

## Responses to Referee #1

(1) This paper presents results from high resolution time-of-flight aerosol mass spectrometer measurements made during the winter of 2012 and 2013 at two different major cities in Greece (Athens and Patras). The composition of the aerosol is presented and discussed. Positive Matrix Factorization (PMF) analysis results are provided to explore the different sources of organic aerosol in the two cities, with a specific aim at better understanding the role of residential wood burning.

Biomass burning is one of the main sources of organic aerosol, but its contribution is still not well understood. In addition, the role of biomass burning as a source is becoming more and more important in many locations. This paper is presenting results from one such location. Therefore, many in the atmospheric community would be interested in this work. Overall, this is a good paper. The authors have done a nice job with the paper. I really appreciate seeing all parts of the PMF analysis to understand how they obtained their results.

We do appreciate the positive assessment of our work and the many suggestions by the referee that led to an improvement of our paper.

### General comments:

(2) I am mainly wondering if the authors have considered trash burning as possibly playing a role in their results and could be part of what leads to the two BBOA factors being identified in Patras.

We have considered the possibility of the use of different fuels (including trash) resulting in two BBOA factors in Patras. To test this hypothesis we have used the results of Mohr et al. (2009) who investigated in the laboratory the organic aerosol emissions of paper and plastic burning. The trash burning AMS spectra were completely different from those of biomass burning in this study with angles exceeding 45 degrees. Based on this trash burning cannot be the main source of any of the two BBOA factors in Patras. This information has been added to the revised paper.

(3) Otherwise, I just have a number of comments to help with the flow of the paper, like clarifying the notation for BBOA-I and BBOA-II in the figures which the authors seem to go back and forth on using. All of this is outlined below in detail and needs to be addressed before the paper can be considered for publication.

We have followed the reviewer's suggestion and made the corresponding corrections to the notation used in the paper.

### Specific Comments:

(4) Line 96 – Suggest changing till to until.

Done.

(5) Line 97 – Suggest changing place at to place on.

Done.

(6) Line 106 – Suggest changing real time to real-time.

Done.

(7) Line 117 – The start of this line should be indented to match the rest of the formatting of this section.

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Done.

**(8)** Line 133 – The chemical abbreviations used are not defined.

We have added the corresponding definitions.

**(9)** Lines 101-142 – If the authors were looking for a way to shorten the paper, they could remove some of the discussion in the instrument section pertaining to data they do not show in the paper. I believe that data from the SMPS, thermodenuder, some of the gas monitors, and filters are not presented or discussed in the text.

We have removed the corresponding discussion just mentioning the other instruments that were used in the study for completeness.

**(10)** Line 160 – *m/z* should be in Italics.

Done.

**(11)** Line 186 – Suggest changing meteorological conditions to air masses observed

Changed.

**(12)** Lines 224-229 – In this line the authors are discussing OA being correlated with VOCs found in petrol fuel. But I am a bit unclear if it is during the same times as when OA was correlated with acetonitrile (18:30-02:30) or the remaining period they are also discussing in this paragraph.

We have rewritten this sentence to clarify that we are referring to the periods during which biomass burning was not the dominant source.

**(13)** Lines 240-244 – The authors mention that chloride has a major morning peak and an evening peak. They mention the pools located near the sampling site as a possible source. But could this also be due to trash burning? I would expect that if trash burning is occurring, it would peak in the morning before the boundary layer started to rise.

This is an interesting suggestion. The data suggest that the chloride peaks occurred on average an hour earlier (at 7:00 LST) than the corresponding OA peaks (at 8:00 LST). This coupled with the major differences between known AMS fingerprints for trash and the estimated PMF factors suggest that trash burning was probably not a major contributor to the morning chloride peaks. This information has been added to the revised manuscript.

**(14)** Line 260 – Suggest adding a the before value.

Added.

**(15)** Line 261 – Suggest adding the word peaked after OA.

Added.

**(16)** Line 266 – The chemical abbreviation used is not defined.

We have added the definition.

**(17)** Lines 276 and 278 – Suggest changing till to until.

Done.

**(18)** Line 313 – Suggest changing time-series to time series

Done.

(19) Lines 367-379 – In this section the authors are discussing the difference between BBOA-I and BBOA-II. I was wondering have the authors checked to see if one of these factors is more correlated with chloride than the other? It was shown that there was an extra chloride peak in the diurnal profile and chloride can come from trash burning.

We checked the corresponding coefficients of determination and they were both quite low. The  $R^2$  between BBOA-II and the AMS chloride was 0.02 and that between BBOA-I and chloride was 0.16. These low correlations do not support the link between one of these factors and trash burning or chloride. These values have been added to the paper.

(20) Lines 413-425 – In this section the authors are comparing the BBOA spectrum obtained at both sites to those from literature. Although this is interesting, the spectra the authors are comparing to include both residential burning and burning from wild- fires and prescribed burning. My understanding is that the data collected from Greece should be impacted by residential burning. I am not sure if the authors have taken this into account in their comparison.

We included for completeness all types of burning in the comparisons. We do clarify now in the revised paper the type of burning dominating in each field study (e.g., residential, agricultural, prescribed, wild fires) and that the fact that residential burning was the main BBOA source in Greece in the present study.

(21) Line 424 – I am not sure if the correct reference is listed. I don't believe there are any AMS spectra provided in that paper.

We have corrected the FLAME-I reference.

(22) Line 434 – Suggest changing summertime at to summertime in.

Done.

(23) Line 438 – The period for the end of the sentence is missing after processes.

Added.

(24) Lines 449-453 – The authors mention that the diurnal concentration of OOA decreased in the morning and then increased again in the afternoon peaking at midnight. They believe this could be due to nighttime SOA production from biomass burning-related VOCs reacting with  $\text{NO}_3$  radicals. I am not sure I am completely following this discussion. For this to be true, wouldn't there have to be organonitrates present then? I believe that it was mentioned earlier that when biomass burning dominated organonitrates were low.

This is an excellent observation that has been added to the manuscript. The increase of OOA during the night is accompanied by increases in mostly inorganic nitrate with a relatively small contribution of organic nitrate. This does not support the hypothesis that the OOA increase is due to the production of SOA from reactions of the biomass burning emissions with nitrate radicals. We have rephrased this sentence adding the above discussion.

(25) Line 454 – Suggest removing the word reasons.

Done.

(26) Lines 467 and 476 – Suggest adding a the before Po.

Done.

(27) Line 546 – Figure 10a is being referenced, but there is no Figure 10. Should it be Figure 8?

We have made the suggested correction.

(28) Line 551 – Suggest adding the words had only before minor.  
Done.

(29) Line 612 – Believe accent marks are missing on Prevot.  
We have added the accent marks.

(30) Line 630 – Should the a before Li be capitalized?  
We have made the change.

(31) Line 639 – Believe accent marks are missing on Prevot.  
We have added the accent marks.

(32) Line 667 – Should the a before Herndon be capitalized?  
Changed.

(33) Line 692 – The a after Prevot should be capitalized.  
Changed.

(34) Line 720 – There is an added hyphen in Seinfeld.  
Removed.

(35) Line 724 - The a after Boreave should be capitalized.  
Changed.

(36) Lines 758 -760 – All the a initials should be capitalized.  
Done.

(37) Line 774 - The a before Knighton should be capitalized.  
Done.

(38) Line 775 - The a before de Foy should be capitalized.  
Done.

(39) Lines 783-785 - All the a initials should be capitalized.  
Done.

(40) Line 793 - Believe accent marks are missing on Prevot.  
We have added the accent marks.

(41) Line 809 – The a after Nenes should be capitalized.  
Done.

(42) Line 842 – The a before Alfarra should be capitalized.  
Done.

(43) Line 843 - Believe accent marks are missing on Prevot.  
We have added the accent marks.

(44) Line 866 – The a before Querol should be capitalized.

Done.

(45) Line 886 – I believe the year is listed in the wrong place.  
Done.

(46) Line 910 – The a before Facchini should be capitalized.  
Done.

(47) Line 925 - Believe accent marks are missing on Prevot.  
We have added the accent marks.

(48) Line 929 – The a before Forster should be capitalized.  
Done.

(49) Line 934 - All the a initials should be capitalized.  
Done.

(50) Line 949 – The a after Chaloulakou should be capitalized.  
Done.

(51) Line 953 – The a before Nitrogen should be capitalized.  
Done.

(52) Lines 963-964 - All the a initials should be capitalized.  
Done.

(53) Lines 973-974 - All the a initials should be capitalized.  
Done.

(54) Table 1 -I believe that not all the chemical abbreviations used in the Table have been defined in the text, for example EVK and MBO.  
We have added all the definitions here too.

(55) Figure 2 -I am not sure what the 124 in the OA plot is referring to. It is not mentioned in the caption.  
During the first day of the measurements the mass concentration of OA was equal to  $124 \mu\text{g m}^{-3}$  and this is not shown in the plot. This information has been added to the figure caption.

(56) Figure 6 -I believe that BBOA is actually BBOA-I and BBOA-I is BBOA-II. To stay consistent with the previous figure and the text it would be best to label them as I and II.  
We have made the corresponding correction.

(57) Figure 7 -I am not sure what the 30 in the HOA plot is referring to. It is not mentioned in the caption.  
During the first day of the measurements the peak concentration of HOA was equal to  $30 \mu\text{g m}^{-3}$  and this is not shown in the plot. This information has been added to the figure caption.

(58) Figure S2 -Suggest adding to caption that the figure is showing the origin of the air mass every 3 h for all the sampling days  
We have added the suggested information.

(59) Figure S3 -In caption, suggest removing the hyphen from back trajectories.

Done.

(60) Figure S4 -In caption, suggest removing the hyphen from back trajectories.

Done.

(61) Figure S5 -Suggest adding to caption that the figure is showing the origin of the air mass every 3 h for all the sampling days.

Done.

(62) Figure S6 -What do the red boxes indicate? It is not mentioned in the caption.

The boxes show the two periods with frequent precipitation. This information has been added to the figure caption.

(63) Figure S8 -What do the dashed lines indicate? Is it the average ratio? It is not mentioned in the caption.

The dashed values are indeed the average ratios. This is now mentioned in the figure caption together with the corresponding values.

(64) Figure S9 -What do the dashed lines indicate? Is it the average ratio? It is not mentioned in the caption. -In caption, suggest removing the comma after Patras.

The dashed values are the average ratios. This is now mentioned in the figure caption together with the corresponding values. The comma has been deleted.

(65) Figure S11 -There are no letters identifying the plots as indicated in the caption

We have added the missing letters to the corresponding graphs.

(66) Figure S12 -There are no letters identifying the plots as indicated in the caption.

We have added the missing letters to the corresponding graphs.

(67) Figure S14 -In caption, I believe there is a word missing after stable. Maybe something like solution or area -In first line of caption, fpeak is misspelled

The word "solution" has been added and the typo has been corrected.

(68) Figure S15 -The label on the right-hand y-axis of plot d is missing.

We have added the label.

(69) Page 17 -I believe the plots shown on this page are a repeat of Figure S11 and show the data for Patras and not Athens.

We have corrected this typo.

(70) Figure S19 -The letters identifying the plots do not match the caption.

We have corrected these letters.

(71) Figure S22 -In order to match the text, I believe that the plot labeled BBOA-1 is really BBOA-II and the plot labeled BBOA is really BBOA-I..

We have made the suggested changes.

(72) Figure S23 -In order to match the text, I believe that for the legend labels BBOA-I is really BBOA-II and BBOA is really BBOA-I.

We have made the suggested changes.

(73) Figure S24 -In order to match the text, I believe that the plot labeled BBOA-I is really BBOA-II and the plot labeled BBOA is really BBOA-I..

We have made the suggested changes.

(74) Table S1 -I believe Table S1 is not referenced in the text and that it is also an exact copy of Table 1.

We have removed Table S1.

(75) Table S2 -I believe Table S2 is not referenced in the text.

A reference to Table S2 has been added in the main paper.

(76) Figure S29 -In the caption and in the graph title, it should be BBOA-II.

Done.

(77) Figure S34 -This figure is referenced before Figures S32 and S33.

We have renumbered the corresponding figures.

(78) Figure S35 -I believe there is no reference to or discussion about this figure in the text.

A reference to Figure S35 has been added in the main paper.

(79) Figure S36 -I believe there is no reference to or discussion about this figure in the text.

A reference to Figure S36 has been added in the main paper.

## References

Mohr, C., Huffman, J. A., Cubison, M. J., Aiken, A. C., Docherty, K. S., Kimmel, J. R., Ulbrich, I. M., Hannigan, M. and Jimenez, J. L.: Characterization of primary organic aerosol emissions from meat cooking, trash burning, and motor vehicles with High-Resolution Aerosol Mass Spectrometry and comparison with ambient and chamber observations, *Environ. Sci. Technol.*, 43, 2443–2449, doi:10.1021/es8011518, 2009.

## Responses to Referee #2

### General comments:

(1) Florou et al. describe aerosol measurements from two Greek cities. Using PMF, the authors resolve factors for BBOA, HOA, COA, and OOA. The authors find that biomass burning is a significant contributor to organic aerosol in both cities. In Patras, the authors resolve two biomass burning factors (BBOA-I and BBOA-II). The time profile and mass spectra of these two factors are quite different. These two factors may result from differences in (a) the degree of atmospheric aging, (b) the composition of the fuel, (c) the burning conditions, or (c) a combination of these processes. The influence of biomass burning emissions on regional air quality is important to assess. This study is particularly interesting because it is focused in a region that has seen large increases in wood stove usage, which may be a result of the recent Greek economic crisis (Paraskevopoulou et al., 2014). Given its relevance, this work will be of general interest to the atmospheric community. Overall, the methods the authors employ are good. I especially commend the authors for the extensive amount of detail they provide to justify the PMF results and the comparisons they make with previous published BBOA factors. We appreciate the constructive comments and suggestions by the referee.

(2) I have one major comment pertaining to the resolution of the two biomass burning factors in Patras (see below). I believe addressing this comment will significantly strengthen the paper. Also, some of the sentences can be improved to help with the overall flow of the paper. Editorial comments are provided at the end of this review. The authors identify two biomass burning factors associated with the Patras data set. The resolution of these two factors is interesting, and I believe the authors may be able to draw more conclusions from these results. The authors hypothesize that the differences between these two factors arise from either (a) different degrees of aging (b) different types of combustion or fuel, or (c) some combination of these mechanisms. The authors seem to emphasize that the differences between BBOA-I and BBOA-II could result from aging. Can the authors elaborate more about the impact of different fuels?

We have followed the reviewer's suggestion (see also Comments 2 and 13 of the first reviewer's suggestion) and added some discussion and analysis of the various hypotheses. In principle, the two factors could correspond to different fuel types. However, the corresponding emissions should have either a different temporal or a spatial profile in order to be separated by PMF. Given that residential biomass burning in a city like Patras is due to tens of thousands of individual sources (fireplaces, wood stoves, etc.) spread all over the city following a similar burning schedule on average it is difficult to see how PMF would be able to separate the BBOA from different fuel types. The null hypothesis is that the BBOA factor represents an average of the emissions of all fuel types. However, there are other potential explanations (e.g., different areas of the city using different fuel types and the emissions arriving at the site at different times) that could involve the use of different fuel types. We have added the above discussion to the revised paper.

(3) The authors note similarities between BBOA-II and AMS spectra of burned olive tree branches. Do people in Patras use multiple types of fuels, whereas those in Athens typically use one? It would be helpful for the reader to know what types of fuels are typically burned for home heating.

In general people in Greece use multiple fuel types that can vary from region to region due to the availability of the corresponding type of biomass. For example, in Patras citizens use either



softwood (pine, fir, etc.) or hardwood (oak, beech, olive tree). In Athens, people use mainly pine and fir. This information has been added to the paper.

(4) I find it striking that nitrate in Patras is so strongly associated with organic aerosol (Fig S7). As the authors note, BBOA is the dominant organic component; therefore, I wonder if BBOA in Patras is also the dominant source of ON. The authors mention that ON was not strongly associated with BBOA plumes in Athens, but do not make a similar statement for Patras (lines 275-279); therefore, I'm assuming the evening nitrate in Patras is indeed affected by ON.

In Patras, ON was 44% of the total nitrate for the period from 16:00 until 24:00 LST (for the whole dataset) that is during the BBOA high concentration periods. This has been added to the paper.

(5) Furthermore, the correlation with evening nitrate enhancements seems to be better for BBOA-I than for BBOA-II (line 359). Could this imply that BBOA-I was a significant source of ON in Patras?

The  $R^2$  between the BBOA-I and the AMS nitrate was 0.55, while that between the BBOA-II and nitrate was 0.09. BBOA-I also correlated better ( $R^2=0.31$ ) with ON in Patras than BBOA-II ( $R^2=0.09$ ). So the reviewer's hypothesis is correct, most of the ON in Patras was associated with BBOA-I and not with BBOA-II. This is now clarified in the revised manuscript.

(6) ON (as well as nitrate) is typically the result of secondary processes; however, the nitrate trace appears to be better correlated with the "fresh" biomass burning factor (BBOA-I) rather than the "aged" biomass burning factor (BBOA-II). This, along with the nearly identical diurnal patterns (I would expect the "aged" factor to have a broader diurnal pattern), leads me to question whether the differences between BBOA-I and BBOA-II are truly due to chemistry. Therefore, could differences in fuel composition explain these observations? I ask about composition because recent work has shown that the emissions of nitrogen containing organic compounds (such as acetonitrile) strongly depend on the composition of the fuel (Coggon et al. 2016). Fuels containing low amounts of nitrogen (e.g. wood) emit lower amounts of N-organics than fuels containing large amounts of nitrogen (e.g. grasses, the boughs of trees). The same behavior has been observed for inorganic nitrogen gasses, such as  $\text{NH}_4$  and  $\text{NO}_x$  (e.g. Burling et al., 2010). Consequently, if different fuels were burned, then different amounts of ON could be formed due to differences in the amount of  $\text{NO}_x$  emitted or, perhaps, differences in emissions of primary organic nitrogen. If this were the case, then it would (1) be very interesting and (2) be an explanation for the different factor profiles for BBOA-I and BBOA-II.

These are good points and have been added to the revised paper together with the corresponding references. We have extended the discussion of the potential explanations of the nature of the two BBOA factors in Patras stressing the possibility that they may due to different fuel types (see also our response to Comment 2 above).

For the case of Athens, during nights (from 20:00 until 3:00 LST) in enhanced OA plumes exceeding  $15 \mu\text{g m}^{-3}$ , the ON fraction of nitrate was approximately 20%. In Patras, ON was 44% of the total nitrate for the period 16:00 until 24:00 LST (for the whole dataset) and 37% for the case of OA plumes exceeding  $30 \mu\text{g m}^{-3}$  (16:00 until 24:00 LST).

(7) Already, it appears that there may indeed be differences due to composition, as the two biomass burning factors appear to have different correlations with acetonitrile (Table S2). Note: The authors seem to mix up notation, as well as the city to which they are referencing (please be consistent with notation.). Is Table S2 for Patras (see caption)? Likewise, does BBOA-fr refer to BBOA-I and BBOA-ox refer to BBOA-II? If so, the BBOA factor with the higher

correlation to acetonitrile (BBOA-fr) could be an indication that this factor originated from a source composed of higher nitrogen.

We agree that these different correlations do support the hypothesis of two different types of fuels. This argument has been added to the paper. Table S2 has been renamed to Table S1. This table refers to Athens. BBOA-fr and BBOA-ox are the new resolved BBOA factors, by extending the solution from a 4 to a 5 factor solution. BBOA-fr is the BBOA-I from section SI.7b and BBOA-ox is the BBOA-II from the same section. The notation in the table has been changed.

**(8)** To tease out differences between composition vs. chemistry, I suggest that the authors do a more thorough comparison of the BBOA factors with the gas-phase compounds measured by PTR-MS. Do the authors also have  $\text{NO}_x$  measurements? If so, this may also help in the interpretation of these data. One way that the authors could tackle this question is by repeating PMF with the inclusion of some gas-phase species. Acetonitrile will likely vary depending on fuel composition, but other biomass burning markers (e.g. 2-furfuraldehyde ( $m/z$  97)) may not show similar dependencies.

This is an excellent suggestion for future work. Unfortunately, there was not a PTR-MS monitoring during the Patras measurements. We will make this a priority in future campaigns in the area.

**(9)** Alternatively, to avoid doing tedious PMF analyses, the authors can do a “plume” analysis to pick out differences in PTR-MS measurements when enhancements of biomass burning is dominated by BBOA-I vs. those when BBOA-II is dominant. Regardless of the outcome, I do believe that the authors need to provide a more complete discussion about the differences between BBOA-I and BBOA-II. As mentioned above, the aging explanation is questionable; therefore, the authors should consider discussing, in detail, other possible mechanisms.

We agree with the aging explanation is weak and that the potential existence of two different sources deserves at least equal discussion. We have added this to the revised paper in response to the comments and suggestions of the two reviewers. Unfortunately, there was not a PTR-MS available during the Patras measurements.

#### Other Comments:

**(10)** Line 122. Please define VOCs.

The definition has been added.

**(11)** Line 135. What do you mean by “main ions.” Are you referring to nitrate, sulfate, and ammonium?

Yes we are referring to the above ions. A clarification has been added.

**(12)** Line 167 – 168. This sentence is unclear. Do you mean that differences in  $m/z$  18, 28, and 39 were the reasons for higher theta values between the BBOA and COA spectra resolved by PMF and those from the HR spectral database? How would this change if you were to remove these ions from the analysis?

Yes. The theta angle is higher because of the differences in these three  $m/z$ . If one excludes these specific  $m/z$  values, the theta angle decreases. We have rephrased the corresponding sentence to avoid confusion.

**(13)** Lines 223-230. Here, the authors discuss the correlation between OA, acetonitrile,  $m/z$  79, and  $m/z$  69. It should be noted that furan ( $m/z$  69) and benzene have large contributions from

biomass burning (Gilman et al., 2015; Hatch et al., 2015; Stockwell et al., 2015); therefore, these masses may be affected by other sources than just petrol.

This point has been added in the revised paper together with a reference to Kaltsonoudis et al. (2016) who have analyzed the wintertime Athens VOC data set in great detail.

**(14)** Line 246. Please add “the” between “in” and “Patras”.

Done.

**(15)** Line 275-277. This sentence is confusing. Do the authors mean that ON fraction at night is high, except during peak OA enhancements? If so, it may be clearer to indicate that ON is high at nights, but not in enhanced OA plumes exceeding  $15 \mu\text{g m}^{-3}$ .

It was clarified in the text. ON was higher (38%) on average during nighttime (17:00 until 3:00 LST for the whole dataset) compared to periods of enhanced OA levels exceeding  $15 \mu\text{g m}^{-3}$  (19%) which were all associated with high BBOA concentrations.

**(16)** Lines 300 – 306. How should the reader interpret the diurnal patterns in O:C, H:C, and OSC? Do the enhancements of H:C during high OA reflect that these periods were affected mostly by primary emissions? The variability of O:C seems to be the inverse of H:C, suggesting that the composition of background OA dominates the observed O:C ratio during “off hours” (e.g. 4:00, 11:00, and 15:00), which is consistent with PMF results. The authors should provide additional discussion describing the cause in O:C, H:C, and OSC variability.

The explanations of the patterns by the reviewer are correct. The primary emissions mainly from traffic lead to reductions of the average O:C and increases in H:C during the corresponding peak periods. We have added the corresponding discussion in the revised paper.

**(17)** Line 322. I believe it’s better to state that the COA factor was resolved, rather than “added”.

We agree and we have made the corresponding change.

**(18)** Line 331. Here, I would state that the PMF model “resolved” four factors rather than “identified” four factors.

We have followed the suggestion of the reviewer.

**(19)** Line 358 and Line 363. Both factors correlate similarly to BC ( $R^2 \sim 0.26$ ), so I would not say that CO correlates modestly with BBOA-I (line 358) and poorly with BBOA-II (line 363).

We have made the suggested change.

**(20)** Fig 6. I believe that the BBOA traces are mislabeled. Is this correct?

We have corrected the typographic error.

**(21)** Table S2. Do these correlations correspond to the Patras data set? Also, what is BBOA-fr and BBOA-ox?

These correspond to the Athens dataset for the case of 5 factors. We have added this information together with the definition of the two BBOA factors (BBOA-1 and BBOA-2) that are resolved by PMF in this case.

## Editorial comments

(22) Section Formatting: The authors delineate sub-sub sections with bold text (e.g. line 86). I recommend numbering sub-sub sections (e.g. 4.1.1 Patras, 4.1.2 Athens, etc). This makes it easier to reference sections in the main text.

We have made the recommended change.

(23) Grammar: There are sentences scattered throughout the manuscript that are difficult to follow. Most of these sentences would improve with better formatting. Below are a few observations of grammatical errors. Addressing these comments will help the manuscript read more fluently. Please use commas around interrupters. For example, at line 221, the sentence would read more clearly as “The nitrate was, on average,  $0.48 \mu\text{g m}^{-3}$  . . .” Similar examples can be found at line 240 and 564.

We have followed the suggestions of the two reviewers to make the corresponding sentences easier to follow.

(24) These authors are inconsistent with the use of commas after introductory elements. For example, at line 179, the authors use a comma to separate the introductory element (During the Athens campaign, . . .); however, a comma is not included after the introductory element at lines 176 – 177 (During February 26-27 and March 5 the air masses. . .). Similar examples can be found at lines 145, 217, 236. Please be consistent and use commas.

We have made the corresponding changes making consistent use of the commas after introductory elements.

(25) The authors write some sentences with multiple dependent clauses. At times, it is difficult to understand what the authors are trying to convey. For example, at lines 70- 72, the authors write “The PMF source apportionment algorithm, used unconstrained, was applied to the corresponding datasets, estimating the contributions of the different OA sources, without assuming any a priori knowledge of their origin.” Here, several clauses are combined into one long, hard-to-read sentence. I recommend splitting up the sentence to clearly state each clause. Other examples include sentences at lines 127- 129, 236 – 238, and 241-243.

We have simplified the suggested sentences splitting them up in simpler sentences.

## References

Burling, I. R., Yokelson, R. J., Griffith, D. W. T., Johnson, T. J., Veres, P., Roberts, J. M., Warneke, C., Urbanski, S. P., Reardon, J., Weise, D. R., Hao, W. M. and De Gouw, J.: Laboratory measurements of trace gas emissions from biomass burning of fuel types from the southeastern and southwestern United States, *Atmos. Chem. Phys.*, 10, 11115–11130, doi:10.5194/acp-10-11115-2010, 2010.

Gilman, J. B., Lerner, B. M., Kuster, W. C., Goldan, P. D., Warneke, C., Veres, P. R., Roberts, J. M., de Gouw, J. A., Burling, I. R. and Yokelson, R. J.: Biomass burning emissions and potential air quality impacts of volatile organic compounds and other trace gases from fuels common in the US, *Atmos. Chem. Phys.*, 15, 13915–13938, doi:10.5194/acp-15-13915-2015, 2015.

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# The contribution of wood burning and other pollution sources to wintertime organic aerosol levels in two Greek cities

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**Abstract.** The composition of fine particulate matter (PM) in two major Greek cities (Athens and Patras) was measured during two wintertime campaigns conducted in 2013 and 2012, respectively. A major goal of this study is to quantify the sources of organic aerosol (OA) and especially residential wood burning, which has dramatically increased due to the Greek financial crisis. A high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) was deployed in both sites. PM with diameter less than 1  $\mu\text{m}$  ( $\text{PM}_{10}$ ) consisted mainly of organics (60-75%), black carbon (5-20%) and inorganic salts (around 20%) in both Patras and Athens. In Patras, during evening hours,  $\text{PM}_{10}$  concentrations were as high as  $100 \mu\text{g m}^{-3}$ , of which 85% were OA. In Athens, the maximum hourly value observed during nighttime was  $140 \mu\text{g m}^{-3}$ , of which  $120 \mu\text{g m}^{-3}$  was OA. 40-60% of the average OA was due to biomass burning for both cities, while the remaining mass originated from traffic (12-17%), cooking (12-16%) and long-range transport (18-24%). The contribution of residential wood burning was even higher (80-90%) during the nighttime peak concentration periods, and less than 10% during daytime. Cooking OA contributed up to 75% during mealtime hours in Patras, while traffic-related OA was responsible for 60-70% of the OA during the morning rush hour.

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## 1. Introduction

Fine particulate matter is associated with premature mortality and cardiovascular disease (Nel, 2005), but also impacts climate, visibility, and ecosystems (Grantz et al., 2003; Hallquist et al., 2009; Watson, 2002). An important fraction of submicrometer PM is organic representing 20 - 90% of PM<sub>1</sub> mass worldwide (Jimenez et al., 2009; Zhang et al., 2007) and originating from various anthropogenic and natural sources. Organic aerosol (OA) can be either emitted directly in particulate form, and is considered primary (POA) or can be produced in the atmosphere by oxidation of organic gaseous precursors and is considered secondary (SOA) (Kanakidou et al., 2005; Turpin and Huntzicker, 1995). POA is emitted during combustion of fossil fuels, biomass burning, cooking, etc. (Hallquist et al., 2009; He et al., 2010; Mohr et al., 2009).

Aerosol mass spectrometry can quantify the mass spectra of OA and helps identify its sources (Jayne et al., 2000). Source apportionment techniques, such as Positive Matrix Factorization (PMF), are widely used in order to estimate the contributions of the various OA sources (Lanz et al., 2007; Mohr et al., 2009; Paatero and Tapper, 1994). Sources that are often identified include oxygenated OA (OOA), hydrocarbon-like OA (HOA), biomass burning OA (BBOA), and cooking OA (COA) (Aiken et al., 2009; He et al., 2010; Herndon et al., 2008; Huang et al., 2011; Mohr et al., 2009; Saarikoski et al., 2012; Zhang et al., 2007).

During wintertime, wood burning for domestic heating purposes is one of the main sources of OA in several countries (Alfarra et al., 2007; Puxbaum et al., 2007). Biomass burning is a major source of gas- and particle-phase air pollution on urban (Robinson et al., 2006; Schauer and Cass, 2000), regional (Watson, 2002; Wotawa and Trainer, 2000) and global (Bond et al., 2004; Lelieveld et al., 2001) scales. Violations of daily PM standards due to wood burning have been reported in major European cities such as London, Paris and Berlin (Fuller et al., 2013; 2014). In several countries while traffic and industrial emissions are decreasing, the impact of residential wood burning emissions on air quality is increasing (Favez et al., 2010).

In Greece, the use of woodstoves and fireplaces has increased dramatically during the last few years. Pikridas et al. (2013) reported that biomass burning for heating purposes was the dominant source of OA during wintertime in Patras, the third biggest city of Greece. Elevated mass concentrations of PM<sub>2.5</sub> (diameter less than 2.5 μm), due to wood burning were also observed in the capital and largest city of Greece, Athens, coinciding with the beginning of the economic crisis in Greece (Paraskevopoulou et al., 2014). Measurements conducted during 2010 and 2012, in the city of Thessaloniki (Pettrakakis et al., 2013) showed an increase of PM<sub>10</sub>

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(diameter less than 10  $\mu\text{m}$ ) by 13% and  $\text{PM}_{2.5}$  by 25% during domestic heating hours. Gaidajis et al. (2014) reported the same behavior in Northern Greek cities (Kavala and Drama), where the daily average  $\text{PM}_1$  concentrations exceeded  $40 \mu\text{g m}^{-3}$ .

In this study, a HR-ToF-AMS was deployed in order to investigate the chemical composition of  $\text{PM}_1$  in two Greek cities (Athens and Patras) during two different wintertime periods (2012 and 2013). The PMF source apportionment algorithm, used unconstrained, was applied to the corresponding datasets, estimating the contributions of the different OA sources, without assuming any a priori knowledge of their origin. Previous studies in these areas have relied on filter-based measurements of the  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  concentrations (Argyropoulos et al., 2012; Chaloulakou et al., 2003; Katragkou et al., 2009; Koulouri et al., 2008; Theodosi et al., 2011) and had limited or no information about the composition of the OA. Few PM source apportionment studies are available for Athens. Dall'Osto and Harrison (2006) using an aerosol time-of-flight mass spectrometer (ATOFMS) found two dominant particle types in Athens: calcium-rich dust particles and secondary carbonaceous ones. The main objectives of this paper is to quantify the contribution of wood combustion and the fine PM levels of additional OA sources in the corresponding areas during wintertime.

## 2. Experimental Section

### 2.1 Measurement Sites

**2.1.1 Patras:** Patras is the third biggest city of Greece with 300,000 inhabitants and is situated at the foothills of Mount Panachaikon, overlooking the Gulf of Patras, 220 km west of Athens. The instruments were housed inside the campus of the Technological Educational Institute (TEI) of Patras ( $38^{\circ}13' \text{ N}$ ,  $21^{\circ}45' \text{ E}$ ) and were monitoring from February 26 to March 5, 2012. The TEI is in the southwest of the city, surrounded by moderate traffic streets and houses and is considered an urban site (Fig. S1). Measurements of black carbon (BC) were made at the center of the city ( $38^{\circ}14' \text{ N}$ ,  $21^{\circ}44' \text{ E}$ ), approximately 4 km away from the main site. A more detailed description of the two sites and the instrumentation used can be found in Pikridas et al. (2013).

**2.1.2 Athens:** Athens is the capital and largest city of Greece, with approximately 5 million inhabitants within the Greater Athens Metropolitan area, which represents almost half of the country's population (Fig. S1). An extensive campaign was conducted in the center of Athens from January 10 until February 9, 2013, the year following the Patras campaign. The measurements took place on the premises of the National Observatory of Athens (NOA) at

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Thiseio (37°58' N, 23°43' E). NOA is situated on the small hill of the Nymphs (100 m above sea level), overlooking the Acropolis and near the city center of Athens.

## 2.2 Instrumentation

The same instruments and sampling approach were used during both campaigns. More specifically:

*HR-ToF-AMS*: The Aerodyne Aerosol Mass Spectrometer (AMS), developed by Jayne et al. (2000), can provide continuous quantitative size and composition information for the non-refractory (NR) components of submicron atmospheric aerosol in real time. The chemical composition of NR-PM<sub>1</sub> organic and inorganic (nitrate, sulfate, ammonium and partially potassium) aerosol particles was monitored using a High Resolution Time of Flight AMS (HR-ToF-AMS) (Canagaratna et al., 2007; DeCarlo et al., 2006; Drewnick et al., 2005). The sample during this study was not dried, and the vaporizer surface temperature was set at 600 °C. The HR-ToF-AMS measurements during both campaigns alternated between the V- and W-modes every 3 min, but only the V-mode data are presented here.

*SMPS*: The ambient aerosol number distribution, for particles smaller than 500 nm in diameter, was measured by a TSI Scanning Mobility Particle Sizer (SMPS) (model 3081) coupled with a Condensation Particle Counter (CPC) (model 3775). The system operated at a sheath flow rate of 5 L min<sup>-1</sup>, a sample flow rate of 1 L min<sup>-1</sup>, without drying the sample.

*MAAP*: PM<sub>2.5</sub> aerosol absorption was monitored continuously during the two campaigns by a Multi Angle Absorption Photometer (MAAP, Thermo Scientific Inc. model 5012). The MAAP estimates black carbon (BC) mass loadings by combining aerosol optical absorption and back scattering (Petzold and Schönlinner, 2004). The MAAP was set at a wavelength of 637 nm during the measurements and its inlet was heated to 70 °C.

*PTR-MS*: The volatile organic compounds (VOCs) during the Athens campaign were measured by a Proton Transfer Reaction- Mass Spectrometry (PTR-MS, Jonicon Analytik Inc.), the operation principles and application details of which can be found in Steinbacher et al. (2004). Additional information and analysis about these VOC measurements during the sampling period are presented by Kaltsonoudis et al. (2016).

*Other instrumentation*: The aerosol volatility was quantified using a thermodenuder (TD), based on the design of An et al. (2007). A detailed analysis of these measurements is presented in Louvaris et al. (2016) and Gkatzelis et al. (2016). The concentrations of carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>) and ozone (O<sub>3</sub>) were measured by

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the corresponding Teledyne monitors (models 300E, T360, T201 and 400E, respectively), Additional information is presented by Kaltsonoudis et al. (2016). Also, three different filter samplers were used in order to collect PM<sub>2.5</sub> samples for the measurement of organic carbon (OC), elemental carbon (EC), water-soluble organic carbon (WSOC) and the main ions (NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, and NO<sub>3</sub><sup>-</sup>). The Met One SuperSASS (Speciation Air Sampler System) was deployed during the Patras campaign, while a dichotomous Partisol sampler (Rupprecht & Patashnick Co., Inc.) and a Partisol FRM (Federal Reference Method) Model 2000 were used in Athens. Both teflon and quartz fiber filters were used in Athens and Patras at daily, 12-hour and 6-hour collection intervals. Details about the filter extraction procedure and chemical analysis can be found in Pikridas et al. (2013) and Paraskevopoulou et al. (2014).

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### 2.3 AMS Data analysis

All HR-ToF-AMS data were analyzed using the standard AMS software toolkit (SQUIRREL v1.53), while the high-resolution data were processed using the Peak Integration by Key Analysis (PIKA v1.10C, Sueper, 2011) software within Igor Pro 6.34A (Wave Metrics). For the determination of the elemental ratios measured by the HR-AMS both the method of Aiken et al. (2008) and the improved ambient calculation approach proposed by Canagaratna et al. (2015) were used. In order to calculate the collection efficiency (CE) of the HR-ToF-AMS, we combined the HR-ToF-AMS mass distributions and the SMPS volume distributions (for particle diameters between 10 and 500 nm) applying the algorithm of Kostenidou et al. (2007) every two hours. The algorithm was modified to take into account the BC measured by the MAAP, assuming that the BC had the same size distribution as the OA.

PMF analysis (Lanz et al., 2007; Paatero and Tapper, 1994; Ulbrich et al., 2009) was performed using as input the HR-AMS organic mass spectra ( $m/z$  values 12-200) in order to determine the different OA sources. The mass spectra of the identified factors and those in the literature have been compared using the theta angle ( $\theta$ ) between the corresponding vectors (Kostenidou et al., 2009). This method treats mass spectra signals as vectors in a multi-dimensional space, with size equal to the number of  $m/z$  values used. Two spectra are considered to be the same, if the angle theta ( $\theta$ ) between them, is zero. In practice, a low value of the theta angle (less than  $10^\circ$ ) indicates high similarity between the two spectra. This approach has been utilized by commercially available software to match unknown mass spectra with references from mass spectral libraries (Stein and Scott, 1994). For the comparison of the OOA and HOA spectra with the ones in the literature, only HR spectra using the fragmentation table by Aiken et

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al. (2009) were used. The BBOA and COA spectra have been compared with almost all the corresponding spectra found in the database (both HR and unite mass resolution), resulting in higher theta values due to differences at  $m/z$  18, 28, and 39. By excluding these specific  $m/z$  values, the theta angle decreased.

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### 3. Meteorology

During the Patras campaign, the ambient temperature ranged from 4 °C to 16 °C, with an average of 11 °C. The average RH was 75% (ranging from 40 to 90%) (Figure 1). Three types of air masses were identified using back trajectory analysis based on FLEXPART (Stohl et al., 2005) during the campaign (Fig. S2). The periods Feb. 25-26 and March 3-4 were characterized by air masses coming predominantly from western Greece (44% of the air masses). During February 26-27 and March 5 the air masses had marine origin (14%), while air masses during February 28 to March 2 were originating from central Greece and the Balkans (42%). Examples of the three categories are depicted in Fig. S3.

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During the Athens campaign, the average ambient temperature was 12.5 °C (ranging from 6 °C to 19 °C), while the average RH was 71% (ranging from 25 to 95%) (Fig. 2). FLEXPART indicated four types of air masses arriving at the site. The first type originated from Africa and passed over the Mediterranean Sea. The second had marine origin coming from the west often passing over Sicily. The third came from the NE passing over the Aegean and the Eastern Balkans. Finally, the fourth type included air masses from central Greece and the western Balkans. Examples of these trajectories are depicted in Fig. S4 and a summary of the air masses observed are provided in Fig. S5.

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## 4. PM<sub>1</sub> chemical composition and characteristics

### 4.1 PM<sub>1</sub> chemical composition

4.1.1 Patras: The estimated mean value of the AMS collection efficiency (CE) during the wintertime campaign in Patras was  $0.77 \pm 0.05$ . The density of organics estimated by the same algorithm was  $1.45 \pm 0.04$  g cm<sup>-3</sup>, which is close to the one for Patras during the summer of 2012 ( $1.34 \pm 0.21$  g cm<sup>-3</sup>, Kostenidou et al., 2015). The one-hour averaged PM<sub>1</sub> mass concentration (estimated as the sum of the concentration of the AMS and MAAP) varied from a few μg m<sup>-3</sup> up to more than 100 μg m<sup>-3</sup> (Fig. 1). The average PM<sub>1</sub> mass concentration was approximately 25 μg m<sup>-3</sup> (of which 2 μg m<sup>-3</sup> was BC and 18 μg m<sup>-3</sup> was OA). The OA mass concentration increased dramatically during the evening hours, reaching levels up to 85 μg m<sup>-3</sup>. The OA also increased

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during the morning rush hour (7:00-10:00 LST), but the corresponding concentrations were less than  $25 \mu\text{g m}^{-3}$ . BC concentrations followed closely the variation of OA, with maximum values around  $8 \mu\text{g m}^{-3}$  during nighttime and  $4 \mu\text{g m}^{-3}$  during rush hour in the mornings. The nitrate mass concentration followed a similar trend with OA with significant peaks occurring both during the morning and evening hours.  $\text{PM}_{10}$  nitrate was on average  $1 \mu\text{g m}^{-3}$ , but higher values up to  $5 \mu\text{g m}^{-3}$  were measured during the second half of the campaign (from March 1 to 5). The sulfate concentration was on average  $2.6 \mu\text{g m}^{-3}$  and had smaller fluctuations during the campaign, indicating a different source origin. Chloride's average mass concentration was  $0.4 \mu\text{g m}^{-3}$ , but reached values up to  $7 \mu\text{g m}^{-3}$  (March 4). The ammonium concentration followed closely that of sulfate, chloride, and nitrate, indicating the formation of the corresponding ammonium salts.

**4.1.2 Athens:** The mass concentrations of OA, sulfate, nitrate, ammonium, chloride, and BC are depicted in Fig. 2. The dominant component of  $\text{PM}_{10}$  was OA (61%). The average AMS CE during the campaign calculated by the Kostenidou et al. (2007) algorithm was  $0.85 \pm 0.23$  and the density of OA was  $1.29 \pm 0.21 \text{ g cm}^{-3}$ . A similar OA density was estimated for Athens during the summer of 2012 ( $1.15 \pm 0.36 \text{ g cm}^{-3}$ , Kostenidou et al., 2015). During the wintertime campaign in Athens, the average total  $\text{PM}_{10}$  mass concentration was approximately  $10 \mu\text{g m}^{-3}$  (of which,  $2 \mu\text{g m}^{-3}$  was BC), while the hourly maximum value was  $140 \mu\text{g m}^{-3}$  (of which  $120 \mu\text{g m}^{-3}$  was OA). OA was the major component of  $\text{PM}_{10}$  with frequent nighttime (after 18:00 LST) peaks, exceeding  $40 \mu\text{g m}^{-3}$  (Fig. 2). During most campaign days  $\text{PM}_{10}$  was lower than  $20 \mu\text{g m}^{-3}$  due to frequent rain and strong winds (Fig. S6). BC during the nighttime was as high as  $15 \mu\text{g m}^{-3}$ , while the corresponding morning rush hour maximum was  $8 \mu\text{g m}^{-3}$ . Average ammonium mass concentration was low and around  $0.4 \mu\text{g m}^{-3}$ , while the average sulfate was  $0.96 \mu\text{g m}^{-3}$ . The nitrate was, on average,  $0.48 \mu\text{g m}^{-3}$ , but its maximum values were up to  $3.4 \mu\text{g m}^{-3}$  during nighttime. The correlation between OA and nitrate was moderate ( $R^2=0.59$ ).

During the high mass concentration days, and between 18:30 to 02:30 LST, OA was highly correlated ( $R^2=0.95$ ) with acetonitrile (PTR-MS,  $m/z$  42,  $\text{CH}_3\text{CN}^+$ ), indicating that biomass burning was the dominant OA source during these hours. As a reference, during the remaining period (02:30 to 18:30 LST) of the measurements the correlation between OA and acetonitrile deteriorated to  $R^2=0.54$ . During the same periods (18:30 to 02:30 LST), isoprene and furan (PTR-MS,  $m/z$  69) as well as benzene (PTR-MS,  $m/z$  79) were also correlated ( $R^2=0.93$  and  $R^2=0.95$ , respectively) with OA, and also with each other ( $R^2=0.95$ ). Isoprene and benzene, both

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constituents of petrol fuel (Borbon et al., 2001), are emitted to the atmosphere by transportation sources (Langford et al., 2009). However, furan and benzene are also emitted during from biomass burning (Gilman et al. 2015, Stockwell et al. 2015, Hatch et al. 2015); therefore, these species may be affected by other sources than just petrol combustion. A detailed analysis of the wintertime Athens VOC data can be found in Kaltsonoudis et al. (2016).

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**4.1.3 Diurnal profiles:** The average diurnal profiles of the main PM<sub>1</sub> species during the two campaigns are shown in Fig. 3. In Patras, the average OA and BC concentrations were characterized by two major peaks during the day (Fig. 3). The first one appeared at around 8:00 LST, coinciding with the morning rush hour traffic, while the second one, which was approximately two times higher for both OA and BC, at 20:00 LST. During the evening, the OA and BC concentrations increased considerably representing almost 90% of the total PM<sub>1</sub> concentration, until around midnight. Sulfate had a relatively flat diurnal profile, while nitrates had two peaks, which coincided with those of OA and BC. This suggests that nitrate and OA had, at least to some extent, similar sources. The diurnal variation of chloride had a major morning peak at around 7:00 LST. This could be due to two Olympic-size swimming pools, located in a major athletic center, less than 1 km from the sampling site. The peak coincides with their opening and cleaning hours. The data suggest that the chloride peaks occurred on average an hour earlier (at 7:00 LST) than the corresponding OA peaks (at 8:00 LST). This coupled with the major differences between known AMS fingerprints for trash burning and the estimated PMF factors here, suggest that trash burning was not a major contributor to the morning chloride peaks. There was also an evening chloride peak (at 20:00 LST) coinciding with those of OA and BC.

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In Athens, the OA average diurnal profile was dominated by an evening peak at around 23:00 LST, almost three hours later than in the Patras campaign. BC exhibited two major peaks, one in the morning at 8:00 LST, which was attributed to the morning rush hour, and another one in the evening at 23:00 LST, coinciding with the OA peak (Fig. 3). This suggests that OA and BC probably originated from the same source during the night. Nitrate also displayed two peaks, one in the morning (9:00 LST) during the peak traffic hours, and another one in the late evening (23:00 LST). Chloride and ammonium exhibited an evening peak at 23:00 LST, while sulfate had once more a rather flat average diurnal profile.

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**4.1.4 Size distributions:** The average size distributions of the different aerosol species (sulfate, organics, ammonium, chloride and nitrate) as a function of the vacuum aerodynamic diameter are depicted in Fig. S7. In Patras, sulfate peaked at 340 nm, whereas OA peaked at 140 nm, implying that the PM<sub>1</sub> particles during the sampling period had different composition at different size ranges. The size distribution of ammonium mainly followed that of sulfate, contrary to nitrate, which followed that of OA suggesting that a major fraction of nitrate was in the form of organonitrates (ON). In Athens, the sulfate size distribution peaked at 310 nm, close to the value during the Patras campaign, whereas OA peaked at 250 nm. Nitrate peaked at the same diameter (250 nm) as OA.

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**4.1.5 Organonitrates:** The NO<sub>2</sub><sup>+</sup>/NO<sup>+</sup> ratio was used in order to estimate the concentration of particulate organonitrates (ON) (Farmer et al., 2010; Fry et al., 2009). Typical values of NO<sub>2</sub><sup>+</sup>/NO<sup>+</sup> for ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) have been reported to be in the range from 0.33 to 0.85 (Alfarra et al., 2006; Cottrell et al., 2008; Fry et al., 2009), while those for organonitrates from 0.07 to 0.1 (Fry et al., 2009; Hao et al., 2014). ON are also known to be components of SOA (Farmer et al., 2010; Fry et al., 2009; Ng et al., 2008).

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In this work, the measured NO<sub>2</sub><sup>+</sup>/NO<sup>+</sup> ratio for pure NH<sub>4</sub>NO<sub>3</sub> in Athens and Patras was equal to 0.59 and 0.75, respectively. The fraction of ON to the total nitrate was estimated using the equation suggested by Farmer et al. (2010). The minimum observed NO<sub>2</sub><sup>+</sup>/NO<sup>+</sup> ratio during each campaign was 0.04 for Athens and 0.055 for Patras.

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The average ON fraction of the total nitrate in Athens and Patras was 42±25% and 45±14%, respectively. In Athens, during nighttime (17:00 until 3:00 LST, for the whole dataset), the average ON fraction was 38±20%. During nights (from 20:00 until 3:00 LST) in enhanced OA plumes exceeding 15 µg m<sup>-3</sup>, the ON fraction was lower, and equal to 19±10%. This suggests that during biomass burning events ON is not the major component of nitrate. In Patras, the average ON fraction for the whole campaign was 45±14%. During evening OA peaks (16:00 until 24:00 LST, for the whole dataset) the average ON fraction was 44±13%.

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## 4.2 OA elemental ratios

The atomic oxygen-to-carbon ratio (O:C) of OA is a useful metric of its extent of oxygenation, which increases as the aerosol is exposed to atmospheric oxidants (Murphy et al., 2011; Sage et al., 2008).

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In Patras, the average O:C ratio was  $0.42 \pm 0.09$  following the Canagaratna et al. (2015) approach, while the hydrogen-to-carbon ratio (H:C) was  $1.71 \pm 0.06$ . For comparison with literature, the corresponding values following the Aiken et al. (2008) approach were  $0.33 \pm 0.07$  and  $H:C = 1.53 \pm 0.05$ . The corresponding time series are shown in Figs. S8 and S9. In Athens, the average O:C ratio based on the Canagaratna approach was  $0.32 \pm 0.11$  ( $0.26 \pm 0.09$  for the Aiken approach) while the H:C was  $1.72 \pm 0.11$  ( $1.59 \pm 0.11$  using the Aiken approach). The temporal evolution of these ratios is also depicted in Fig. S8 and Fig. S9. The relatively low average values of O:C for both Athens and Patras are consistent with the OA being dominated by primary emissions (Allan et al., 2010). The corresponding O:C ratios during summertime for Athens and Patras were approximately 0.2 units higher (Kostenidou et al., 2015).

Another metric of the extent of oxidation of atmospheric OA, is the average carbon oxidation state  $OS_C$ , a value that increases upon oxidation (Kroll et al., 2011). The  $OS_C$  for both cities was calculated by applying the equation proposed by Kroll et al. (2011):

$$OS_C = 2(O:C) - (H:C) \quad (1)$$

In Patras,  $OS_C$  was  $-0.88 \pm 0.19$ , ranging from -1.40 to -0.37 while in Athens, the  $OS_C$  was on average  $-1.08 \pm 0.28$  varying from -1.8 to 0.98 (Fig. S10).

The average diurnal profiles of O:C, H:C and  $OS_C$  for both cities are shown in Fig. 4. The changes in the O:C ratio were in the opposite direction from those of the H:C. The decreases in O:C and the corresponding increases in H:C during the morning and midday rush hours were due to the fresh traffic emissions. The corresponding changes during the evening were due to the biomass burning activity. Background OA dominated the observed O:C ratio during the rest of the day. In Patras, the O:C ratio increased significantly during the late night hours (2:00 to 7:00 LST), during the morning (peak at around 10:00 LST), and also at around 15:00 in the afternoon. The primary emissions, mainly from traffic, led in this case too to reductions of the average O:C and increases in H:C, during the corresponding peak periods. In Athens, the O:C increased during the early morning hours and after 10:00 LST. The diurnal profiles of  $OS_C$  were similar to those of O:C. The mean organic-aerosol-to-organic-carbon ratio (OA/OC) for Athens was  $1.50 \pm 0.1$ , and varied between 1.2 and 2.2 during the measurement period, while in Patras it was  $1.59 \pm 0.1$ , ranging from 1.3 to 2.

## 5. Source apportionment of OA

Positive matrix factorization (Paatero and Tapper, 1994) (PMF) was applied to the HR-ToF-AMS organic mass spectra ( $m/z$  up to 200), using the PMF Evaluation Tool, PET (Lanz et al., 2008;

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Ulbrich et al., 2009). The selection of the PMF solution was based on the comparison of the spectra of the calculated factor to those of the literature, their physical meaning, their diurnal cycles, the correlation of the factor's resulting time series with each other and other variables (usually concentrations of other pollutants), and finally based on the PMF-model residuals. For both campaigns, biomass burning was identified as the dominant source of OA, and was responsible for 40-60% of the total OA. The results for both cities are summarized in Figure 5.

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For Patras, PMF solutions with 1 to 7 factors were investigated. In the three factor solution a BBOA, a HOA and an OOA factor were obtained, with BBOA being the dominant component of OA. Moving to a 4-factor solution, a second factor related to biomass burning was obtained quite different from the other, with a noticeable difference in both fractions of  $m/z$  44 and 60 ( $f_{44}$  and  $f_{60}$ ), and a high value of theta angle ( $\theta=32^\circ$ ) between the mass spectra. In the five-factor solution, a COA factor was resolved. The increase in the number of factors up to this point was accompanied by a reduction in the residuals. On the other hand, the model residuals for the 6 and 7-factor solutions were almost identical to those of the 5-factor solution. Thus, a 5-factor solution was selected for the specific dataset (SI, Section 7, Figures S11-S17). The two biomass burning-related OA factors (BBOA-I and BBOA-II) dominated the measured OA composition (almost 60%) during the evenings. The OOA factor accounted on average for 18% of the OA, while HOA and COA represented 12% and 11%, respectively. The BBOA, HOA and COA factors correspond to primary sources, whereas the OOA to atmospheric processing (secondary OA). The PMF factors' time series and contribution to  $PM_{10}$  OA are depicted in Fig. 6.

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In Athens, the PMF model resolved four sources of OA: BBOA (43%), OOA (24%), HOA (17%) and COA (16%). Their time series and contribution to  $PM_{10}$  organic aerosol are depicted in Fig. 7. A PMF analysis with up to 6 factors was performed (SI, Section 7, Figures S18-S24). By extending the solution from a 4 to a 5-factor solution, another biomass burning-related (BBOA-II, 14% of OA) factor was generated in addition to the first one (BBOA-I, 39% of OA). The PMF residuals in this case decreased slightly and only for the two first days of the campaign (Fig. S19). For this solution, the mass concentrations of OOA (20% of OA), HOA (13% of OA), and COA (14% of OA) were pretty similar to those in the 4-factor solution. The 5-factor solution was not used because the two biomass burning factors resembled each other ( $\theta=15^\circ$ ) and had similar diurnal profiles. The correlations between the BBOA-I and BBOA-II factors in Athens and VOCs as measured by the PTR-MS are shown in Table S1.

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Figure 8 summarizes the mass spectra of the identified OA components. Each one of these is analyzed in the following sections.

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## 5.1 Biomass Burning-related Organic Aerosol

**5.1.1 Patras:** The selected five-factor PMF solution for Patras included two BBOA-like factors named BBOA-I and BBOA-II. Both of these factors had significant contributions at the  $m/z$  values 60 ( $C_2H_4O_2^+$ ) and 73 ( $C_3H_5O_2^+$ ), that are typical fragments of levoglucosan, a marker for biomass burning (Aiken et al., 2009; Alfarrá et al., 2007; Cubison et al., 2011; Heringa et al., 2011). However, their mass spectra were quite different ( $\theta=32^\circ$ ) and their time series had a rather low correlation to each other ( $R^2=0.2$ ). The two BBOA factors represented 60% of the OA on average during the campaign with their contribution rising to 90% or so during the night hours (18:00 to 24:00 LST). The fraction of  $m/z$  60, characteristic of levoglucosan, was  $f_{60}=0.025$  in BBOA-I and much lower,  $f_{60}=0.009$ , in BBOA-II.

The BBOA-I factor was by far the most important component of  $PM_{10}$  OA (40%) in Patras with a mean concentration of  $7 \mu g m^{-3}$ . BBOA-I levels increased after 16:00 LST, reaching values exceeding  $35 \mu g m^{-3}$  at around 20:00 LST (Fig. 6). During the evening periods it represented up to 75% of the total OA while during the day its concentration was always less than  $5 \mu g m^{-3}$ . The BBOA-I factor correlated well with the AMS  $K^+$  ( $R^2=0.76$ ), and had modest correlations with the total nitrate measured by the AMS ( $R^2=0.55$ ), CO ( $R^2=0.40$ ), and BC ( $R^2=0.27$ ). The corresponding coefficient of determination between the BBOA-I and chloride, was low and equal to 0.16. Also, BBOA-I correlated better ( $R^2=0.31$ ) with ON, than BBOA-II ( $R^2=0.09$ ), implying that BBOA-I was a more significant source of ON in Patras.

BBOA-II was the second biomass burning-related factor, contributing 19% to the  $PM_{10}$  OA in Patras. Its contribution was higher during the first days when it represented up to 40% of the total OA. Its mean value for the whole campaign was equal to  $3.3 \mu g m^{-3}$ , and its hourly maximum value almost  $21 \mu g m^{-3}$ . BBOA-II correlated poorly with all the major AMS species ( $R^2 < 0.1$ ) and modestly with BC ( $R^2=0.26$ ). The  $R^2$  between BBOA-II and the AMS chloride was 0.02. The low correlations between both BBOA-I and BBOA-II to chloride, did not support the link between one of these factors and trash burning or chloride. Its diurnal cycle exhibited two evening peaks, one at 18:00 (LST) and a smaller one at 22:00 (LST). Its mean mass concentration during daytime was less than  $3 \mu g m^{-3}$ , and close to the corresponding one of BBOA-I.

The BBOA-I and BBOA-II spectra for Patras are shown in Fig. 8. The BBOA-I spectrum had strong signals at  $m/z$  29 ( $CHO^+$ ), 43 ( $C_2H_3O^+$ ), 60 ( $C_2H_4O_2^+$ ) and 73 ( $C_3H_5O_2^+$ ), while the spectrum of BBOA-II was characterized by lower signal in  $m/z$  values attributed to fresh OA, like 43, 55 and 57, and a stronger signal at  $m/z$  44, which is usually related to aged organic

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aerosol ( $f_{44}$  was equal to 0.043). The contribution of  $m/z$  60 in the BBOA-II spectrum was 67% lower compared to the BBOA-I spectrum, suggesting the evaporation of levoglucosan. There was also a prominent peak at  $m/z$  39, as well as a peak at  $m/z$  91, which can be indicative of aromatic species such as benzyl ions (Allan, 2003; Allan et al., 2004; Trainer et al., 2012). This  $m/z$  91 peak was also present in the spectrum of the BBOA factor from the burning of olive tree branches in Greece (Kostenidou et al., 2013). In general, people in Greece use multiple fuel types that can vary from region to region due to the local availability of the corresponding type of biomass. For example, in Patras softwood (pine, fir, etc.) or hardwood (oak, beech, olive tree) is used. In Athens, people burn mainly pine and fir. The O:C ratio of BBOA-I was 0.3 while that of BBOA-II was equal to 0.26. The corresponding OA/OC ratios were 1.55 and 1.47. These two factors could correspond to different degrees of aging of the BBOA, different types of combustion or fuel, or a combination of these mechanisms.

Another explanation is that the two BBOA factors in Patras could correspond to different fuel types with one of them including trash burning. To test this hypothesis, the spectra of the BBOA factors were compared to those of OA resulting from paper and plastic burning (Mohr et al., 2009). The trash burning AMS spectra were completely different from the BBOA spectra in this study, with angles exceeding 45 degrees. This suggests that trash burning cannot be the main source of any of the two BBOA factors in Patras.

Recent work has shown that the emissions of nitrogen containing organic compounds (such as acetonitrile) strongly depend on the composition of the fuel (Coggon et al. 2016). Fuels containing low amounts of nitrogen (e.g. wood) emit lower amounts of N-organics, than fuels containing large amounts of nitrogen (e.g. grasses, the boughs of trees). The same behavior has been observed for inorganic nitrogen gasses, such as ammonia (NH<sub>3</sub>) and NO<sub>x</sub> (e.g. Burling et al., 2010). Consequently, if different fuels were burned, then different amounts of ON could be formed due to differences in the amount of NO<sub>x</sub> emitted or, perhaps, differences in emissions of primary organic nitrogen.

The BBOA factor in ambient studies is believed to represent an average of the emissions of all fuel types. However, the corresponding emissions should have either a different temporal or a spatial profile in order to be separated by PMF. Given that residential biomass burning, in a city like Patras, is due to tens of thousands of individual sources (fireplaces, wood stoves, etc.), spread all over the city following a similar burning schedule on average, it is difficult to explain how PMF would be able to separate the BBOA from different fuel types. However, there are other potential explanations (e.g., different areas of the city using different fuel types and the

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The null hypothesis is that the BBOA factor represents an average of the emissions of all fuel types. However, the corresponding emissions should have either a different temporal or a spatial profile in order to be separated by PMF. Given that residential biomass burning, in a city like Patras, is due to tens of thousands of individual sources (fireplaces, wood stoves, etc.), spread all over the city following a similar burning schedule on average, it is difficult to explain how PMF would be able to separate the BBOA from different fuel types. However, there are other potential explanations (e.g., different areas of the city using different fuel types and the

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[emissions arriving at the site at different types](#)), that could involve the use of different fuel types. Therefore, even if the support of this hypothesis is rather weak, it cannot be eliminated as a potential explanation.

**5.1.2 Athens:** The biomass burning OA (BBOA) factor contributed almost half (43%) of the OA in Athens. BBOA reached hourly levels up to  $80 \mu\text{g m}^{-3}$  during the evening of the first day of the campaign, which was the coldest day during the period of the measurements. The average BBOA concentration was  $2.3 \mu\text{g m}^{-3}$ . However, when meteorological conditions were favorable (no precipitation), BBOA sharply increased after 17:00 LST, frequently reaching values greater than  $20 \mu\text{g m}^{-3}$  at approximately 23:00 LST (Fig. 7). This peak was observed a few hours later in Athens than in Patras, potentially due to the different lifestyles in the two cities. A peak was also observed between 1:00 and 4:00 LST. A similar peak was present in the diurnal profile of the AMS  $\text{K}^+$  (Fig. S25), implying a common source for both. This could be due to either morning combustion or vertical mixing. During the rest of the day the BBOA levels remained lower than  $1 \mu\text{g m}^{-3}$ .

BBOA and gas-phase acetonitrile (PTR-MS,  $m/z$  42,  $\text{CH}_3\text{CN}^+$ ) were strongly correlated ( $R^2=0.81$ ). BBOA was also well correlated with AMS chloride ( $R^2=0.88$ ). Moderate correlations were observed with CO ( $R^2=0.58$ ), AMS  $\text{K}^+$  ( $R^2=0.40$ ), BC ( $R^2=0.33$ ) and  $\text{NO}_x$  ( $R^2=0.29$ ). High correlations were observed between BBOA and 2-methyl-3-buten-2-ol (MBO) (PTR-MS,  $m/z$  87) ( $R^2=0.93$ ) which is related with emissions from pine trees (Vlasenko et al., 2009), isoprene and furan at  $m/z$  69 ( $\text{C}_5\text{H}^+$  and  $\text{C}_4\text{H}_5\text{O}^+$ ) ( $R^2=0.88$ ), methacrolein and methyl vinyl ketone ( $\text{C}_4\text{H}_7\text{O}^+$ ) at  $m/z$  71 ( $R^2=0.81$ ), and  $m/z$  85 (among possible candidates, ethyl vinyl ketone) ( $R^2=0.9$ ). These results are consistent with previous studies suggesting that wood burning is major source of biogenic VOCs (Akagi et al., 2011). The correlation between the BBOA factor in Athens and VOCs as measured by the PTR-MS are shown in Table 1.

The AMS spectrum of BBOA in Athens had prominent peaks at  $m/z$  39 ( $\text{C}_2\text{H}_2^+$ ), 60 ( $\text{C}_2\text{H}_4\text{O}_2^+$ ) and 73 ( $\text{C}_3\text{H}_5\text{O}_2^+$ ). A high contribution to the total organic signal was present at  $m/z$  29 mostly due to  $\text{CHO}^+$ , consistent with the findings of Alfara et al. (2007). The BBOA high resolution (HR) spectra, with the dominant ion families are represented in Fig. 8. The BBOA O:C ratio in Athens was 0.27.

**5.1.3 BBOA factor comparisons:** The mass spectra of BBOA in Athens and BBOA-I in Patras had some similarity ( $\theta=14^\circ$ ) with main differences at  $m/z$  29, 39, 43, 57, 60 and 73 (Fig. S26).

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Their  $f_{44}$  were close to each other (0.02). The mass spectra of BBOA in Athens and BBOA-II in Patras were quite different ( $\theta=25^\circ$ ). Their major differences were presented at  $m/z$  values 26, 28, 29, 39, 41, 43, 44, 55, 57, 60, and 73.

Residential burning was the main BBOA source in Greece, during the present study. The BBOA-I spectrum in Patras was similar to that of residential wood combustion in Fresno ( $\theta=11^\circ$ ) (Ge et al., 2012), and to BBOA from the MILAGRO (Megacity Initiative: Local and Global Research Observations) campaign ( $\theta=14^\circ$ ) (Aiken et al., 2009) (Fig. S27). The BBOA-II spectrum in Patras was quite different (angles  $\theta$  exceeding  $24^\circ$ ) than those in the literature (Fig. S28). All types of wood burning were included in the comparisons, for completeness. Comparisons of the spectrum of BBOA-II with those of OOA factors revealed lower theta values (less than  $30^\circ$ ) (Fig. S29). This indicates a moderate resemblance of BBOA-II and chemically aged aerosol. The theta angle was lower than  $30^\circ$  with the spectra of OOA in Fresno (Ge et al., 2012), New York (Sun et al., 2012), and SV-OOA from SOAR (Study of Organic Aerosols at Riverside) (Docherty et al., 2011).

The Athens BBOA spectrum was similar ( $\theta=12^\circ$ ) to that of the BBOA factor in Fresno (Ge et al., 2012), had some similarity with spectra from the burning of ponderosa pine duff ( $\theta=21^\circ$ ) and the Alaska core Tundra duff ( $\theta=22^\circ$ ) during FLAME-I (Fire Lab At Missoula Experiment, Phase 1). For the rest of the database spectra, the theta angle values exceeded  $25^\circ$  (Fig. S30).

## 5.2 Oxygenated Organic Aerosol (OOA)

OOA is often considered as a surrogate for secondary OA due to both local production processes and long range transport of chemically aged OA from other areas.

**5.2.1 Patras;** The OOA factor dominated daytime OA and its mass concentration increased during the last days of the campaign. The mean OOA was  $3.2 \mu\text{g m}^{-3}$ , while its hourly maximum value, around midday of March 5, was equal to  $10.8 \mu\text{g m}^{-3}$ . It represented 18% of the OA on average. For comparison, during summertime at the same area, OOA accounted for almost 80% of OA due to the higher photochemical activity (Kostenidou et al., 2015). The average diurnal pattern of OOA was relatively stable (Fig. 9) with small peaks during the morning (8:00 and 11:00 LST) and a little lower values after 18:00 LST. This pattern is consistent with a regional source and domination by atmospheric transport rather than local sources and processes The

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OOA was correlated reasonably well with particulate ammonium ( $R^2=0.61$ ) and sulfate ( $R^2=0.56$ ), while its correlation to nitrate was moderate ( $R^2=0.38$ ).

**5.2.2 Athens:** In contrast to other measurement datasets in Athens, in which two OOA types were obtained by PMF (a very oxidized OA, V-OOA, and a moderately oxygenated, M-OOA) (Kostenidou et al., 2015), only one type was clearly present in Athens during the winter. The OOA factor dominated OA during the daytime and its mass concentration increased after January 28, when air masses originated from the Balkans and not from the Mediterranean Sea (SI, Sections 3 and 4). The average OOA concentration was  $1.3 \mu\text{g m}^{-3}$ , with a maximum value of  $9.7 \mu\text{g m}^{-3}$ . OOA was correlated with  $\text{NH}_4^+$  ( $R^2=0.65$ ),  $\text{SO}_4^{2-}$  ( $R^2=0.58$ ), and  $\text{NO}_3^-$  ( $R^2=0.67$ ). The average diurnal concentration of OOA decreased smoothly during morning hours, reaching a minimum at around 15:00 LST, and increased again during the afternoon and evening, peaking at midnight (Fig. 9). Biomass burning-related VOCs such as monoterpenes ( $\text{C}_{10}\text{H}_{16}$ ), and isoprene ( $\text{C}_5\text{H}_8$ ) can react with  $\text{NO}_3$  radicals (Hodzic et al., 2009; Ng et al., 2008; Rollins et al., 2009) resulting in nighttime SOA production. In the case of Athens, the increase of OOA during the night is accompanied by increases in mostly inorganic nitrate, with a relatively small contribution of organic nitrate. This does not support the hypothesis that the OOA increase is due to the production of SOA from reactions of the biomass burning emissions with nitrate radicals.

In Patras, the wintertime OOA O:C ratio was 0.65 (Fig. 10). For comparison reasons, the O:C ratio for V-OOA, M-OOA and b-OOA during the summer campaign of 2012 in Patras, were 0.81, 0.54 and 0.48, respectively (Kostenidou et al., 2015). In Athens, the O:C ratio for OOA during the winter of 2013 was equal to 0.47 (Fig. 10). The corresponding values of O:C during summertime in Athens, for V-OOA and M-OOA were 0.68 and 0.56, respectively (Kostenidou et al., 2015).

**5.2.3 OOA factor comparisons:** Both OOA spectra were characterized by a strong peak at  $m/z$  44 (mostly due to  $\text{CO}_2^+$ ), related to the thermal decomposition of organic acids (Alfarra et al., 2004), a lower one at  $m/z$  43 (mostly due to  $\text{C}_2\text{H}_3\text{O}^+$ ), and another one at  $m/z$  28, all consistent with the OOA spectra reported in other studies. The OOA spectra in the two cities were quite similar to each other ( $\theta=11^\circ$ ), with the greater differences at  $m/z$  values 28 and 44. The OOA in Athens exhibited lower  $f_{44}$  (0.12) compared to the one in Patras (0.15).

The spectrum of OOA in Patras was quite similar to the OOAa and OOAb ( $\theta=7^\circ$ ) for the Po Valley (Saarikoski et al., 2012), the summertime spectra of moderate oxygenated OA (M-

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OOA) in Athens and Patras ( $\theta=10^\circ$ ) (Kostenidou et al., 2015), and the LV-OOA spectra ( $\theta=11^\circ$ ) from both the DAURE (Mohr et al., 2012) and SOAR campaigns (Docherty et al., 2011). Higher theta angles ( $\theta=20-40^\circ$ ) were found when comparing it to spectra of SV-OOA (Fig. S31) from different studies, implying that the OOA in Patras was closer to LV-OOA, instead of SV-OOA.

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The spectrum of OOA in Athens was similar to both the M-OOA identified in Athens ( $\theta=8^\circ$ ) and Patras ( $\theta=10.6^\circ$ ) during the summer (Kostenidou et al., 2015). It was also similar with the proposed biogenic OOA factor (b-OOA) ( $\theta=10^\circ$ ) in Patras for the same summer period, and all the OOA (OOA-a, OOA-b, OOA-c) factors ( $\theta=11-13^\circ$ ) identified in the Po Valley (Saarikoski et al., 2012). The values of the theta angles between the OOA spectra in this study and spectra in the literature are summarized in Figure S31.

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### 5.3 Hydrocarbon-Like Organic Aerosol (HOA)

**5.3.1 Athens:** The HOA mean value was  $0.95 \mu\text{g m}^{-3}$ , and its hourly maximum concentration was  $34 \mu\text{g m}^{-3}$ . HOA was correlated with CO ( $R^2=0.69$ ) and  $\text{NO}_x$  ( $R^2=0.74$ ), suggesting that traffic was a major HOA source. HOA was also correlated to AMS chloride ( $R^2=0.75$ ), and VOCs, such as methacrolein (MACR) (PTR-MS,  $m/z$  71) ( $R^2=0.74$ ), naphthalene (PTR-MS,  $m/z$  129) ( $R^2=0.73$ ), benzene (PTR-MS,  $m/z$  79) ( $R^2=0.65$ ), and the xylenes (PTR-MS,  $m/z$  107) ( $R^2=0.7$ ). The HOA correlation with BC for the whole campaign was low ( $R^2=0.15$ ). However, splitting the BC and HOA time series into two periods, one during the day (6:00-18:00 LST) and another one during the night (18:00-6:00 LST), led to high correlation during daytime ( $R^2=0.78$ ) and low during the nighttime ( $R^2=0.15$ ). A potential explanation of this behavior is that BC originated mainly from diesel combustion sources during the day, while during the night it was associated with both traffic and wood burning. The average diurnal pattern of HOA in Athens was the same as that of BC (Fig. S32), and exhibited strong morning (8:00-9:00 LST) and evening peaks (20:00-23:00 LST) consistent with increased traffic during morning and evening rush hours.

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**5.3.2 Patras:** HOA contributed on average approximately 10% to the total OA, while the corresponding HOA contribution during summer was 7% (Kostenidou et al., 2015). Its mean concentration was  $2.1 \mu\text{g m}^{-3}$ , and its hourly maximum value was  $15.8 \mu\text{g m}^{-3}$ . The strongest peak in the average diurnal cycle occurred in the morning (06:00–09:00 LST) and was probably associated with traffic emissions (Fig. 9). A peak in the afternoon (17:00–22:00 LST) was also present, but was frequently smaller. The time series of HOA showed poor correlation with  $\text{NO}_x$

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( $R^2 = 0.16$ ), and BC ( $R^2 = 0.2$ ). Splitting again the BC and HOA time series into two periods, one during 4:00-16:00 LST, and another one during 16:00-4:00 LST, led to a higher correlation during the first period ( $R^2 = 0.48$ ). However, no perfect correlation was expected, as the MAAP was monitoring 4 km away from the HR-ToF-AMS.

In Athens, the O:C for HOA was equal to 0.13, while its H:C ratio was 1.74. In Patras, the O:C ratio was 0.1, while the H:C ratio was equal to 1.83. The corresponding values of O:C during summertime of 2012 were 0.07 (Athens) and 0.1 (Patras) (Kostenidou et al., 2015).

**5.3.3 HOA factor comparisons:** The two HOA spectra were also quite similar to each other ( $\theta=13^\circ$ ) having characteristic peaks at  $m/z$  41, 43, 55, 57, 69, 71, 83 and 85 etc., with little signal at  $m/z$  44 (Fig. S26). Their major differences were observed at  $m/z$  28, 43, 44, 57, 69, and 71, which were generally higher in Patras. The  $f_{44}$  in the Athens spectrum was two times higher (0.04) compared to the Patras one. These mass spectra of HOA are similar to those of diesel exhaust (Canagaratna et al., 2004; Schneider et al., 2005), and other incomplete combustion sources.

Comparing the identified HOA spectra with literature values, the spectrum of HOA in Athens was similar ( $\theta=6^\circ$ ) with the corresponding one from HOA in Patras during summertime (Kostenidou et al., 2015), and relatively similar with the majority of the other HOA spectra in the literature (Aiken et al., 2009; Crippa et al., 2013; Ge et al., 2012; Saarikoski et al., 2012; Sun et al., 2011) ( $\theta=10-15^\circ$ ), as presented in Fig. S33. The spectrum of HOA in Patras was very similar ( $\theta=5-8^\circ$ ) with the spectra of HOA in New York City (Sun et al., 2012), Fresno (Ge et al., 2012), and the MILAGRO campaign (Aiken et al., 2009).

## 5.4 Cooking Organic Aerosol (COA)

**5.4.1 Patras:** The COA factor identified during the campaign had a mean value of  $2.1 \mu\text{g m}^{-3}$ , and an hourly maximum value of  $33.6 \mu\text{g m}^{-3}$ . The diurnal profile of COA in Patras exhibited two peaks during the day. The major peak occurred in the late evening (18:00–24:00 LST) probably modulated by the boundary layer height, and a second significant peak around midday (10:00–15:00 LST), both corresponding to typical Greek mealtimes (Fig. 9). Its contribution was similar to that of HOA.

**5.4.2 Athens:** In Athens, COA exhibited strong night peaks at around 23:00 LST, with values up to  $15 \mu\text{g m}^{-3}$ . Its mean value was  $0.9 \mu\text{g m}^{-3}$ . COA correlated moderately with BC ( $R^2=0.32$ ), and

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CO ( $R^2=0.27$ ), while the correlation between COA and individual VOCs measured by the PTR-MS was relatively poor, with  $R^2$  varying from 0.01 to 0.32. The temporal trend of COA in Athens is different from those in previous studies. Contrary to what was expected, no peak was found during lunch hours (12:00-15:00 LST). However, no other factor displayed a peak during these hours because of the meteorology (strong winds). A large nighttime peak, clearly affected by the decrease of the planetary boundary layer was present at around 23:00 LST (Fig. 9). This late peak is consistent with the late dinner hours of Athenians.

In Patras, the COA O:C ratio was 0.14, while the H:C ratio was equal to 1.71. In Athens, the O:C was 0.11, while the H:C was equal to 1.68. The corresponding values of COA O:C during summertime of 2012 were 0.21 (Athens) and 0.24 (Patras) (Kostenidou et al., 2015).

**5.4.3 COA factor comparisons:** The COA spectrum, like the HOA one was characterized by the ion series  $C_nH_{2n-1}^+$ , but also by  $C_mH_{2m-1}CO^+$  ( $m/z$  41, 55, 69, 83, etc.) and  $C_mH_{2m+1}CO^+$  ( $m/z$  29, 43, 57, 71, 85, etc.) (Crippa et al., 2013; He et al., 2010) (Fig. 8a). The COA spectra in both cities had some similarity to the HOA spectra but were dominated by  $m/z$  41 and 55 instead of  $m/z$  43 and 57. The signal of  $m/z$  55 in the COA spectrum in Patras was almost twice that of the  $m/z$  57. This characteristic of the COA spectrum was first reported by Lanz et al. (2007) and is related to charbroiling (Allan et al., 2010). The COA mass spectra in the two cities were very similar to each other with a theta angle of  $8^\circ$ , and **had only** minor differences at  $m/z$  27, 28, 39, 41 and 43 (Fig. S26). The COA spectrum in Athens and Patras were rather similar to the COA spectra reported in Fresno (Ge et al., 2012), New York City (Sun et al., 2012) and Paris (Crippa et al., 2013) with theta angles values varying between  $11-17^\circ$  (Fig. S34). Comparing the identified COA spectra with the corresponding ones (called HOA-2) in Athens and Patras during summertime (Kostenidou et al., 2015), theta values higher than  $20^\circ$  were observed.

The location of COA and the other factors in the  $m/z$  43 fraction versus  $m/z$  44 fraction ( $f_{43}$  and  $f_{44}$ ) plot suggested by Ng et al. (2009) are shown in Figures S35 and S36.

## 6. Conclusions

An aerosol mass spectrometer (HR-ToF-AMS) accompanied by a suite of aerosol and gas related instruments were used for the characterization of the OA in two of the biggest cities of Greece (Athens and Patras), during two consecutive winter periods (2012 and 2013). The carbonaceous PM (BC+OA) accounted for about 80% of the measured  $PM_{10}$  during the two field campaigns, and up to 95% during morning (5:00-10:00 LST) and evening (16:00-24:00 LST) hours.

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During both wintertime campaigns, domestic wood burning was in general a more significant OA source than traffic. The PM<sub>1</sub> consisted of biomass burning-related OA (25-42%), a contribution from traffic (BC =7-20% and HOA=9-10%), cooking activities (COA= 8-9%), and also oxygenated OA (OOA=13-15%). Sulfates, ammonium and nitrate accounted for 10%, 4% and 5%, respectively, of the total PM<sub>1</sub> in both cities. The OA O:C ratio for the Athens campaign was equal to 0.32, while in Patras 0.42, suggesting that the atmosphere in Athens had higher contributions on average by less processed emissions.

Biomass burning related factors were the dominant source (40-60%) of OA in both cities, and their mass concentration levels increased sharply during the evening hours (18:00- 24:00 LST). The BBOA-I in Patras reached its maximum value at 20:00 LST, three hours earlier than the BBOA factor in Athens. A second biomass burning-related factor (BBOA-II) was also identified in Patras. The BBOA-II (19% of OA) had a quite different mass spectrum than the dominant BBOA-I factor. The two BBOA factors (BBOA-I and BBOA-II) exhibited different diurnal profiles, but comparable O:C ratios (0.30 and 0.26). In Athens, only one BBOA factor was identified by the PMF analysis.

In both sites the HOA factor had two peaks during morning (8:00-9:00 LST) and evening (20:00-23:00 LST) hours respectively. Its diurnal pattern in Athens followed closely that of BC, exhibiting strong peaks consistent with increased traffic during morning and evening rush hours. The two HOA spectra were quite similar to each other ( $\theta=13^\circ$ ) and to the corresponding spectra reported in campaigns in New York City, Fresno, and Mexico City.

The diurnal profile of COA in Athens exhibited strong peaks only during the night at around 23:00 LST, while COA in Patras exhibited two major peaks during the day (13:00 and 22:00 LST). The two COA mass spectra were very similar to each other with a theta angle of  $8^\circ$  and also quite similar to COA spectra in Fresno, New York City, and Paris.

The OOA factor dominated daytime OA in both sites and correlated reasonably well with inorganic compounds. In comparison to summertime campaigns in the same cities during 2012, only one type of OOA was present during wintertime. The spectra of OOA in the two cities were quite similar ( $\theta=11^\circ$ ) to each other, and were quite comparable ( $\theta=8-11^\circ$ ) to the more oxygenated OA (M-OOA) identified during the summertime study.

Both measurement campaigns were conducted during relatively warm wintertime periods. The average ambient temperature for both campaigns was close to 12°C, ranging from 6-19 °C in Athens, and 4-16°C in Patras. The contribution of wood burning is expected to be even higher for lower temperatures.

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**Table 1.** Coefficient of determination  $R^2$  between the OA factors in Athens and VOCs

<i>m/z</i>	BBOA	HOA	COA	OOA
42 (acetonitrile)	0.81	0.67	0.35	0.20
43	0.71	0.59	0.30	0.19
47 (formic acid)	0.43	0.49	0.11	0.06
59 (acetone, glyoxal)	0.70	0.61	0.31	0.25
61 (acetic acid)	0.52	0.33	0.18	0.14
69 (isoprene, furan)	0.88	0.63	0.31	0.14
71 (methyl vinyl ketone, methacrolein)	0.81	0.74	0.32	0.13
73 (methyl ethyl ketone)	0.75	0.61	0.31	0.21
75 (hydroxyacetone)	0.69	0.41	0.24	0.09
79 (benzene)	0.71	0.70	0.31	0.16
81 (terpenes)	0.85	0.63	0.30	0.15
85 (ethyl vinyl ketone)	0.90	0.68	0.34	0.14
87 (2-methyl-3-buten-2-ol, C5, methacrylic acid)	0.93	0.59	0.33	0.15
93 (toluene)	0.37	0.54	0.18	0.09
95 (2 vinyl furan, phenol)	0.43	0.36	0.12	0.02
99 (hexenal)	0.87	0.58	0.34	0.21
101 (isoprene hyperoxides)	0.71	0.48	0.25	0.12
105 (styrene)	0.54	0.55	0.19	0.09
107 (xylenes)	0.47	0.65	0.23	0.11
113 (chlorobenzene)	0.80	0.53	0.31	0.21
115 (heptanal)	0.66	0.48	0.22	0.13
121 (C9 aromatics)	0.47	0.66	0.21	0.10
129 (octanal, naphthalene)	0.70	0.73	0.21	0.07
135 (C10 aromatics)	0.50	0.64	0.21	0.12
137 (monoterpenes)	0.71	0.52	0.25	0.19
139 (nopinone)	0.67	0.41	0.22	0.18

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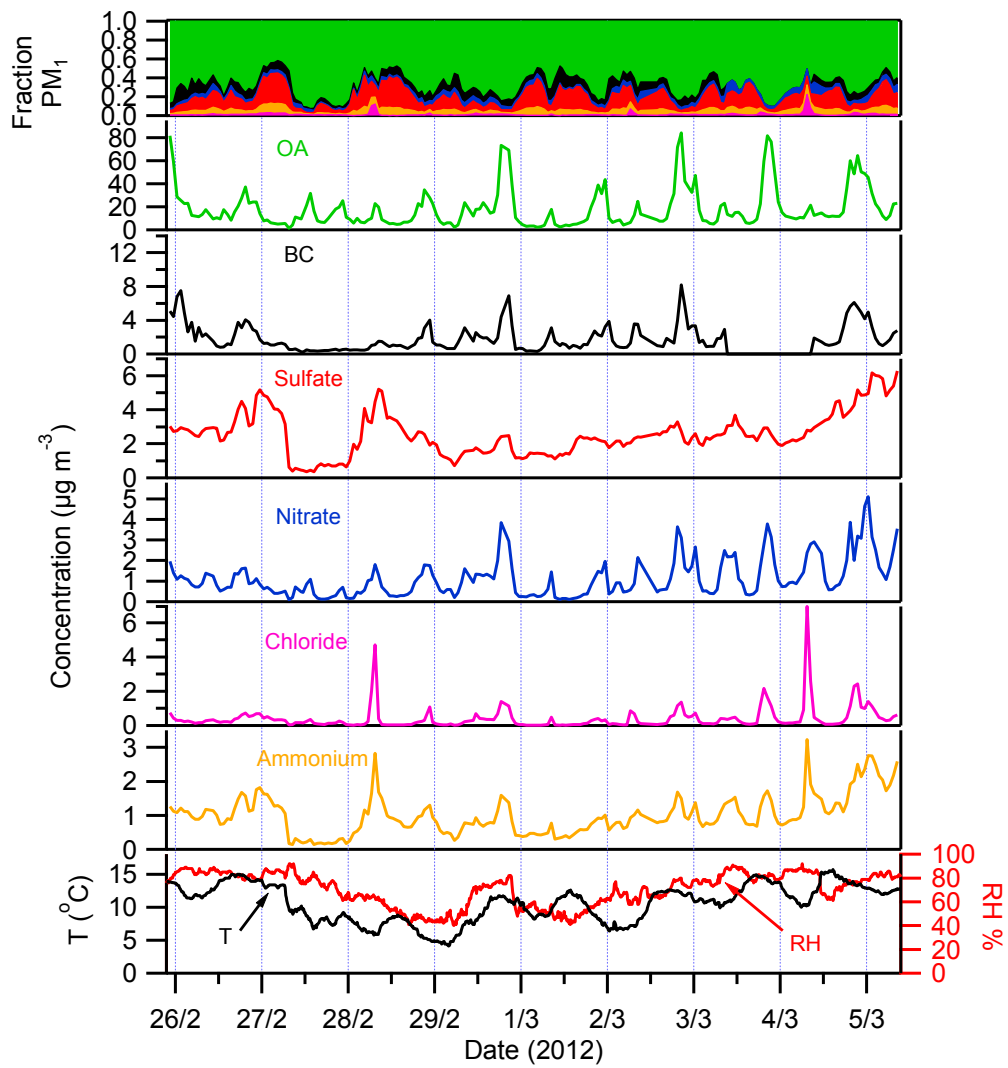
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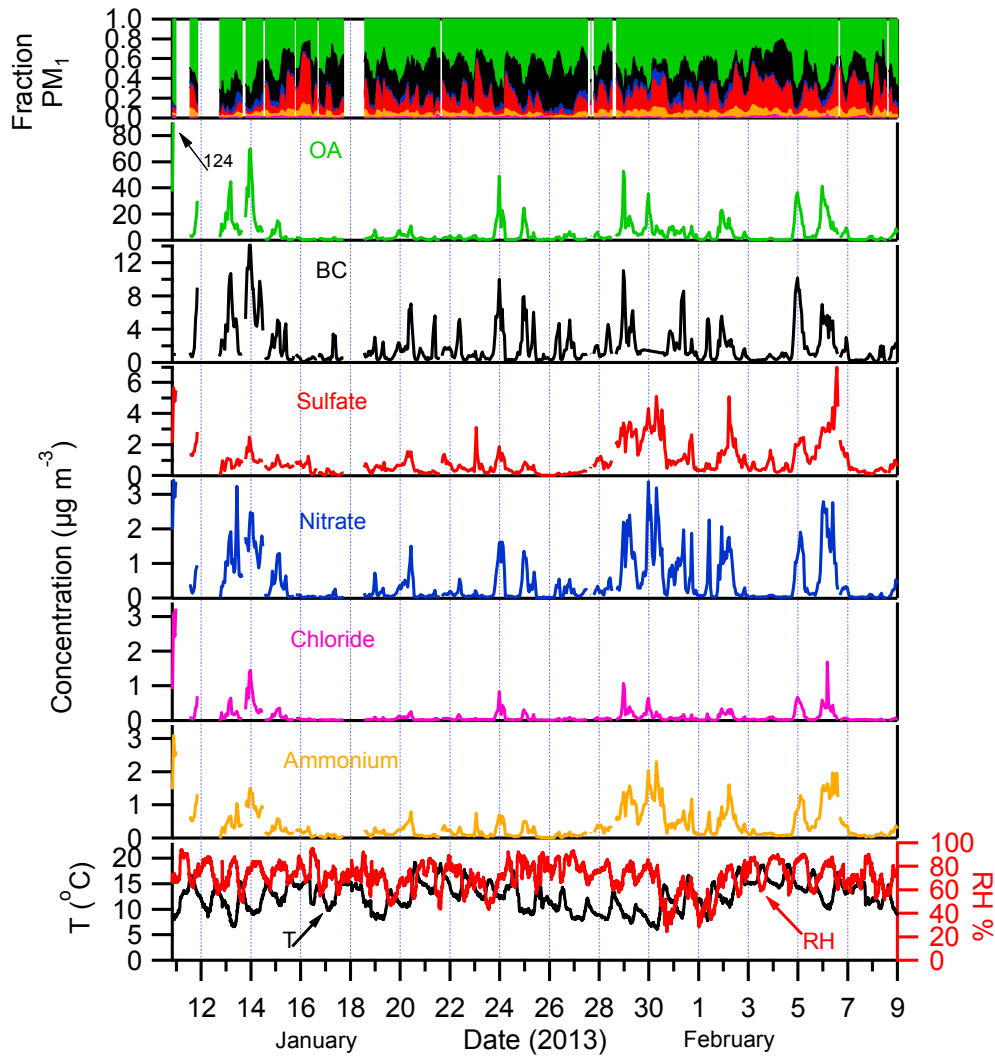
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<del>151 (pinonaldehyde)</del>	<del>0.32</del>	<del>0.26</del>	<del>0.09</del>	<del>0.06</del>
<del>163 (C12 aromatics)</del>	<del>0.38</del>	<del>0.27</del>	<del>0.10</del>	<del>0.12</del>

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**Figure 1.** Fractional contribution and time series (hourly averages) of the main PM<sub>1</sub> species during the Patras campaign. Also shown the temperature and RH.

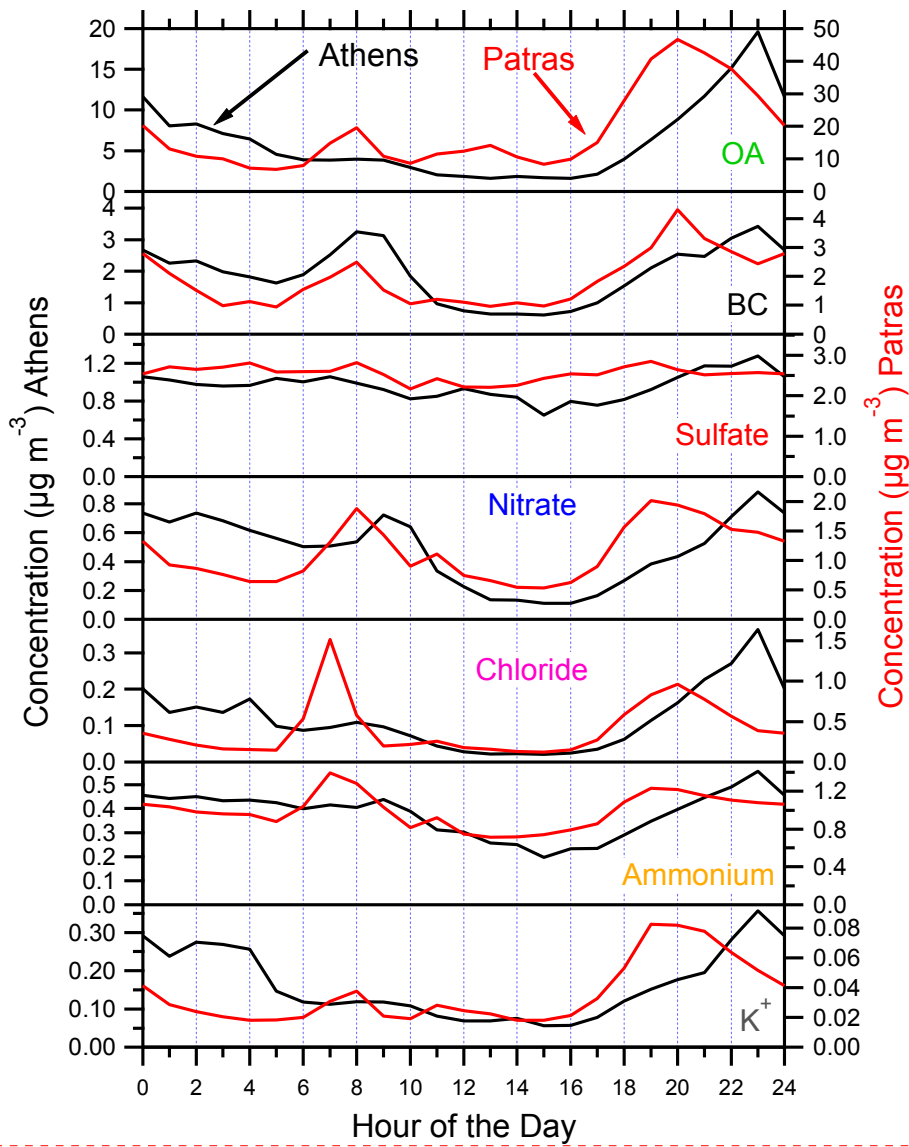


**Figure 2.** Fractional contribution and time series (hourly averages) of the main PM<sub>1</sub> species during the Athens campaign. Also shown the temperature and RH. During the first day, OA reached levels up to 124  $\mu\text{g m}^{-3}$ .

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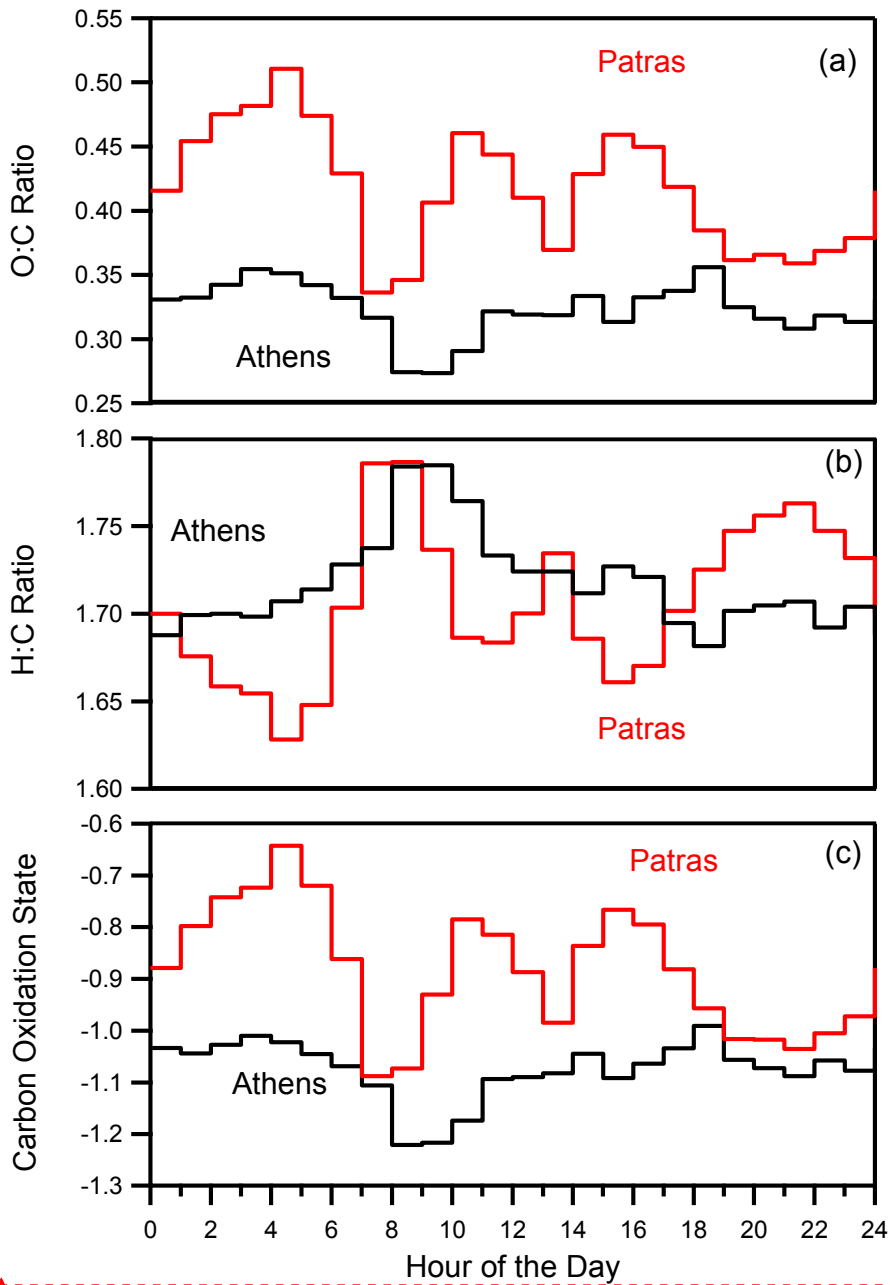




**Figure 3.** Average diurnal profiles for the main PM<sub>1</sub> components during the Athens and Patras campaigns. The concentration of PM<sub>1</sub> in Patras is shown in the right y-axis.

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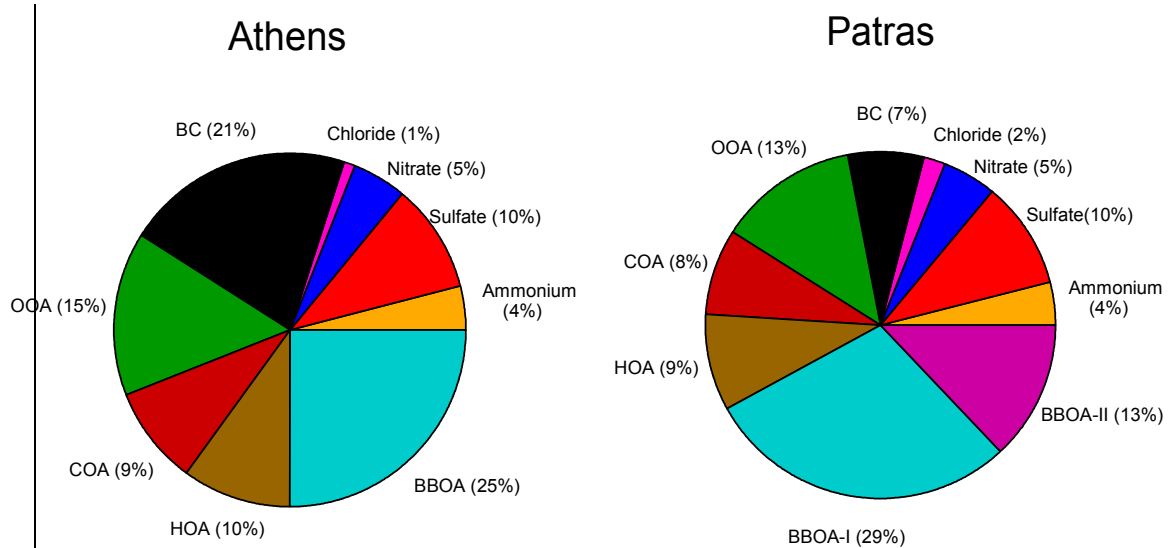


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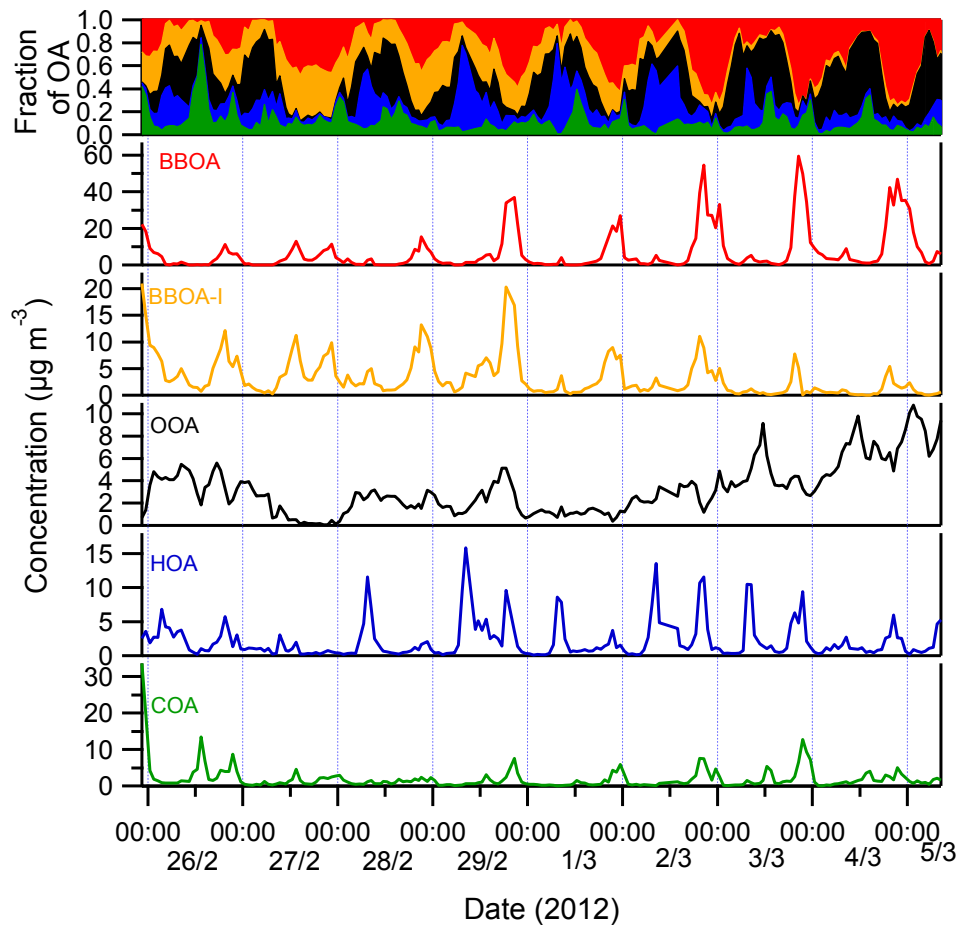
**Figure 4.** The diurnal profile of (a) the O:C ratio; (b) the H:C ratio; and (c) the OS<sub>c</sub> in Athens (black) and Patras (red). The ratios have been calculated following the Canagaratna et al. (2015) approach.

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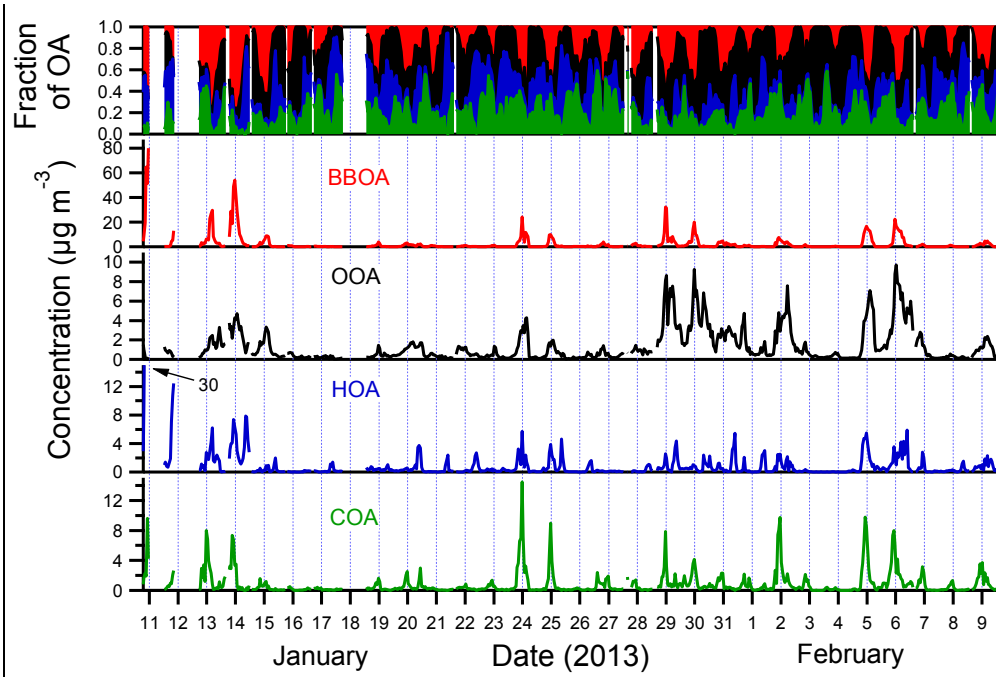
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**Figure 5.** Average PM<sub>1</sub> composition and organics source apportionment for Athens and Patras. The average PM<sub>1</sub> for Athens was 10 µg m<sup>-3</sup> while for Patras 25 µg m<sup>-3</sup>.



**Figure 6.** Fractional contribution to OA and time series of the PMF factors during the Patras campaign.



**Figure 7.** The hourly fractional contribution to OA and time series of the PMF factors during the Athens campaign. During the first day, HOA reached levels up to 30  $\mu\text{g m}^{-3}$ .

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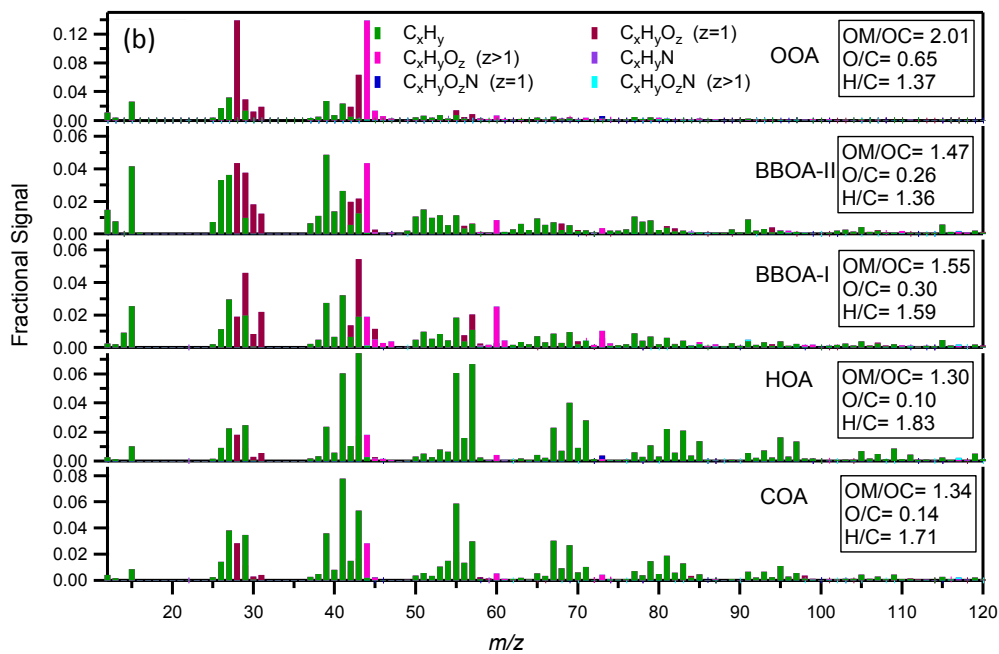
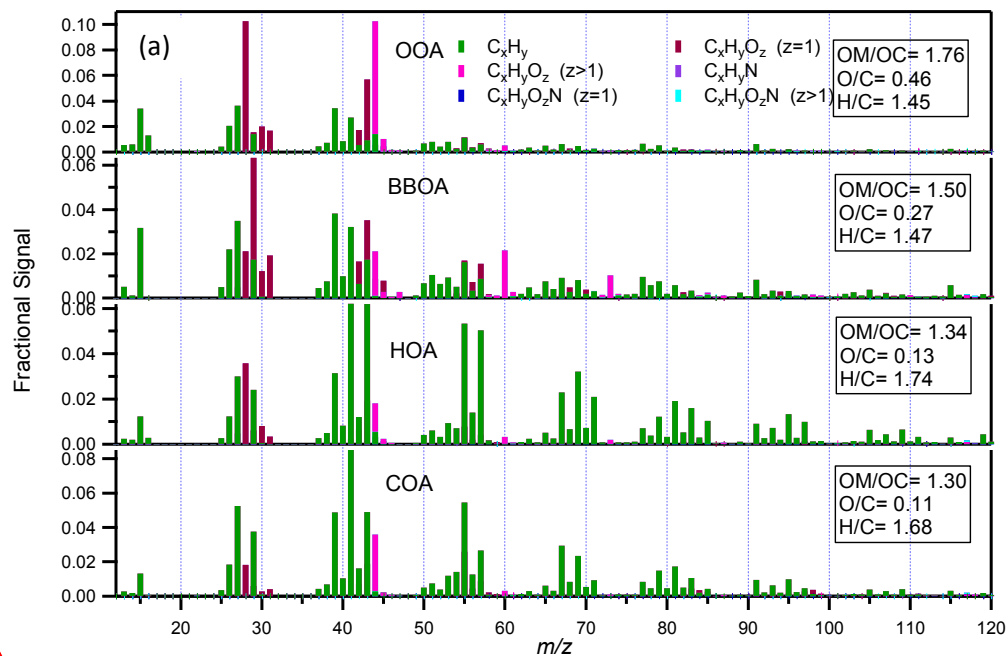
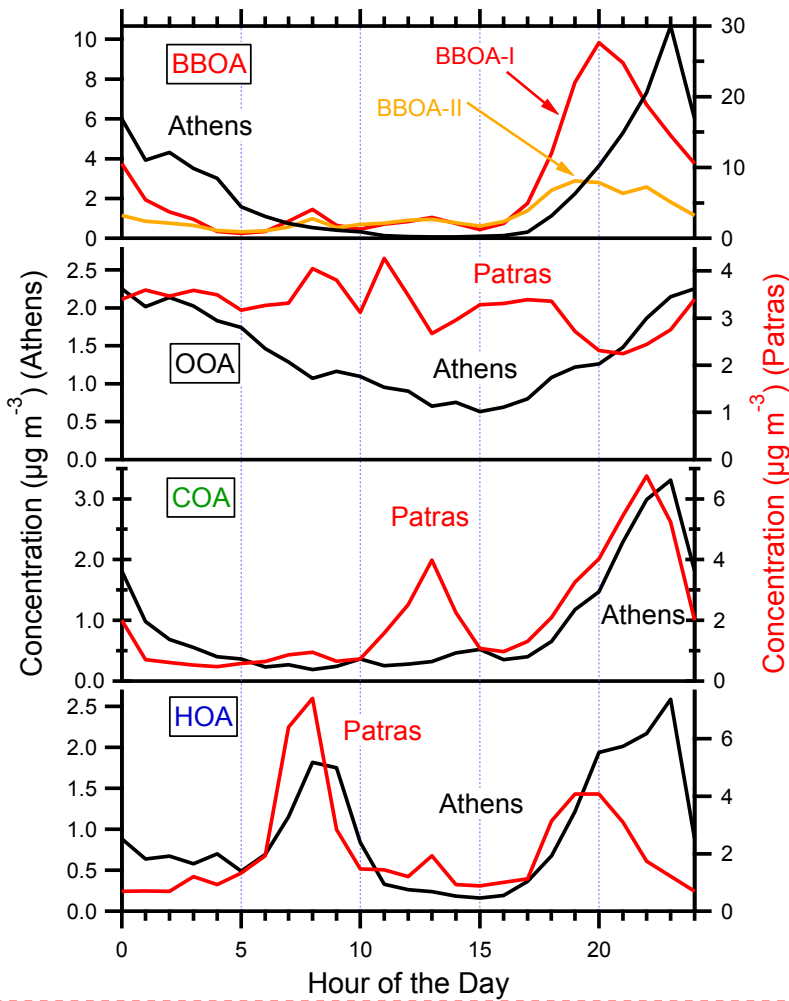


Figure 8. High resolution AMS mass spectra for the factors in (a) Athens and (b) Patras.



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**Figure 9.** Diurnal profiles of the PMF factors in Athens (black) and Patras (red-yellow). On the left y-axis are the concentrations of Athens' PMF factors, while on the right y-axis those of Patras.

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The null hypothesis is that the BBOA factor represents an average of the emissions of all fuel types. However, the corresponding emissions should have either a different temporal or a spatial profile in order to be separated by PMF. Given that residential biomass burning, in a city like Patras, is due to tens of thousands of individual sources (fireplaces, wood stoves, etc.), spread all over the city following a similar burning schedule on average, it is difficult to investigate how PMF would be able to separate the BBOA from different fuel types. However, there are other potential explanations (e.g., different areas of the city using different fuel types and the emissions arriving at the site at different types), that could involve the use of different fuel types.

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69 (isoprene, furan)

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71 (MVK, MACR)

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<b>Page 45: [12] Deleted</b> 73 (MEK)	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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<b>Page 45: [13] Deleted</b> 75 (hydroxyacetone)	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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<b>Page 45: [14] Deleted</b> 79 (benzene)	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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<b>Page 45: [15] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>

81 (terpenes)

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	0.85	

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	0.63	

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	0.30	

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	0.15	

Page 45: [16] Deleted	Spyros	12/17/2016 1:08:00 PM
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E

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V

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K

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85 (EVK)

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	0.90	

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	0.68	

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	0.34	

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	0.14	

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87 (MBO, C5, methacrylic acid)

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	0.93	

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	0.59	

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0.33

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0.15

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93 (toluene)

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0.37

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0.54

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0.09

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95 (2 vinyl furan, phenol)

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0.43

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0.36

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0.12

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0.02

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99 (hexenal)

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0.87

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0.58

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0.34

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0.21

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	0.71	
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	0.48	
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	0.25	
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	0.12	
<b>Page 45: [23] Deleted</b> 105 (styrene)	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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	0.09	
<b>Page 45: [24] Deleted</b> 107 (xylenes)	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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	0.47	
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	0.65	
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	0.11	
<b>Page 45: [25] Deleted</b> 113 (chlorobenzene)	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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0.80

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	0.53	

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	0.21	

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115 (heptanal)		

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	0.66	

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	0.48	

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	0.22	

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	0.13	

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121 (C9 aromatics)		

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	0.66	

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	0.21	

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	0.10	

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129 (octanal, naphthalene)		

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	0.70	

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0.73

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	0.07	
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135 (C10 aromatics)		
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	0.50	
<b>Page 45: [29] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
	0.64	
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	0.21	
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	0.12	
<b>Page 45: [30] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
137 (monoterpenes)		
<b>Page 45: [30] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
	0.71	
<b>Page 45: [30] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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	0.19	
<b>Page 45: [31] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
139 (nopinone)		
<b>Page 45: [31] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
	0.67	
<b>Page 45: [31] Deleted</b>	<b>kalli</b>	<b>12/16/2016 6:31:00 PM</b>
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0.22

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kalli

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0.18