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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 #1 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 #1 (22 Apr 2021)**

*General comments*

*The manuscript presents one month of hourly measurements of major and trace elements in PM2.5 and PM10 in Warsaw, Poland, in February and March 2016. The data are thoroughly discussed, and three different receptor models are applied to determine the sources and origins of the elements. Five sources in PM10 and seven sources in PM2.5 are found, demonstrating the advantages of high time resolution for appropriate source identification. Furthermore two cases of Saharan dust transport are discussed..*

*The structure of the manuscript, the results and the presentation of the material are very detailed and carefully worked out. The topic is relevant and well worth publication in ACP. I would, however, suggest a few minor changes and additions before publication.*

*Specific comments*

*The advantage of +/- hourly time resolution of elemental concentrations is nicely demonstrated, but biased towards Streaker sampling and PIXE analysis. Recent studies with XRF method have also achieved hourly resolution and size segregation for source apportionment, e.g. see references for Beijing and Delhi in Rai et al. (2021):*

*Rai, P., Slowik, J. G., Furger, M., El Haddad, I., Visser, S., Tong, Y., Singh, A., Wehrle, G., Kumar, V., Tobler, A. K., Bhattu, D., Wang, L., Ganguly, D., Rastogi, N., Huang, R. J., Necki, J., Cao, J., Tripathi, S. N., Baltensperger, U., and Prévôt, A. S. H.: Highly time-resolved measurements of element concentrations in PM10 and PM2.5: comparison of Delhi, Beijing, London, and Krakow, Atmos. Chem. Phys., 21, 717-730, 10.5194/acp-21-717-2021, 2021.*

Thank you for this comment. We have added in the manuscript more references showing the use of hourly sampling as well as a statement clarifying that the examples are referred to both the streaker sampler and Xact metals monitor. This paragraph (lines 64-80) has been modified as follows (changes to the manuscript are indicated in red font):

"Only limited number of studies have applied receptor modelling techniques to the high-time resolution PM samples, particularly applying Positive Matrix Factorization (PMF) model to the hourly elemental composition of $PM_{2.5}$ and $PM_{2.5-10}$ samples. There are only few measurement devices allowing for the sampling of the concentrations of the elements with high time resolution, with the wide application of the streaker sampler (PIXE International Corporation) (Calzolai et al., 2015) and the semi-continuous X-ray fluorescence spectrometer Xact Ambient Metals Monitor (Cooper Environmental Services) (Rai et al., 2020). Identification of PM sources based on aerosol samples in 1-hour resolution has been carried out in Southern Europe,

e.g.: at 4 urban sites in Barcelona (Spain), Porto (Portugal), Athens (Greece) and Florence (Italy) (Lucarelli et al., 2015); at an urban site in Elche (Spain) (Nicolás et al., 2020); at 6 sites of different types in Tuscany (Central Italy) (Nava et al., 2015), in a small town in the Po Valley (Italy) (Belis et al., 2019); and in an industrial area of Taranto (Italy) (Lucarelli et al., 2020). Outside this region, hourly-resolved PM samples have been investigated e.g.: in London (Crilley et al., 2017) in the United Kingdom and at 4 different sites in Alexandra in New Zealand (Ancelet et al., 2014). There has been also the wide application of both measurement devices in Asia, e.g.: in Wuhan (Acciai et al., 2017), the Pearl River Delta region (Zhou et al., 2018), Shanghai (Chang et al., 2018) and Beijing (Rai et al., 2021) in China; as well as in the capital of India, Delhi (Rai et al., 2020). However, according to our knowledge, receptor modelling studies based on hourly elemental composition of PM has not been carried out in Central Europe previously".

The following references has been added:

Acciai, C., Zhang, Z., Wang, F., Zhong, Z., and Lonati, G.: Characteristics and source analysis of trace elements in PM2.5 in the urban atmosphere of Wuhan in spring, Aerosol Air Qual. Res., 17, 2224–2234, doi: 10.4209/aaqr.2017.06.0207, 2017.

Chang, Y., Huang, K., Xie, M., Deng, C., Zou, Z., Liu, S., and Zhang Y.: First long-term and near real-time measurement of trace elements in China's urban atmosphere: temporal variability, source apportionment and precipitation effect, Atmos. Chem. Phys., 18, 11793–11812, doi:10.5194/acp-18-11793-2018, 2018.

Rai, P., Furger, M., El Haddad, I., Kumar, V., Wang, L., Singh, A., Dixit, K., Bhattu, D., Petit, J. -E., Ganguly, D., Rastogi, N., Baltensperger, U., Tripathi, S. N., Slowik, J. G., and Prévôt, A. S. H.: Real-time measurement and source apportionment of elements in Delhi's atmosphere, Sci. Total Environ., 742, 140332, doi:10.1016/j.scitotenv.2020.140332, 2020.

Rai, P., Furger, M., Slowik, J. G., Zhong, H., Tong, Y., Wang, L., Duan, J., Gu, Y., Qi, L., Huang, R. -J., Cao, J., Baltensperger, U., and Prévôt, A. S. H.: Characteristics and sources of hourly elements in PM10 and PM2.5 during wintertime in Beijing, Environ. Pollut., 278, 116865, doi:10.1016/j.envpol.2021.116865, 2021.

*In Figs. 4 and 5 the right hand axes are labelled as 'contribution [%]', while in the captions you call this 'explained variation'. Would it not be more consistent (and more correct) to just use 'contribution' in both places?*

The captions of both figures have been changed as follows:

Figure 4: Left panel: PMF profiles (bars, left y axis) and contributions (black diamonds, right y axis) of the identified sources for the fine fraction. Right panel: Daily patterns of the identified sources (in arbitrary units).

Figure 5: 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).

*In Fig. 6 the traffic source area is quite different from the road salt source. Shouldn't we expect more similarity between the two factors, as in both cases probably resuspension would be the mechanism for 'creating' the sources? Or is deicing salt not evenly distributed along the Warsaw road system? Please discuss this a bit more.*

Thank you for this comment. In the case of "Road salt" source, the origin area is located in the part of Warsaw where main road from the city's airport and one of the main expressway running through Warsaw are located. This road could be intensively de-iced during the measurement campaign. Moreover de-icing activities at the airport could also influence the concentrations as sodium-based compound are used for the de-icing of airports' runways and roads. Also daily profiles of this source suggest the resuspension processes. In contrary, "Traffic" source identified for both fine and coarse fractions shows bi-modal daily profiles characteristic for the traffic sources. The CPF analyses show different location than for "Road salt" sources. The measurement site was located in the city center where exhaust emission is probably playing a key role.

*Fig. 8 shows trajectories originating in or crossing over parts of the Sahara desert and ending above Warsaw in 1500 and 3000 m asl. It is not straight forward that PM arriving at these elevations is measured with ground-based samplers, and possible downward mixing processes should be discussed in more detail. This is especially the case on 18 Feb, where at 500 m asl the airmass is advected from SE, indicating a completely different source location than the Sahara. While I find these two cases interesting and plausible, a strong connection is not given. I recommend adding one or two sentences discussing the uncertainties (Saharan dust composition, vertical mixing from upper layers, concentrations aloft).*

Thank you for this comment. Following the recommendation we provide additional support to the occurrence of the African dust outbreak of February 2016 over Warsaw. We have included several plots as Supplementary Material: the MODIS true color image for 15 February, the output of the DREAM prediction model for 18 February and the radiosounding launched near Warsaw on 18 February. Cloud cover prevented from having optical remote sensing measures that would provide information regarding the vertical load/distribution of dust in this episode. In addition, we have included in the main text (subsection 3.5) a more detailed description that allows relating the presence of dust at low levels with the height of the trajectories carrying African dust.

"The advection of dust-laden airflows out of Africa toward Warsaw is evidenced by satellite imagery on 15 February (Fig. S24), and both back trajectories (Fig. 8) and dust prediction models (Fig. S25) indicate they reach the study area aloft by 18 February. However, the small-scale downward movement and mixing of dust particles is not represented in the back trajectory model and therefore it cannot explain the dust impact at the ground level. Besides, cloud coverage prevented optical remote sensing monitoring. Therefore the presence of dust at low levels is supported primarily by the $PM_{2.5}$ chemical composition and the PMF analysis as shown below. The operational radiosounding launched on 18 February in Legionowo (12374), around 20 km to the north of Warsaw, and surface meteorological instruments confirm the near-surface southeasterly winds in association with a high-pressure system located to the northeast of Poland, consistent with the trajectories found at 200 m a.s.l. Half-hourly wind speed at the Warsaw Okęcie Airport was over 20 km $h^{-1}$ only from 00:00 UTC to 02:30 UTC, so the $PM_{2.5}$ soil contribution registered on 18 February (Fig. 9) is unlikely due to local/regional soil particulate resuspension. The radiosounding (Fig. S26) shows the presence of a dry layer from near 1 500 m asl to over 2 500 m asl where wind have veered to southerly. This layer overlaps the top of a temperature inversion layer with base (the atmospheric boundary layer) at 750 m a.s.l., located in a cloud top. Above the dry layer, a thick ice cloud extends above 5000 m a.s.l. The trajectories corresponding to the dry layer have a North African origin; therefore it can be identified as a dust layer. As the dust layer overlaps the top of the inversion layer and the

entrainment zone is at the base of the inversion, near the cloud top, the dust layer is then near the entrainment zone making possible the mixing of dust within the ABL down to the ground."

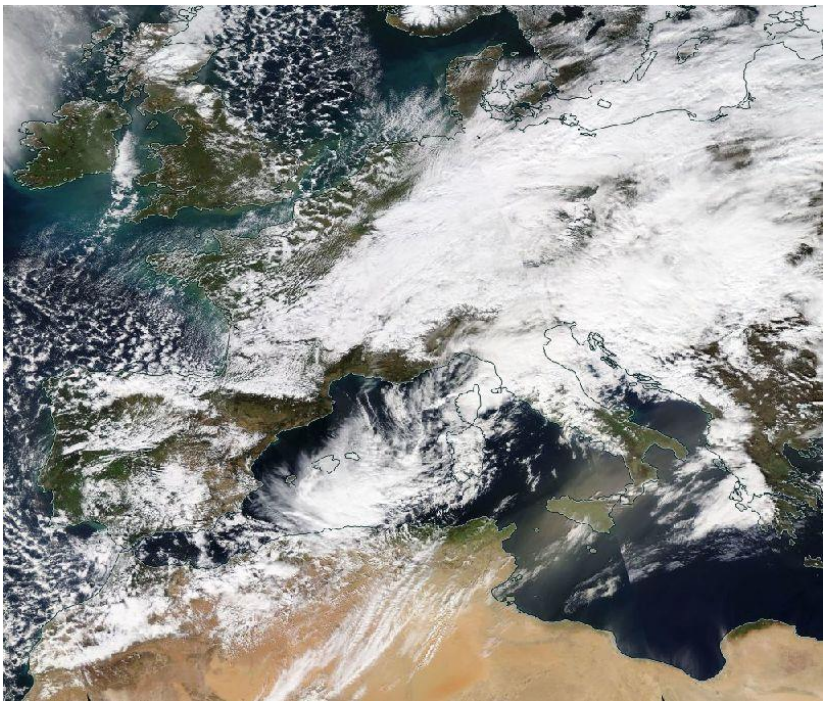

Figure S24. Aqua MODIS corrected reflectance (true color) on 15 February, 2016. NASA EOSDIS Worldview (https://worldview.earthdata.nasa.gov/).

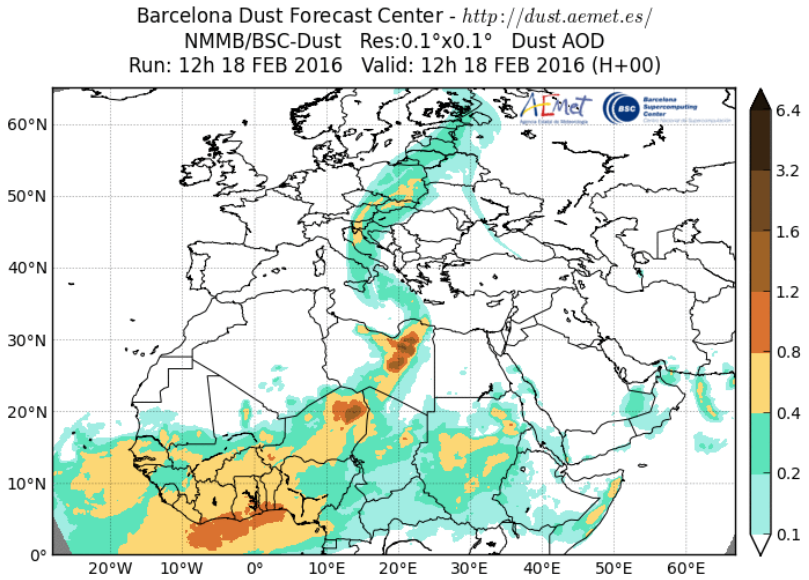


Figure S25. Aerosol Optical Depth (AOD) forecast for 18 February, 12 UTC. Image from the NMMB/BSC-Dust model operated by the Barcelona Supercomputing Center, Spanish National Supercomputing Center (https://www.bsc.es/ess/bsc-dust-daily-forecast/).

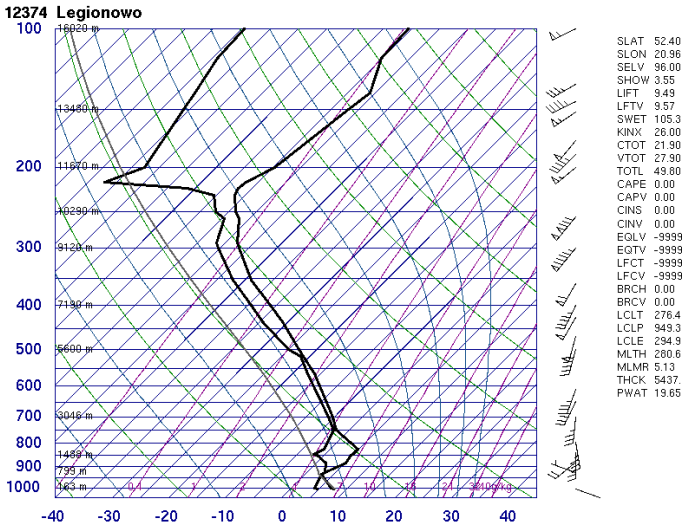


Figure S26. Skew T-log p diagrams for Legionowo on 18 February, 12 UTC. University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html).

*In Fig. 9, I do not see an advantage of comparing the elemental time series in ng/m3 with the PMF time series in arbitrary units, as the main elements comprising the soil dust are those in eq. (8), so we basically just add O (in stoichiometric ratios) to these elements. It should be possible from a linear regression to estimate the fraction of the two (I guess it would be something around the value of 2). With respect to the Saharan dust, it would be the same to just compare the five relevant elements, not their oxides, as long as we do not have quantitative information on the amount of dust (mass) transported.*

Thank you for this comment. Our main goal of this part of the manuscript was to support the apportion of the "Soil dust" source identified by PMF. For this purpose, the soil dust component was chemically reconstructed by using the conversion of elemental concentrations of typical crustal species Al, Si, Ca, Ti and Fe following the approach widely reported in the literature (e.g.: Chow et al. (2015)). As these elements are predominantly present in the soil in the oxidized forms this approach assumes the conversion of elements to their oxides.

Chow, J. C., Lowenthal, D. H., Chen, L.-W. A, Wang, X. and Watson, J. G.: Mass reconstruction methods for $PM_{2.5}$: a review, Air Qual. Atmos. Hlth., 8, 243–263, doi:10.1007/s11869-015-0338-3, 2015.

*Technical corrections*

*L118 device (delete s)*

It has been corrected.

*L121 substrata (insert s)*

It has been corrected.

*L126 beam (delete s)*

It has been corrected.

*L131 write Micromatter in one word*

It has been corrected.

*L141 Reorder the sentence. Write '… is a widely used (…) multivariate factor analysis model in air quality studies based on …'*

It has been corrected.

*L142 weighted least squares fit (add an s)*

It has been corrected.

*L369 Can you explain the term 'bioavailable' a bit better? The way you use it, it appears to be something like a quantitative entity with a time dependence.*

Bioavailability is defined as the degree and rate of absorption of a substance by the living systems or the degree which the substance is available to physiologically active sites. The following explanation 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)."

*L373 … emitting a substantial amount … (insert a)*

It has been corrected.

*L377 … attributed to wood combustion … (delete the)*

It has been corrected.

*L387 In the case of the coarse … (insert the)*

It has been corrected.

*L393 … the pattern of the source … (insert the)*

It has been corrected.

*L504 … points towards two … (insert towards)*

It has been corrected.

*L516 … all ranges of wind … (insert s)*

It has been corrected.

*L518 … are used for the maintenance … (replace to with 'for the')*

It has been corrected.

*L533 write '…of the sources identified by PMF for….'*

It has been corrected.

*L592 widely (instead of wide)*

It has been corrected.

*L633 … receptor modeling based … (insert ing)*

It has been corrected.