

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 #1

Received and published: 29 February 2016

This paper uses mobile measurements to spatially map aerosols and trace gases in two Estonian cities. The use of high time resolution instruments means that good spatial resolution is obtainable and the results are systematically analysed to present statistics on the urban increments. The use of source apportionment techniques on the AMS and Aethalometer data adds extra depth to the results. Overall, the analysis appears solid and the results are well presented. I recommend publication subject to minor (mostly technical) revisions.

Page 1, line 1: Please be more specific when referring to 'polluted continental areas'.

Changes in text:

Page 1, Line 30: A strong increase in the secondary organic and inorganic components was observed during periods with transport of air masses from [northern Germanypolluted continental areas](#), while the primary local emissions accumulated during periods with temperature inversions.

Page 2, line 18: *PM2.5 has been the major focus for over 20 years now, so can hardly be described as 'recent'.*

Author's response: We fully agree with the reviewer's comment and have removed "recently" in the revised manuscript.

Changes in text:

Page 2, Line 21: ~~Recently, m~~Major attention has been devoted to the study of the PM_{2.5} fraction (particulate matter with an aerodynamic equivalent diameter $d_{aero} \leq 2.5 \mu\text{m}$), which has been linked...

Page 6, line 20: *Remove the word 'highly'.*

Author's response: Removed in the revised manuscript.

Page 11: *More explanation and justification of the P05 method should be given. Why was the 5th percentile chosen? What specific effect was the subtraction expected to achieve? Given that the base of the sigmoid fits was allowed to vary, why is this even necessary?*

Author's response: This is a good point raised by the reviewer that needs additional explanation. The subtraction of P05 is needed in order to decrease the background variability of each single loop before averaging. This step could be skipped if the sigmoid fit could be applied to single loops, but this is not possible due to the high variability in the signal within each single loop. We tested the sensitivity of the method by subtracting P10 instead of P05, and no major changes were observed in the results. This information has been added in the revised manuscript.

Changes in text:

Page 14 Line 7: In most ~~of the~~ cases the base of the sigmoid functions is slightly above zero. This indicates that the ~~P05 previously~~ subtracted P05 didn't represent the full regional background, which is therefore given by the sum of the average P05 and the base of the sigmoid function. Note that the initial subtraction of P05 would not be necessary if the longitudinal profile of each single loop could be fitted. However, this is not possible due to the high concentration variability within each single loop. A sensitivity analysis was performed by using P10 instead of P05 and no major changes were observed in the final results.

Page 12, line 13: *The use of the median does not completely rule out the influence of the kerbside increment because street canyon effects (e.g. when the local wind is perpendicular to a road) can cause on-road emissions to persist in this microenvironment and the increment to no longer manifest itself as discrete spikes in the data. This would cause the median to increase over what would be expected of the urban background. Because these measurements are taken on-road, it is perhaps inevitable that estimates of the urban background will be biased slightly high because of the influence of traffic sources, so this should be added as a caveat. It may be possible to exclude this by selectively averaging the less-busy roads.*

Author's response: This has been discussed further in the revised manuscript.

Changes in text:

Page 13, Line 27: While the averaged profiles take into account the effects of the measured point sources in the urban area (mostly traffic and residential emissions), the use of the median profiles is expected to represent more selectively ~~exclude these effects, making the results more representative of~~ the urban background concentrations. We note that the influence of curbside increments may not be completely removed when using median increments (e.g. accumulation of traffic emissions due to street canyon effects), and therefore these increments might be biased high and should be regarded as our highest estimates of urban background concentrations.

Page 12, line 3: Replace 'component' with 'components'.

Author's response: Replaced in the revised manuscript

Page 15: *How was the temperature difference between 0 and 22m measured?*

Author's response: The temperature is measured at different heights in a meteorological tower at the Tallinn Zoo monitoring station.