

General Comments:

Elser et al. describe gas and aerosol measurements conducted in two Estonian cities. The authors use a mobile platform to investigate the extent to which pollution concentrations within the city limits exceed regional background levels. Via source apportionment, the authors attribute the observed organic aerosol loadings to four primary sources: traffic emissions, biomass burning, primary residential emissions, and secondary aerosol formation. The authors map the spatial distribution of each component to identify source “hot spots.” In both cities, traffic-related components were most variable in the city center while biomass burning and primary residential emissions were concentrated in populated regions. Secondary components were well distributed throughout each city and had the least impact by point sources; however, increases in secondary aerosol were most strongly influenced by long-range transport of air masses originating from polluted regions west of Estonia.

The paper presents a useful and effective methodology for studying the impact of point sources on local air quality. Furthermore, these measurements are important as they assess the pollutant levels and source contributions in an understudied region of Europe. The manuscript is very well organized and many of the conclusions are well supported. I have some recommendations that would improve the quality of the manuscript. Upon addressing these comments, I recommend the manuscript for publication.

First, I believe that further details of the source apportionment should be included to strengthen the argument of a four-factor solution. Questions and comments pertaining to this aspect of the manuscript are summarized below. Second, parts of the methodology section employ sentence structure and wording that, at times, is awkward and/or difficult to follow. While I do not wish to interfere with stylistic choices, I believe that some rewording of these sections may help the manuscript read more fluently. Suggestions are provided in the Minor Comments.

Source Apportionment

The authors identify four factors that sufficiently describe the variation in the data. The authors are thorough with the comparison of factors with external tracers; however, there is little discussion and no figures demonstrating the model residuals as the PMF solution is pushed to higher factors. The authors describe their observations (Page 10, lines 12 – 24), however a figure should be included demonstrating the behavior of Q/Q_{exp} as a function of the number of factors. Furthermore, the authors present a 5-factor solution, but argue that the fifth “unknown” factor exhibits a primary emission temporal pattern (which is uncharacteristic of a LV-OOA factor) and therefore does not significantly improve the interpretation of data. While this may be true, I believe it is necessary to demonstrate that the temporal residuals are not significantly improved for a 5-factor solution. It may be that the “unknown” factor results from factor splitting or some other mathematical construct. In any case, the PMF discussion should better describe the factor residuals.

The authors invoke bootstrapping as a means of constraining the error in the PMF solution. The author’s note that bootstrapping inherently varies the algorithm starting point (i.e., seed) and therefore accounts for model uncertainties; however, the PMF solution may also be strongly affected by variations in f_{peak} (Ulbrich et al. 2009). There is little discussion about the rotational ambiguity of the PMF solution. I believe this discussion is necessary in order to evaluate the robustness of the PMF solution.

Finally, I believe it would be useful to compare the factor profiles to published spectra. This comparison would provide additional justification for the resolved factors. Specifically, I have some questions regarding the RIOA factor. The RIOA factor exhibits a temporal pattern that appears to be unique; however, the RIOA factor only exists in a 4 or higher factor solution and is primarily resolved from the BBOA and HOA factors (Fig S3). Consequently, the factor associated with RIOA results from the contribution of two other primary emission factors. While this may be simply due to the fact that

RIOA, BBOA, and HOA are common in residential areas, this result may also be a result of factor splitting.

There are a number of ways the authors can provide additional evidence in support of the RIOA factor. The simplest option would be to compare the factor profiles to published spectra. The authors provide some comparison in the text, however a supplemental figure would be more illustrative. A more thorough analysis would be to perform PMF on subsets of the data and determine if the RIOA factor is still resolved. For example, if one were to remove time periods when the RIOA component is dominant, does PMF still resolve an RIOA factor? I believe these additional tests would strengthen the authors' PMF solution.

Specific Comments:

Page 4, Lines 29-30: Are there any sources that outline the spatial distribution of heating systems within the city? For example, can the authors comment on why BBOA emissions are higher and more dispersed in region (2) of Tallin (Fig 1) as opposed to region (7)?

Page 8, Line 5: What studies have used the eBC source apportionment method? Please provide references.

Page 12, Lines 24-26: From what directions do emissions in Tartu/Tallin drain? It would seem to make most sense to take the upwind concentrations as your regional background. Perhaps a discussion of the topography and typical springtime meteorological conditions would help orient the reader to understand which airspaces reflect background conditions.

Page 13, Lines 1-3: Here, you state that BBOA is most enhanced during the evening hours, while on Page 11, Line 7 you state that higher loadings during the day are attributed to an increase primary sources (including BBOA). These statements tend to contradict each other. Please clarify.

Page 13, Lines 1-13: Here, you discuss diurnal patterns. If possible (perhaps with the stationary measurements), it would be most illustrative if the diurnal patterns were included as a figure.

Figures 5 and 6: Consider adding the labeling from Fig 1 to these plots in order to facilitate the identification of source regions.

Minor Comments

The following are wording suggestions that may help improve the fluency of the methodology section

Section 2.1

Page 4, Line 13: "... Tartu, with an area of 38.8 km²..."

Page 4, Line 21: "...to strongly enhance the signal of traffic emissions ..."

Page 4, Line 23: "... with low stacks ~~in both cities. In this regard,~~ a detailed...."

Section 2.2

Page 5, Line 17: "...For this work, the AMS ..."

Page 5, Line 20: "...lens efficiently transmits particles with 80 nm < D_p ≤ 3 μm and has been tested in previous chamber and ambient studies (Williams et al., 2013; Wolf et al., 2015; Elser et al., 2015)"

Page 6, Line 1: "... measurement method automatically corrects for the loading effect ..."
The following are additional comments related to the manuscript.

Page 6, Lines 3-6: "The concentrations of trace gases were measured by a Picarro-G2301 CO₂/CH₄ analyzer and a Licor-6262 CO monitor."

Section 2.3

Page 6, Lines 15-16: "...collection efficiency (CE) algorithm by Middlebrook et al. (2012) was used in the calculation of ambient mass concentrations (Middlebrook et al., 2012)."

Section 2.4

Page 4, Line 24: "...allows the representation of a two-dimensional ..."

Page 7, line 4: "In our case, the model input are the data and error matrices of OA..."

Page 7, Line 6: "...contain the fits to the high-resolution data (292 ions)..."

Page 7, Line 7: "...agrees with the mass calculated from the unit mass resolution integration..."

Page 7, Line 13: "...directly calculated from the CO₂⁺ fragment using the organic ..."

Page 7, Line 14: "... were excluded from the PMF analysis..."

Page 7, Line 15: "... variability of the CO₂⁺; these ions were reinserted post-analysis"

Page 7, Line 18: "... replicate datasets resulting from the perturbation of the original data..."

Page 7, Line 20: "... while other rows are removed (Paatero et al., 2014)..."

Page 7, Line 23: "Note that ~~as~~ each bootstrap..."

Page 7, Line 24: "... initialization point; thus, this methodology inherently includes the investigation of the classic seed variability..."

Page 7, Line 25: "...consistent, suggesting that the solution is robust."

Section 2.4.2

Page 8, Line 8: "... for the correlations with the external tracers, ~~but their spatial distributions couldn't be explored~~..."