

Response to the comments of Referee # 2

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General comments

This study investigated the sources of ambient carbonaceous aerosol, particularly PAHs, in the area of Athens, Greece. The authors made good use of filter-based measurement methods for PMF analysis with organic molecular markers to link PAH concentrations and composition with specific sources, e.g. biomass burning. The analysis is thorough and generally clear. A few issues should be addressed: (a) figure quality could be improved, (b) the authors should more clearly state the novelty of the study relative to previous studies in the region, and (c) ACSM data should be discussed in more detail and in the context of the filter samples.

We thank the reviewer for the positive assessment of our work. We have carefully considered the general and specific suggestions and incorporated them in the revised manuscript. Please find below our detailed response to the reviewer's comments.

Specific comments

All figures: The figures are pixelated and hard to see/read – please save the images in higher resolution

We believe that the pixelation was due to the online production of the pdf file. All figures have been now produced as high resolution image files and can be submitted separately as well. A better color selection was also made in the map figure (Fig. 4).

Seasonal trends: How does Dec/Jan 2016-2017 compare with Dec/Jan 2017-2018 in terms of PAH concentrations, source contributions, etc.?

This point was also raised by the reviewer 1 and addressed accordingly.

Lines 74-84: There have been several previous studies of ambient air quality in Greece and how it is impacted by domestic biomass burning in particular, as the authors reference earlier in the introduction. Please explicitly state the novelty of the present study: other than being more recent, how do the sampling techniques/locations/times and analyses employed provide new insight? This would also be a good opportunity to discuss the use of complementary filter-based and online (?) techniques and what additional insight these provide, as using both of these is a strength of the study.

Thank you for providing the opportunity to describe more clearly the novel aspects of our work. The elements of originality of our work are now presented in detail in the last paragraph of the introduction.

Lines 128-133: Describe ACSM monitoring in more detail. Was this conducted at the same site as the filter sampling? What were the sampling dates/duration?

More details on ACSM measurements (operating principle, QA/QC, PMF methodology etc.) are provided in the revised manuscript.

Lines 133-136: Describe the method used to apportion BC into fossil fuel and wood burning contributions, as this is a secondary calculation for interpretation rather than primary data from the instrument

The Aethalometer model (Drinovec et al., 2015; Sandradewi et al., 2008) that is used internally by the AE33 7- λ aethalometer to apportion BC into source-specific components, is now briefly described.

Lines 180-183: Include numbers from this study for reference, and change “which however included 7” to “7 of which were”

We included indicative numbers and also discussed that the cited study reported street-site concentrations higher by 44 and 55% compared to urban background sites, for Σ -PAHs and BaP, respectively.

Figure 1: Stacked bar plots with this many different categories/colors do not provide useful information. I recommend presenting the individual PAH concentrations in a table, which would facilitate direct comparisons with other data sets in the future. The barplot could instead show monthly averages of HMW/MMW/LMW PAHs using the same colors as in the pie charts below, which would provide the reader with an overview of both absolute and relative concentrations of general PAH categories.

The figure was redesigned according to the suggestions of the reviewer.

Lines 234-242: Why is the ACSM data only discussed here? If it is included in the manuscript, it should be discussed further: for example, how the ACSM SoFi and filterbased PMF source apportionment compare, and the temporal trends in ACSM data. Which ACSM sources would likely include PAHs based on chemical signatures, and do these source contributions correspond to PAH concentrations? Also, clarify exactly what m/z 60 and 73 are thought to represent (for readers less familiar with AMS data). Do they correspond with filter-based levoglucosan measurements?

Regarding the potential inclusion of PAHs in ACSM measurements based on the mass spectrometric chemical signatures, that is suggested by the reviewer, we note that the upper m/z peak that is detected by our Q-ACSM is m/z 140 and therefore it wasn't possible to associate specific fragments with PAHs (naphthalene at m/z 128 is used by the ACSM as an internal standard to monitor detector performance) (Ng et al., 2011). The identification of distinctive PAH m/z signals in mass spectra has been reported by measurements with more advanced instrumentation, such as the ToF-ACSM (Zheng et al., 2020) and aerosol mass spectrometers (AMS) (Dzepina et al., 2007; Herring et al., 2015).

However, following his/her suggestion, we included in the revised manuscript additional ACSM data and performed correlation analysis with the source contributions from filter-based PMF, to explore commonalities in the temporal variabilities of the two approaches. The above analysis was incorporated in the revised manuscript.

Lines 293-295: How was CO measured?

CO was measured using a reference-grade NDIR monitor (APMA 360, Horiba Inc.). All details regarding measurement of regulatory pollutants are now included in the last paragraph of section 2.2.

Figure 3: Given that the carcinogenic risk of PAHs is summarized in the analysis as BaP equivalents, what is the purpose of showing both “c” and “d” pies (carcinogenic PAHs and BaPeq)? Though individual PAHs have differing degrees of toxicity, the numbers end up being very similar between c and d, so it doesn’t seem necessary to present both in the main manuscript. Individual pies for OC and EC apportionment would be more interesting from a chemical perspective. It would also be helpful to label each pie with a short title, in addition to the more detailed descriptions in the figure legend.

The Figure was amended according to the suggestion of the reviewer, omitting panel (c). Pie charts for source contributions to OC, EC were included in the supplement (since these are already shown in the source profiles of Figure 2).

Technical corrections

Line 26: Change “effective” to “present”

Amended.

Line 27: Change “lead” to “leads”

Amended.

Line 32: This refers to total measured PAHs? Clarify

It refers to the ensemble of the 12 PAHs included in the PMF analysis. It is now specified.

Line 41: Add “such as” before “power and industrial plants”

Amended.

Line 46: Change “Particular PAH members” to “Several PAHs”

Amended.

Line 49: BaP equivalents? Clarify.

While the directive highlights the necessity of measurements for additional PAHs, the set target value refers only to BaP, which is selected as a marker for the carcinogenic risk of polycyclic aromatic hydrocarbons in ambient air. The part was clarified.

Lines 54-56: Wouldn’t it make sense to cite Saffari et al 2013 here, too?

We agree, this was indeed the first publication to directly address the RWB issue in Greek cities. It is now included.

Lines 178-180: Need references

We added references to (Andreou and Rapsomanikis, 2009; Mantis et al., 2005) that measured at traffic sites and also to (Pateraki et al., 2019) where the urban industrial site recording the highest concentrations (Table S3) is in fact close to a major avenue in the center of Athens.

Lines 329-330: Change “time-series” to “from non-local sources” (I believe that is what this is referring to, but currently it is unclear)

Amended.

Line 336: change “lighter members” to “LMW PAHs”

Amended.

Lines 382-383: “transport” should be followed by a semicolon (;), not a comma (,) and “its variability and origin” should be “their variability and origins”

Amended.

Line 414: Shouldn't BaPeq be written with a subscript, i.e. BaP_{eq}? (here and throughout the rest of the manuscript)

Amended.

Line 441: Do “GAA” and “central Athens basin” refer to the same region? If not, clarify, and if so, be consistent

The GAA includes areas outside of the central Athens basin, namely the Thriassion Plain to the west and the Mesogeia Plain to the east. That is why we mention in section 2 that it has a larger population (3.8 million), compared to the 3 million stated in Line 441. We chose to make the projection just for the population of the Athens basin because we can't claim that levels at Thissio are representative of conditions in the Thriassion (industrial area) or Mesogeia (mostly rural background conditions).

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