

## **Response to the comments of anonymous referee #2**

We thank the referee for handling our paper carefully and for providing valuable comments. The corrections were implemented in the main text and can be distinguished with the “track changes” tool of MS-Word. We addressed all the comments (in italic typeset) and prepared a point-to-point response (in regular typeset). Changes to the manuscript are indicated in **blue font**. Please note that the line numbers are with reference to the submitted manuscript and not the revised manuscript.

*The authors present highly time-resolved measurements of size-fractionated elements in four cities in Asia and Europe. The high time resolution and size-segregated elemental dataset are indeed a powerful tool to assess aerosol composition, sources, health effects in complex urban environments. However, this kind of studies are not widespread in the literature. The full source apportionment was already presented in other articles, but the authors present an interesting and simple approach for the analysis of the dataset which allows a first characterization of the major sources, site-to-site similarities or differences and the identification of key information required for efficient policy development. Therefore I suggest it for publication after minor revisions:*

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.

### **Comment #1**

*L. 38: add concentration before above.*

Done.

### **Comment #2**

*L. 77: The sampling period in Krakow is a little different respect to the one in the other three sites; it should be taken into account.*

We have added the following text at L. 76:

**It is important to notice that the sampling period in Krakow is different from the rest of the sites.**

### **Comment #3**

*L. 130-132: Such a low EF for Si in all the sites is quite strange. Differences in the soil composition relative to the assumed values for the continental crust in all the sites does not seem to be a reasonable explanation. XRF is known not to be the best analytical technique to detect low Z-elements like Al or Si; probably Si is under-estimated by the instrument. The authors should add some comments.*

We appreciate the concern raised by Referee#2. We have modified L. 130-132 as follows:

The unexpectedly low EFs observed for Si (0.41–0.45) and compared to previous studies (Majewski and Rogula-Kozłowska, 2016; Tao et al., 2013), are likely due to self-attenuation issues in XRF analysis for lighter elements (atomic number < 19), which may cause underestimation in their concentrations (Maenhaut et al., 2011; Visser et al., 2015). Therefore, the measurements of Al and Si from Xact need to be treated with caution. However, low EFs for Si is also probably due to crust-air fractionation in the wind-blown generation of crustal aerosol particles (Rahn, 1976).

Maenhaut, W., Raes, N., and Wang, W.: Analysis of atmospheric aerosols by particle induced X-ray emission, instrumental neutron activation analysis, and ion chromatography, Nucl. Inst. Meth. Phys. Res. Sect. B: Beam Interact. Mater. Atoms., 269, 2693–2698, <https://doi.org/10.1016/j.nimb.2011.08.012>, 2011.

Visser, S., Slowik, J. G., Furger, M., Zotter, P., Bukowiecki, N., Dressler, R., Flechsig, U., Appel, K., Green, D. C., Tremper, A. H., Young, D. E., Williams, P. I., Allan, J. D., Herndon, S. C., Williams, L. R., Mohr, C., Xu, L., Ng, N. L., Detournay, A., Barlow, J. F., Halios, C. H., Fleming, Z. L., Baltensperger, U., and Prévôt, A. S. H.: Kerb and urban increment of highly time-resolved trace elements in PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1.0</sub> winter aerosol in London during ClearLo 2012, Atmos. Chem. Phys., 15, 2367–2386, <https://doi.org/10.5194/acp-15-2367-2015>, 2015.

Rahn, K. A.: Silicon and aluminum in atmospheric aerosols: crust air fractionation?, Atmos. Environ., 10, 597–601, [https://doi.org/10.1016/0004-6981\(76\)90044-5](https://doi.org/10.1016/0004-6981(76)90044-5), 1976.

#### **Comment #4**

*L. 141: This is not true for Krakow, see comment above.*

We have modified L. 141 as follows:

Although the measurement periods do not overlap, they were all performed during the colder months of the year ([partially true for Krakow](#), see Section 2.1), and characteristic features of each site are evident.

#### **Comment #5**

*L. 181: "Si is selected as the Group 1 element", pleas add typical or representative element.*

Done.

#### **Comment #6**

*Conclusions: I think the information reported here are interesting, but the authors should stress the importance of a complete source apportionment to obtain a quantitative apportionment of the different sources.*

We have added the following text on L. 315:

Although the method proposed in this work allows for a comparison of the characteristics in different cities, a full SA analysis is necessary if more quantitative information (e.g. source contributions) is desired.