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Authors: Magdalena Reizer, Giulia Calzolai, Katarzyna Maciejewska, José A. G. Orza, Luca Carraresi, Franco Lucarelli, Katarzyna Juda-Rezler

Title: Measurement report: Receptor modelling for source identification of urban fine and coarse particulate matter using hourly elemental composition

We would like to thank the Anonymous Reviewer #2 for the assessment of our manuscript and for sound and constructive comments. The authors appreciate a lot the work that Reviewer put to help us in improving our paper. We took into account comments and suggestions of the Reviewer, and performed revision of the manuscript, trying to clarify all issues. The Reviewer's comments are in italics; our responses are in dark blue.

Response to the comments of Anonymous Referee #2 (21 May 2021)

The manuscript presents a study to identify the sources of fine and coarse particulate matter based on high resolution sampling of elemental components in the city of Warsaw. The sample collection and the data treatment were carried out according to state-of-the art methodologies. The manuscript is well organized and complete. The figures are of good quality and summarize the data properly. The discussion of some of the sources needs to be improved as pointed out in the specific comments. The text should be revised by a native speaker.

Thank you for this comment. The manuscript will be checked by English native speaker before final submission.

Specific comments

Line 81: the year should be between parentheses

It has been corrected.

Line 141: Maybe Belis et al., 2020 (https://doi.org/10.1016/j.aeaoa.2019.100053) is a more appropriate citation for this paragraph

The reference Belis et al. (2019) has been replaced by Belis et al. (2020).

Line 171: The authors should discuss what's the impact of the absence of such important components on the ability of the analysis to quantify sources

This paragraph has been modified as follows (changes to the manuscript are indicated in red font):

"In this study, concentrations of PM mass and its macro components, i.e., organic carbon, elemental carbon, secondary inorganic aerosols and other water-soluble inorganic ions, were not available with the hourly temporal resolution. Therefore, the analysis of the hourly concentrations of the elements have been used for the detailed identification of PM sources supplementing the previous source apportionment study performed by Juda-Rezler at al. (2020). Due to the lack of PM mass in the measurement campaign, the contribution of the identified sources to the total aerosol mass cannot be provided and the source time series will be expressed in arbitrary units (see e.g., Lucarelli et al., 2020)".

Line 222: Please, discuss if the soluble ions are comparable with the elemental determinations

The following discussion has been added to the text:

"Simultaneously, to the streaker data, daily concentrations of $PM_{2.5}$ and its components were collected. The daily elemental concentrations (averaged from the hourly data) and daily concentrations of the respective water-soluble ions (SO_4^{2-}, K^+, Cl^-, Na^+) was compared. A general good agreement was found, with a strong correlation between elemental sulfur and SO_4^{2-} (r = 0.97), as well as between elemental and water-soluble potassium (r = 0.74). For elemental and water-soluble chlorine and sodium moderate correlations (r = 0.69 and r = 0.61, respectively) was observed. Missing data and data below the detection limit did not allow the comparison of the concentrations of calcium and magnesium."

Lines 220 – 228: This paragraph should go to materials and methods

The paragraph has been moved to "Materials and method" to the chapter 2.2 ("PM sampling and elements determination").

Lines 249-251: This paragraph should go to materials and methods

The paragraph has been moved to "Materials and method" to the chapter 2.1 ("Study area").

Line 369: The bioavailability issue should be explained/discussed more in detail

The definition of the bioavailability has been added to the text (changes to the manuscript are indicated in red font):

"It is noteworthy that in wintertime in Warsaw, As and K appeared to be highly bioavailable, indicating the high degree and rate of absorption of a substance by the living systems or the high degree which the substance is available to physiologically active sites. During the days of elevated air pollution levels higher bioavailability of those element was observed, suggesting a higher risk to humans posed by emission from this source during those days (Juda-Rezler et al., 2021)."

Line 396: Although sulfate is a common component of biomass burning profiles, claiming this fuel is a major contributor to secondary sulfate should be better supported by evidence. Especially in an area where coal combustion, a well-known SO2 source, is well documented.

This paragraph has been modified as follows (changes to the manuscript are indicated in red font):

"This confirms that the fine fraction is dominated by regional rather than local transport of SO_2 and sulfate from Warsaw's outskirts with individual residential heating, mainly with the use of coal and biomass, whereas in the coarse fraction the presence of sulfate is probably due to the local combustion activities in the city itself."

Line 494: Consider calling this source mixed wood and coal combustion

Thank you for this comment. We agree, the name of the source has been changed to "Wood and coal combustion".

Line 568: The aged sea salt contribution may be higher than the authors' estimations considering that what is identified as "road salt" has the highest shares in the three clusters originated in the North Atlantic

This paragraph has been modified as follows (changes to the manuscript are indicated in red font):

"This may suggest that although road salt is the main source of atmospheric aerosol in Warsaw, the partial contribution of sea spray is also highly probable."

Line 580: I guess you mean "m above ground level" please, specify

The trajectories arriving to Warsaw were calculated at all 3 heights (i.e. 200 m, 1 500 m, 3 000 m) above sea level. This sentence has been corrected:

"The trajectories reaching the sampling point at 200 m a.s.l., however, came from the east (Fig. 8)."

Lines 615-617: This sentence is not clear, please, rephrase

The sentence has been changed as follows (changes to the manuscript are indicated in red font):

"Such high contribution of transport of secondary sulfates was also found in the daily $PM_{2.5}$ data showing unusual for the Central European urban area content of secondary inorganic aerosols (35%) during the measurement campaign."

Line 622: excluding the secondary aerosol

In the study Juda-Rezler et al. (2020) the source representing residential sector was identified as "Residential combustion (fresh and aged aerosol)", including PM primary emitted within the city as well as aged aerosol transported from the outskirts of the city. Therefore, this sentence (lines 621-624) has been modified as follows (changes to the manuscript are indicated in red font):

"We can conclude that presented findings are consistent with the previous study performed at the same urban background site (Juda-Rezler et al., 2020), which demonstrated combustion in the residential sector within the city and in the surrounding suburban areas, followed by road transport as the predominant sources for $PM_{2.5}$ pollution in Warsaw, using daily concentrations of $PM_{2.5}$ and its constituents, i.e., 8 ions, carbonaceous matter (EC, OC) and 21 trace elements".

Table 1: you should point out when min values are below the detection limit here

The minimum concentrations below the detection limits have been indicated by the asterisks, as follows:

Table 1: Descriptive statistics for the hourly concentrations (ng m^{-3}) of the elements measured in fine and coarse fractions. Minimum concentrations below the detection limit are indicated by an asterisk.

Element	Fine					Coarse				
[ng m^{-3}]	Mean	Median	Min	Max	SD	Mean	Median	Min	Max	SD
Al	25.3	18.9	3.9*	206.0	24.1	70.2	50.9	2.2*	477.6	64.9
As	0.5	0.3	0.1*	2.3	0.4	0.2	0.1	0.04*	0.9	0.1
Ba	7.8	6.5	5.1*	28.1	3.7	6.5	3.4	2.1*	42.9	5.4
Br	2.4	2.2	0.4	9.7	1.3	0.2	0.1	0.05*	3.0	0.2
Ca	48.8	31.5	4.1*	287.2	45.4	108.1	84.7	4.3	449.8	88.8
Cl	113.3	60.1	5.5*	1 492.9	168.1	141.8	35.5	1.5*	1 682.5	251.3
Cr	2.5	2.3	0.1*	14.4	1.3	1.3	1.1	0.2*	6.9	0.9
Cu	6.7	4.5	0.7	190.2	13.2	4.9	3.6	0.2	41.3	4.8
Fe	114.7	95.4	13.2	1 296.9	105.1	193.1	154.3	11.1	1 281.7	148.8
K	166.8	153.2	18.1	639.6	85.1	29.9	24.5	2.1*	152.9	20.1
Mg	17.4	17.0	4.9*	48.4	8.6	36.8	25.4	3.0*	207.3	33.5
Mn	2.4	2.1	0.4*	21.1	1.9	2.3	2.0	0.2*	11.8	1.6
Mo	0.4	0.4	0.2*	2.1	0.2	0.2	0.2	0.1*	0.9	0.1
Na	80.4	57.2	8.6*	1 538.5	91.8	170.6	71.7	3.5*	1 347.8	231.5
Ni	0.8	0.7	0.02*	8.2	0.6	0.3	0.2	0.2	2.4	0.2
P	20.9	19.8	1.8*	48.7	8.3	7.0	6.5	1.3*	21.7	3.3
Pb	11.7	9.9	0.5*	146.2	11.5	1.0	0.3	0.2*	20.5	1.5
Rb	0.3	0.2	0.1*	3.1	0.2	0.2	0.1	0.1*	10.7	0.5
S	1 020.8	907.6	185.2	2 612.8	548.0	73.9	55.7	6.1	468.0	62.2
Se	0.4	0.4	0.1*	1.2	0.2	0.1	0.1	0.05*	0.5	0.04
Si	51.4	51.4	0.7*	349.1	39.2	166.1	126.4	8.9	995.5	134.4
Sr	0.5	0.3	0.2*	15.8	0.9	0.4	0.3	0.1*	3.1	0.4
Ti	2.9	1.7	1.3*	20.6	2.4	5.7	4.4	0.6*	34.6	4.8
V	1.2	1.0	0.8*	3.3	0.5	0.6	0.4	0.3*	2.5	0.3
Y	0.3	0.3	0.2*	2.3	0.1	0.1	0.1	0.1*	1.1	0.1
Zn	38.7	33.0	6.7	239.5	25.3	6.5	4.5	0.4	52.2	6.0
Zr	0.4	0.3	0.2*	2.2	0.2	0.4	0.2	0.1*	3.3	0.4

Figures 4 and 5: Please, put the labels of the x axis categories also at the top for a better visualization.

The change has been done. As an example the new Figure 5 is presented below. Please note that according to the next comment the colors corresponding to the sources in Figure 5 has been also changed as they were analogous in Figures 4, 5 and 7.

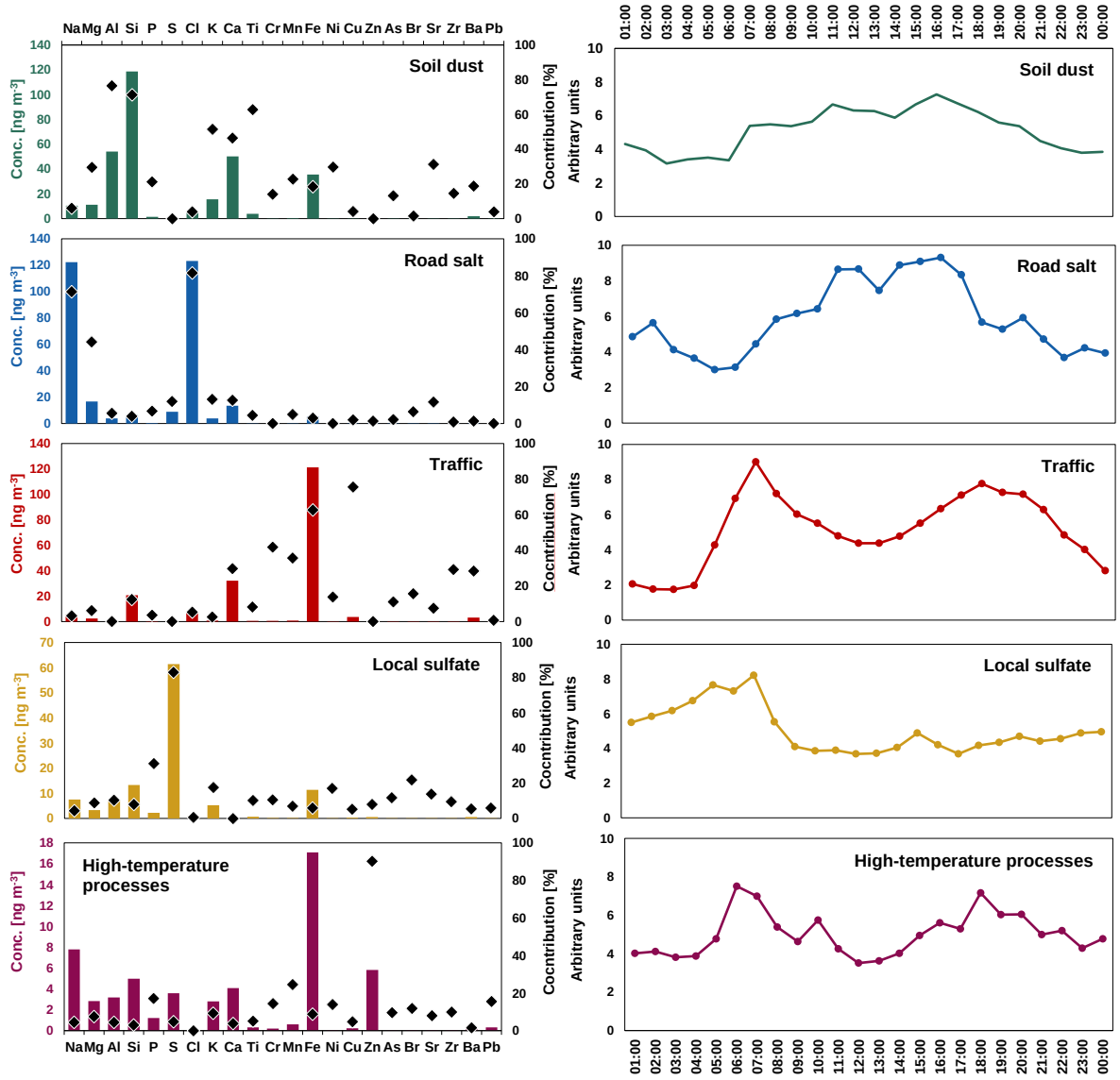

Figure 1: Left panel: PMF profiles (bars, left y axis) and contributions (black diamonds, right y axis) of the identified sources for the coarse fraction. Right panel: Daily patterns of the identified sources (in arbitrary units).

Figure 7: I suggest to use the same colour for corresponding sources in the fine and coarse fraction.

The colors has been changed as presented in the new Figure 7 below. Please note that also colors corresponding to the sources in Figure 5 has been also changed as they were analogous in Figures 4, 5 and 7.

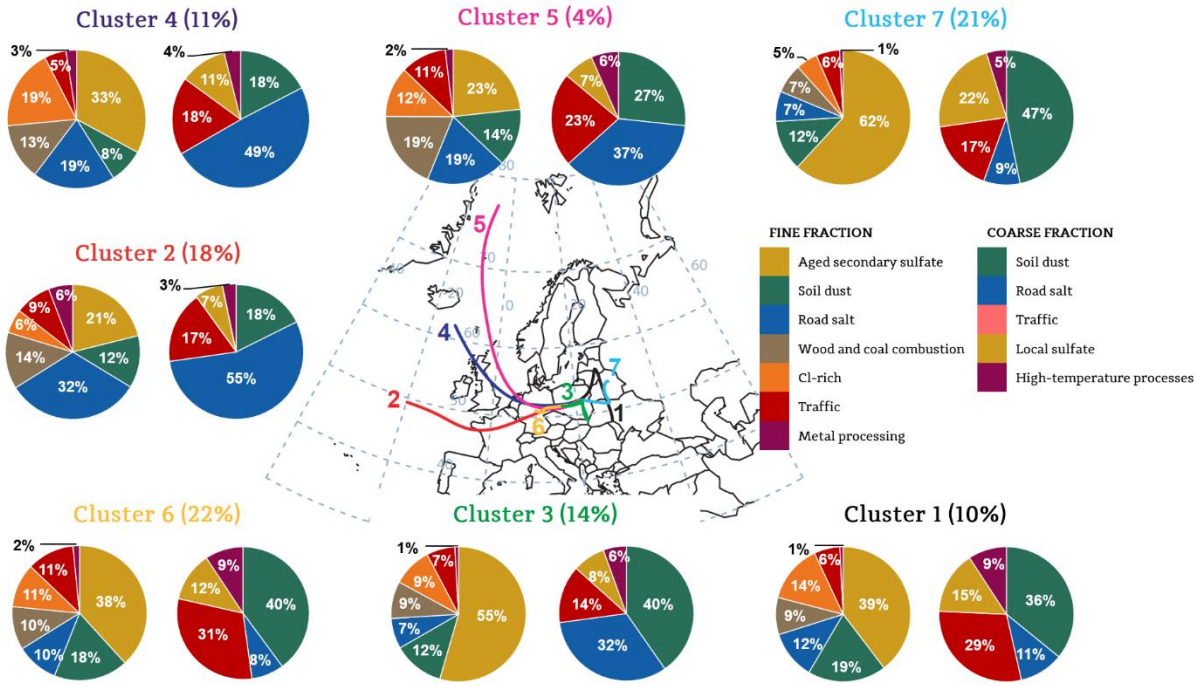

Figure 2: Trajectory cluster centroids arriving to Warsaw at 200 m (center map) with PMF factor contribution in different clusters for fine (left pie charts) and coarse (right pie charts) fraction. Percentage of the trajectories classified into each cluster is given in the parentheses.