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Response to Anonymous Referee #2 comments

This manuscript presents a long-term dataset of near real time chemical composition of submicron aerosols in Athens, Greece. It is completed by two intensive campaigns during winter time. Statistical analysis was performed in order to apportion the sources of organic matter. The subject of this paper is of interest and falls within the scope of ACP, although in its current form neither the methodology (PMF) nor the results bring strikingly new outputs in this region. I am still favorable to publication after major revisions.

Response: *We thank the anonymous referee for the review and have incorporated his/her suggestions and comments in the revised version of the manuscript.*

Major Comments

1) Overall, the result section is too descriptive. Describing the angles between profiles impinges upon the actual results. The authors should re-focus the discussions on how this study slots into previous knowledge in Greece (Athens and their cities, such as Patras) and, why not, in the Eastern Mediterranean.

Response: *We would like to thank the reviewer for this comment. Given the fact that matters of clarity and focus have been also pointed out by some of the other reviewers, the results section and especially §3.3, has now been revised in the new version of the manuscript. Similarity with literature spectra is now presented in a more systematic and clear way, while the discussion incorporates a more in depth analysis of the novel findings that complement the knowledge obtained regarding submicron aerosols and more specifically the organic fraction, for the region of the Eastern Mediterranean.*

Moreover, strong assessments are made regarding SOA, simply from diurnal variations. The authors should either tone down these statements or add much more discussion and figures.

Response: *We do not feel that our assessment of the nature/origin of SOA is simply based on diurnal variability. SV-OOA which is believed to originate from the fast oxidation of primary combustion sources is correlated during winter-time both with external tracers such as* BC_{wb} *and fine-mode nss-*K^+*, as well as with mass spectra from oxidized biomass burning. Additionally, the contribution of m/z=60 and m/z=73, well known fragments of levoglucosan, is significant to this secondary factor. Additional information for compounds such as VOCs would be useful, but this is outside the purpose of this manuscript. Nevertheless,*

the fast oxidation of fresh biomass burning in plumes within just a few hours after emission has been documented in literature (Lathem et al., 2013; Cubison et al., 2011), supporting our present assessment. Furthermore, the LV-OOA spectrum exhibits a highly oxidized nature, correlating very well with literature spectra of OOA attributed to regional processes from earlier studies in the region (Bougiatioti et al., 2014). In any case, we have tried in the revised version to tone down the statements, where not fully supported by experimental results.

Then, organonitrates have been found to have significant contributions in Greece (Florou et al., 2017). Can the authors add some more knowledge about that? (especially regarding the role of primary combustion sources in their formation?)

Response: *We would like to thank the reviewer for pointing out the issue of organic nitrate contribution to nitrate measured by the ACSM, a matter of rising concern among the scientific community. For pure ammonium nitrate, Fry et al. (2009) report a NO^+:NO_2^+ ratio of 2.7, while Farmer et al. (2010) report 1.5. Older studies using AMS, report ratios ranging from 1.18 (Cottrell et al., 2008) to 2 or 3 (Alfarra et al, 2006), while more recently Kindler-Scharr et al. (2016) report values ranging from 2.04 to 3.45. Using data from the ammonium nitrate calibration of our instrument we calculate a NO^+:NO_2^+ ratio of 3.84, slightly higher than those presented in the literature. Given that the NO^+:NO_2^+ ratio highly depends on each instrument and it's tuning, and also due to the fact that the instrument's response - in terms of this ratio - when sampling specific organic nitrate standards was not explored during this study, some uncertainty is expected when calculating the fraction of nitrate signal attributed to organonitrates (R_{ON}). These uncertainties are expected to be even larger since the limited sensitivity of the instrument does not allow to distinguish the interference of the CH_2O^+ ion at m/z=30, thus potentially leading to an overestimation of the measured NO^+:NO_2^+ ratio.*
In our study, for the 2016-2017 period the NO^+:NO_2^+ ratio ranged roughly from 3.20 (10^{th} percentile) up to 9.83 (90^{th} percentile), thus yielding several negative values for R_{ON} and a few values larger than 1, when calculated according to Farmer et al. (2009). Eventually the R_{ON} data acquired had to be treated so that negative values correspond to a fractional contribution of organonitrates to NO_3 signal of 0, meaning that we assume that those negative values correspond to pure ammonium nitrate particles. Furthermore, we assume that larger values of NO^+:NO_2^+responsible for elevated R_{ON} values are not due to other inorganic salts such as $NaNO_3$ and $Ca(NO_3)_2$. In the above context we find that during the cold season R_{ON} is pretty low with an average value of 0.19. Diurnal variability during the cold season shows minima during night time while a maximum is evident in the afternoon. The opposite trend is observed in the warm season were R_{ON} averages at 0.62, exhibiting maxima during night time. Given all the points presented above we believe that although organonitrates is a very important subject, we feel that first it is beyond the scope of this manuscript and second that the overall uncertainties of the ACSM do not allow an accurate and qualitative determination of their levels in the present work.

2) The method used by the authors to select the appropriate solution is not clearly stated, although it was inspired by Crippa et al. (2014). From the first ACSM intercomparison, Frohlich et al. (2015) proposed a methodology of find optimal solution, and stated some recommendations. Why did the authors prefer Crippa et al.?

Response: *We agree with the anonymous referee that the presentation of the PMF strategy used, which is indeed the one proposed by Crippa et al. (2014), has to be described in a more clear and systematic way. In the original manuscript the results of each step of the process were not presented, in an effort to keep the manuscript short and easy to follow. Both those issues are addressed in the revised version. . The steps of the strategy are clearly presented and PMF solutions of a) the unconstrained runs, b) runs with a constrained HOA factor and c) runs with both HOA and BBOA factor constrained for the cold season dataset are now presented in the revised supplement.*
Regarding the methodology selected, we believe that Frohlich et al. (2015) performed such a detailed optimization of the alpha values used to constrain each factor, due to the scope of their work, which was specifically to explore the performance of the ME-2 method itself across different instruments, thus trying to assure a high level of compatibility amongst 15 different ACSM datasets. However, in order to fully address the reviewer's comment, the revised version of the manuscript and supplement now incorporates results from a sensitivity analysis performed on the alpha value approach.

3) The local vs regional vs advected features are not well characterized, and I would strongly recommend the authors to perform a wind analysis, especially for the "local" sources (eg traffic & biomass burning).

Response: *According to the reviewer's suggestion, wind effects have been examined in the revised manuscript, focusing on the sources of local character, namely traffic and biomass burning, as expressed by the HOA and BBOA time-series. The concentrations of these parameters, when regressed against wind speed, decline exponentially for winds exceeding 2 m sec^{-1}, indicating the locality of the respective sources (Fourtziou et al., 2017). The examination of bivariate wind direction and speed plots (Grivas et al., 2018) further corroborate that organic aerosols characterized as HOA and BBOA are mainly produced in the vicinity of the sampling location, rather than advected from the extended area of Athens. This information, is included in the revised manuscript, accompanied by relevant supplementary figures.*

Minor comments
P3 l78: the introduction mainly focuses on wintertime biomass burning, so why would you need long-term datasets?

Response: *The introduction has now been revised so as to be much more balanced and provide discussion also related to the warm periods of the year. Furthermore, the importance of long-term chemical composition datasets with high temporal resolution, in the urban setting, is highlighted.*

P4 l120: please indicate the calibration values

Response: *Calibration values have been added to the supplement.*

P4 l103: the ACSM does not measure “aerosol mass” but only the chemical composition; it is not equivalent to a TEOM-FDMS.

Response: *This phrasing has been corrected in the revised manuscript.*

P5 l123: it is not clear why a chemical-dependent CE has not been applied. Although it is discussed later on, it could be quickly stated here.

Response: *We would like to thank the reviewer for pointing this out. A chemical dependent CE according to Middlebrook et al., 2011, has been now applied on the dataset and will be incorporated in the revised version. Concentrations have been updated accordingly.*

P5 l125: did the authors use denuders ahead of the PILS in order to prevent nitric acid, sulfuric acid and ammonia to be respectively confused with particulate nitrate, sulfate and ammonium?

Response: *Two denuders are placed in front of the PILS inlet, which is now added as information in the revised version of the manuscript. Note also that PILS was operating in cation mode and mainly nss-K were used in this manuscript.*

P5: filter samplings are not presented.

Response: *Details on filter sampling and analysis has been added.*

P10 l292: See major comment. I don’t think that the only fact that nitrate has a morning peak similar to BC is enough to link it with morning traffic. More discussion would be needed.

Response: *We agree that a direct comparison between nitrate and BC is not enough to link with morning traffic. Nitrate is an oxidation product, probably what we see is the combined result of several factors like nighttime oxidation, pH, BL. and therefore we have decided to remove this sentence from the revised text.*

P11 l318-319: Datasets have been separated into cold and warm months prior to PMF. Why not seasonally? Not just cold and warm months influence the characteristics of secondary organic aerosols, it could also be related to air masses. So the approach chosen here is not well justified.

Response: *According to studies on the climatology of Southern Greece, the transient period (spring and fall seasons) in Athens doesn't exceed 60 days on average (Argyriou et al., 2004), covering mainly the months of April and October - which were excluded from the seasonal analysis. As a result, the vast majority of air pollution studies in the area have examined the seasonal variability on a cold-warm season basis (Grivas et al., 2008 and reference therein). This classification reflects the contrast between dry/sunny summer and humid/cloudy winter conditions which are typical in the Mediterranean climate setting.*

Moreover, this split is representative of the relative intensity of local sources and processes, since the cold period effectively includes the time-span when residential heating is active as a source. On the other hand, the warm period essentially coincides with the photochemical season, also being characterized by an absence of precipitation and therefore a larger impact from regional sources.

P11 l329: why the HOA from Ng et al. has been used? Why not any other profiles, especially gotten from previous studies in Greece?

Response: *The HOA profile from Ng et al. (2011) is an average of profiles derived from different environments in Europe, Asia and North America, covering different seasons of the year and countries with different types of vehicular fleet. Consequently, we consider that the HOA factor of Ng et al. (2011) can be suitable as an anchor profile for constrained runs in the present study. The results obtained using this factor as an anchor are proven to be representative. Correlation with HOA factors from other studies in Greece is very good for all seasons. Correlation with the wintertime HOA obtained by Florou et al. (2017) at the same measuring site yields an R^2=0.92 while correlation of this study's deconvolved factors for HOA during summertime of 2016 and 2017, with the HOA factor of Kostenidou et al. (2015) measured at a suburban site in Athens during the summer of 2012, was also excellent (R^2=0.92 and R^2=0.94 respectively). All of this information has now been added to the revised manuscript.*

P11 l333: Do the authors have any hint of how representative the BBOA of Ng et al. is in Greece?

Response: *We agree with the reviewer that the nature of the biomass burning fuel may vary considerably depending on the location. Especially in Athens, Fourtziou et al. (2017) have reported that a wide variety of hardwood and softwood types are commonly used for heating purposes. This is why an average BBOA spectrum has been selected, and a large alpha value imposed. The BBOA spectrum from Ng et al. (2011) exhibits very good correlation with BBOA factors identified in previous studies for two major Greek cities. Comparison with the Florou et al. (2017) BBOA factor for Athens yields an R^2=0.91 while comparison with the factor for Patras yields an R^2=0.90. BBOA from this study for the 2016-2017 winter, also correlates very well with R^2=0.94. Finally BBOA from Ng et al. (2011) correlates well with BBOA obtained at Finokalia, a regional background site (Bougiatioti et al., 2014) yielding R^2=0.78. The results obtained during this study justifies our decision, as the derived BBOA is in excellent correlation with the respective ones found in the literature. one obtained during wintertime by Florou et al. (2017) in Athens (R^2=0.89), and in Patras (R^2=0.89), even with the BBOA from Bologna during winter (R^2=0.85; Gilardoni et al., 2016).*

P12 l340-341: it could be appreciable if the authors provide a bit more details on the metrics used through the correlation of PMF timeseries with external tracers.

Response: *Even though it is stated at the supplementary material, we agree with the reviewer that this piece of information was obscured by the way the correlation tables were presented. This matter has been considered in the revised version of the supplement.*

P12 l351: a correlation coefficient of 0.86 corresponds to a r2 of 0.63, which is still a good statistical correlation. Ranges of r2 are by the way not consistent throughout the manuscript.

Response: *The inconsistency in terms of reported correlations was also pointed out by some of the other referees. In the revised text all correlations are given in terms of the square of the Pearson correlation coefficients (R^2).*

P13 l397 and p14l400, r2 of 0.32, 0.36 and 0.39 are considered as "moderate", which should rather be a poor correlation. Later, p16 l477, a r2 of 0.53 is considered as "very well". I strongly suggests the authors to use a consistent description of correlation coefficients.

Response: *Both of the abovementioned comments have been addressed in the revised manuscript. Characterization of the derived correlations is now uniform in the revised version. Correlations are presented in a more systematic way. Only one metric is used when investigating both affinity with other mass spectra and correlation with external time series. Quantitative criteria regarding the description of correlations are also set.*

P13 l388-390: See major comment. Linking SVOOA with primary sources only from mean diurnal variations is not convincing. Please add more discussion.

Response: *The phrasing of this sentence has been changed in the revised version of the manuscript, even though the assumption made, does not refer to the diurnal variability of the factor, but rather to its comparison with reference mass spectra obtained in other studies. In this context, it is now made clear, that since the warm season SV-OOA factor spectrum, obtained during this study, correlates better to factors related with the oxidation of biogenic VOCs together with SOA factors related to primary sources such as cooking and traffic an assumption linking the factor to primary sources can be made.*

P14 l400: See major comment. Same comment, the statement "SVOOA may, to some extent, partially originate from a combustion source" seems random and hardly quantifiable.

Response: *We agree with the reviewer that the fact that SV-OOA may originate from a combustion source is not quantifiable, but, nevertheless, it must originate from the oxidation process of some kind of primary organic aerosol. Its good correlation with external spectra of aged combustion emissions (aged diesel exhaust, as well as aged BBOA), supports the validity of the statement.*

P14 l407: "HOA emissions are very low", compared to what?

Response: *This sentence should rather read "HOA concentrations are very low compared to the cold season" since HOA concentration during the warm months is on average more than five times lower than during the cold season. This information has been added to the revised manuscript.*

P14 l422: the authors would need to prove the link between SOA and regional biomass burning.

Response: *The link between SOA from the oxidation of primary biomass burning emissions is not new in the literature. It has been demonstrated that OOA of a mixed nature, but which is also made up of aged BBOA, has been observed in a regional background site in Spain, 100 km away from a wildfire (Minguillón et al. 2015) and also in a regional background site in Greece, affected by plumes from wildfires over even longer distances (Bougiatioti et al., 2014). With air masses during the warm period originating mostly from the north, northeastern sector, it is clear that part of the transported OOA is bound to include to some extent (nonetheless non-quantifiable) processed, regional biomass burning from the Balkans and the area of the Black Sea, where hotspots of fires are peaking during July-September (Sciare et al., 2008).*

P15 l444-448: how does BBOA compare with BBOA profiles from other studies in Greece? Or in other Mediterranean sites?

Response: *BBOA correlates very well with the spectra reported by Florou et al.,2017 both for the BBOA factor obtained at the same site (R^2=0.88) and BBOA in Patras (R^2=0.89). It also correlates well with the BBOA factor obtained at Finokalia (Bougiatioti et al., 2014) exhibiting an R^2 of 0.81 as well as with the BBOA obtained in Bologna by Gilardoni et al., 2016 with an R^2 of 0.85.*

P16 l477-479: HOA correlates moderately with CO, BC and NO. So is HOA representative of traffic?

Response: *HOA is representative for fossil fuel combustion processes, and it may originate from both traffic and domestic heating oil emissions. Performing a linear regression exercise for the cold period data set, excluding values obtained during night-time (19:00 – 05:00) the HOA factor exhibits stronger correlation with tracers that are related to traffic emissions, than when using the entire dataset. Indicatively, during the day-time period including the traffic rush hour peak, HOA correlates well with BC_{ff} (R^2=0.71).*

Technical corrections and suggestions

P1 l22: replace "fine" by "submicron"

Response: *Amended.*

P1 l24: rephrase to "with concentrations during wintertime sporadically reaching up to 200 μg/m3". Please also indicate the time resolution for this (daily/hourly concentrations?)

Response: *Amended.*

P2 l50: replace "namely" by "such as"

Response: *Amended.*

P4 l92: “105 m above sea level”

Response: *Amended.*

P4 l105-109: these information are redundant and/or well known. It could be removed.

Response: *This information has been now removed.*

P6 l165-172: I don’t think a thorough description of PMF and ME-2 is necessary here. Please shorten or remove this section.

Response: *Amended*

P8 l237: rephrase to “The other striking feature is that”

Response: *Amended.*

P8 l241: rephrase to “average 8 of such”

Response: *Amended.*

P8 l242: please add “to our knowledge”

Response: *Amended.*

P8 l243: rephrase to “highlight the strong impact”

Response: *Amended.*

P10 l286: rephrase to “to the regional character”

Response: *Amended.*

P11 l231: one could cite here Canonaco et al.(2015): Canonaco, F., Slowik, J. G., Baltensperger, U., and Prévôt, A. S. H.: Seasonal differences in oxygenated organic aerosol composition: implications for emissions sources and factor analysis, Atmos. Chem. Phys., 15, 6993-7002, https://doi.org/10.5194/acp-15-6993- 2015, 2015. -

Response: *The proposed references are now added in the revised text.*

P12 l343-350: I think this has already been presented elsewhere, so I don’t think it is necessary here.

Response: *Given the fact that a new approach in describing correlations, has been adopted for the revised manuscript, omitting the use of the theta angle, this description has now been removed.*

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