but also on the mass spectra characteristics and the stability of the different f_{peak} solutions calculated by both the ME-2 and PET algorithms. We have modified the SI discussing in more detail the factor selection.

(8) P. 3462, section 2.6: Concerning the CE in Patras, do you have any information on ambient RH? Can you see a dependency of the CE on RH?
The R^2 between CE and the ambient RH was very low and equal to 0.02. This low correlation and the high CE values can be explained by the fact that the particles were acidic most of the time and probably contained water during practically all the campaign.

(9) P. 3469, l. 18: What about the influence of boundary layer height? Temperature?
If there was an influence of the boundary layer height it would be visible in all factors. However while M-OOA and HOA-2 increased, V-OOA and HOA-1 decreased during the night. The average temperature at noon was around 32^{o} C and during the night it decreased to around 21^{o} C. The change in temperature could be one of the explanations for the increase in M-OOA. We have added this potential explanation and the corresponding discussion to the revised manuscript.

(10) P. 3472, l. 15 – 16: Add more information on the meaning of this plot and your data in this plot. Just showing it is of no scientific value.
We moved Figure 6 to the SI and added some brief discussion.

Technical comments:
(11) P. 3456, l. 2: Should read "The concentration and chemical composition of non-refractory [: : :] (delete "the")
Corrected.

(12) P. 3456, l. 11: Should read “In both cases PM1 [: : :]” (delete “the”, and check throughout manuscript for more of such “thes”)
Done.

(13) P. 3456, l. 25: Should read “by causing cardiovascular [: : :]”
Done.

(14) P. 3457, l. 1: Update reference to latest IPCC report
Done.

(15) P. 3457, l. 4: Should read “contributes to [: : :]” (check throughout manuscript for more)
Replaced with “represents”.

(16) P. 3457, l. 6: Use HR-ToF-AMS for instrument abbreviation
Done.

(17) P. 3457, l. 6: Should read “high time resolution [: : :]”
Corrected.

(18) P. 3457, l. 25: Typo, should read “spray”
Corrected.

(19) P. 3457, l. 27: Concretize studies, e. g. “Most of the studies on air quality [: : :]”
Done.

(20) P. 3457, l. 29: Delete “just”
Done.

(21) P. 3458, l. 11: Replace “higher with “more”
Done.

(22) P. 3458, l. 16: Should read “[: : :]was of industrial origin.”
Corrected.

(23) P. 3458, l.16 – 20: Very complicated sentence, consider breaking it up into 2.
We broke that sentence into 2.

(24) P. 3458, l. 29: Replace “very hard”.
Replaced with “quite difficult”.

(25) P. 3459, l. 7 – 9: Consider adding a couple of sentences more on the objectives of the paper.
We added information about the objectives of this study.

(26) P. 3459, l. 12: Replace “results” by “measurements”.

Done.

(27) P. 3459, l. 18: Major anthropogenic activities?
We added “anthropogenic” before “activities”.

(28) P. 3461, l. 2: Most densely populated?
Corrected.

(29) P. 3461, l. 12: The aethalometer measures absorption and derives BC concentrations
We replaced “measured” with “provided”.

(30) P. 3461, l. 13: Delete comma after “classifier”
There is no comma after classifier. However we deleted the comma after SMPS.

(31) P. 3462, l. 6: Should read” throughout”
Corrected.

(32) P. 3469, l. 1: Delete ”while”; “Similar” instead of “similarly”
We rephrased these two sentences.

(33) P. 3470, l. 3: Should read “while BC originated from [: : :]”
Corrected.

(34) P. 3460, l. 19 – 20: Replace “correspondingly” by “, respectively”
Corrected.

(35) Supplemental information, figure caption S3: “suggests” instead of “suggest”
Corrected.