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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 #3 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 #3 (16 June 2021)

Authors present results of high time resolution measurement of elements in fine and coarse urban aerosols with subsequent identification of sources using a combination of three receptor models.

The paper provides interesting results with detailed insights into the winter pollution sources in Warsaw area.

The paper is suitable for publication in the journal Atmospheric Chemistry and Physics, however, there are a few issues that need to be addressed before acceptance for publication. Minor revisions of the paper taking into consideration the comments reported below are requested.

Comments:

line 19: Exhaust traffic emissions are mostly prevailing in fine (and especially in submicrometre) fraction.

Traffic-related sources identified in the coarse fraction include soil dust mixed with road dust, road dust, as well as exhaust and non-exhaust traffic emissions. The latter emission sources were identified as more general source "Traffic" as elements characteristic for exhaust and non-exhaust emissions were presented in the PMF profile and detailed separation of both types of emission were not possible. However, most probably the share of non-exhaust emission is prevailing in this fraction, but our study is not able to confirm this.

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

"In the fine fraction, aged sulfate aerosol related with emissions from solid fuel combustion in the residential sector located outside the city was the largest contributing source to fine elemental mass (44%), while traffic-related sources, including soil dust mixed with road dust, road dust, as well as ~~exhaust and non-exhaust~~ traffic emissions, had the biggest contribution in the coarse elemental mass (together accounting for 83%)."

line 34-35: Statement that the greatest health risk is from PM_{2.5} is relative. It depends on considered particle size. The statement is true if you compare PM₁₀ and PM_{2.5}. But it is not true for the comparison of PM_{2.5} and PM₁. PM₁ and especially UFP are more dangerous than PM_{2.5} due to their ability to penetrate deeper into the lung than PM_{2.5} particles.

Thank you for this comment. We have focused on the PM₁₀ and PM_{2.5} as those fractions are commonly studied in the cohort studies worldwide. This paragraph has been modified as follows (changes to the manuscript are indicated in red font):

“Many epidemiological studies have shown strong relationship between PM and adverse health effects, focusing on either short-term or long-term exposure (e.g., Pope and Dockery, 2006; Pope et al., 2018). Majority of the worldwide cohort studies used PM₁₀ and/or PM_{2.5} (particles with aerodynamic diameter smaller than 10 μm and 2.5 μm , respectively) as the exposure metric. Comparing these two main fractions of PM, the greater risk to health is posed by PM_{2.5} (~~particles with aerodynamic diameter smaller than 2.5 μm~~), as it can penetrate the respiratory system via inhalation, causing or aggravating respiratory and cardiovascular diseases, reproductive and central nervous system dysfunctions, as well as cancer (e.g., Manisalidis et al., 2020). Globally, ambient PM_{2.5} air pollution contributed to 4.14 million deaths in 2019 (Murray et al., 2020).”

line 73: It is not true, you have overlooked some papers, for example, Pokorná et al., Sci. Total Environ. 2015, 502, 172–183.

Thank you for this comment. We have focused on the source apportionment using PMF model applied to the hourly elemental composition of PM₁₀, PM_{2.5-10} and/or PM_{2.5}. In the given reference (Pokorná et al., 2015) the coarse and fine fractions were defined in different way, as PM_{0.15-1.15} and PM_{1.15-10}, respectively. Thus, this reference was not included in the paper. However, this paragraph has been clarified as follows (changes to the manuscript are indicated in red font):

“However, according to our knowledge, receptor modelling studies based on hourly elemental composition of PM_{2.5} and PM_{2.5-10} has not been carried out in Central Europe previously.”

line 118: Add sampling flow rate and volume of passed air per sample.

The information has been added (changes to the manuscript are indicated in red font):

“The aerosol was collected by a sampling device (PIXE International Corporation (Calzolari et al., 2015)) designed to separate the fine (<2.5 μm aerodynamic diameter) and the coarse (2.5–10 μm) modes of atmospheric aerosol at an air flow rate of 1 l min⁻¹.”

line 281: Cl originates also from various combustion sources, more details see for example in Mikuška et al, Atmosphere 2020, 11, 688.

We agree that sea/road salt is not the only source of Cl emission. However, no or weak correlation with other elements and no diurnal pattern of Cl concentrations do not allow identifying any particular source of this element in the fine fraction. Only moderate correlation with Br and K, may suggest the combustion processes as a source, but other analyses performed in the study do not confirm this. The information on Cl sources has been added (changes to the manuscript are indicated in red font):

“Cl is usually attributed either to the sea salt in areas close to the coasts or to the road salt in continental areas of Central and Northern Europe (Belis et al., 2013). It is also emitted from the

combustion of coal, wood and solid waste, in particular in residential sector (e.g., Mikuška et al., 2020). ~~The latter case is the most probable source of Cl in Warsaw is road salt.~~ Recorded time series of Cl in Warsaw are different in the fine and coarse fractions with no correlation between concentrations in the two modes ($r = 0.08$).”

Mikuška, P., Vojtěšek, M., Křůmal, K., Mikušková-Čampulová, M., Michálek, J., and Věčera, Z.: Characterization and source identification of elements and water-soluble ions in submicrometre aerosols in Brno and Šlapanice (Czech Republic), *Atmosphere-Basel*, 11(7), 688, doi: 10.3390/atmos11070688, 2020.

line 363-384: Component contribution and time profile of this factor in coarse fraction suggest considering renaming this factor to residential heating.

Sources in both fractions are connected with residential sector as was explained in the text (Chapter 3.3.1). Both sources has similar chemical profile, with sulfur as the main component and thus they were named in similar way. However, in order to differentiate these sources in both fractions, we have used more precise names as: “Aged secondary sulfate” and “Local sulfate” for the fine and coarse fraction, respectively.

line 466-469: Mentioned metals could also indicate emissions from waste incinerator. Is there any incinerator in the vicinity?

There is a small municipal waste incinerator in Warsaw, however it is located north-east of the measurement point and cannot influence the measurement point what was confirmed by the CFP analyses. Moreover, lack of Cl in the profile of the sources identified in both fractions do not suggest that sources in both fractions can be related with emissions from waste incinerator.

line 495-501: Br, Se, As are strong markers of coal combustion, so this factor looks rather like combined combustion of coal and biomass (wood).

This issue was also raised by Reviewer #2. The name of the source has been changed to “Wood and coal combustion”.

line 608-609: As far as I know other studies are providing high time resolution measurement in Central Europe, see comment for line 73.

As was stated in the response to the comment for line 73, our analyses and literature review have been focused on the PMF source apportionment applied to the hourly elemental composition of PM₁₀, PM_{2.5-10} and/or PM_{2.5}. The studies with different definition of fine and coarse fraction as well as studies using 2-h time resolution have been excluded. This paragraph has been clarified as follows (changes to the manuscript are indicated in red font):

“The analysis of the composition of trace elements in the fine (PM_{2.5}) and coarse (PM_{2.5-10}) fractions of particulate matter at an urban background site in central Warsaw during a high time resolution wintertime measurement campaign has been carried out for the first time in Central Europe.”

line 626-627: According to my experience, parallel measurement of different PM fractions with shorter time resolution can also provide the same results as high time resolution measurement.

Thank you for this comment.