Review of

“Urban increments of gaseous and aerosol pollutants and their sources using mobile aerosol mass spectrometry measurements”

By Elser et al.

General comments

This paper presents mobile measurements of gas- and particle-phase pollutants at two Estonian cities of Tartu and Talinn. Detail chemical composition of NR-$PM_{2.5}$ as well as BC and trace gases (CO, CO_2, and CH4) were observed in high time resolution and OA characteristics were found to be similar (HOA, BBOA, RIOA, and OOA) at both cities. Primary types of OA (HOA, BBOA and RIOA) were high during the day, whereas secondary OA (OOA) enhanced at night. In summary, the mobile measurements allowed the authors to observe time and spatial distribution of primary and secondary pollutants, as well as influences of long-range transport and local temperature inversion to aerosol and gases pollutants in Tartu and Talinn.

The manuscript is well structured and written. I have some comments regarding research methods, results and discussion as well as suggestions that would improve quality of the manuscript. There are also some typing errors in the text, tables and figures summarized in technical comments. Overall, I support publication of the manuscript after my comments are addressed.

Specific comments

OA source apportionment:

I believe the authors have done rigorous analysis in selecting the best PMF factor solution. I expect some of the analysis figures and/or tables are provided in the SI. PMF diagnostic plots, such as those presented by Zhang et al. (2011), are important for understanding the analysis and discussion. Residuals of time series and mass spectra for 3-, 4-, and 5-factor solutions and correlations between factors time series and external tracers (i.e., CO, eBC, SO_4, NO_3) are useful in understanding selection of the best factor solution. I recommend adding this information in the SI at the least.

eBC source apportionment:

It is not clear why do the authors choose to calculate Angstrom exponent using absorption at 470 and 950 nm. Since Zotter et al. paper has not yet published, I could not verify how the calculation was done. I suggest adding a brief description of this calculation in the main text or SI. Also, brief description of calculation of eBC_{wb} and eBC_{tr} would be a useful addition in the SI.

Results and discussion:

Page 10

Ln 1-2: Could the authors provide approximate uncertainty value of ratio of BBOA and eBC_{wb} relative to change in Angstrom exponent?

Ln 2-10: From information provided, it seems reasonable to assign RIOA as COA. The lack of diurnal variability does not mean that the factor is not a COA factor. It is possible that the lack of diurnal variability is due to homogeneous source of cooking emission, or stagnant atmosphere during measurements Have the authors look at meteorological conditions, such as wind direction and speed, when RIOA concentrations were high? I would recommend adding meteorological information in the SI.

Identification of RIOA or COA could be further assessed by plotting the factor mass spectra side-by-side with reference mass spectra from previous studies, such as by Mohr et al. (2012). The authors will need to provide more evidence to support identification of the RIOA factor.

Ln 16-24: The 5th factor, LV-OOA, may not show certain process or source, but it can show whether highly oxidized OA is important in the study locations. The LV-OOA can be formed from oxidation of primary OA (e.g., BBOA) or transported into the study location. The authors can discuss this further.

Page 11 Ln 12-15: Increase of RIOA in Talinn is relatively small. I think distribution of RIOA in Talinn is more homogeneous compared to Tartu. Thus, enhancement of RIOA in urban area of Talinn is not well supported.

Page 13 Ln 1-3: Spatial distribution of eBC, CO, and CO_2 are consistent not only with HOA but also with BBOA. Thus, they may come from BBOA as well. It would be easier to show consistency or inconsistency by correlation coefficient (R^2) between those tracers and HOA and BBOA. Also, in general I disagree that CO_2 is mostly traffic because it can be emitted from vegetation and other sources. The authors will need to provide more evidence to support CO_2 from traffic.

Technical comments
Page 4 Ln 22: Add reference for these statements.

Page 5
Ln 10: Unit for flow rate is $m^3\ s^{-1}$ or L min^{-1}
Ln 12: What is the size of particle in the aerosol inlet before particles are divided into different aerosol measurements.
Ln 28: eBC has been defined in the introduction.

Page 8 Ln 28-30: The statement about enhancement of negative health impacts is not well supported, as it was not within the scope of this study. I suggest the authors to omit the part or revise the sentence.

Page 10 Ln 29: Delete "secondary" or change it to "secondary source (OOA)"

Page 11 Ln 24: "the 5th percentile (P05) ~~of~~"

Page 13 Ln 20: Add space "… is 4.2 …"

Table 2: Superscript for the unit $\mu g\ m^{-3}$.

Figure 2:
(a) What does the different shade of purple for CO_2 mean?
(b) If the average pollutant concentrations exclude those from special events, this needs to be included in the title.

Figure 3: I think mass spectra relative contribution is not in %. For comparison, relative contribution in Figure S3 is unitless.

Figure 8: Add in the title that the back-trajectories is from 10:00 at 10 March 2014 to 8:00 at 11 March 2014.

Reference

Mohr, C., DeCarlo, P.F., Heringa, M.F., Chirico, R., Slowik, J.G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Penuelas, J., Jimenez, J.L., Crippa, M., Zimmermann, R., Baltensperger, U. and Prevot, A.S.H.: Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data, Atmos. Chem. Phys., 12, 1649-1665, doi:10.5194/acp-12-1649-2012, 2012.

Zhang, Q. Q.: Understanding atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, Analyt. Bioanalyt. Chem., 401, 3045-3067, 2011.