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 #2

Received and published: 26 February 2016

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 fpeak (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.

Author's response:
Based on the reviewer comment, we have additionally performed a thorough residual analysis as a function of the number of factors. These diagnostics are presented in the new figure Fig. S5 in the supplementary of the revised manuscript (see below). This figure includes the change in Q/Q_{exp}, in the correlation coefficients (R^2) of the resolved factors with the external markers and the change in the residuals time series and profiles for solutions with increasing number of factors. We show that the correlation coefficients (R^2) between factors and markers increase when a fourth factor is included, but are not improved when a fifth factor is added. The addition of the fourth factor, which enabled the extraction of RIOA, allows explaining additional structures in the residuals' time series and unsaturated fragments in the residuals mass spectrum. Including a fifth factor also improves the model mathematical quality, by additionally explaining $C_xH_yN_w$ and biomass burning (at *m/z* 60 and 73) related fragments. The additionally extracted factor in the five-factor solution, referred to as 'unknown', has elevated contributions from oxygenated fragments often related to SOA (*m/z* 44) and BBOA (*m/z* 60 and 73), but a time series that unambiguously relates this factor to a spatially variable primary emission source. In effect, the majority (62%) of this factor contribution arises from a split in the BBOA factor from the four-factor solution (the rest comes from the residuals and the OOA). Moreover, the sum of the contributions of the 'unknown' factor and the BBOA from the five-factor solution matches the BBOA contributions from the four-factor solution (R^2 = 0.97 and slope = 1.15 as shown in Fig. S6). This split in the BBOA is very likely a direct consequence of the variable nature of this combustion source, but the two BBOA-like factors extracted in the five-factor solution could not be related to different emission processes. The addition of this factor did not affect the spectral profiles and time series of the other factors and their correlations with their respective markers and did not aid the interpretation of the data. Therefore, we considered the four-factor solution as an optimal representation of our data. This discussion is now added in the text.

The f_{peak} approach has been used in the past to study the rotational uncertainty of the source apportionment solution. However, varying the fpeak parameter allows to trace only one dimension through the rotationally accessible domain (which is multi-dimensional), and therefore provides only a lower limit for rotational uncertainty (Paatero et al., 2014). Bootstrap is a more effective approach to explore the rotational ambiguity and provides an upper limit for the rotational uncertainty.

A table containing the correlation coefficients (R^2) between the OA profiles from the four-factor solution and literature profiles has been added in the main text of the revised manuscript (Table 2). The high correlations retrieved for the RIOA with cooking spectra from literature (R^2 of about 0.8), strengthens the use of a four-factor solution and the link between our RIOA spectra and cooking emissions.

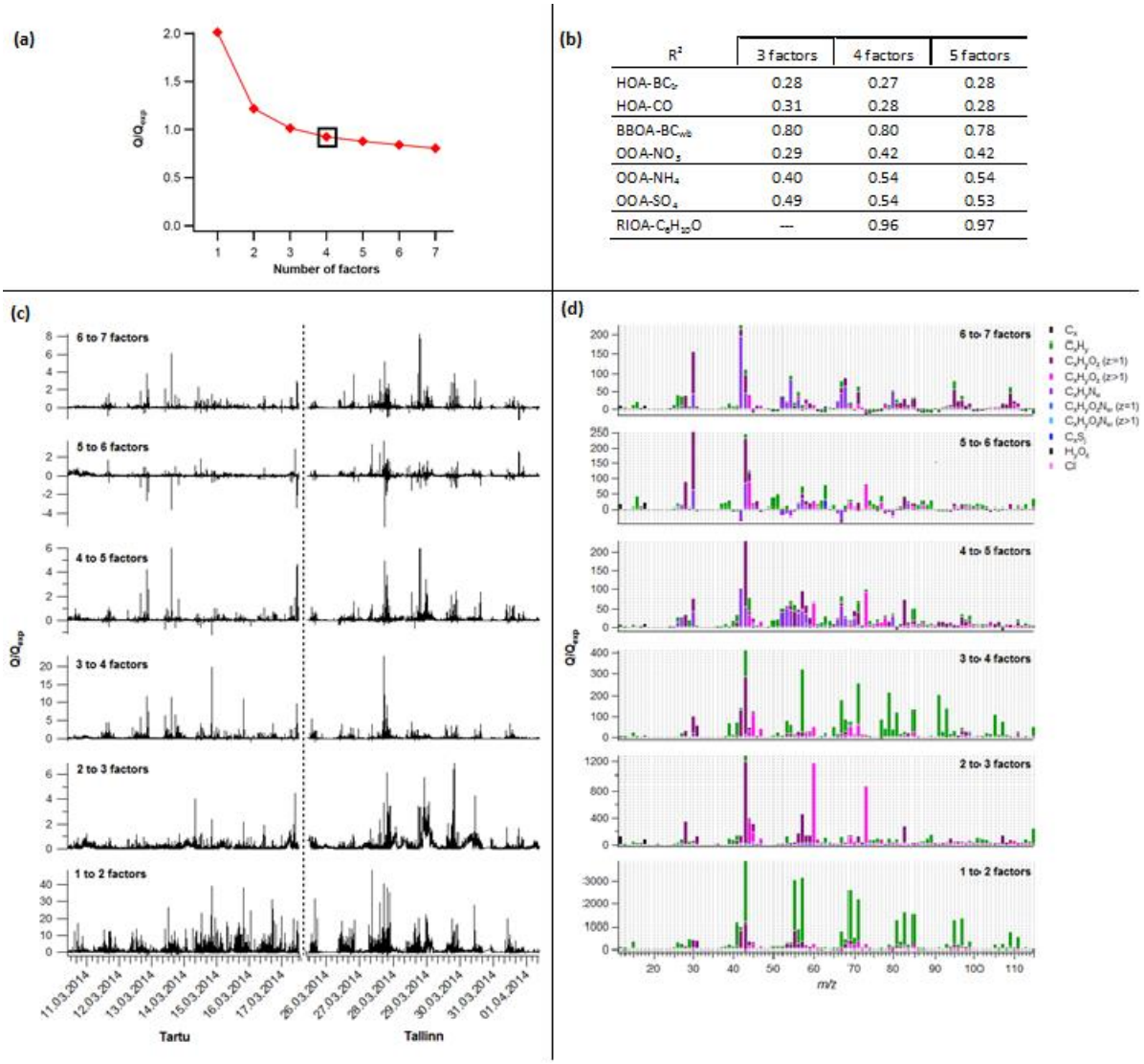
As described in the text, within the bootstrap method 64 % of the original points are used in each replicate of the input matrices. For all one-hundred bootstrap runs the RIOA was retrieved and the solution was very stable. Moreover, we also performed some PMF runs using only the data from Tallinn, where RIOA is more homogeneous compared to Tartu, and this factor was still always resolved. Therefore, this factor will always be resolved even when performing PMF on smaller subsets of the data.

Changes in text:

Page 10, Line 31: Some important diagnostic parameters of the source apportionment (including Q/Q_{exp}, factor-marker correlation, and time-series and profiles residuals for solutions with different number of factors) are reported in Fig. S5. The correlation coefficients

(R^2) between factors and markers significantly increase when a fourth factor is included, but are not improved when a fifth factor is added. The addition of the fourth factor, which enabled the extraction of RIOA, allows explaining additional structures in the residuals' time series and unsaturated fragments in the residuals mass spectrum. Including a fifth factor also improves the model mathematical quality, by additionally explaining $C_xH_yN_w$ and biomass burning (at *m/z* 60 and 73) related fragments. The additionally extracted factor in the five-factor solution, referred to as 'unknown', has elevated contributions from oxygenated fragments often related to SOA (*m/z* 44) and BBOA (*m/z* 60 and 73), but a time series that unambiguously relates this factor to a spatially variable primary emission source. In effect, the majority (62%) of this factor contribution arises from a split in the BBOA factor from the four-factor solution (the rest comes from the residuals and the OOA). Moreover, the sum of the contributions of the 'unknown' factor and the BBOA from the five-factor solution matches the BBOA contributions from the four-factor solution (R^2 = 0.97 and slope = 1.15 as shown in Fig. S6). This split in the BBOA is very likely a direct consequence of the variable nature of this combustion source, but the two BBOA-like factors extracted in the five-factor solution could not be related to different emission processes. Furthermore, the addition of this factor did not affect the spectral profiles and time series of the other factors and their correlations with their respective markers and did not aid the interpretation of the data. Therefore, we considered the four-factor solution as an optimal representation of our data. Table 2 contains the correlation coefficients (R^2) between the OA profiles from the four-factor solution and available literature profiles (Aiken et al., 2009; Mohr et al., 2012; Setyan et al., 2012; Crippa et al., 2013b). The high correlations obtained in all cases support the use of a four-factor solution and strengthen the link between the RIOA and cooking emissions (R^2 of about 0.8 between RIOA and cooking tracer).

~~If the number of factors is decreased, the RIOA factor is not resolved and the OOA time-series becomes contaminated by local spikes, which is unexpected for a regional component (see Fig. S3 and S4). In contrast, if a five-factor solution is considered an additional highly oxygenated factor is obtained ("unknown" factor in Fig. S3 and S4). The mass spectrum of this additional factor resembles a low-volatility OOA (LV-OOA), as resolved in many previous works (Jimenez et al., 2009), but its time series exhibits the typical characteristics of the primary factors, i.e. strong increases in emission areas. Therefore, this further increase in the number of factors doesn't seem to improve the interpretation of the data, as the new factor cannot be explicitly associated to distinct sources or processes. Accordingly, a four-factor solution was considered as optimal and is utilized below.~~



R^2	3 factors	4 factors	5 factors
HOA-BC_y	0.28	0.27	0.28
HOA-CO	0.31	0.28	0.28
BBOA-BC_{wb}	0.80	0.80	0.78
OOA-NO_3	0.29	0.42	0.42
OOA-NH_4	0.40	0.54	0.54
OOA-SO_4	0.49	0.54	0.53
RIOA-$C_8H_{20}O$	---	0.96	0.97

Figure S5 (new): Source apportionment diagnostics for increasing number of factors: (a) Q/Q_{exp}; (b) Correlation coefficient (R^2) between OA sources and markers; (c) Decrease in Q/Q_{exp} time series; (d) Decrease in Q/Q_{exp} profiles.

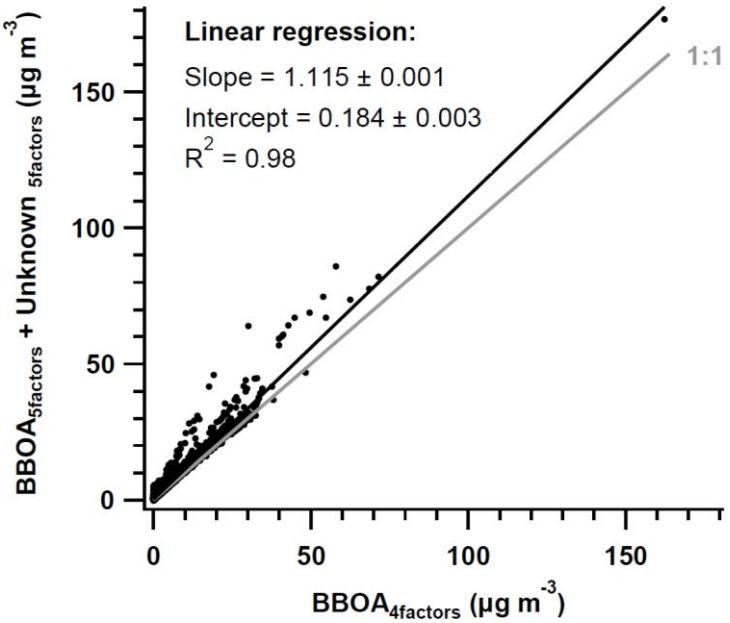


Figure S6 (new): Correlation between the BBOA time series from the four-factor solution and the sum of the BBOA and the 'unknown' time series in the five-factor solution.

Table 2: Correlation coefficients (R^2) between the OA profiles from the four-factor solution and literature profiles. Note: The different nomenclatures used in the literature for the different OOA factors have been homogenized to a semi-volatile OOA (SV-OOA) and a low-volatility OOA (LV-OOA).

R^2	Aiken et al., 2009	Mohr et al., 2012	Setyan et al., 2012	Crippa et al., 2013b
HOA-HOA	0.82	0.96	0.72	0.78
BBOA-BBOA	0.86	0.68	---	---
RIOA-COA	---	0.83	---	0.81
OOA-SVOOA	0.96	0.72	0.90	0.71
OOA-LVOOA	0.91	0.93	0.94	0.96

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)?*

Author's response:
To give an overview of the local PM sources, we have added emission maps of the major sources (including residential wood combustion, industry and traffic) in the revised manuscript. These maps are reported in the new Fig. S1 (see below), where residential wood combustion sources are marked with green dots, industrial sources (mainly local boiler houses) with blue markers and main streets are colored based on the traffic emission rates. It is clear that the density of sources of residential wood combustion is much higher in the southern part of the driving route in Tallinn (region 2), which is in agreement with the higher BBOA concentrations observed in this area.

Changes in text:

Page 4, Line 30: The measurements took place from 10 to 17 March 2014 in Tartu and from 25 March to 1 April 2014 in Tallinn. ~~The GPS trace of the driving routes in the two cities is shown in Fig. 1.~~ Emission maps including residential wood combustion and industrial sources and the traffic emission rates in the major streets of the two cities are reported in Fig. S1.The driving routes ~~paths~~ were chosen in order to cover heavily trafficked roads, residential areas ~~where different heating systems are used (wood/coal burning, central heating or mixed)~~ and background sites with little local emissions.

Page 14, Line 31: BBOA is ~~more~~ strongly enhanced in the residential areas, consistent with the distribution of residential wood combustion sources shown in Fig. S1. ~~and t~~The maximum BBOA enhancement is seen in the evening hours (15:00 to 21:00, LT) when domestic heating is more active.

Page 16, Line 5: BBOA (Fig. 5d~~6b~~) has higher contributions in the ~~two~~ residential areas, especially in region 2 of the driving route, where there is a very high density of residential wood combustion sources (see Fig. S1). ~~while c~~Compared to Tartu, in Tallinn the spatial distribution of RIOA (Fig. 5e~~6c~~) is more homogeneous, with only slight enhancements in the residential area and in the city center.

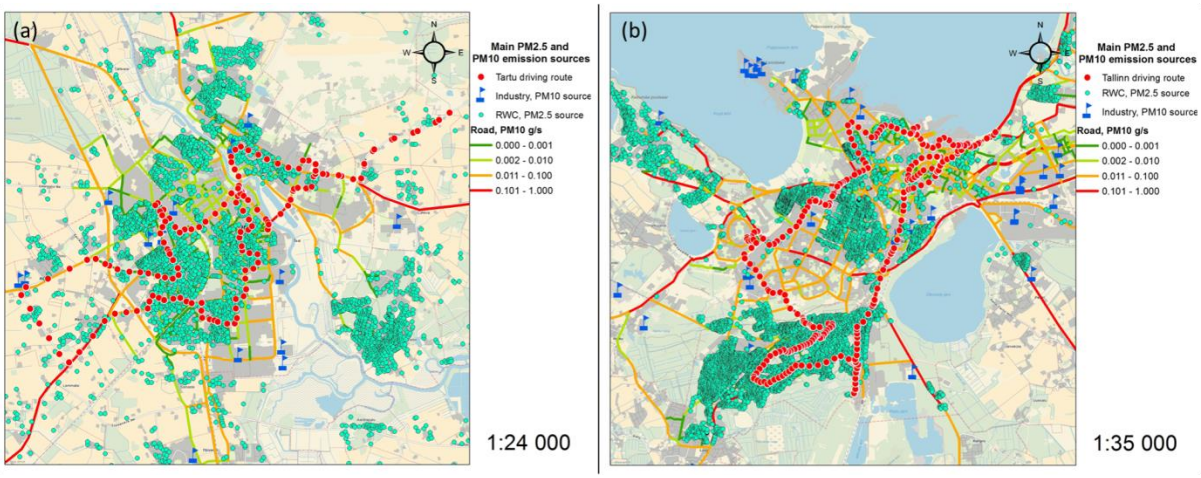


Figure S1 (new): Emission maps for (a) Tartu and (b) Tallinn. Green dots indicate residential wood combustion sources, blue markers indicate industrial sources (mainly local boiler houses) and the color of the main streets represents the traffic emission rates.

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

Author's response: We thank the reviewer for the valuable remark. We have added the following references in the modified manuscript: Favez et al., (2010), Herich et al. (2011), Sciare et al. (2011) and Crilley et al. (2015).

Changes in text:
Page 8, Line 14: This method is described in detail in Sandradewi et al. (2008) and has been successfully applied at many locations across Europe (Favez et al., 2010; Herich et al., 2011, Sciare et al., 2011; Crilley et al., 2015).

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.*

Author's response:
As mentioned in the introduction of this review, we have included detailed wind direction and wind speed analyses for the measurement periods in both cities in the revised manuscript. The time series of the wind direction, wind speed and the wind roses showing the average wind direction and speed during each loop are reported for the two measurement locations in the revised manuscript (Fig. S2, Fig. 4b and Fig. 5b). During the mobile measurements, the wind was predominantly from the west in Tartu, and from west and east in Tallinn.
The west winds observed during the drives in Tartu (with speeds between 1 and 2.6 m s^{-1}), don't seem to influence the background concentrations measured in the east side of the loop, as the base values obtained for the east side are always equal or lower than those obtained in the west (see Table 3). As the differences between the east and west base values (from the sigmoid fits) are in most cases minor, the west-east averages were used to calculate the urban increments concentrations.
In Tallinn, in order to identify possible processes influencing the spatial distributions of the measured pollutants for the two different wind patterns, the average spatial distributions were calculated for loops with west winds (7 loops) and east winds (16 loops), excluding drives during accumulation events. The results of these analyses are reported in the supplementary of the revised manuscript (Fig. S14 and S15) and show that, in general, the wind direction doesn't have a visible effect on the identified source areas and similar enhancements are found for both wind directions. A detailed analysis of the spatial distributions shows that BBOA, SO_4 and NO_3 show considerably higher enhancements for west winds, while HOA is more increased for east wind conditions. This difference is most probably related to the presence of west winds during the weekend (enhanced residential emissions) and east winds during the week-day measurements (enhanced traffic emissions).

Changes in text:
Page 5, Line 10: Meteorological data were recorded on~~in~~ a meteorological ~~-~~tower in Külitse (around 10 km south-east from Tartu) and in the Tartu and Tallinn-Zoo meteorological stations. The most relevant parameters (including wind direction and speed, temperature and precipitation) are reported in Fig. S2.

Page 14, Line 14: As shown by the wind rose in Fig. 4b, during the drives in Tartu the wind was predominantly from the west. However, the background concentrations measured at the east side of the loop don't seem to be affected by the transport of pollutants from the urban area, as the base values obtained for the east side are equal or lower than those from the

west side (see Table 3). ~~Moreover, the fits on the west side of Tartu show always higher base values than those for the east, indicating the influence of local sources in the considered regional background area west of Tartu. However,~~ As the~~se~~ differences between the west and east fits are in most cases rather low, ~~and therefore~~ we use the west-east averages of the base values to calculate the urban increments concentrations in Table 2.

Page 16, Line 14: Winds from west and east were observed during the mobile measurements in Tallinn (Fig. 5b). In order to identify possible processes influencing the spatial distributions of the measured pollutants for the two different wind patterns, the average spatial distributions were calculated for al loops with west wind (7 loops) and loops with east wind (16 loops, excluding drives during accumulation events). The results of these analyses are reported in the supplementary information (Fig. S14 and S15) and show that, in general, the wind direction didn't have an effect on the identified source areas and similar enhancements were found for both types of winds. A detailed analysis of these spatial distributions shows that BBOA, SO_4 and NO_3 are stronger enhanced during west winds, while HOA is more enhanced for east wind conditions. This difference is most probably related to the presence of west winds during the weekend (enhanced residential emissions) and east winds during the week-day measurements (enhanced traffic emissions).

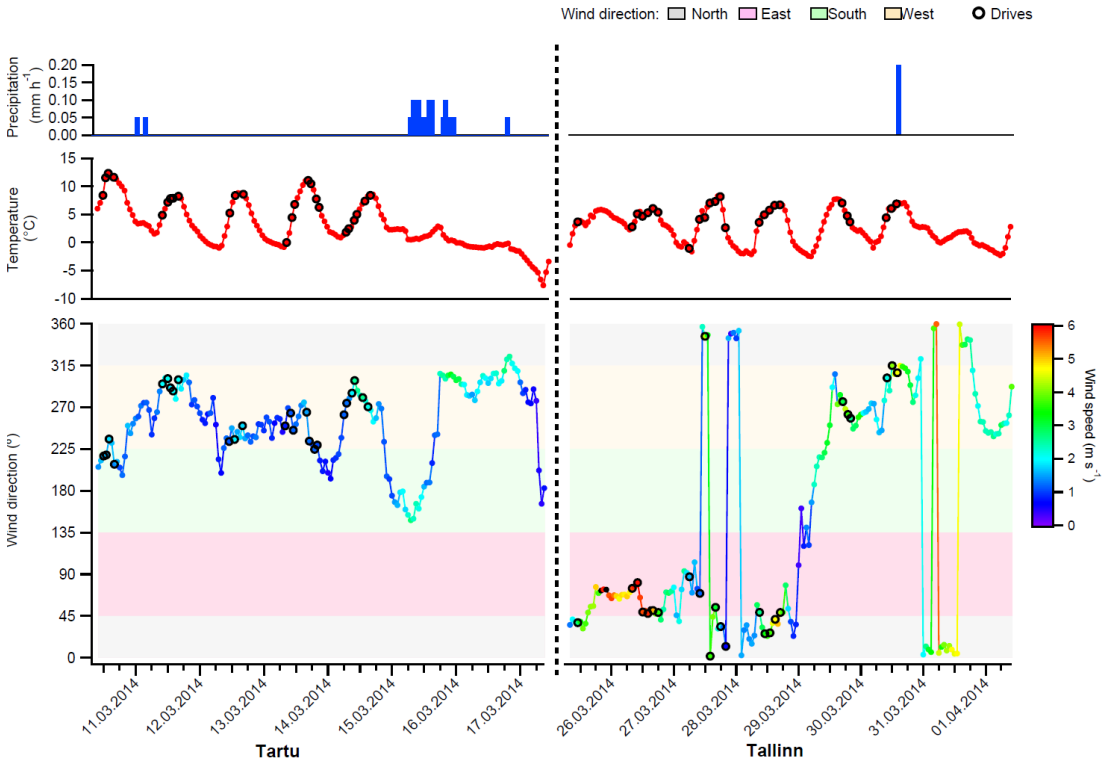


Figure S2 (New): Meteorological conditions during measurements periods in Tartu (data from the Tartu monitoring station) and Tallinn (data from the Zoo monitoring station).

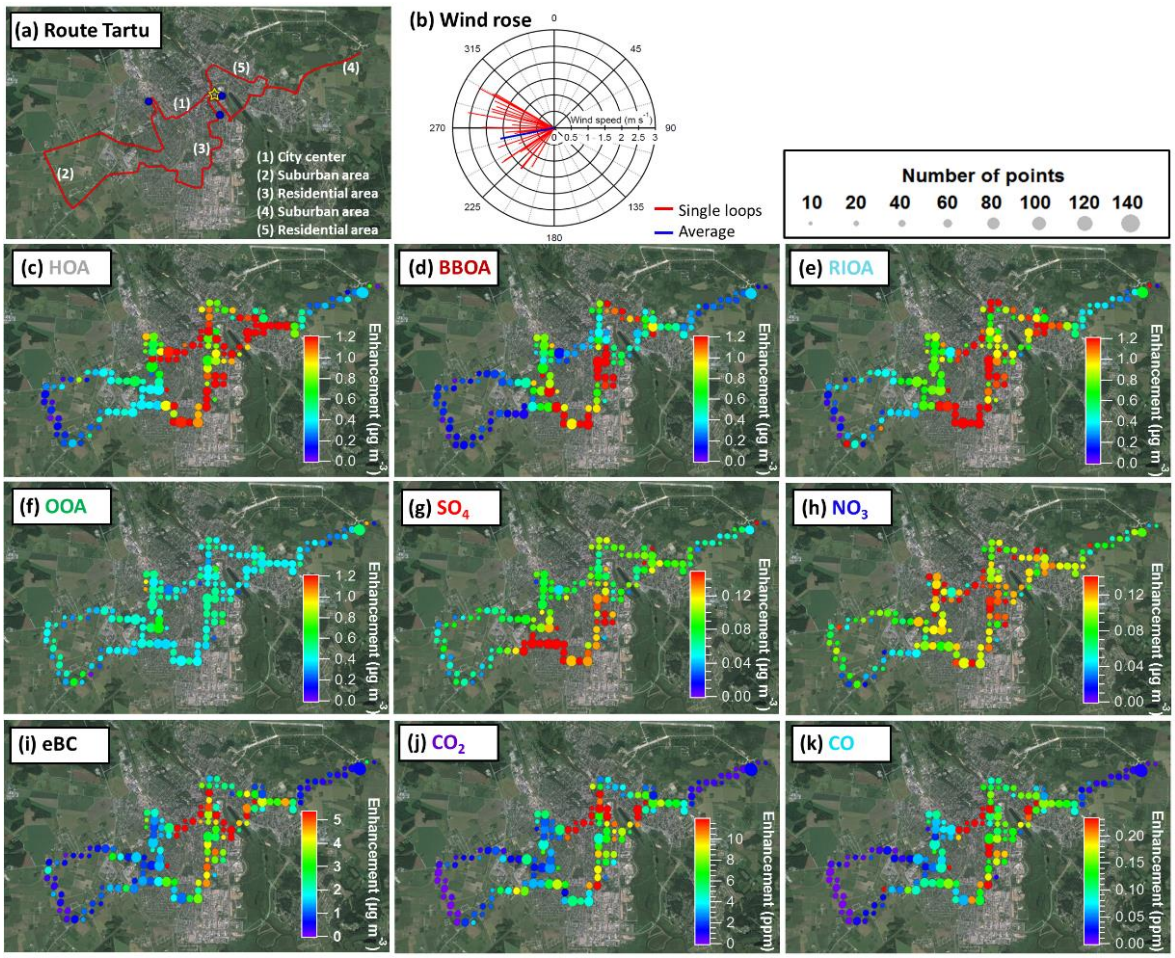


Figure 4 (Modified): (a) Driving route in Tartu: the red trace represents the GPS data, the yellow star the stationary measurements location and the blue dots the monitoring stations of the Estonian Environmental Research Institute (EERC); (b) Wind conditions during the mobile measurements in Tartu: red traces represent the wind direction and speed for the single loops and the average of all loops is represented in blue; (c to k) Average spatial distributions of all identified OA sources (panels c to f) and other measured components (panels g to k) in Tartu. The color scales represent enhancement over the background concentrations; the maximum of the color scales is fixed to the 75th percentile of the average enhancement of each component in panels g to k and to the highest 75th percentile among all OA sources in panels c to f. The sizes of the points represent the number of points that were averaged in each case.

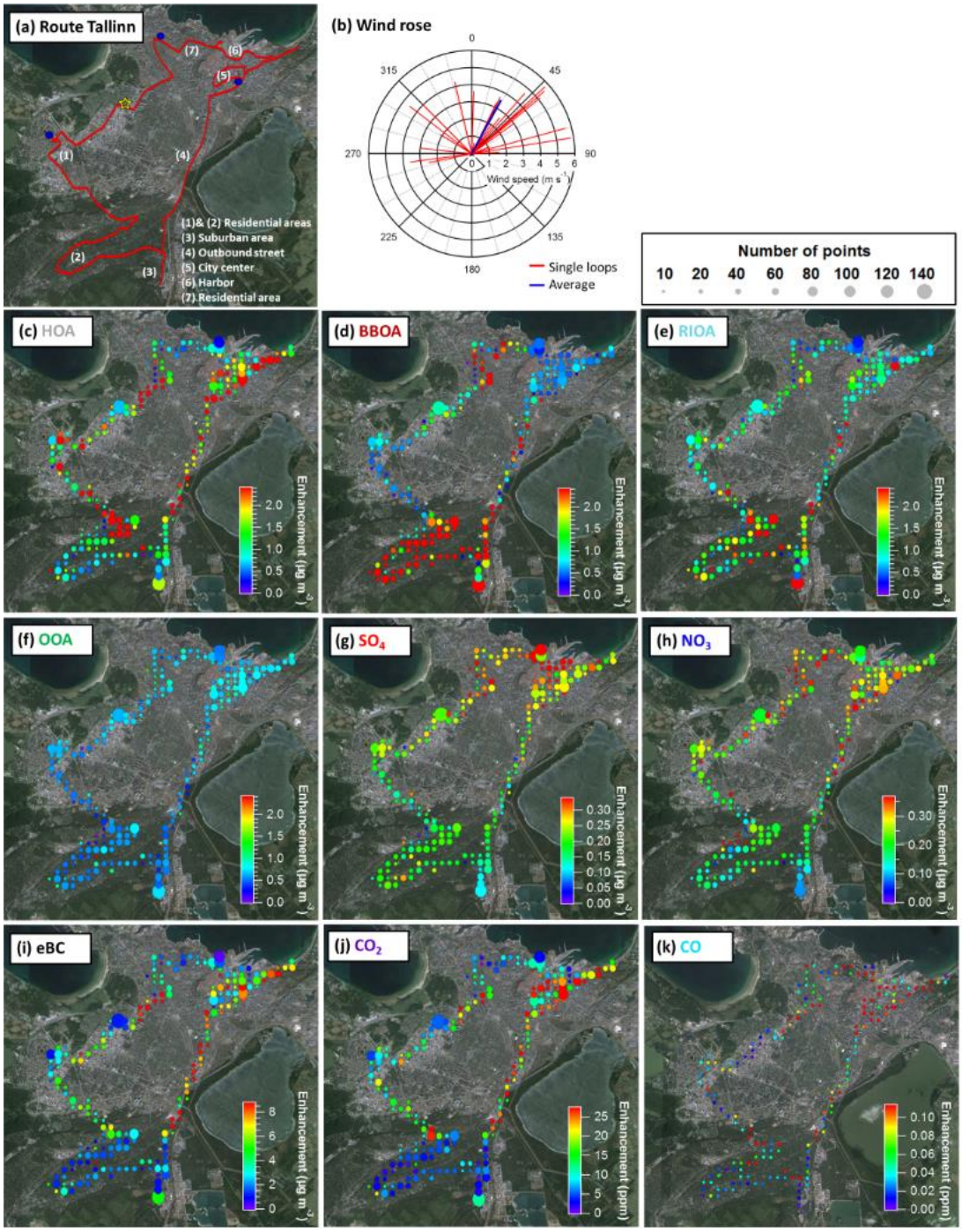


Figure 5 (Modified): (a) Driving route in Tartu: the red trace represents the GPS data, the yellow star the stationary measurements location and the blue dots the monitoring stations of the Estonian Environmental Research Institute (EERC); (b) Wind conditions during the mobile measurements in Tartu: red traces represent the wind direction and speed for the single loops and the average of all loops is represented in blue; (c to k) Average spatial distributions of all identified OA sources (panels c to f) and other measured components (panels g to k) in Tallinn. The color scales represent enhancement over the background concentrations; the maxima of the color scales have been fixed to the 75th percentile of the average enhancement of each component in panels g to k and to the highest 75th percentile among all OA sources in panels c to f. The sizes of the points represent the number of points that have been averaged in each case (Note: less data available for CO).

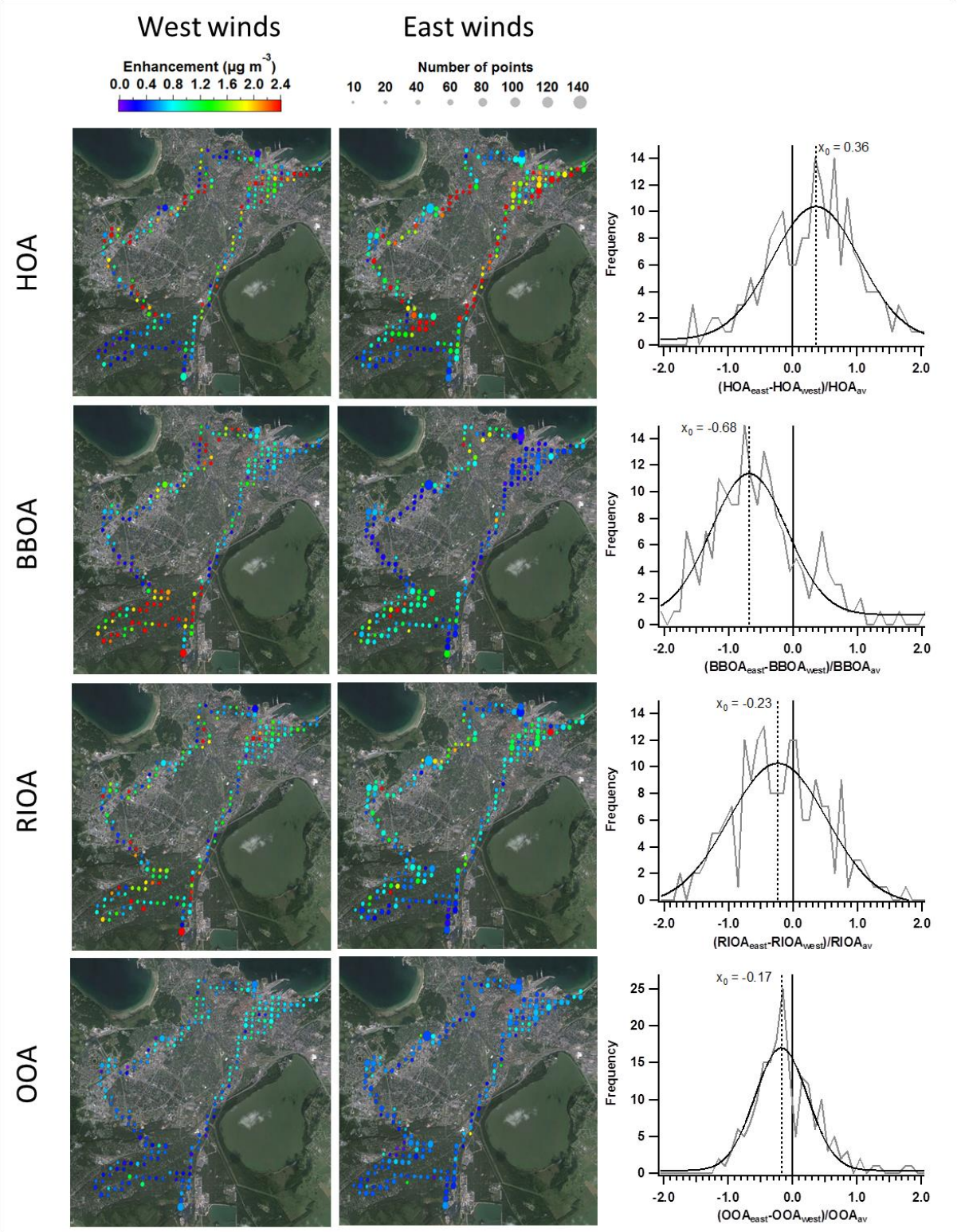


Figure S14 (New): Left: Average spatial distributions of the OA sources in Tallinn for west and east winds. The color scale represents the enhancement over the background concentrations and the size the number of points that have been averaged in each case. The data related to special events was excluded for these analyses. Right: Distribution of the normalized differences between the east- and west-related spatial distributions for each compound. X_0 indicates the center of the gauss function used to fit each distribution.

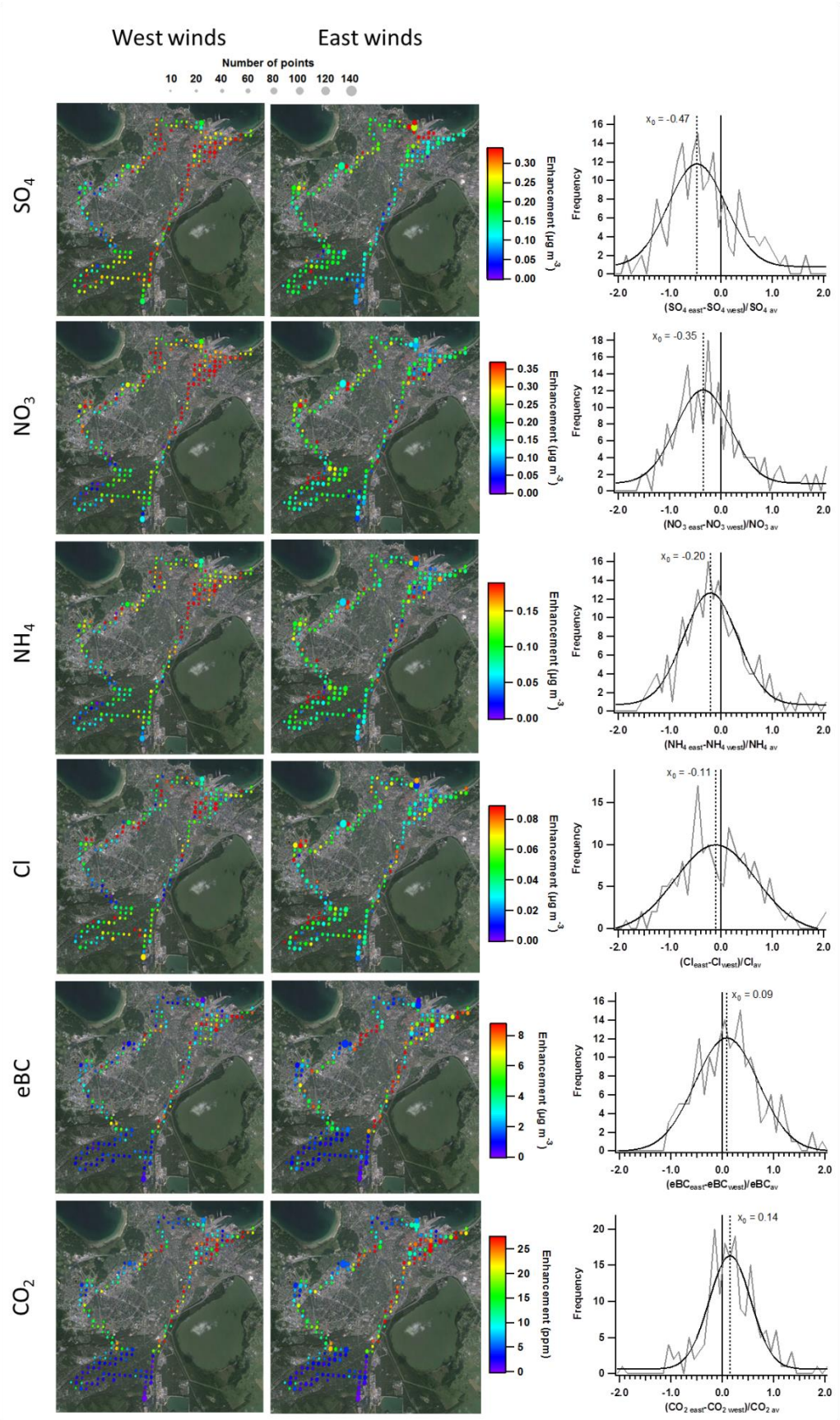


Figure S15 (New): Left: Average spatial distributions of the inorganic components (SO_4, NO_3, NH_4 and Cl), eBC and CO_2 in Tallinn for west and east wind conditions. The color scales represent the enhancement over the background concentrations and the size the

number of points that have been averaged in each case. The data related to special was excluded for these analyses. Right: Distribution of the normalized differences between the east- and west-related spatial distributions for each compound. X_0 indicates the center of the gauss function used to fit each distribution.

Page13, 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.*

Author's response: The hours between 7:00 to 19:00, LT and 19:00 to 7:00, LT are referred to as day-time and night-time respectively, while we consider evening hours when biomass burning contribution is highest during the hours between 15:00 to 21:00, LT. So, there is an overlap between evening and day-time hours. This has been clarified in the revised manuscript.

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.*

Author's response: We agree with the reviewer that the diurnal patterns would help in our analysis. However, such analyses were not possible with our data, as stationary measurements were performed mostly overnight and mobile measurements are strongly affected by point sources. Nevertheless, similar analyses were performed using mobile conditions only, upon averaging the data over longer time periods (i.e. 2h, see Fig. S13). The discussion in Lines 1-13 in Page 13 is derived from these analyses.

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

Author's response: We have deleted Fig. 1 and have added the driving routes for Tartu and Tallinn in Fig. 4 and 5 of the revised manuscript.

Minor Comments

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

Author's response: We thank the reviewer for the useful recommendations. The following suggestions have been introduced in the revised manuscript:

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 6, Line 1: *"… measurement method automatically corrects for the loading effect …"*
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…"*
Page7, 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…"*

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

Author's response: As mentioned in the text and described by Williams et al. (2013), the $PM_{2.5}$ lens efficiently transmits particles between 80 nm and up to at least 3 µm. The fact that particles larger than 3 µm could also be transmitted efficiently with this system is an important detail that needs to be considered in the presence of large particles. Therefore we prefer to keep this information in the text.

References:

Aiken, A. C., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D., Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner, E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L.: Mexico City aerosol analysis during MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) – Part 1: Fine particle composition and organic source apportionment, Atmos. Chem. Phys., 9, 6633-6653, 2009.

Crilley, L. R., Bloss, W. J., Yin, J., Beddows, D. C. S., Harrison, R. M., Allan, J. D., Young, D. E., Flynn, M., Williams, P., Zotter, P., Prevot, A. S. H., Heal, M. R., Barlow, J. F., Halios, C. H., Lee, J. D., Szidat, S., and Mohr, C.: Sources and contributions of wood smoke during winter in London: assessing local and regional influences, Atmos. Chem. Phys., 15, 3149–3171, 2015.

Crippa, M., El Haddad, I., Slowik, J. G., DeCarlo, P. F., Mohr, C., Heringa, M. F., Chirico, R., Marchand, N., Sciare, J., Baltensperger, U., and Prévôt A. S. H.: Identification of marine and continental aerosol sources in Paris using high resolution aerosol mass spectrometry, J. Geophys. Res., 118, 1950–1963, 2013b.

Favez, O., El Haddad, I., Piot, C., Boréave, A., Abidi, E., Marchand, N., Jaffrezo, J.-L., Besombes, J.-L., Personnaz, M.-B., Sciare, J., Wortham, H., George, C., and D'Anna, B.: Inter-comparison of source apportionment models for the estimation of wood burning aerosols during wintertime in an Alpine city (Grenoble, France), Atmos. Chem. Phys., 10, 5295–5314, 2010.

Herich, H., Hueglin, C., and Buchmann, B.: A 2.5 year's source apportionment study of black carbon from wood burning and fossil fuel combustion at urban and rural sites in Switzerland, Atmos. Meas. Tech., 4, 1409–1420, 2011.

Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U. and Prévôt, 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, 2012.

Paatero, P., Eberly, S., Brown, S. G., and Norris, G. A.: Methods for estimating uncertainty in factor analytic solutions, Atmos. Meas. Tech., 7, 781–797, 2014.

Sciare, J., d' Argouges, O., Sarda-Estève, R., Gaimoz, C., Dolgorouky, C., Bonnaire, N., Favez, O., Bonsang, B., and Gros, V.: Large contribution of water-insoluble secondary organic aerosols in the region of Paris (France) during wintertime, J. Geophys. Res. Atmos., 116, D22203, 2011.

Setyan, A., Zhang, Q., Merkel, M., Knighton, W. B., Sun, Y., Song, C., Shilling, J. E., Onasch, T. B., Herndon, S. C., Worsnop, D. R., Fast, J. D., Zaveri, R. A., Berg, L. K., Wiedensohler, A., Flowers, B. A., Dubey, M. K., and Subramanian, R.: Characterization of submicron particles influenced by mixed biogenic and anthropogenic emissions using high-resolution aerosol mass spectrometry: results from CARES, Atmos. Chem. Phys., 12, 8131-8156, 2012.