

We kindly thank the referee for taking our manuscript into consideration and we value the comments raised to improve the manuscript. A point-to-point response to the issues raised is enclosed below.

The manuscript contains results obtained in an extensive study at urban and rural areas in London, UK. The analysis is focused (but no limited) to elemental contents in several fractions of atmospheric particulate matter. Although the general quality of the paper is good, it is necessary to discuss and improve several aspects, which I explain below.

Comment #1:

1. I am concerned about the agreement of the measurements using RDI and PM₁₀ filters, in particular for some elements. The question here is: what are the truly accurate values to be considered in the paper? This problem would not exist if certified reference materials had also been analyzed. Please, do not confuse this accuracy determination with the calibration process, which is thoroughly described in section 2.2.1. The main drawback is that the forthcoming discussions in the manuscript may not have a strong basis.

Response:

The referee refers to the disagreement between measurement techniques in Sect. 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 PM₁₀ filters within $\pm 50\%$ with good Pearson's R of > 0.78 .
2. Quantitative agreement between RDI and PM₁₀ 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 SO₄²⁻ 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 PM₁₀ filter-ICP-MS analysis, increasing the uncertainty of the PM₁₀ filter analyses. 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). We also note that Mg lies in an area of the XRF spectrum that is free of overlapping lines, resulting in

low fitting errors. Thus, while the absolute concentrations (i.e. the accuracy) differ by a factor of 2.5, relative changes (i.e. the precision) are considered to be 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 (precision), and thus neither the analysis nor the main conclusions are undermined by method reliability.

We agree that the use of certified reference materials would, in theory, be helpful (please note that the filter measurements are calibrated with NIST standards). However, these materials are typically available only in concentrations that are orders of magnitude higher than those attained with ambient sampling, as well as being mounted on a different foil or filter substrate. Both issues can significantly perturb XRF measurements, rendering these materials unsuitable for use. Meanwhile, after this manuscript went to press, we were able, for the first time, to obtain a set of certified single element standards in concentrations relevant to the ambient atmosphere on the same $6\ \mu m$ PP foil used for RDI sampling. XRF measurements were performed in a recent beamtime and are not directly transferable to beamtimes in which the current data samples were analysed (i.e. different geometry and irradiation conditions). Preliminary results indicate agreement within 25 % with no positive or negative bias between RDI results using the multi- and single-element standards for the elements Na, Ca, Fe, Co, Sr and Ba in the area concentration range $3-5\ \mu g\ cm^{-2}$ on the PP foil. The use of these single-element standards will not influence the current comparison between SR-XRF and PM_{10} filters since the deviations of XRF to PM_{10} filters and to single-element standards go in opposite directions from a 1-to-1 ratio.

Comment #2:

2. Some of the elements affected by the above situation are very important for studies related to elemental concentrations in aerosols. For example, S and K, which are valuable for tracing human activities and/or biomass burning, in page 15907, present doubtful values. Regarding those of Na and Mg, it is not clear to me which values should be taken as more accurate (XRF or ICP).

Response:

We agree with the referee that the precise measurement of S and K is of great help in understanding the distribution of these elements across larger geographical regions. As stated in the response to Comment #1 the quantitative disagreement between RDI and PM_{10} filters for S, K, Sn and Pb is expected based on differences in measured size ranges. The size distributions of S and K make it therefore unsurprising (and not alarming) that previous XRF studies (Bukowiecki et al., 2005 and Richard et al., 2010), to which the referee refers at p. 15907, did not achieve good agreement with PM_{10} filter techniques.

To clarify these points in the text we intend to make the following changes.

1. We will change the last sentence of the RDI description in Sect. 2.2.1 beginning at p. 15902 into:
“This results in sampling of a smaller fraction of $PM_{1.0}$ particles than previously expected and influences those elements that occur predominantly in this size range, notably S, K and Pb.”
2. We include the following sentence in Sect. 4.1 towards the end of the first paragraph to repeat the explanation of the mass underestimation in the results section:

“According to Supplement A and B, elements with considerable mass in the PM_{1.0} fraction (S, Pb, potentially K, Zn, Br, Sn) may be significantly underestimated due to a higher than expected small-end cut point, relative to analysis methods with a smaller cut point towards 0 nm (290-410 nm, rather than the previously estimated value of 100 nm, Bukowiecki et al., 2009c; Richard et al., 2010).”

The referee also points to the disagreement of Na and Mg between both analyses methods, which differ by a factor of 2.5. This is well outside the estimated uncertainty (~25 %) of the RDI-SR-XRF analysis for these elements (see Comment #7), and the reason for the discrepancy is not clear (see Comment #1). In the absence of additional independent measurements of Na and Mg, it is indeed difficult to determine which method provides accurate absolute values. However, we note that both methods provide internally consistent measurements of these elements as evidenced by Na-to-Mg ratios and sensible time series. The critical criterion for the analysis herein is therefore met, providing precise relative changes for each element.

Comment #3:

3. Although the authors made the evaluation of the concentrations using the DE data as a reference, there may be other procedures that provide information about the origin of the elements. A simple method is the use of the Enrichment Factor (EF). Using the figures given in Table 2, I calculated the EF for the elements Fe and K, usually associated to particles with a geological (soil) origin, using Si as reference element and average Earth crust composition. It is possible to find that in the three sites, the EF for Fe in the coarse fraction is very high (of the order of 10), which may agree with the hypothesis given by the authors relating Fe to brake wear. Moreover, I would expect the contribution of brake wear in the rural site to be very low, approximating EF to unity and suggest a soil origin. Instead, the EF is higher at the DE site than the urban one. The authors should try to explain this. EF for Fe in the finest fraction is closer to 1, associating it to a crustal origin. As for K, for the coarse fractions the EF is almost equal to 1. However, for the finest fraction in the three sites it is higher than unity, in particular for DE, where the value is almost 20, showing it has a different origin than soil. I am including a table with the EF (with the last three columns giving the average EF for each site). It may be advisable to calculate the EF for the other elements, not necessarily to include in the manuscript, but as a guide to better understand the origin of each element.

Response:

We greatly appreciate this extended comment on the Enrichment Factor (EF) and the efforts that went into creating the table in the supplement to this comment. We will incorporate EF analysis into the paper by introducing it in Sect. 4.2.1 ‘Urban increment’. We will compare EF values of several elements that are grouped together in traffic-related and mineral dust groups based on both urban and kerb increment results.

We stress that urban/kerb increment calculations specifically relate concentrations of multiple sites to each other per element, which we believe is very valuable in exploring differences in human exposure at these sites. With EF, each element is related to Si as an indication for anthropogenic or soil origins. A comparison of the EF per element at multiple sites is probably as robust as the current increment calculations, but thus provides different information. The absolute magnitudes of EF are less meaningful since individual element concentrations are biased by the RDI small-size cut off of 300 nm and differences in uncertainties regarding absolute and relative calibrations.

Although we are not able to reproduce the exact EF values as given by the referee, we have calculated the EF for our data based on the upper continental crust composition as given by Wedepohl (1995).

For Fe we obtain EF of close to 40 for the kerbside, 20 for the urban background and 12 for the rural site, independent of size fraction. These results clearly indicate anthropogenic influences of Fe to all sites. The size independency can be understood when realizing that both Si and Fe are reduced by the same factor from the coarse to the fine mode, resulting in the same EF for each size range. The EF of 12 at the rural site is indeed high, but the concentration of Fe has decreased by a factor of 16 compared to 6 for Si between the kerbside and rural site. This strong decrease in Fe relative to Si is in line with an expected strongly reduced anthropogenic influence of mainly traffic at the rural site, and thus a larger soil fraction of Fe.

The EF of K is indeed highest in the fine fraction with values of 14, 26 and 28 at the kerbside, urban background and rural site, respectively, and indicates anthropogenic influence. We group fine fraction K as regionally influenced. The increased EF towards the rural site likely indicates stronger influences of biomass burning relative to the city sites.

Incorporating EF analysis will lead to the following paragraph on p. 15915 of Sect. 4.2.1:

“We group elements together based on similar urban increment values as an indication for co-emission. Mn, Fe, Cu, Zn, Zr, Mo, Sn, Sb and Ba show urban increments on average of 3.5 in the coarse, 3.1 in the intermediate and 2.0 in the fine fraction (Fig. 4). These have been identified as traffic-related elements by e.g. Amato et al., 2011; Bukowiecki et al., 2010; Minguillón et al., 2014; Richard et al., 2011 and Viana et al., 2008. Zr can also have a mineral dust origin (Moreno et al., 2013) as can be seen by the use of Enrichment Factors (EF). EF is a measure of the enrichment of elements relative to the upper continental crust (UCC) and is defined as ppm metal in sample / ppm metal in UCC with Si as reference material (UCC from Wedepohl, 1995). Zr is the only element in this traffic group that is depleted in the atmosphere relative to their UCC concentrations, but with concentrations at NK higher than at DE. Most other elements clearly indicate anthropogenic origin with $EF > 10$. The urban increments are similar to that of NO_x , where concentrations at NK were on average a factor 4.9 higher than at DE (mean concentration at NK was 68 ppb, at DE 14 ppb). Black carbon (BC), a marker for both traffic and wood burning emissions, had an urban increment of only 1.1 (concentration at NK 757 ng m^{-3} , at DE 633 ng m^{-3}), likely due to local wood burning emissions around DE (Mohr et al., 2013). Al, Si, Ca, Ti and Sr as markers for mineral dust (e.g. Amato et al., 2009; Lin et al., 2005; Lucarelli et al., 2000) show a factor 2.0 higher concentrations at NK relative to DE in the coarse, 1.9 in the intermediate and 1.6 in the fine fraction ($EF < 10$). These results indicate that moving from rural to urban backgrounds yield a larger relative increase in traffic than in mineral dust elements. Surprisingly, sea salt elements (Na, Mg, Cl) show higher concentrations at NK than at DE of up to a factor of 2 for the coarse mode, despite the expected dominance of regional over local sources. This highlights the potential importance of sea or road salt resuspension by traffic. Similar urban increment values for traffic-related, resuspension and sea salt elements have been observed by Lee et al. (1994) for particles below a few μm . Theodosi et al. (2011) also found higher increments (> 2) for trace elements in PM_{10} aerosol from local anthropogenic sources like fossil fuel combustion (V, Ni, Cd) and traffic (Cu), relative to long range transported Saharan dust (Fe, Mn) with increments close to 1. However, our study suggests that the non-size-resolved increment values reported in the cited studies do not fully capture the urban/rural differences.”

Comments #4 and #5:

4. *In the conclusions, Fe is initially associated to traffic, then it is excluded from the list of brake wear related elements. In the text, however, it was suggested to be produced by brake wear and even a reference is given. Please clarify this point.*

5. *In several cases, the association among the elements does not look strongly justified. For example, why V and Ni are related to traffic? In many studies they are related to industrial*

sources (fuel oil burning and they are strongly correlated to S). Also, Zr is often linked to soil-derived particles in other papers.

Response:

There might be some misunderstanding about the grouping of elements we use in the paper. We note that we empirically group elements according to their similarities in urban/kerb increments and diurnal/weekly cycles, rather than that we associate them to specific emission sources. With other analysis methods, such as the Enrichment Factor (EF) as proposed in Comment #3, it might be possible to find another distinction between groups of elements related to specific emission sources.

We agree that the group of elements labelled as traffic-related in the kerb increment analysis contains also elements potentially emitted by other sources, such as V and Ni mainly from industrial sources and heavy-oil combustion. We realize that this label is confusing and will change it into anthropogenically-influenced (ANTH). This label is introduced, together with a source justification of all elements in this group, in Sect. 4.2.2 'Kerb increment' in the 2nd paragraph on p. 15919:

“The second group consists of V, Cr, Mn, Fe, Ni, Zn and Pb with increments of 5.7-8.2 (PM_{10-2.5}) in the SW sector (2.6-3.0 for NE) (Fig. 6). V and Ni are typically assigned to industrial sources and heavy-oil combustion (e.g. Mazzei et al., 2007; Viana et al., 2008), Zn is usually associated with tire wear (e.g. Harrison et al., 2012b; Lin et al., 2005), and the other elements are commonly associated with traffic-related emissions (e.g. Amato et al., 2013; Bukowiecki et al., 2009a; Richard et al., 2011). We empirically label this group as anthropogenically-influenced (ANTH). The EF of V and Ni are much lower than those of the other elements in this group (2 vs. > 10), indicating at least to some extent different source origins. These kerb increments are similar to the ones for NO_x of 8.5 for SW and 2.4 for NE, confirming the anthropogenic influence (traffic and other sources) on these elements. The high braking frequency at MR due to congested traffic probably resulted in increased kerb increments of brake wear relative to ANTH elements that are also influenced by local traffic and other sources around NK. Increments of these ANTH elements are higher than previously reported values of 1.8-4.5 for studies with low time resolution and non-size segregated particles (Boogaard et al., 2011; Janssen et al., 1997). The high increments presented here might be caused by street canyon effects, trapping pollutants emitted at street level and preventing dilution to the urban background. The enhanced kerb increments for brake wear relative to ANTH elements are apparent in all three size fractions, although increments become more similar towards smaller sizes with a factor 1.7 between both element groups in the coarse, 1.5 in the intermediate and 1.4 in the fine mode. Both groups show the additional information gained with size-segregated aerosol, where exposure to trace elements in the street canyon relative to the urban background increases with particle size, either caused by increased traffic-related emissions with particle size or by more efficient transport of submicron particles from street sites to the urban background. Furthermore, the highly time-resolved element measurements presented here enabled us to resolve the systematic, wind direction dependent variability in kerb increments.”

The referee points out that Zr is often linked to soil-derived particles, whereas we group it with elements typically related to brake wear based on its urban increment. Both origins have been proposed in previous literature. In line with the response to Comment #3, Zr shows a different EF than the other elements in this group, potentially indicating a different source origin.

In the urban increment analysis no distinction was observed for brake wear and other traffic-related elements, and therefore just formed 1 group. In the analysis of the kerb increment higher values were observed for brake wear (Cu, Zr, Mo, Sn, Sb and Ba) than for other traffic-related (V, Cr, Mn, Fe, Ni, Zn and Pb) elements, and were treated as two different groups. Fe falls in

this second category with lower kerb increment values. Some studies (e.g. Harrison et al., 2012b) assign Fe to brake wear, but we assign Fe to the traffic-related group purely based on its lower kerb increment value.

Following the discussion above, we believe the label of the traffic-related group is confusing and this group of elements will be relabelled as anthropogenically-influenced (ANTH). The paper will be changed accordingly.

We will rephrase the 2nd paragraph of the Conclusions at p. 15927-15928 according to the new labelling of the groups:

“Urban and kerb increments were defined as the concentration ratios of urban background to rural, and kerbside to urban background, respectively, and the kerb increments were further explored as a function of wind direction. The group with the largest kerb increments consisted of elements typically associated with brake wear (Cu, Zr, Mo, Sn, Sb, Ba). The second largest increments were formed by anthropogenically-influenced elements typically assigned to non-brake wear traffic emissions (Cr, Mn, Fe, Zn, Pb) but also by elements like V and Ni. This could indicate either a traffic source for these elements or a similar kerbside-to-urban emission gradient. Kerb increments were larger for the brake wear group and under SW winds due to local street canyon effects, with coarse fraction increments between 10.4 and 16.6 for SW winds (3.3-6.9 for NE winds) against increments for the anthropogenically-influenced group between 5.7 and 8.2 for SW winds (2.6-3.0 for NE winds). The kerb increments for all these elements in the PM_{10-2.5} size fraction are roughly 2 times that of the PM_{1.0-0.3} fraction. Urban increments (no distinction between both groups) were around 3.0. In addition to direct emissions, traffic-related processes influence the concentrations of other elements by resuspension, with mineral dust (Al, Si, Ca, Ti, Sr) increments of 1.3-3.3.”

Comment #6:

6. The paper is very long, and maybe some parts may be reduced, as the comparison with other techniques.

Response:

We appreciate the referee’s suggestion and have taken the following steps to condense the manuscript:

1. As suggested by the referee, the method intercomparison will be significantly shortened. We will move Fig. 2 and most of the associated discussion to the Supplement. Only a brief summary 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.
6. We also note that the Introduction was condensed at the “technical corrections” stage prior to publication in ACPD by reducing the detail in which trace element measurements

across Europe are discussed, and by shortening the introduction of the concept of urban micro-environments.

We feel that the remainder of the paper presents our results with an appropriate level of detail and explanation regarding the discussion of concentration levels of 25 elements in 3 size fractions at 3 sites as a function of increments, wind direction and time.

Comment #7:

7. Finally, a major problem in the entire manuscript is that there is no mention to experimental uncertainties, except for a few elements (Na, Mg or Mo), but there is not a careful explanation on how they were estimated. It is mandatory to present uncertainties in every experimental/laboratory work.

Response:

We agree that presenting uncertainties of this type of work is essential. During the “technical corrections” stage we added a paragraph at the end of Section 4.1 ‘Trace element concentrations’ giving an overview of the main sources of uncertainty in our study. We will extend this in the revised manuscript and add a table to the Supplement reporting element-specific detection limits and size-dependent element uncertainties.

The following text summarizes the major sources of experimental uncertainty:

“The RDI-SR-XRF technique is subject to various sources of uncertainty, which change in importance depending on whether the data are described in terms of absolute/fractional concentrations (as above) or in terms of relative changes/ratios (as in the remainder of the manuscript). A brief overview is presented here:

1. RDI sampling: the fluctuations in the flow rate are negligible within 5 % (Richard et al., 2010) and the uncertainties in the size cut off are discussed in Supplement A.
2. SR-XRF accuracy: uncertainties in the absolute and relative calibrations affect absolute/fractional concentrations, but cancel out for relative changes/ratios, because all samples were measured under the same calibration conditions.
3. Issues such as imperfect flatness of the sample foils and detector dead time corrections (Richard et al., 2010) reduce measurement precision but affect all elements with the same scaling factor.
4. SR-XRF measurement precision is affected by sample inhomogeneity and spectral analysis uncertainties. Sample inhomogeneity was assessed by Bukowiecki et al. (2009c) and found to contribute ± 20 % uncertainty.

For most elements, except Mn, sample inhomogeneity is the largest source of uncertainty. Mn is affected by spectral analysis uncertainties due to peak overlap with Fe, present in much higher concentrations. Therefore, a small change in the energy calibration as function of detector channel leads to a large change in the peak area of Mn. All data points lie well above their element detection limits, resulting in negligible uncertainties from the signal strength. Table S3 in Supplement B provides an estimate of the total uncertainty per size fraction and detection limits for each element. In addition, RDI-SR-XRF measurements (both absolute/fractional and relative/ratio) are affected by atmospheric variability. This variability is likely the predominant source of the data spread evident in Table 2 and the following analyses.”

The following table (in the Supplement) lists estimated total uncertainties per size fraction and detection limits for each element. Note that the sample inhomogeneity dominates the uncertainties for most elements.

Table S3. Estimated total uncertainty (% of measured value) of the calculated element concentrations per size fraction, and detection limits for each element (ng m^{-3}).

Element	PM _{10-2.5} (%) ^a	PM _{2.5-1.0} (%) ^a	PM _{1.0-0.3} (%) ^a	DL (ng m^{-3}) ^b
Na ^c	25	25	25	2.552
Mg	24	24	24	0.962
Al	24	25	25	1.709
Si	24	24	24	0.420
P	25	25	25	0.118
S	24	24	24	0.503
Cl	24	24	24	0.158
K	24	24	24	0.031
Ca	21	21	21	0.267
Ti	24	26	27	0.024
V	30	30	24	0.008
Cr	27	27	26	0.015
Mn	83	69	46	0.042
Fe	21	21	21	0.033
Ni	22	22	21	0.005
Cu	21	21	21	0.028
Zn	21	21	21	0.058
Br	21	21	21	0.117
Sr	21	21	21	0.036
Zr	21	21	21	0.036
Mo	21	21	21	0.037
Sn	21	21	21	0.061
Sb	21	21	21	0.052
Ba	21	21	21	0.254
Pb	21	21	21	0.137

^a Combination of uncertainties regarding sample inhomogeneity (20 %), RDI flow rate (5 %), absolute and relative calibration (Na-K 13 %, Ca-Pb 2 %) and spectral analysis specific per element and size fraction (median uncertainties for all data points).

^b As 3x the standard deviation of the spectra signals used for continuum corrections.

^c Na uncertainties might be underestimated due to the overlap with the L lines of Ni, Cu and Zn. In the current analysis the ratio of the L α to K α lines are determined empirically, and quantification of the associated uncertainties is under investigation.

In the Supplement regarding the data intercomparison the first paragraph will introduce the intercomparison and this table, together with a more detailed explanation of the sources of uncertainty in this study. The table will serve as a guide to interpret the comparison between RDI and PM₁₀ filter data, as described in the rest of this Supplement.

References

Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Perez, N., and Hopke, P. K.: Quantifying road dust resuspension in urban environment by Multilinear Engine: A comparison with PMF2, *Atmos. Env.*, 43, 2770-2780, 2009.

Amato, F., Viana, M., Richard, A., Furger, M., Prevot, A. S. H., Nava, S., Lucarelli, F., Bukowiecki, N., Alastuey, A., Reche, C., Moreno, T., Pandolfi, M., Pey, J., and Querol, X.: Size and time-resolved roadside enrichment of atmospheric particulate pollutants, *Atmos. Chem. Phys.*, 11, 2917-2931, 2011.

Bukowiecki, N., Hill, M., Gehrig, R., Zwicky, C. N., Lienemann, P., Hegedus, F., Falkenberg, G., Weingartner, E., and Baltensperger, U.: Trace metals in ambient air: Hourly size-segregated mass concentrations determined by synchrotron-XRF, *Environ. Sci. Technol.*, 39, 5754-5762, 2005.

Bukowiecki, N., Richard, A., Furger, M., Weingartner, E., Aguirre, M., Huthwelker, T., Lienemann, P., Gehrig, R., and Baltensperger, U.: Deposition uniformity and particle size distribution of ambient aerosol collected with a rotating drum impactor, *Aerosol Sci. Technol.*, 43, 891-901, 2009c.

Bukowiecki, N., Lienemann, P., Hill, M., Furger, M., Richard, A., Amato, F., Prevot, A. S. H., Baltensperger, U., Buchmann, B., and Gehrig, R.: PM₁₀ emission factors for non-exhaust particles generated by road traffic in an urban street canyon and along a freeway in Switzerland, *Atmos. Env.*, 44, 2330-2340, 2010.

Lee, D. S., Garland, J. A., and Fox, A. A.: Atmospheric concentrations of trace elements in urban areas of the United Kingdom, *Atmos. Env.*, 28, 2691-2713, 1994.

Lin, C. C., Chen, S. J., Huang, K. L., Hwang, W. I., Chang-Chien, G. P., and Lin, W. Y.: Characteristics of metals in nano/ultrafine/fine/coarse particles collected beside a heavily trafficked road, *Environ. Sci. Technol.*, 39, 8113-8122, 2005.

Lucarelli, F., Mando, P. A., Nava, S., Valerio, M., Prati, P., and Zucchiatti, A.: Elemental composition of urban aerosol collected in Florence, Italy, *Environ. Monit. Assess.*, 65, 165-173, 2000.

Minguillón, M. C., Cirach, M., Hoek, G., Brunekreef, B., Tsai, M., de Hoogh, K., Jedynska, A., Kooter, I. M., Nieuwenhuijsen, M., and Querol, X.: Spatial variability of trace elements and sources for improved exposure assessment in Barcelona, *Atmos. Env.*, 89, 268-281, 2014.

Mohr, C., Lopez-Hilfiker, F. D., Zotter, P., Prévôt, A. S. H., Xu, L., Ng, N. L., Herndon, S. C., Williams, L. R., Franklin, J. P., Zahniser, M. S., Worsnop, D. R., Knighton, W. B., Aiken, A. C., Gorkowski, K. J., Dubey, M. K., Allan, J. D., and Thornton, J. A.: Contribution of nitrated phenols to wood burning brown carbon light absorption in Detling, United Kingdom during winter time, *Environ. Sci. Technol.*, 47, 6316-6324, 10.1021/es400683v, 2013.

Moreno, T., Karanasiou, A., Amato, F., Lucarelli, F., Nava, S., Calzolari, G., Chiari, M., Coz, E., Artinano, B., Lumberras, J., Borge, R., Boldo, E., Linares, C., Alastuey, A., Querol, X., and Gibbons, W.: Daily and hourly sourcing of metallic and mineral dust in urban air contaminated by traffic and coal-burning emissions, *Atmos. Env.*, 68, 33-44, 10.1016/j.atmosenv.2012.11.037, 2013.

Richard, A., Bukowiecki, N., Lienemann, P., Furger, M., Fierz, M., Minguillon, M. C., Weideli, B., Figi, R., Flechsig, U., Appel, K., Prevot, A. S. H., and Baltensperger, U.: Quantitative sampling and analysis of trace elements in atmospheric aerosols: impactor characterization and synchrotron-XRF mass calibration, *Atmos. Meas. Tech.*, 3, 1473-1485, 2010.

Richard, A., Gianini, M. F. D., Mohr, C., Furger, M., Bukowiecki, N., Minguillon, M. C., Lienemann, P., Flechsig, U., Appel, K., DeCarlo, P. F., Heringa, M. F., Chirico, R., Baltensperger, U., and Prevot, A. S. H.: Source apportionment of size and time resolved trace elements and organic aerosols from an urban courtyard site in Switzerland, *Atmos. Chem. Phys.*, 11, 8945-8963, 2011.

Theodosi, C., Grivas, G., Zarpas, P., Chaloulakou, A., and Mihalopoulos, N.: Mass and chemical composition of size-segregated aerosols (PM₁, PM_{2.5}, PM₁₀) over Athens, Greece: local versus regional sources, *Atmos. Chem. Phys.*, 11, 11895-11911, 10.5194/acp-11-11895-2011, 2011.

Viana, M., Kuhlbusch, T. A. J., Querol, X., Alastuey, A., Harrison, R. M., Hopke, P. K., Winiwarter, W., Vallius, A., Szidat, S., Prevot, A. S. H., Hueglin, C., Bloemen, H., Wahlin, P.,

Vecchi, R., Miranda, A. I., Kasper-Giebl, A., Maenhaut, W., and Hitzenberger, R.: Source apportionment of particulate matter in Europe: A review of methods and results, *J. Aerosol Sci.*, 39, 827-849, 10.1016/j.jaerosci.2008.05.007, 2008.

Wedepohl, K.: The composition of the continental crust, *Geochim. Cosmochim. Acta*, 59, 1217-1232, doi:[http://dx.doi.org/10.1016/0016-7037\(95\)00038-2](http://dx.doi.org/10.1016/0016-7037(95)00038-2), 1995.