Responses to the Editor's Comments

Scientific comments:

(1) Line 221: Were the filters analysed for Na^+ which could be used to estimate the sea-salt sulphate that would not have been detected by the AMS?
The $PM_{2.5}$ Na^+ concentration of the filters for Patras was below the detection limit, so if there was any sea-salt in this size range its contribution was extremely low. This is now mentioned in the paper.

(2) Line 264: I am not convinced that the correlation coefficient would necessarily be poor if a significant proportion of the organics were contained in the submicron fraction. You should instead look at the quantitative comparison between organics in $PM_{2.5}$ and those reported by the AMS, based on the C/OM ratio from the AMS.
We have performed the recommended comparison between the organics of the $PM_{2.5}$ filters and PM_1 from HR-ToF-AMS. First the $PM_{2.5}$ filters organic carbon (OC) was converted to organic mass (OM) using for each filter the corresponding value of OM:OC ratio provided by the HR-ToF-AMS. Then the $PM_{2.5}$ filters OM was compared to the OM of PM_1 HR-ToF-AMS. We have rephrased the corresponding text (lines 262-264) in order to avoid any confusion.

(3) Line 357: Please introduce the meaning and calculation of the angle theta or at least add reference Kostenidou et al. (2009). It's only been used in a few papers and the reader may also not have a feel for what angle reflects close agreement. Has this ever been compared with the more typically applied comparison of spectra using the correlation coefficient? Maybe it is worth also adding R2 values to Table 2 also? Or at least re-iterating the relationship between theta and R? Given that R2 and theta are related, neither is a perfect descriptor for factor similarity as both are biased towards the main peaks. Section 3.3.4 may benefit from such caveat.
The meaning and the calculation of the angle theta, as well as the reference of Kostenidou et al. (2009) can be found at the end of section 2.6, lines 242-245: *"The angle theta (θ) between the mass spectra vectors was used as a measure of their similarity (Kostenidou et al., 2009). The mass spectra are treated as vectors and the angle θ is calculated by using their internal product. The lower the angle θ is the higher the similarity between the two spectra."*. We have added the R^2 values in Table 2 to facilitate the readers.

(4) Factor names: I suggest you make it even clearer in the manuscript why you deviate from the standard description of the factors of LV-OOA/SV-OOA and HOA/COA. In particular, the diurnal pattern of HOA-2 looks very similar to that reported for (what was called) COA in other studies. The lunchtime peak in COA looks early in other studies also. You state that this is inconsistent with lunch time in Greece (Line 461). Could larger canteens and outside catering starting cooking earlier and dominate the lunchtime emissions? With the exception of COA from Barcelona, there is still marked similarity between your HOA-2 and any of the literature factors used in Table 2. Also, in the UK, COA has been attributed mainly to vegetable oils rather than meat cooking (Allan et al., 2010); not sure how this may relate to Greek conditions. There has always been a concern

that what was reported as COA may include other (possibly traffic related) sources, but it is unclear why these would not be distinguishable and consistently lumped into one single factor given their different diurnal behaviour. In summary, I suggest you expand the discussion on HOA-2 and its relationship to COA factors (and temporal profiles) in the literature and review your decision not to call it COA.

We named the factors V-OOA and M-OOA instead of LV-OOA and SV-OOA based on their degree of oxygenation. PMF analysis does not provide volatility characteristics and since we did not have volatility measurements (e.g. a thermodenuder system) it a little dangerous to make assumptions about their volatility. In addition, recent unpublished work from our group (Paciga et al., ACPD, in press) indicates that two OOA factors with different O:C ratios may have similar volatility distributions (e.g., the LV-OOA may include some semivolatile components and vice versa the SV-OOA may include low volatility components). We added these clarifications in the revised manuscript.

We agree that the HOA-2 factor may also contain cooking emissions from canteens or catering services which start cooking earlier than the typical Greek lunch time. However, this does not explain the correlation of HOA-2 with BC (R^2=0.57). Recent experiments from our group (not yet published) showed that BC emissions from meat (souvlaki) charbroiling are low, with a BC/OA less than 0.02. Thus there is some evidence that additional sources, potentially traffic emissions, may be included in the HOA-2 together with COA. One reason that PMF could not distinguish these sources in a separate factor could be the similarity of their mass spectrum with cooking emissions. A second reason could be the timing of these sources. We have updated the manuscript summarizing the above discussion.

(5) HOA-1 / BC correlation: How important are emissions from two-stroke engines in Greek cities as a potential source of HOA-1 (without generating BC) and are there other possible sources of BC other than diesel trucks?

According to the Hellenic Statistical Authority (EL.STAT., 2014) there were 5.1 million passenger cars in 2013 in Greece and 1.6 million motorcycles. We assume that 80% of the passenger vehicles are gasoline cars and that 70% of the motorcycles are two-stroke scooters. Platt et al. (2014) found that the primary OA emissions from two-stroke engines vary from 0.5 to 9 g C kg^{-1} fuel, while other studies (e.g., Platt et al., 2013) indicated that primary emissions from gasoline vehicles were in the range of 0.01 to 0.05 g C kg^{-1} fuel. So these numbers indicate that two-stroke engines can be an important source of HOA in Greece without generating BC. An additional potential BC source is the ships in the ports of Piraeus (15 km south-west of the site) and Rafina (17 km to the east). However, the highest BC concentrations were observed when the winds were coming from the north. Thus, the main source of BC was probably diesel trucks. We modified the manuscript including the above discussion

(6) Line 377 and Table 1. Can you please indicate which correlations are significant? Maybe, in Table 1, you could show those R2 values that reflect statistically significant correlations (state P value) in bold.

All the correlations in Table 1 are significant at the p=0.05 level due to the large sample sizes used (these are hourly averages). This is now mentioned in a footnote.

Clarifications:

(7) Line 145. Please provide a rationale. Was there another AMS operated in the city centre? If so why was this excluded?
There was no AMS deployed in the city centre. Most of our instrumentation was deployed at the ICE-HT institute, due to space limitations in the city centre station. We made this clear in the text.

(8) Line 208. At this point you should spell out more explicitly that you are performing high resolution PMF as the references point to unit mass resolution analyses.
We made it ever clearer in the manuscript.

(9) Line 216: Please clarify whether an average CE was used or whether this varied on a half-hourly basis.
We compared the 2-hour resolution CE with a 2-hour average RH. We changed the text accordingly.

(10) Line 566: Could you make your reasoning more explicit here? Artifact for what? You are probably implying that organic S is not a good quantification of MSA aerosol, but you should spell this out.
We have explained it better.

(11) Figure S11 and related text. If the disagreement between filter and AMS organics is due to uncertainties in the CE (as you suggest), why does this not affect the comparison for sulphate?
We deleted this confusing explanation.

Technical corrections:

(12) Line 102: characterization of OA sources
Done.

(13) Line 115: Maybe this could be rewritten: "… in the dark in less than a day …" does not read quite right as it is only dark for less than half a day in summer.
We replaced the phrase with "..less than 12 hours.."

(14) Line 213: Suggestion: "was significantly higher compared with most other studies"
Corrected.

(15) Line 227: "at the site."
Corrected.

(16) Line 315: redundant "that".
We deleted "that".

(17) Line 395: Add Allan et al. (2010) as the first study that identified COA in ambient air?
According to the literature the first study that identified cooking aerosol in the ambient is Lanz et al. (2007), which is stated in the introduction (lines 65-66). For the comparisons of the factors' mass spectra we choose those mass spectra from the literature which applied the new fragmentation table of Aiken et al. (2009). Allan et al. (2010) used an older fragmentation table so any comparisons would be misleading.

(18) Figs. S13, S14, S25 and S26: please set offsets for vertical axes to zero.
Unfortunately a lot of values are negative, so if we set offsets to zero (for the vertical axes) part of the graphs will disappear. To make these graphs better looking we completely deleted the zero lines form the vertical axes.

(19) Line 532. "Organic contributions in Patras and Athens were around"
Done.

References:

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