Author's response:

We thank the reviewers for a careful reading and correction of our manuscript. Their suggestions have strongly helped improving the quality of the manuscript.

Following the suggestions of the anonymous referees 2, 3 and 4 we have added in the revised manuscript the description of the meteorological conditions during the measurement periods in both cities. A figure with the time series of wind direction and speed, temperature and precipitation has been added in the supplementary information (Fig. S2) and is described in the methodology section. Moreover, the average wind directions and speed during each measurement loop are now reported in a wind rose plot in Fig. 4 and 5 (spatial distributions for Tartu and Tallinn, respectively) and are fully discussed in the manuscript.

As suggested by anonymous referees 2 and 4, a detailed analysis of the source apportionment diagnostics has been added in the revised manuscript. A figure including (a) Q/Q_{exp} as a function of the number of factors, (b) correlations between OA sources with external factors as a function of the number of factors and the decrease in Q/Q_{exp} time series (c) and profiles (d) for increasing number of factors has been added in the supplementary information (Fig S5). Moreover, a table reporting the correlations between the OA sources from our four-factor solution and literature profiles has been added in the main text.

Moreover, following the suggestion of anonymous referee 4, we have added the correlation coefficients (R^2) between the spatial distributions of all sources and compounds in Tartu in the revised manuscript (Table S1).

Lastly, in order to give an overview of the major local PM sources, we have added emission maps in the revised manuscript (Fig. S1). The wood combustion and industrial sources and the traffic emission rates of the main streets are reported in these maps.

Anonymous Referee #3

Received and published: 23 February 2016

The paper by Else et al. summarizes mobile springtime measurements of aerosol concentrations and several gas phase species in two Estonian cities. The measurements allowed the authors to identify 4 classes of OA in both cities. Overall, aerosol composition in both cities was similar and was dominated by higher concentration of primary types of OA during the day and by lower amount of secondary OA at night. Contribution of the secondary inorganic species was low except during a transport event.
The manuscript is very well-organized and well-written. There are two aspects that need some work in my opinion. One is related to wind direction and its variability during day and night sampling and how it might affect the interpretation of the results (see my comment below). The other aspect is that since the measurement was done in two cities, I think more can be done to compare quantitatively aerosol air quality in these two cities. Since measurements of CO are already available, I think it will be valuable to look at the enhancement ratios (not by subtracting a background) but considering scattering plots of say OA vs. CO, BC vs CO (or the PMF-resolved factors or other species vs. CO) in comparable times of the day to separate out the differences in dynamics, boundary layer heights, dilution, etc. and be able to determine a more valuable comparison of the aerosol sources in these cities. This will also allow the authors to compare the measurements with other

measurements (ground based on airborne) in other cities around the world. I support publishing the paper after my comments (above and below) are addressed.

Author's response:
Indeed, ratios of different OA components to CO are generally used to take into consideration the effect of the PBL and dilution in order to investigate the evolution of a plume with photochemistry. However, here we lack measurements to estimate the photochemical lifetime of the sampled air masses (e.g NOx/NOy or VOCs).

In the figures below we display the scatter plots of different aerosol components versus CO to investigate differences between the two locations and with the time of the day. Such plots mostly reflect the profiles of combustion sources that dominate CO emissions. Similar ratios are found for the two cities (Fig. R1), which is consistent with similar sources of CO and similar emission profiles at both locations. HOA and eBC show higher ratios to CO during daytime (Fig. R2), as traffic (which dominates eBC emissions) is more enhanced during daytime compared to CO, which can be also affected by other sources (i.e. BBOA and RIOA).

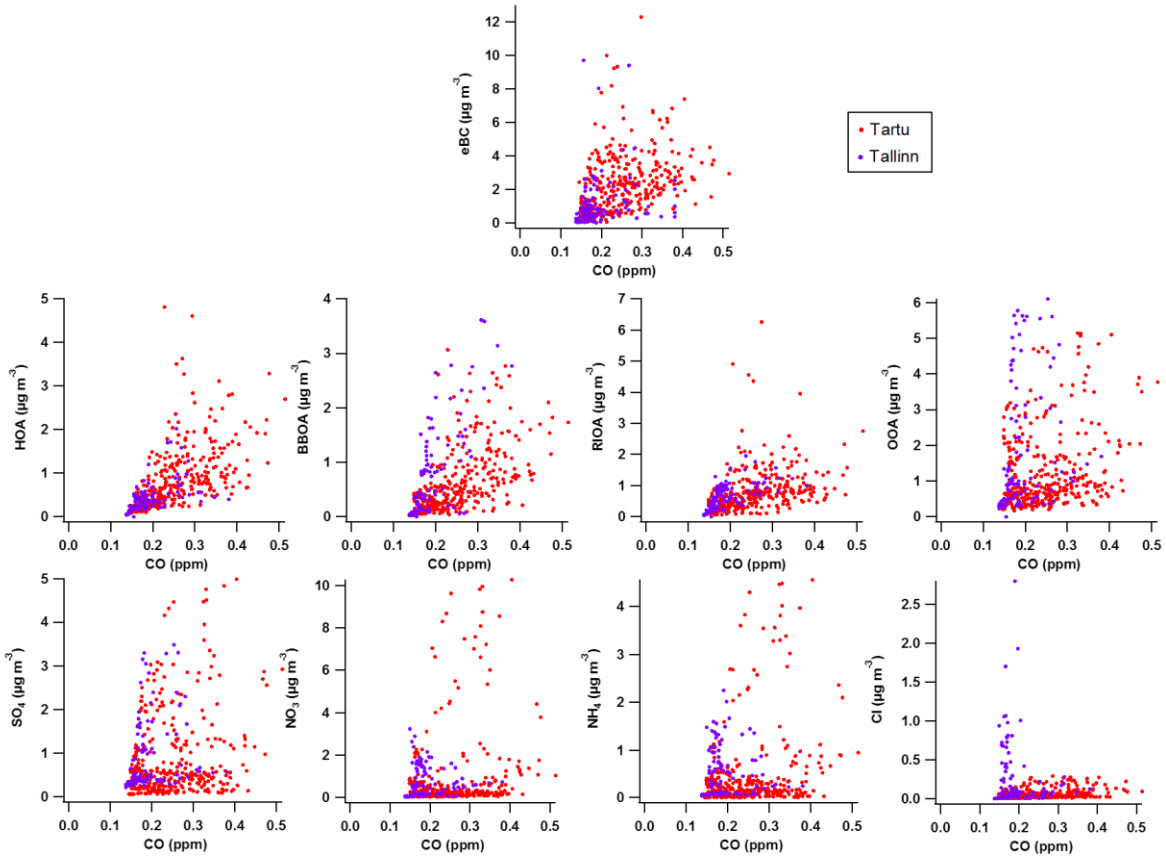

Figure R1: Scatter plots of all particle phase components versus CO, color-coded by measurement location.

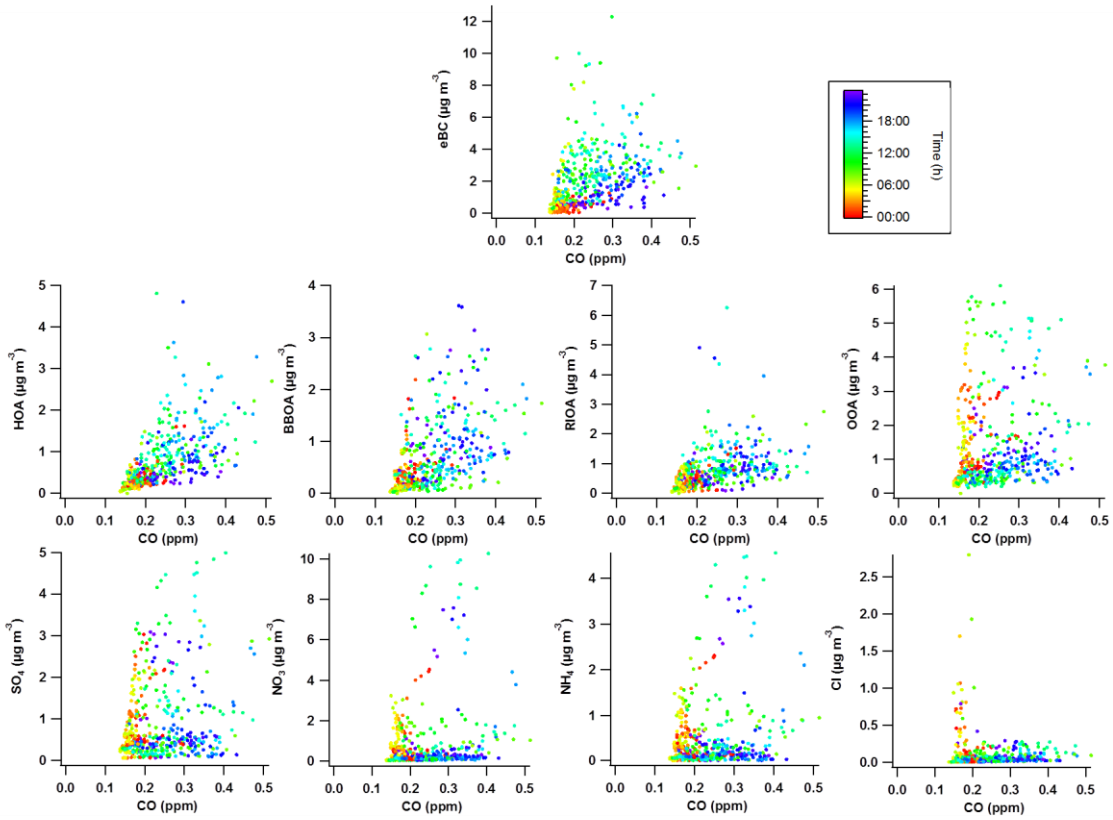

Figure R2: Scatter plots of all particle phase components versus CO, color-coded by measurement time.

Abstract: *indicate which month/season the measurements were carried out.*

Author's response: This information has been added in the abstract of the revised manuscript.

Changes in text:

Page 1, Line 14: This work presents the first spatially-resolved detailed characterization of the $PM_{2.5}$ in two major Estonian cities, ~~(~~Tallinn and Tartu~~), using mobile measurements~~. The measurements were performed in March 2014 using a mobile platform. In both cities, the non-refractory (NR)-$PM_{2.5}$ was characterized by a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) using a recently developed lens which increases the transmission of super-micron particles.

P5, L3: *It is mentioned that stationary measurements were made at night. Were there any mobile measurements also carried out at night?*

Author's response: The mobile measurements were performed between 6:00 and 23:00 LT, including only few drives during the early morning and at nighttime.

P8, L2: *Explain why A(abs)=1.7 was used for wood burning BC? And why was it that the lower wavelength of 370 nm was not used? Doesn't it make sense to use 370 nm since BrC would be stronger there?*

Author's response: The choice of the wavelengths and of the angstrom exponents used in this work are based on the findings in Zotter et al. (In prep.), where radiocarbon (^{14}C) measurements of elemental carbon (EC) are combined with Aethalometer data to determine the best Angstrom exponents for wood burning (α_{WB}) and traffic (α_{TR}). The best α values were evaluated by fitting the source apportionment results of the Aethalometer (in particular BC_{tr}/BC) against the fossil fraction of EC (EC_f/EC) derived from ^{14}C measurements. This analysis resulted in α_{tr} = 0.9 and α_{wb} = 1.68 fitting best the data when using the attenuation measured at 470 and 950 nm. Other wavelength combinations were also tested but in all cases, especially when 370 nm was used, the residuals of the fit were worse. Moreover it is known that the 370 nm channel of the Aethalometer is more sensitive to artefacts, including response to light absorbing SOA and the adsorption of VOCs on the filter. A brief description of the Aethalometer source apportionment method and the findings in Zotter et al. (In prep.) has been added in the revised manuscript.

Changes in text:
Page 8, Line 9: The Aethalometer measurements can be used to separate eBC from wood burning (eBC_{wb}) and from traffic (eBC_{tr}), by taking advantage of the spectral dependence of absorption, as described by the Ångström exponent (Ångström, 1929). Specifically, the enhanced absorption of wood burning particles in the ultraviolet and visible wavelengths region (370–520 nm) relative to that of traffic particles is used to separate the contributions of the two fractions. This method is described in detail….

Page 8, Line 19: The absorption Ångström exponent was calculated using the absorption measured at 470 and 950 nm and Ångström exponents of 0.9 and 1.7 were used for traffic and wood burning, respectively.~~,~~ These parameters were chosen following the suggestions in Zotter et al. (In prep.), where the comparison between radiocarbon (^{14}C) measurements of elemental carbon (EC) and the Aethalometer source apportionment results allowed the identification of the best wavelengths and Ångström exponents pairs.

P8, L16-17: *Just looking at Figure 2, it seems standard deviations of the averages would be really high, and maybe that's why they're not indicated along with the average values in Panel B. I wonder if estimates of the median values (or to be more complete, box and whisker plots) of the tracers will be more valuable than the average values.*

Author's response: We decided not to report the standard deviations in Fig. 2b (now Fig. 1b) as the big variability of the data can be seen from the time series and in our case it's simply reflecting the driving conditions. Thus, we believe that the standard deviation of the time series would not provide any useful information in our case, especially because this variability can be explained by the spatial distribution of the sources. Moreover, the addition of standard deviations of the time series could introduce some confusion between the meanings of the errors in different cases, as for the source apportionment results (see Fig. 3 of the revised manuscript) we report standard deviations among the 100 bootstrap runs (which is an indication of the model uncertainty and not of the temporal variability of the sources).

P11, L15: *missing a word ": : :of ?? (data??)..."*

Author's response: We have modified this paragraph in the revised manuscript.

Changes in text:
Page 12, Line 26: RIOA is also enhanced during day-time in Tartu (27% compared to 20% during night-time), and has similar relative contributions for day- and night-time in Tallinn (20 and 22%, respectively). In contrast, BBOA shows similar relative contributions for day- and night-time in Tartu (~~explaining~~ representing about 17 % of the OA mass), and slightly lower contributions during ~~the~~ day-time in Tallinn (20 % during day-time and 25 % at night-time).

P12, L1: *replace kurbside with curbside*

Author's response: Replaced in the revised manuscript.

Figure 2: *I suggest having the inorganics on a separate axis, with max _10 ug/m3, so you can see the tracers better.*

Author's response: The plot has been modified accordingly in the revised manuscript.

Figure 5: *For some species, it appears that the conc. were very different on different sides of the loop, suggesting that the sources are towards the center of the loop (as opposed to one side, e.g., BBOA and sulfate). In other words there is gradient in the latitudinal direction as well as longitudinal direction. To further investigate the source regions, it makes sense to consider wind direction data with these distribution maps. Were wind directions consistent during the day and night sampling time? It seems the averages include both daytime and nighttime. Could you add average wind barbs representatives for daytime and nighttime or at least discuss the wind patterns in the text? Correct interpretation of the mean and median values in Table 2 with relation to the source regions also needs some knowledge of the wind direction.*

Author's response:
As mentioned above, we have added a time series with the meteorological parameters during the measurement periods (including wind direction and speed, temperature and precipitation) in the supplementary information of the revised manuscript. No systematical difference can be observed between day and night wind conditions.
In Tartu, during the mobile measurements west winds (with speeds between 1 and 2.6 m s^{-1}) were predominant (see wind rose in Fig. 4 in the revised manuscript). However, the wind doesn't seem to influence the background concentrations measured in the east side of the loop, as the base values obtained for the this side of the loop were always equal or lower than those obtained in the west (see Table 3). Therefore, we also exclude a big influence of the transport of pollutants within the urban area and we expect that the identified source areas will not be strongly biased by this effect.

Figure 7: *Indicate in the legend that enhancement is relative to P05 values.*

Author's response: The Figure has been modified in the revised manuscript to indicate that the enhancement is relative to the P05 values. This information has also been added in the figure caption for further clarification.

Changes in text:

Figure 7 caption: Average longitude profiles of the enhancements (above P05) of all measured components and sources in Tartu.

Table 2 legend: *Indicate which city the stats refer to.*

Author's response: Added in the revised manuscript.

Changes in text:

(A) Average longitude profiles (Tartu):
(B) Median longitude profiles (Tartu):