Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-356-AC1, 2018

Interactive comment on "Sources and processes that control the submicron organic aerosol in an urban Mediterranean environment (Athens) using high temporal resolution chemical composition measurements" *by* Iasonas Stavroulas et al.

Iasonas Stavroulas et al.

kbougiatioti@gmail.com

Received and published: 23 August 2018

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-356-RC1, 2018

Response to Anonymous Referee #1 comments

The manuscript of Stavroulas et al., presents and analyzes the organic aerosol sources over Athens (Greece) using long term observations (1 year) and 2 intensive campaigns

measurements. The source analysis is based on the Aerosol Chemical Speciation Monitor (ACSM). The finding are very important for the region of Athens but also crucial for the atmospheric science community as they found that during the winter a significant fraction of the secondary organic aerosol is clearly connected to the biomass burning organic aerosol. The paper is very well written and organized. I definetely recommend publishing of this paper, after some minor changes:

Response: We thank the anonymous referee for the thoughtful review. We have further elaborated on these points in the revised manuscript.

(1) The introduction contains information about Athens topography and biomass burning aerosol (BBOA). However, since in the paper are discussed additional sources found (e.g., LV-OOA, SV-OOA, HOA and COA) I suggest adding a paragraph giving some information about these sources and provide previous measurements for Athens. For example Kostenidou et al., 2015 have measured the OA sources for Athens suburban area during an intensive summer (2012) camping using HR-AMS data.

Response: Indeed, the introduction focuses mostly on winter time conditions and mainly discusses the biomass burning related atmospheric chemistry and its effect on air quality. The introduction in the revised version of the manuscript now adopts a wider perspective and addresses the referee's comment.

(2) Page 3, line 62: please add Florou et al. 2017 after Kalogridis et al. 2017 reference.

Response: Reference added.

(3) I suggest replacing the sentence: "Respective measurements using high resolution techniques are scarce and limited in time (Florou et al., 2017)" with: "Florou et al., 2017 have measured the chemical composition and the OA sources during a wintertime intensive campaign in the center of Athens using an HR-AMS an however their data are limited in time."

Response: Amended.

(4) Page 4, line 119: Please provide the RIENH4 and RIESO4 with the corresponding standard deviations.

Response: Response factor and relative ionization efficiencies of NH4+ and SO4= are now added to the revised supplementary material.

(5) Page 6, line 162: the right parenthesis should not be in bold form.

Response: Amended.

(6) Page 9, line 261: please add domestic or residential before heating.

Response: Amended.

(7) Pages 12-14, lines 353-425 (warm period section): It would be interesting to compare the HOA, COA, SV-OOA and LV-OOA mass spectra of the warm period in terms of angle theta with the corresponding spectra of Kostenidou et al. (2015) that they found for a suburban area of Athens during summer.

Response: A comparison with the summertime factors from Kostenidou et al. (2015) has now been added. Primary factors correlate well, namely HOA with $R^2=0.92$ for the 2016 dataset and $R^2=0.94$ for 2017, COA with $R^2=0.75$ for 2016 and $R^2=0.77$ for 2017. On the other hand, the semi-volatile component correlates moderately ($R^2=0.50$ and 0.56 for 2016 and 2017 respectively), mainly because of the least oxidized nature of this study's SV-OOA which is probably related with the fact that the Thissio station is urban (city-center) in contrast to the suburban Demokritos station where the measurements of Kostenidou et al. (2015) took place. Finally, the LV-OOA factor shows a slightly better correlation ($R^2=0.61$ and 0.59 for 2016 and 2017 respectively). The fact that the LV-OOA factor does not exhibit better correlation, is driven mostly by the elevated signal at m/z=18 (H_2O^+) attributed to this factor in this study, in contrast to its almost complete absence in the Kostenidou et al. (2015) spectra, performed with a High Resolution Time-of-Flight Aerosol Mass Spectrometer. If m/z=18 of this study is excluded from the correlation exercise the derived values for R2 are of 0.85 and 0.84 for 2016 and 2017

respectively.

(8) Pages 14-15 lines 426-459 (cold period section): It would be nice to also compare the BBOA, HOA, COA and LV-OOA mass spectra from the winter period with the BBOA, HOA, COA and OOA mass spectra of Florou et al. 2017 that they measured during the winter at the same site. Florou at al. (2017) found two BBOA factors during the winter for a study in Patras (Greece); one of them (BBOA-II) was less oxygenated and its origin was not fully explained i.e., it could be due to the different types of fuel orcombustion or due to different degree of BBOA aging. What is the angle theta between the SV-OOA mass spectrum of this study (that is linked to aged BBOA) and the BBOA-II of Florou et al. (2017)?

Response: A comparison of the mass spectra for HOA, COA, BBOA and LV-OOA to the ones obtained by Florou et al (2017) for Athens has been added to the revised version. Correlations are very good for all primary factors, e.g. HOA with $R^2=0.92$, COA with $R^2=0.96$ and BBOA with $R^2=0.88$. For the LV-OOA factor the same issue as when comparing with the summertime factors arises. When taking into account the m/z=18 fragment R2 is 0.57 while when excluding it correlation is stronger with $R^2=0.78$. For the Patras site, as well, the correlation of the primary factors is very good (HOA: $R^2=0.97$, COA: $R^2=0.93$ and BBOA: $R^2=0.89$). LV-OOA correlates well when not taking into account the contribution of m/z=18 ($R^2=0.88$) while correlation less strong otherwise ($R^2=0.68$)

(9) Page 17, line 496: there is a “t” alone in that sentence which it should be deleted.

Response: Amended.

(10) For the winter case you show in Table 2 that the BBOA mass fraction is around 8-10%. How about the absolute mass concentration? Did you see any correlations with temperature?

Response: As seen from the factor's time-series, BBOA exhibits the highest concentrations during nighttime, when temperatures are lower and people resort to biomass burning for heating purposes. However, temperature alone cannot explain the high values of BBOA as high values have been also observed during smog periods (SP, Fourtziou et al., 2017), characterized by low wind speed, which does not allow dispersion of air masses.

(11) I believe that a comparison with summer sulfate measurements from previous years should be made. It could be a paragraph before the conclusion part. How did the PM1 sulfate mass concentration and mass fraction change over the years? You could use the data of Kostenidou et al. (2015), for Athens during the summer and Bougiatioti et al., (2014) and Hildebrandt et al., (2010), which are for summer but for the Finokalia station (different location) given the fact that the sulfate concentration is similar in many locations above Greece during the summer (Tsiflikiotou, Master thesis). Using this trend, could you make any implications? For example how did the economical crisis in affect the air quality (less industry that produces SO2, which is converted to particulate sulfate)?

Response: We would like to thank the reviewer for his/her suggestion., The studies of Bougiatioti et al. (2014), Kostenidou et al. (2015) and this study were performed well within the economic recession, while only the short-term study of Hildebrandt et al. (2010) was performed before. . Thus by comparing levels at different locations, different months and given the limited amount of data such comparison could be biased. There is, indeed, an apparent reduction in the mean annual submicron sulfate levels, when compared with previous filter-based studies conducted in the area during the previous decade (Theodosi et al., 2011; Pateraki et al., 2012). We will keep however his/her suggestion and we plan in the future to collect all available data set from Greece to examine the impact of economic recession on SO4 levels.

(12) Figure 5. Please improve the resolution of this figure. Top graph: The left y axis should say “mass concentration (g m-3)” once. The right y axis should also say “mass concentration (g m-3)” once and for each sub-axis just indicate the name of the

species without g m-3. Bottom graphs: again “mass concentration (g m-3)” or “% mass concentration and for each sub-axis just indicate the mane of the species without g m-3 or Contrib.(%). Please take care the numbers on the y axis, some fall on other and it is difficult to be read. Avoid gaps.

Response: Figure 5 has now been redrawn in order to make the diurnal variability of each factor clearer. The referee’s suggestions/comments have been taken into account.

(13) Figure 6. The same as for Figure 5.

Response: Figure 6 has been redrawn in the same manner as Figure 5.

(14) Figure 8. You should consider using a lighter green for the LV-OOA in order to be more distinguishable from the SV-OOA. May be use another color for the map behind (light blue)?

Response: Figure 8 has been redrawn in the revised manuscript, so the information depicted is more clear.

References

Bougiatioti, A., Stavroulas, I., Kostenidou, E., Zarmpas, P., Theodosi, C., Kouvarakis, G., Canonaco, F., Prévôt, A. S. H., Nenes, A., Pandis, S. N., and Mihalopoulos, N.: Processing of biomass-burning aerosol in the eastern Mediterranean during summertime, Atmos. Chem. Phys., 14, 4793-4807, https://doi.org/10.5194/acp-14-4793-2014, 2014.

Florou, K., Papanastasiou, D. K., Pikridas, M., Kaltsonoudis, C., Louvaris, E., Gkatzelis, G. I., Patoulias, D., Mihalopoulos, N., and Pandis, S. N.: The contribution of wood burning and other pollution sources to wintertime organic aerosol levels in two Greek cities, Atmos. Chem. Phys., 17, 3145-3163, https://doi.org/10.5194/acp-17-3145-2017, 2017.

Fourtziou, L., Liakakou, E., Stavroulas, I., Theodosi, C., Zarmpas, P., Psiloglou, B., Sciare, J., Maggos, T., Bairachtari, K., Bougiatioti, A. and Gerasopoulos, E., 2017. Multi-tracer approach to characterize domestic wood burning in Athens (Greece) during wintertime. Atmospheric Environment, 148, pp.89-101.

Hildebrandt, L., Kostenidou, E., Mihalopoulos, N., Worsnop, D.R., Donahue, N.M. and Pandis, S.N., 2010. Formation of highly oxygenated organic aerosol in the atmosphere: Insights from the Finokalia Aerosol Measurement Experiments. Geophysical Research Letters, 37(23).

Kostenidou, E., Florou, K., Kaltsonoudis, C., Tsiflikiotou, M., Vratolis, S., Eleftheriadis, K., and Pandis, S. N.: Sources and chemical characterization of organic aerosol during the summer in the eastern Mediterranean, Atmos. Chem. Phys., 15, 11355-11371, https://doi.org/10.5194/acp-15-11355-2015, 2015.

Pateraki, S., Assimakopoulos, V.D., Bougiatioti, A., Kouvarakis, G., Mihalopoulos, N. and Vasilakos, C.: Carbonaceous and ionic compositional patterns of fine particles over an urban Mediterranean area, Sci. Total Environ., 424, 251–263, 2012.

Theodosi, C., Grivas, G., Zarmpas, P., Chaloulakou, A., and Mihalopoulos, N.: Mass and chemical composition of size-segregated aerosols (PM1, PM2.5, PM10) over Athens, Greece: local versus regional sources, Atmos. Chem. Phys., 11, 11895-11911, htt

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-356/acp-2018-356-AC1-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-356, 2018.