

Interactive comment on “Six-year source apportionment of submicron organic aerosols from near-continuous measurements at SIRTa (Paris area, France)” by Yunjiang Zhang et al.

Anonymous Referee #1

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General comment: The manuscript presents the results from 6 years of almost uninterrupted ACSM measurements in the metro area of Paris, and their analysis for OA sources-fractions, inter-annual trends and potential geographic origins. The study is unique as it combines high temporal resolution with a long timeframe, allowing for the examination of variability on different scales. Long-term studies on aerosol characterization and source apportionment are really sparse, so these results are particularly valuable. The paper is generally well written, however the discussion could be expanded with respect to underlying processes of OA formation. The suggested effect of biomass burning on the two resolved OOA factors should be clarified and explained better. The impact of urban emissions and their transport to the receptor site of the

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study is minimally presented and should be argued in more detail. Finally, the study presents a real opportunity to assess the actual impact of emission reduction policies enacted in European countries during the last decade. Even though the authors briefly discuss these, I believe that these implications require more focus. Addressing these issues and also the specific comments listed below, the authors can provide a substantially improved version of the manuscript.

Specific comments: Lines 19-20: By now there are numerous annual high-res studies on chemical composition. What is really scarce is long-term studies of the kind. Put emphasis on the long-term aspect here. Line 23: Define here what type of background site (e.g. urban, suburban etc.). Lines 47-56: More or less well known facts. I would suggest shortening and instead adding the more recent advances in understanding the SOA processes. Line 59: Duration of cited studies is up to two years, so better rephrase “multi-year”. Also consider citing some filter-based long-term studies for carbonaceous aerosol characterization. Line 61-65: Mention the importance of the high temporal resolution aspect for the study of OA aerosols. Lines 79-82: There is also the SV-LV categorization. However, there is no need to include all the alternative characterizations here. Lines 86-88: Repetitive, can be removed. Line 89-93: Here the transition from emission reductions to the need for long-term observation isn’t very clear. Rephrase or omit this, the necessity for long-term has been already stated. Line 93: Probably “robust technology” isn’t correct here. Correct this to indicate why the ACSM is more suitable for long-term unattended operation. Line 98: Again, 2 years don’t likely qualify for “long-term”. Use another term (e.g. “time-extended”). Lines 110-119: Probably the identification and attribution of potential long-terms should be defined as the primary objective. Line 111: Not clear what you mean here. Specify the duration of each PMF analysis and how these results have been integrated for the 6-year period. Lines 122-128: Although references are provided, some details are necessary here regarding siting, especially regarding the traffic and residential characteristics in the surrounding area. Given the distance from the city, this is important for understanding the origins and role of POA. Line 131-133: Provide data capture in

%. Line 145: Add information about corrections for particle collection efficiency. Lines 151-168: Is there any inter-comparison information between AE31-AE33? Discuss the uncertainties expected due to different instrumentation. Lines 166-167: How are these AAE values selected? Based on literature data or some analysis specific to BC aerosols in the area? Lines 184-185: Although it is not the main focus in the study, are there any results from filter comparisons regarding the remaining anions quantified by the ACSM? Sulfate and nitrate concentrations are used in the subsequent analysis. Line 190: Indicate the methodology for the estimation of BLH. Line 195: All the underlying PMF methodological principles are well-known and ubiquitously present in related literature. I suggest removing the details and equations and keep only the specific parametrizations you applied in your analysis. Line 211: Mention why you restricted m/z up to 100 (naphthalene interference etc.). Line 243-247: This sensitivity analysis would be more complete if you had also checked against longer than tri-monthly time windows, to confirm the intuitive approach of seasonal variability. Line 262: Briefly describe the model and discuss temperature related uncertainties. Line 287: The t-test is of minor importance for trend analysis. Also, what is meant by censored data? Better remove the sentence. Lines 290-295: This part has to be clarified. First specify if you use the Theil-Sen estimator for slope estimation. Second, the Kruskal-Wallis test may not suffice to comprehensively assess seasonality for the purposes of trend analysis (it is also not clear which category is used for the test; month, season? any post-hoc evaluations applied? at which level are you testing for significance). It would be better to apply the MK and TS tests to deseasonalized data in all respects. Line 319: Since the choice of weights is empirical and probably study-specific, I suggest removing this. Line 323: The approach here regarding the absence of the COA factor is consistent with past results for ACSM studies in Paris. However, AMS results have indicated that occasionally the COA contribution could be comparable to that of pure BBOA. If it is understood here that COA is incorporated in the BBOA factor, the extent of its potential participation in BBOA should be discussed. Line 367: Correlations with eBC_{wb} and eBC_{ff} should be utilized for verification of BBOA, HOA and especially

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the BB-related MO-OOA. Check time-lagged cross-correlations between eBC_{wb} and MO-OOA for indications of the aging process. You should also use more your OC, EC data (specifically the OC/EC ratios) for validation of the different OA factors. Lines 377-380: This is somewhat of a stretch. The morning peaks are most probably related to modelling uncertainties. Support with references or remove. However, it might be worth mentioning the highest levels observed during weekends for the BB-related parameters, probably due to recreational use of wood-burning. Lines 381-400: Again, examine the HOA-NO_x correlations. Lines 388-390: Is this pattern constant across all seasons? Is there a possibility of HOA emissions from heating oil combustion in winter (if this is a significant source in the Paris region)? Lines 397-400: This is in contrast with the primary nature of hydrocarbon-like OA Consider if this result warrants indicating that extracted HOA in fall/winter is a mixed-factor (in abstract-conclusions). Lines 401-408: Indicate the correlations of MO-OOA with sulfate. Lines 423-427: These have to be reasoned against the fact that maximum LO-OOA levels are observed during nighttime. Line 429: What do you mean by unclear formation schemes? Explain if substantial BSOA formation is plausible based on general vegetation characteristics in the area. Lines 440-443: Rudimentary comments, remove. Lines 453-456: This is a further indication that you should probably deseasonalize HOA as well. Line 459: Test for significance level. Lines 485-487: This is not sufficient to prove that MO-OOA is BB-related. You should examine the associations with BB-tracers. Lines 487-489: There is number of studies that have associated BBOA with the less oxidized OA fraction as a result of rapid processing during nighttime conditions. The apparent different mechanism here (higher degree of aerosol processing) should be discussed. Lines 490-494: Could low temperatures be associated with increased precursor emissions from biomass burning for heating? Lines 511-514: A major influence from biomass burning has been mentioned in line 417, however it is not considered here. Line 522: I suggest that you perform the trend analysis also for total OA concentrations as well as for the ACSM-derived submicron aerosol concentrations. This can be important from a regulatory standpoint. Also provide numerical results for emission reductions

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during the study period, based on national and regional emission inventories. Lines 529-531: Add a reference regarding the relative dependence of OA, BC emissions on woodstove efficiency. Lines 533-535: Check if this exclusion is necessary when using the seasonal test for trend. Lines 544-545: Give the regional character of MO-OOA, you should probably take into account the impact of emission variability on a much larger spatial scale. Lines 562-565: I think that you should formulate this argument the other way round. Lines 610-612: The role of photochemical processing in SOA formation has to be considered here. Line 594: Results from S14 on primary OA should be discussed in more detail. The impact of the city is downplayed, when it should be a primary feature of the study. Line 595: “more stable conditions with anticyclonic conditions”. Unclear, clarify. Also add a reference for the synoptic meteorology of the Paris region. Lines 599-601: Based on Fig. 8A, could it be the case that the BB-associations observed during winter for this factor is related to processed BB aerosols originating in central-eastern Europe? Line 610: Figure 8g is essentially the only one presenting a contrasting pattern. Is this association with the Southern trajectories source-related or due to climatology? Lines 610-612: This is very speculative at present. Support with arguments or remove. Line 634: Indicate possible mitigation measures on the local administrative scale. Also that residential biomass burning is assuming Europe-wide importance as a pollution source, but remains largely unregulated. Figure S1a: Check if intercept is statistically significant. If not, run it through the origin. Also not sure that the color scale is informative here. Figure S14: I suggest keeping only the primary factors, move it to the main text and expand the discussion for local sources. Also include wind roses to show the relative prevalence of wind directions.

Technical edits: Line 56: Start new paragraph (“Although...”). Line 130: “...using a quadrupole ACSM. These measurements were performed...”. Line 131: Delete “during the investigated period”. Lines 133-135: Already mentioned, remove. Line 218: Delete “recently”. Line 221: Delete “so-called”. Line 231-234: Check citation here. Does this study use PTR-MS? Line 235: “of several”. Line 265: “ambient T”. Line 387: “ratios are”. Line 503: “in more detail” Line 634: “strategies”. Line 638:

“contributions to total OA”. Line 641: “contributions to OA in wintertime”.

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