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Interactive comment on “Investigating the annual behaviour of submicron secondary inorganic and organic aerosols in London” *by* D. E. Young et al.

Anonymous Referee #1

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The paper summarizes almost year-long measurements of aerosol composition in the suburbs of London using aerosol mass spectrometry. The period of study overlaps with two intensive measurement periods in winter and summer. Seasonal trends in aerosol composition are discussed and measurements are compared with previous data. In addition, PMF analysis is performed on 2-sets of organic mass spectra. The uniqueness of the paper is the PMF analysis for source characterization of organic aerosol in the region. In other words, to me, the inorganic data presented don't add any new and significant scientific knowledge to the literature. However, I am troubled with the fact that the PMF results from a c-ToF and HR-ToF mass spectrometers during the winter period are not the same. This brings me to question the validity of PMF analysis for the rest of the period. It is obvious that the authors have worked hard to present the PMF analysis; however, I'm not sure if the analysis is justified because two similar instruments result in different solutions and could lead to two different conclusions. To strengthen the paper (in addition to addressing the major and minor comments listed below), I recommend including auxiliary gas phase data and backtrajectory analysis to support and explain the observed patterns in inorganic and organic composition. Otherwise, the paper appears to be just another AMS paper with a summary of observations, without real meaningful context to it. I recommend accepting the manuscript after major revisions.

-Calling the measurements background measurements is misleading in my opinion. If the sampling site is located 7 km west of central London, it qualifies as more of a sub-urban area and not background environment. Please consider rephrasing throughout the manuscript (including tables and figures). - Line 12 of abstract: considering that the site is more of a suburban area and not background or true urban, the conclusions from this study cannot explain urban behavior of OA, so if indeed, SOA oxidation at the site is not changing in the year, this doesn't explain how SOA in an urban environment will behave. This sentence in the abstract and last paragraph in section 5.2 need to be removed. - Have you tried running PMF on different seasons separately to see how similar/different the factors are? - Table 2: there is no discussion in the discrepancy between PMF winter COA estimates from c-ToF and HR-ToF analysis (slope of only 0.52). In addition, there was no discussion on why only 1 OOA is resolved for HR-ToF and 2 OOAs for c-ToF. Since it seems OOA2(mod) has a very small concentration, the logic of considering a 5-factor solution for c-ToF analysis doesn't seem to hold true. This is not good news that the PMF results from winter from two co-located AMS instruments is not the same. - Is the summer data removed from yearly averages as well because of bad tuning of the mass spec? for example line 24, p. 18757 that indicates 38% contribution to primary OA from SFOA doesn't include summer months? If yes, it's better to re-emphasize that again when discussing 'annual' averages. (also in Figures, e.g, Fig. 2-3). To me, it makes sense to have all the inorganic and total organic data removed from the paper for summer as well because the heater bias may affect detection efficiency of different ions differently. - Line 15, p. 18756 is confusing. From reading the previous section, I was under the impression that SFOA factor from HR-ToF is double that of SFOA of c-ToF. So why is it stated here that ‘nearly all’ of SFOA factor is assigned to OOA2? Do the authors mean that all of OOA2 is in fact SFOA (as is suggested by the very low OOA2(mod)? If yes, line 15 here needs clarification. - In applying eqn 1, I’m not convinced that investigating the daily averages of OOA2 vs. SFOA is better than analyzing the recorded data as is, i.e., before averaging for longer periods. - According to eqns 1 and 3, OOA2(mod) is the same as intercept in eqn 1, i.e., OOA2(noSF)? - Typically OOA2 represents ‘locally’ produced SOA, which one might expect to depend strongly on photochemical processing whereas OOA1 is the strongly oxidized, long range transported OA. Since concentration of OOA2 is very small throughout the analyzed months, it appears that contribution of locally produced SOA to total OA at this site is small. How is the transport pattern of urban pollutants over the site? If the site is mostly upwind of London urban emissions, then it is not surprising that OOA2 and contribution of locally processed OA is small, and also not surprising that the oxidation characteristic of OA is not varying throughout the year. In addition, because summer months were not included in the PMF analysis of C-ToF data, ‘yearly’ discussion of SOA oxidation, which is expected to depend strongly on photochemical activity, is fruitless. These are important points that need to be addressed in the abstract and conclusion. - Line 4, p. 18761: since temperature affects the boundary layer mixing height and therefore concentration of pollutants, it is not straightforward to consider concentrations across different seasons without normalizing to correct for dilution. Isn’t there CO (or CO2) measurements available at the site which can be used to normalize OA (or SOA) by (e.g., in Fig. 6)? - line 13, p. 18761: it has not been shown that the aerosol observed at the site is really London SOA. - line 19, p. 18761: The O/C ratios of OOA1 don’t make sense to me. From the c-ToF data, O/C for all months except summer is 0.79, but for HR-ToF, the O/C for summer (which should represent the highest degree of oxidation) is just 0.52. I believe the HR-ToF PMF analysis is not separating out the factors properly either such that there is some contribution of less oxygenated factors to OOA1. Wasn't there an OOA2 factor resolved for HR-ToF analysis? - Lines 27-29, p. 18761: Because of the comment above, I don't think the authors can comment on seasonality of SOA oxidation. There is no consistent dataset throughout the year to assess how oxidation of SOA components had changed. - Line 27, p. 18762: do the authors mean in the summer high concentration events are dominated by OA? Please clarify. Also, because of mistuning of the mass spec, how would the summer data be jeopardized.

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