We kindly thank the referee for taking our manuscript into consideration and we appreciate the comments raised on the manuscript. The referee's main concerns regard the reliability of the measurement technique and the novelty of the results, which we address in detail below.

The manuscript reports on a study on the aerosol composition in three sites in the London area by DRUM impactor & SXRF analysis. The text is extremely long and full of details, however I have major and basics concerns so that I cannot recommend it for publication.
The manuscript can be divided roughly in two parts: 1) technologies and methods, 2) results and discussion

Comment #1:

Part 1: this is potentially the more interesting part of the paper and actually it could/should re-arranged in a separate technical note (or similar...it is now 10-page long) since most of the details now given are likely not necessary in regular article focused on the experimental results (I mean: the joint use of DRUM & SXRF has been already introduced in previous papers).

Response:

We appreciate the referee's suggestion and we agree that some of the technical aspects in this paper are not directly needed to understand the results sections. We have considered submitting a separate technical note including the technical advancements regarding SR-XRF analysis (Sect. 2.2.1, points 1-4 on p. 15904-15906) and the data intercomparison between RDI and PM_{10} filters (Sect. 3, p. 15907-15910). However, as the referee points out, the current analysis is built on previous papers following a similar methodology (Bukowiecki et al., 2005, 2008, 2009c and Richard et al., 2010). Therefore, we believe that the advancements described in this paper represent an incremental improvement on an existing technique, rather than a reinvention of the method, and are thus best treated within the current paper. However, as noted in the response to Comment #2, the Methods section has been significantly shortened by moving the detailed discussion of the method intercomparison to the Supplement.

Comment #2:

On the other hand, this long discussion fails, in my opinion, in demonstrating the reliability of the adopted methodology and poses the discussion of the experimental results on a "icy ground" (the comparison vs. other techniques show a quite poor agreement).

Response:

The referee refers to the disagreement between measurement techniques in Section 3 (Data intercomparison) and specifically Fig. 2, and suggests that this calls into question the experimental results. However, much of this disagreement is expected, resulting from known differences in the measurement techniques (e.g. different size ranges) and thus does not reflect data quality. In addition, we note that this section should be interpreted as a method intercomparison rather than RDI-SR-XRF validation, and that the extent of the agreement is similar to other intercomparison studies of trace element measurement techniques. We have clarified these points by significantly condensing the intercomparison section of the manuscript, with Fig. 2 and the accompanying detailed discussion moved to the Supplement. An overview of the main points is provided below (please note that Cr has been removed from the intercomparison as this element was rejected from the filter analysis during the final quality assurance checks):

1. Most elements (i.e. all except those discussed below) show good agreement between RDI and PM_{10} filters within ± 50 % with good Pearson's R of > 0.78.

2. Quantitative agreement between RDI and PM_{10} filters should not be obtained for elements with significant mass below the RDI small-size cut off of 300 nm. This includes S, K, Sn and Pb. For S, further investigation is possible by adding the mass from the backup filter to the $PM_{1.0-0.3}$ mass measured by the RDI. Quantitative agreement with the AMS SO_4^{2-} data is then achieved, suggesting the RDI provides accurate $PM_{1.0-0.3}$ values for all these elements.
3. V, Ni and Mo are well-constrained in the RDI-SR-XRF analysis and are well above detection limits, but have low or unknown extraction efficiencies in the PM_{10} filter-ICP-MS analysis, increasing the uncertainty of the PM_{10} filters. Further, the RDI measurements of these elements are internally consistent (strong correlations with co-emitted elements). This suggests that the RDI measurements are correct, and the disagreement does not reflect RDI data quality issues.
4. RDI and filter measurements of Na and Mg are strongly correlated but disagree on the absolute magnitude. The RDI relative calibration of these elements is somewhat uncertain (around 13 %), while the filters have unknown extraction efficiency for Na (Mg is well extracted with 90 % efficiency). However, both techniques provide internally consistent results (e.g. correct Na-to-Mg ratios and sensible time series). Thus, while the absolute concentrations can be questioned, relative changes should be considered robust.

In conclusion, the intercomparison analysis suggests that the RDI provides robust measurements of nearly all trace elements within the $PM_{10-0.3}$ size range. The issues that do exist apply to absolute magnitudes, not relative changes. The analysis in this paper (e.g. urban/kerb increments and diurnal/weekly patterns) relies predominantly on these relative changes, and thus neither the analysis nor the main conclusions are undermined by method reliability.

Comment #3:

I have also to note that, despite of the length, two times the reader is forwarded to future papers which should complete the methodological section: this could be accepted in a letter but not in this case.

Response:

In addressing the comments #1 and #7 of Referee #2, we have included sufficient information on the analysis method for the reader to grasp the relevant issues. These references are thus no longer needed and have been removed.

Comment #4:

Neglecting the concerns at point 1), the section "results" is a extremely detailed list of "raw" data which are extremely valuable at local level but, in my view, not of general interest since they do not improve our knowledge of atmospheric aerosols.

Response:

We believe that the results sections in this paper are very valuable for the following reasons:

1. The dataset is unique: to our knowledge, no previous study achieves size-resolved measurements of trace elements with fast 2 h time resolution at kerbside, urban background, and rural sites simultaneously. We would in general like to make the remark that there are too few increment studies published for various pollutants. Such combined measurements allow a better assessment which components of the pollution are rather driven by regional, urban or very local pollution.

2. Trace element measurements are directly relevant to human health, both in terms of quantifying exposure to toxic metals and as chemical tracers for exposure to other emission sources. Real-world human exposure depends on the temporal characteristics of size-dependent particle concentrations in specific micro-environments. Accurate exposure assessments therefore require measurements of the unique type performed herein.
3. The present study in London can be conceived as a model study for the assessment of micro-environment (kerbside, urban background, rural) effects on trace element concentrations in cities around the world. This is poorly constrained in most locations, although existing results in Europe suggest important differences can exist depending on the local or regional environment. Such investigations are urgently needed for assessing public health risks and evaluating pollution mitigation strategies.

Comment #5:

A real Source Apportionment study is missing and the added-value of the very demanding DRUM+SXRF analysis remains only partially demonstrated (i.e. the possibility to catch transients and episodes with a high-resolution sampling has been introduced and discussed several times in previous papers on DRUM and other high-resolution samplers/impactors).

Response:

A source apportionment study on this data set is definitely interesting, and will be the subject of a future paper. The source apportionment analysis is complex by itself, as we basically deal with 9 datasets (3 sites and 3 size ranges). The inclusion of the source apportionment would definitely overload the paper. In the current study, we do by far not only demonstrate the possibility to catch transients, but we go much further. The present focus is on exploiting the measurement time resolution to investigate the detailed issues governing local trace element concentrations (e.g. wind direction, street canyons, regional air masses). These are only in a broad sense source-dependent; we feel it is an advantage to investigate these issues without the statistical blurring that inevitably occurs within factor analysis.

Comment #6:

I really don't find "the message" (or better the information) in this 18-page long text, which could and should be considered as a technical report in preparation of an article with a real and full source apportionment exercise.

Response:

In addressing this comment and the comment #6 of Referee #2 we have taken the following steps to condense the manuscript, mainly by rephrasing and repetition removing:

1. In line with the response to comment #2, the method intercomparison will be significantly shortened. We will move Fig. 2 and most of the associated discussion to the Supplement. Only a brief summery will remain in the main text.
2. Furthermore, the discussion about the local wind direction influence at kerbside, urban background and rural sites will be condensed. We are convinced that this analysis is interesting and important in understanding wind direction effects on pollution levels at different micro-environments. However, less detail is possible in this part of the paper and some repetition from the urban increment discussion regarding element grouping can be removed.
3. We will condense the discussion about the kerb increment by removing repetition regarding grouping of elements, already discussed in the urban increment and local wind direction influence discussions. We also condense the comparison of increment values to previous studies in this section.

4. We will condense the discussion about the three case studies regarding regional influences by rephrasing this section.
5. Finally, we will condense the discussion regarding daily/weekly cycles, specifically by removing repetition concerning reasons for enhanced element concentrations during rush hour due to increased braking processes.

We feel that the remainder of the results sections provide a clear overview of urban/kerb increments and diurnal/weekly cycles for the broad range of trace elements, which are needed to understand human exposure levels at multiple micro-environments as a function of size and time. We show strongly enhanced element concentrations at the kerbside, especially for coarse fraction particles up to a factor of 17 relative to urban background levels and being heavily affected by wind direction. All elements influenced by traffic, either by wearing processes or by resuspension exhibit elevated concentrations during rush hours and on weekdays compared to weekends. These occur predominantly at the kerbside but are also clearly observed at the urban background site, indicating largely enhanced health risks during these periods throughout a city.

References

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