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RESEARCH ARTICLE

Spatial-temporal variability of aerosol sources based on chemical composition and particle number size distributions in an urban settlement influenced by metallurgical industry

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Abstract

The Moravian-Silesian region of the Czech Republic with its capital city Ostrava is a European air pollution hot spot for airborne particulate matter (PM). Therefore, the spatiotemporal variability assessment of source contributions to aerosol particles is essential for the successful abatement strategies implementation. Positive Matrix Factorization (PMF) was applied to highly-time resolved $PM_{0.15–1.15}$ chemical composition (1 h resolution) and particle number size distribution (PNSD, 14 nm – 10 μm) data measured at the suburban (Ostrava-Plesná) and urban (Ostrava-Radvanice) residential receptor sites in parallel during an intensive winter campaign. Diel patterns, meteorological variables, inorganic and organic markers, and associations between the chemical composition factors and PNSD factors were used to identify the pollution sources and their origins (local, urban agglomeration and regional). The source apportionment analysis resolved six and four $PM_{0.15–1.15}$ sources in Plesná and Radvanice, respectively. In Plesná, local residential combustion sources (coal and biomass combustion) followed by regional combustion sources (residential heating, metallurgical industry) were the main contributors to $PM_{0.15–1.15}$. In Radvanice, local residential combustion and the metallurgical industry were the most important $PM_{0.15–1.15}$ sources. Aitken and accumulation mode particles emitted by local residential combustion sources along with common urban sources (residential heating, industry and traffic) were the main contributors to the particle number concentration (PNC) in Plesná. Additionally, accumulation mode particles from local residential combustion sources and regional pollution dominated the particle volume concentration (PVC). In Radvanice, local industrial sources were the major contributors to PNC and local coal combustion was the main contributor to PVC. The source apportionment results from the complementary datasets elucidated the relevance of highly time-resolved

Main finding
The source apportionment results using complementary datasets elucidated the relevance of highly time-resolved parallel measurements given the variable local meteorology with the possibility of replication at any receptor.

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parallel measurements at both receptor sites given the specific meteorological conditions produced by the regional orography. These results are in agreement with our previous studies conducted at this site.

Keywords Highly time-resolved parallel measurements · Elemental composition · Regional transport · Local heating · Metallurgical industry · Nanoparticles

Introduction

Suspended particulate matter (PM) is complex with respect to its mass concentration, particle size distributions, chemical composition and its sources. Anthropogenic activities including industrial effluent, combustion emissions and physical processes such as wind-blown dust and traffic emissions are typically factors dominating the spatial and temporal PM variations (DeGaetano and Doherty 2004). With respect to small-scale spatial variations in urban areas, the largest variations occur in the ultrafine (< 0.1 μm) and the coarse mode ($PM_{2.5–10}$, re-suspended dust) (Lagudu et al. 2011; Kumar et al. 2012; Wang et al. 2012). Secondary aerosols that contribute to the accumulation mode (0.1–1 μm) show more homogenous spatial distributions (Moon 2001). Despite the limited spatial variations of fine PM within the city, understanding and controlling air pollution in highly polluted areas require multiple-site measurements. During pollution events, it is important to assess the origin of the higher PM concentrations to be able to employ measures to improve air quality (Petit et al. 2017; Waked et al. 2018). Despite the local origin of some PM, the observed PM is also influenced by medium and long-range transport processes. Therefore, for mitigation strategies to be effective, they must be scaled at regional, national and even continental levels (Guerreiro et al. 2014; Guan et al. 2017).

The Moravian-Silesian region, located in the eastern part of the Czech Republic, represents an EU air pollution hot spot. Epidemiological studies from Ostrava, the major city of the region, displayed negative health impacts for the local population, particularly for children, caused by deteriorated air quality (Dostál et al. 2013; Šram et al. 2013; Topinka et al. 2015). The area has been historically burdened with extensive industrial activity in the Upper Silesian basin. The key factors influencing the air quality in the Moravian-Silesian region are (i) exploitation and use of the high quality black coal, (ii) extensive heavy industry, (iii) high density of built-up areas with local heating by solid fuels, (iv) dense transport infrastructure on both sides of the Czech-Polish border, and (v) orography influencing the meteorological conditions with prevailing wind direction from south-west and north-east as well as long-lasting periods of stable conditions (from autumn to spring) with subsequent worsened dispersion conditions. Currently, approximately 760 facilities, large and medium stationary sources of air pollution, are registered in the region (CHMI 2018). Only several dozens of facilities—power plants and enterprise energy generation—and metallurgical production facilities have a substantial effect on overall emissions (TSP, SO_2 and NO_x); nevertheless, fugitive TSP emissions produced e.g. from landfills, handling of bulk materials and halls with dusty operations are not taken into consideration in the emission inventories. Therefore, the planning and implementation of effective abatement strategies to improve air quality in Ostrava are challenging due to the presence of miscellaneous sources, some of them situated near residential areas.

In order to analyse the spatial and temporal changes in airborne PM regarding to the effective abatement strategy implementation, the application of receptor model is highly effective. Receptor models analyse data matrices of aerosol chemical and physical characteristics of samples collected at a given locality, the receptor, to apportion their sources (Polissar et al. 2001). In the literature, a wide range of statistical models and modelling approaches is available e.g. CMB, PCA, PMF, UNMIX, ME and COPREM, and one of the main differences between the models is the degree of knowledge required about the pollution sources prior to the application of receptor models (Viana et al. 2008). PMF is a powerful tool for source resolution using highly time resolved aerosol composition data and has been used successfully in identifying the sources of airborne particles in many studies (e.g.; Ancelet et al. 2012, 2014; Pancras et al. 2013; Moreno et al. 2013; Hovorka et al. 2015; Hopke 2016). Particulate matter toxicity depends on its chemical and physical characteristics: composition, size, morphology or solubility (Heal et al. 2012). Thus, source apportionment studies focus not only on chemical composition but also on particle number size distribution data (Kim et al. 2004; Zhou et al. 2004, 2005a; Ogulei et al. 2007a, b; Kasumba et al. 2009; Pey et al. 2009; Harrison et al. 2011; Cusack et al. 2013; Beddows et al. 2015; Sowlat et al. 2016; Masiol et al. 2016; Squizzato et al. 2019). By investigating particles in various size ranges, it is possible to more clearly identify and apportion contributions from those sources that contributed more to the particle number than to the particle mass (Vu et al. 2015). The two approaches are often complementary (Zhou et al. 2005b; Beddows et al. 2015).

The aim of this study was to access the spatial and temporal variability of winter air pollution sources including nanoparticles in the districts of Ostrava-Radvanice and Ostrava-Plesná, examples of industry affected urban and suburban residential receptor sites. The source apportionment was performed using highly time-resolved elemental composition of

$PM_{0.15–1.15}$ and particle number size distributions (PNSD, 14–10 μm) measured from February 9 to March 3, 2014. The factors resolved from mass chemical compositions and the PNSDs by PMF were compared and associations were determined. The study complements previous source apportionment studies performed at the sites (Pokorná et al. 2015 and 2016; Leoni et al. 2018; Kozáková et al. 2019). The results from the parallel highly time-resolved chemical and PNSD measurements at these two different site types provided further insights into the spatial and temporal variability of the source contributions to PM at this site.

Methods

Experimental

A twin intensive sampling campaign was performed from February 6 to March 6, 2014 in Ostrava-Radvanice and Ostrava-Plesná as urban and suburban residential receptor sites, respectively (Fig. SI1). The distance between the two sites was approximately 16 km.

Five-minute integrated PNSD was measured with a Scanning Mobility Particle Sizer (14–730 nm, SMPS-3936 L25, TSI Inc. and 14.6–710.5 SMPS-3936 L75, TSI Inc.) and an Aerodynamic Particle Sizer (0.542–20 μm, APS-3321, TSI Inc.).

A standard reference sampler (LVS-3, Sven Leckel Ingenieurbüro GmbH, Berlin, Germany; flow rate of 2.3 m^3 h^{-1}) with quartz fibre filters (47 mm; Whatman QMA, GE Healthcare, Maidstone, UK) was used to measure 24-h PM_1 concentrations in Plesná. A high-volume sampler (DHA-80, Digitel, Switzerland, flow rate 30 m^3 h^{-1}) with quartz fibre filters (150 mm diameter; Whatman QMA, GE Healthcare, Maidstone, UK) was used to determine the 24-h concentrations of PM_1 in Radvanice. The samples were analysed for both cations and anions using a Dionex ICS-5000 system (Dionex, Sunnyvale, CA, USA) and for elemental carbon (EC) and organic carbon (OC) using a semi-continuous analyser (Sunset Laboratory Inc., Tigard, OR, USA) operated in an offline mode and using Eusaar2 protocol. PM_1 samples from Plesná were also analysed for monosaccharide anhydrides (levoglucosan, mannosan and galactosan) using a Dionex ICS-5000+ system (Dionex, Sunnyvale, CA, USA). Detailed descriptions of the methods and limits of detection (LODs) along with quality assurance and control for the used methods were reported by Kozáková et al. (2019).

Size segregated and time resolved PM were collected with a Davis Rotating-drum Uniform-size-cut Monitor – 3DRUM (DELTA Group UC-Davis), from February 19 to March 3 and were used to provide 1-h PM elemental compositions. The particles were collected on Mylar substrates lightly greased with Apiezon™. The 3DRUM collects particles in 3 size ranges from 150 to 10 μm. The two size range samples from 0.15 to 1.15 μm were analysed for 24 elements using synchrotron X-ray fluorescence (S-XRF) by the Air Quality Research Center, University of California (Leoni et al. 2018). For the source model, data from the two size ranges of 3DRUM were merged to obtain $PM_{0.15–1.15}$ to replicate the size range of the PNSD.

In Radvanice, 5-min meteorological data—wind speed (WS) and wind direction (WD), temperature (T), relative humidity (RH) and precipitation (P)—were recorded directly at the site during the whole sampling campaign. In Plesná, the meteorological data were available from the nearest representative automatic monitoring stations managed by the Czech Hydrometeorological Institute (Kozáková et al. 2019).

Data analysis

Descriptive statistics

Measured data (online measurement and chemical analysis of filter samples) and modelling results were statistically analysed using descriptive statistics, Pearson correlation and regression analysis to discover underlying patterns and trends regarding the monitoring campaign.

PMF Modelling

PMF (USEPA version 5.0) was applied to $PM_{0.15–1.15}$ chemical composition and PNSD data (15–10 μm). A 1-h integration time $PM_{0.15–1.15}$ chemical composition as a sum of two size fractions was used for PMF input along with missing mass (MM) for missing data for OC/EC (Pokorná et al. 2013). Hourly aerosol mass concentrations assigned as total variable were calculated from 5-min integrates of the number size distributions with a particle density of 1.5 g cm^{-3} (Shen et al. 2002) recorded by an APS and a SMPS. Due to the importance of the local meteorology given by the orography of the region (Pokorná et al. 2015; Černikovský et al. 2016; Leoni et al. 2018; Kozáková et al. 2019), the matrices of the chemical composition from the two sites were modelled separately by PMF.

For the PNSD PMF, the time resolutions from 5 min to 1 h were examined and it was determined that a 15-min resolution was optimal as the best compromise to maintain a high time resolution but avoid unwanted noise. Additionally, the input data were handled by merging three consecutive bins. Since the size segregation of SMPS is based on particle electrical mobility while the APS and 3DRUM impactor use particle aerodynamic properties, the mobility diameter was converted into aerodynamic. The uncertainties were calculated according to Vu et al. (2015). The total variable was calculated summing all the bins. The detailed information was reported in Leoni et al. (2018). PMF was also applied to PNSD from

Plesná (modelled with and without chemical composition data) to extract those factors governing particle number down to the nanometric scale.

The model was run several times using (i) different number of factors, (ii) different extra modelling uncertainties and (iii) for PNSD data different uncertainty input matrices, different C3 value (Vu et al. 2015) in order to obtain the highest S/N ratio and the Q_{true} closest to $Q_{theoretical}$, to determine the most physically meaningful result and the best diagnostics.

The polar plots and daily patterns were obtained using the Openair R Package (Carslaw and Ropinks 2012).

Schematic picture synthetizing all methods (measurement techniques, chemical analysis and data analysis) utilized in this study could be found in the Supplementary Information (Fig. SI2).

Results and discussion

Campaign overview

During the sampling campaign, the average PM_{10} concentrations calculated from the merged SMPS and APS number size distributions in Plesná and Radvanice were 29.2 ± 4.5 μg m^{-3} and 61.5 ± 8.7 μg m^{-3}, respectively. The $PM_{0.15–1.15}$ comprised 90% and 48% of PM_{10} in Plesná and Radvanice, respectively. These PM ratios indicate important differences in the PM size distribution and likely major sources at these locations. The elemental concentrations of almost all elements were lower in Plesná relative to Radvanice (Tables SI1 – 2). On average, the higher arsenic concentrations in Plesná point to the importance of coal combustion there (Tab. SI1). The average particle number concentrations were distributed in four particle modes: nanoparticles (15–30 nm), Aitken mode particles (30–80 nm), accumulation mode particles (80–1 μm) and coarse particles (1–10 μm) are presented in Table 1. The campaign was characterized by prevailing southwesterly and northeasterly winds at both sites due to the topography of the Ostrava region. Wind speed averaged 1.7 ± 0.6 m s^{-1} in Plesná and 1.2 ± 0.5 m s^{-1} in Radvanice.

Factors resolved with chemical composition

To estimate the optimal number of sources, the PMF model was run several times (units of tens for different model settings) and 3 to 8 factors were tested for each data set. The Q values, the resulting source profiles and the scaled residuals were examined. For each data set, the optimum number of factors was chosen based on an adequate fit of the model to the data as shown by the scaled residual histograms and physically interpretable results. The most stable solutions were found for 6-factor and 4-factor solutions for Plesná and Radvanice, respectively. Extra modelling uncertainties of 13.5% for Plesná and 17.9% for Radvanice were included to encompass errors not considered in the uncertainty assessment. All runs converged, the scale residuals were normally distributed, and no unmapped factors were detected with the bootstrap error estimation. No swaps were observed with the displacement error analysis, indicating that there was limited rotational ambiguity (Table SI3).

Table 1 Average particle number concentrations (# cm^{-3}) of the sampling campaign at Plesná and Radvanice site

Site [# cm^{-3}]	Nanoparticles	Aitken	Accumulation	Coarse
Plesná	0.9×10^3	2.2×10^3	3.1×10^3	2
Radvanice	3.5×10^3	6.4×10^3	6×10^3	3.7

The six factors in Plesná were assigned as *coal combustion, mixed factor secondary inorganic aerosol (SIA) and biomass burning, residential heating, biomass burning, re-suspended dust and raw iron production*. The 4 factors in Radvanice were ascribed to *mixed factor SIA and coal combustion, residential heating, raw iron production and sinter/steel production*. The factor elemental profiles and the time-series plots of the estimated contributions from each factor to the $PM_{0.15–1.15}$ mass are shown in Figs. 1 and 3.

Plesná

Factor 1. The first factor, local residential coal combustion, was associated with high concentrations of As, MM, Pb, Cu and Br (Rogula-Kozłowska et al. 2012, 2013a, b; Rogula-Kozłowska and Klejnowski 2013). This factor dominated during the monitoring period with an average contribution of 49% to $PM_{0.15–1.15.}$ The high factor contributions with WS up to 2 m s^{-1} point to local sources primarily situated from the sampling site to the NW/SE (Fig. 2) directions corresponding to the residential area. The time series show a local heating pattern, with a peak in the morning around 8 a.m. and in the evening starting from 5 p.m., with a maximum at 10 p.m. and lasting all night (Fig. SI3). This pattern can be also linked to the stable atmospheric conditions during winter nights, trapping the emissions near the ground. The regression analysis shows moderate relationship between the coal combustion factor and PM_1 OC ($R^2 = 0.40$). Surprisingly, this factor does not correspond to the coal combustion factor presented in Kozáková et al. (2019). This difference is likely due to the differences in the monitored periods (from February 6 to March 6, 2014), lack of chemical species (PM_1 water-soluble ions and OC/EC) and merged data matrix for Ostrava-Radvanice and Ostrava-Plesná sites.

Factor 2. This mixed factor represented mainly by S and K was ascribed to SIA and biomass burning (Leoni et al. 2018). This factor contributed 18% to the mean $PM_{0.15–1.15}$ mass on average. The polar plot points to the source/s of regional

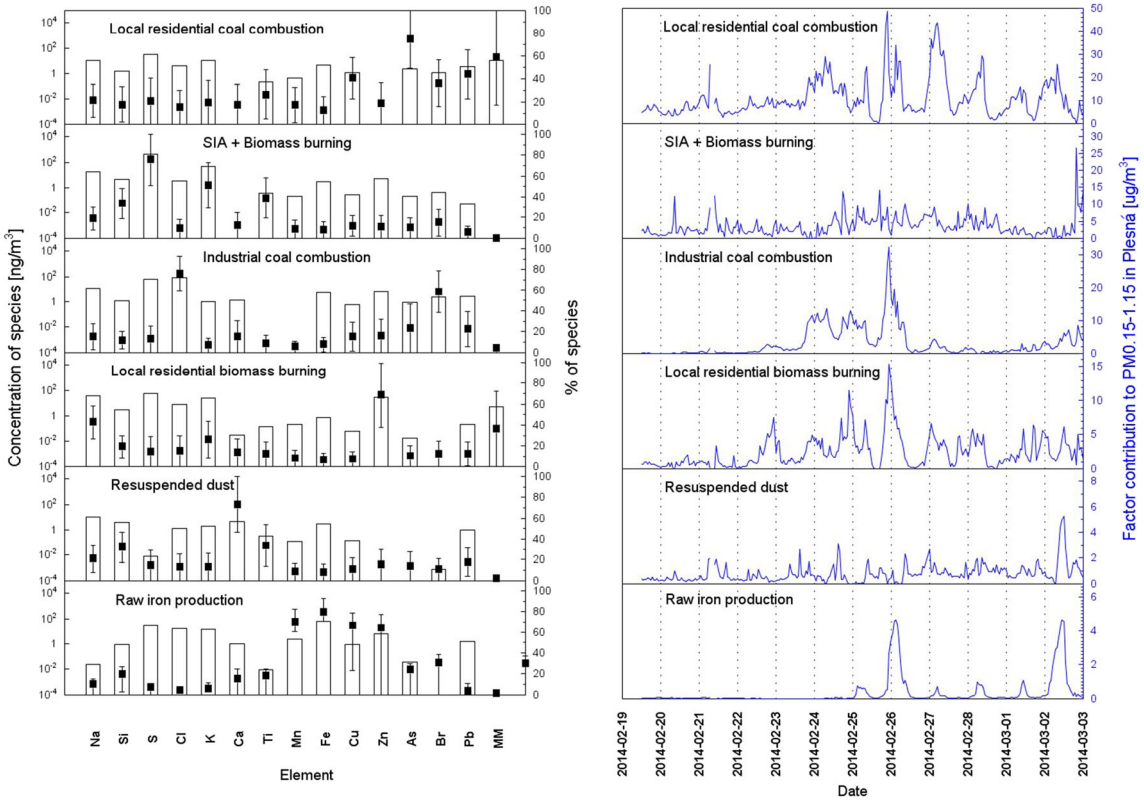

Fig. 1 Plesná factor profiles (concentration of the species, bars; percentage of the species, markers; DISP Average, box; DISP Max and DISP Min, whiskers) and time series resolved for $PM_{0.15–1.15}$ by PMF

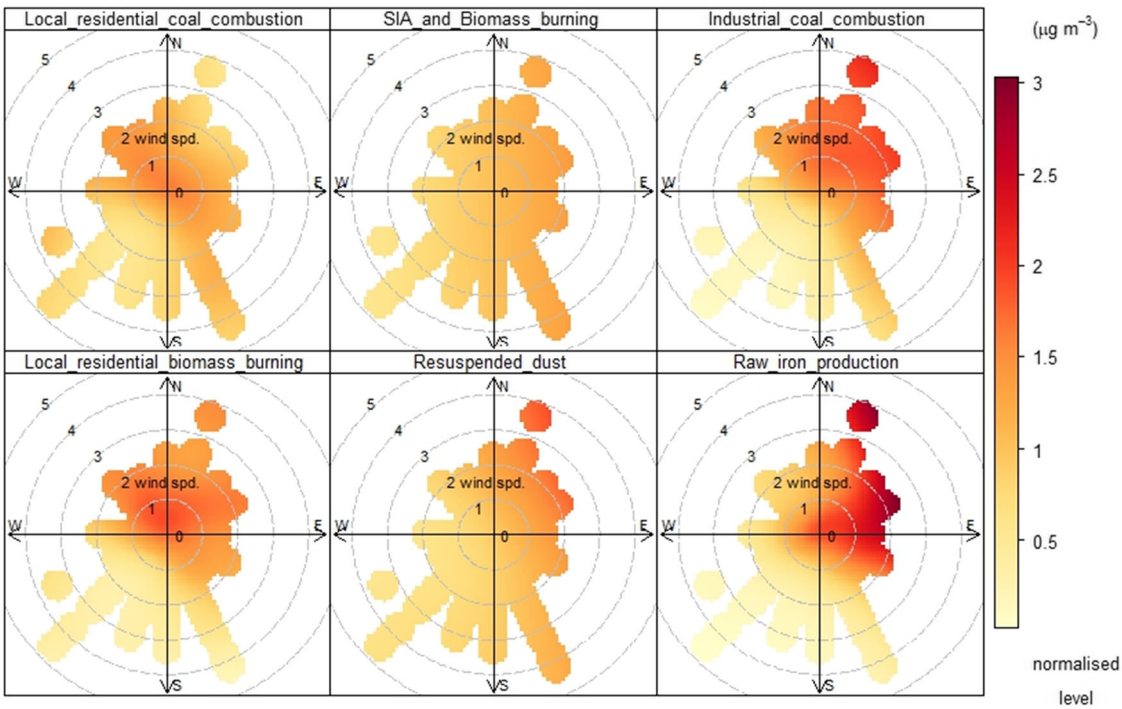

Fig. 2 Polar plots for Plesná $PM_{0.15–1.15}$ factors

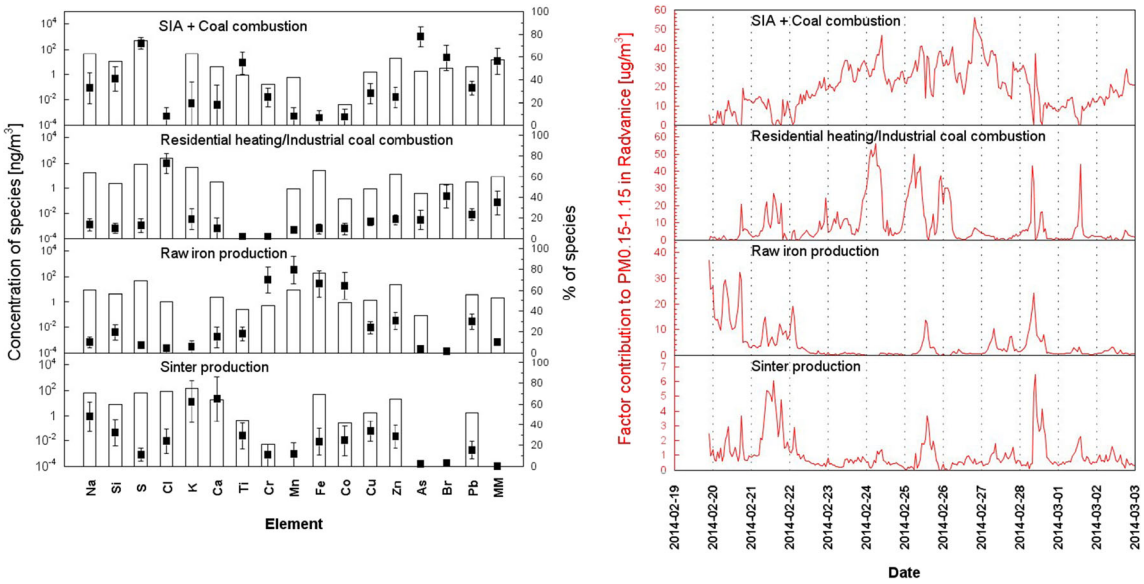

Fig. 3 Radvanice factor profiles (concentration of the species—bars, percentage of the species—markers; DISP Average—box, DISP Max and DISP Min—whiskers) and time series resolved for $PM_{0.15–1.15}$ by PMF.

origin from N over E to S (Fig. 2). The factor shows a moderate and strong relationship with PM_1 K^+, levoglucosan, NH_4^+ and SO_4^{-2}, respectively ($R^2 = 0.40$, $R^2 = 0.44$, $R^2 = 0.48$, $R^2 = 0.80$). In the source apportionment study by Kozáková et al. (2019), secondary sulphate and nitrate of regional origin and biomass burning of local origin contributed to PM_1 by 30%, 26% and 24%, respectively. The diverse result is the consequence of the lack of water-soluble ions in the data matrix and a different monitored period since the SIA and biomass burning peaks from 4 to March 6 are missing in the current study (3DRUM data series from February 19 to March 3).

Factor 3. The third factor was associated with high Cl and Br. There were moderate and strong correlations with the combustion factors (Factor 1 $r = 0.53$ and Factor 4 $r = 0.74$), and statistically significant correlations with PM_1, Cl^-, EC and K^+ ($R^2 = 0.73$, $R^2 = 0.80$, $R^2 = 0.62$). This factor was assigned as industrial coal combustion of regional origin; however, the residential combustion sources should be also considered due to good correlation with EC 24-PM_1, as well as with local residential combustion sources in Plesná. Cl and Br are naturally present in coal as trace elements as well as Hg, As and Se (Finkelman et al. 1993). These elements mostly evaporate during combustion and condense either homogeneously as sub-micron ash or heterogeneously onto already existing fine ash, the former one being more difficult to be captured (Clarke 1993; Vejahati et al. 2010). Zíková et al. (2016) and Li et al. (2019) show the NH_4Cl footprint of coal combustion in Beijing as an ageing product of HCl emissions from local combustion appliances and heating plants. The high factor contributions with WS of > 2 m s^{-1} from the NE point to sources of regional origin (Fig. 2). The three distinct midnight peaks on February 24, 25 and 26 reflect the stagnant atmospheric conditions during winter nights and points to the medium to large combustion source/s of a regional origin. This factor contributed 15% to $PM_{0.15–1.15}$ and corresponds to the coal combustion factor of regional origin apportioned in Kozáková et al. (2019) with contribution of 18% to PM_1.

Factor 4. This factor assigned as local biomass burning contained Zn, Na, MM and K. K also derives from other sources including waste incinerators and cooking (Duan et al. 2004; Wang et al. 2007; Caseiro et al. 2009; Mkoma et al. 2013). The campaign average contribution of this factor to the $PM_{0.15–1.15}$ mass was 12%. The high factor contributions during periods of low WS (< 2 m s^{-1}) point to local sources situated from NW over N to SE from the sampling site. The factor shows a strong relationship with PM_1 levoglucosan and K^+, respectively ($R^2 = 70$ and 0.81, respectively). The daily pattern corresponds to the daily pattern of factor coal combustion (Fig. SI3). Kozáková et al. (2019) present a biomass-burning factor with a contribution of 24% to PM_1.

Factor 5. This factor, re-suspended dust, was associated with the major crustal elements. (Almeida et al. 2005; Rogula-Kozłowska et al. 2012) and on average, contributed 4% to the $PM_{0.15–1.15}$ mass. The dust source was located in the NE quadrant from the sampling site and became more important when WS was of > 2 m s^{-1} (Fig. 2). The re-suspended dust factor was not resolved by Kozáková et al. (2019) due to the limited elemental analyses of the PM samples. Therefore, they lacked the tracers of dust emissions.

Factor 6. The last factor was ascribed to raw iron production. Its profile shows a high concentration of metals especially Mn, Fe and Cu (Querol et al. 2007; Zhou et al. 2004; Cohen et al. 2010). The campaign average of the raw iron factor

contributions was 2%. Source/s of rather regional origin from the NE and E (Fig. 2), and therefore from across the Polish border. However, the local sources in Ostrava could not be overlooked. The regression analysis shows a weak and strong relationship between the raw iron factor and PM_1 Na^+, and K^+ ($R^2 = 0.32$ and $R^2 = 0.71$, respectively). Since the tracers for raw iron factor were missing in the Kozáková et al. (2019) study, the factor was not resolved.

The difference in the number and origin of the factors resolved by PMF based on $PM_{0.15–1.15}$ and PM_1 chemical composition reflects the different measurement period, analysed variables, sampling integration time and modelling approach (two separate matrices in this study and one common matrix for both sites due to the limited number of 24 h samples in Kozáková et al. 2019).

Radvanice

Factor 1. The first factor was associated with high concentrations of S, As, Br and MM. Thus, this factor represents secondary inorganic aerosol (SIA) and coal combustion. The polar plot points primarily to the residential area of Ostrava-Radvanice district situated to the north of the sampling site (Fig. 4). This mixed factor dominated during the measurement campaign (59%). The highest factor contributions to $PM_{0.15–1.15}$ occurred for low WS (< 2 m s^{-1}) and at a temperature lower than the campaign average. The factor shows weak and moderate relationships with PM_1 EC, SO_4^{2-}, and OC ($R^2 = 0.25$, $R^2 = 0.26$, $R^2 = 0.40$). The mixed factor secondary inorganic aerosol/coal combustion/biomass burning (SIA/CC/BB) by Leoni et al. (2018) was also resolved based on the chemical composition of $PM_{0.09–1.15}$. Although the results presented in Leoni et al. (2018) corresponded to a sampling period from February 10 to March 2, 2014, and the present study ran from February 19 to March 3, 2014, the agreement between the two analyses is good.

Factor 2. This factor characterized by Cl, Br and MM was ascribed to local residential heating. However, the industrial coal combustion source/s should be considered since the polar plot shows a high concentration from the north for a WS < 2 m s^{-1} and from the southwest when the WS was between 2 and 4 m s^{-1} (Fig. 4). The chemical profile represents coal and wood combustion in residential boilers and high temperature coke combustion in the metallurgical industry (blast furnace and steel production). The three distinct peaks on February 24, 25 and 26 corresponded to the peaks of the industrial coal combustion factor identified in Plesná. The mean residential heating and industry factor contributed 29% to $PM_{0.15–1.15}$. The regression analysis shows moderate relationships with PM_1 Cl^- and OC ($R^2 = 0.46$, $R^2 = 0.47$, respectively). This factor was not resolved by Leoni et al. (2018). However, the three main peaks between February 24 and 26 correspond to high contributions of the mixed factor SIA/CC/BB of $PM_{0.09–1.15}$.

Factor 3 was identified as raw iron production since its profile was primarily associated with Cr, Mn and Fe. The campaign average of the factor contributions to $PM_{0.15–1.15}$ was 11% that is in agreement with the 10% value reported

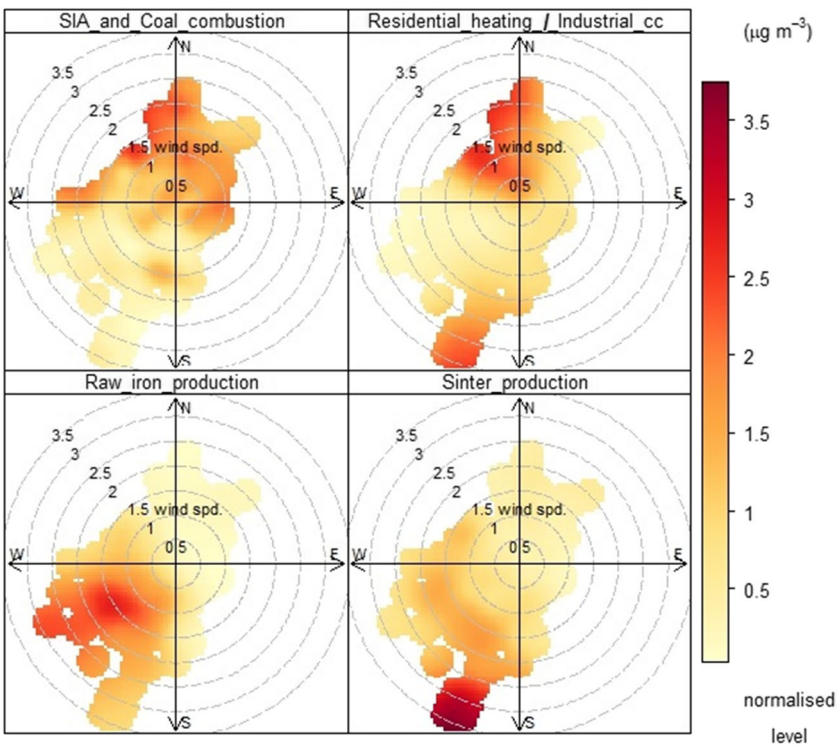

Fig. 4 Polar plots for the Radvanice $PM_{0.15–1.15}$ factors

by Leoni et al. (2018). The high factor contributions from the SW point to local metallurgical sources.

Factor 4. The fourth factor was ascribed to sinter production because of its profile shows high concentration of Na, K and Ca. According to our previous studies (Pokorná et al. 2015), the chemical profile corresponds to sinter production. It does not correspond well to the sinter/steel production factor resolved by Leoni et al. (2018). This factor contributed 1% to $PM_{0.15–1.15}$ with high contribution from SE with WS of 3 m s^{-1}, and therefore, it likely represents the nearby metallurgical complex (Fig. 4). The sinter production factor showed a significant correlation with the raw iron production factor (Factor 4, $r = 0.72$) and weak and very strong correlations with PM_1 Na^+ and K^+ ($R^2 = 0.21$ and $R^2 = 0.83$, respectively). The road dust factor identified by Leoni et al. (2018) was not resolved in this study of $PM_{0.15–1.15.}$ The difference in the number of the factors

Resolved from the $PM_{0.15–1.15}$ and $PM_{0.09–1.15}$, chemical composition was probably driven by the different measurement periods. Additionally, the Pokorná et al. (2015) source apportionment presented five $PM_{0.15–1.15}$ factors at the Radvanice site; coal combustion, sinter production—hot phase, traffic, raw iron production and desulfurization slag processing. The 2012 measurement campaign was affected by a long-term smog period. Therefore, the meteorological conditions were not comparable with the present study. However, there was an overall agreement in the main sources influencing the air quality at the receptor site.

Plesná and Radvanice source comparisons

To assess the relationship between the resolved factor contributions at the two sites, the Pearson correlation coefficients (r) were computed. The mixed factor SIA and coal combustion (*Factor 1, Radvanice*) shows a low correlation with the factors of local residential biomass burning (*Factor 4, Plesná; r = 0.16*), local residential coal combustion (*Factor 1, Plesná; r = 0.21*) and industrial coal combustion (*Factor 3, Plesná; r = 0.25*). Additionally, the mixed factor local residential heating and industrial coal combustion (*Factor 2, Radvanice*) correlated weakly with industrial coal combustion (*Factor 3, Plesná; r = 0.27*). These limited pairwise correlations between the factors time series suggest that different sources were contributing to $PM_{0.15–1.15.}$ These results reflect the important effects of the local meteorology given by the region's orography.

Factors resolved with PNSD and association with chemical composition factors

Plesná

The model was run approximately 100 times to find the most physically meaningful result and the best diagnostics. Due to the low S/N ratio, four variables were set as weak (18.4 nm, 5.1 μm, 6.7 μm and 7.8 μm; midpoint of the size bins) and their uncertainty tripled. Four variables in the coarse size were excluded from the matrix given the high noise level in the data (9.7 μm, 11.9 μm, 14.9 μm, 18.5 μm, midpoint of the size bins). The model was run with different factor numbers (3–9) with 0% extra modelling uncertainty. The most stable solution was found for 6 factors. All runs converged, the scaled residuals were normally distributed, no unmapped factors were detected with bootstrap error estimation and no swaps were observed with displacement error analysis, indicating that the solution was stable (Table S1). The six factors in Plesná were assigned as *residential combustion, urban background, traffic/metallurgical industry, new particle formation (NPF)/metallurgical industry, regional pollution and residential heating/re-suspended dust*. The non-normalized PNSD (N cm^{-3}) was analysed by the model. For the result presentation, the model outputs were converted to dN/d (log Dp) and dV/d (log Dp). The factor NSD and volume size distributions (VSD) are shown in Fig. 5.

Factor 1. This factor includes particles in the Aitken mode and accumulation mode (Aitk2) with the number mode diameter (NMD) at 75 nm and distinct volume mode diameter (VMD) at 274 nm (small VMD at 2 μm, Fig. 5). The average concentration was 2.5×10^3 particle cm^{-3} with a maximum of 1.2×10^4 particle cm^{-3}. This factor is the important contributor for PNC (34%) and a minor contributor to volume concentration (11%). The highest concentrations were under an eastern WD at a WS of 2–3 m s^{-1} (Fig. 6). The diel heating pattern and/or the stagnant atmospheric conditions during winter nights is also obvious (Fig. SI4). There is a moderate correlation with $PM_{0.15–1.15}$ factors of industrial coal combustion ($r = 0.47$), local residential biomass burning ($r = 0.46$), raw iron production ($r = 0.44$) and local residential coal combustion ($r = 0.41$). This factor represents urban background also identified in Radvanice by Leoni et al. (2018) (NMD 93 nm) and similar to the London urban background site reported by Vu et al. (2016) (NMD 93 nm). An urban background factor with morning and evening maxima (Fig. SI4), the same factor daily pattern, attributed to road traffic and residential heating, was observed in the studies by Beddows et al. (2015) and by Masiol et al. (2017).

Factor 2. This includes accumulation mode particles (Acc1) and is the major contributor to PNC (30%) and to volume concentration (38%). The size distribution has a predominant mode, with NMD at 144 nm and VMD at 379 nm (Fig. 5). The average concentration was 2.6×10^3 particle cm^{-3} and peaks up at 6.7×10^3 particle cm^{-3}. The daily variability reflects the heating pattern with morning and afternoon peaks. However, this pattern was also linked to the stagnant atmospheric conditions during winter nights (Fig. SI4). The highest concentrations were observed with a low WS (< 2 m s^{-1}) mainly from the northeast (Fig. 6). In comparison

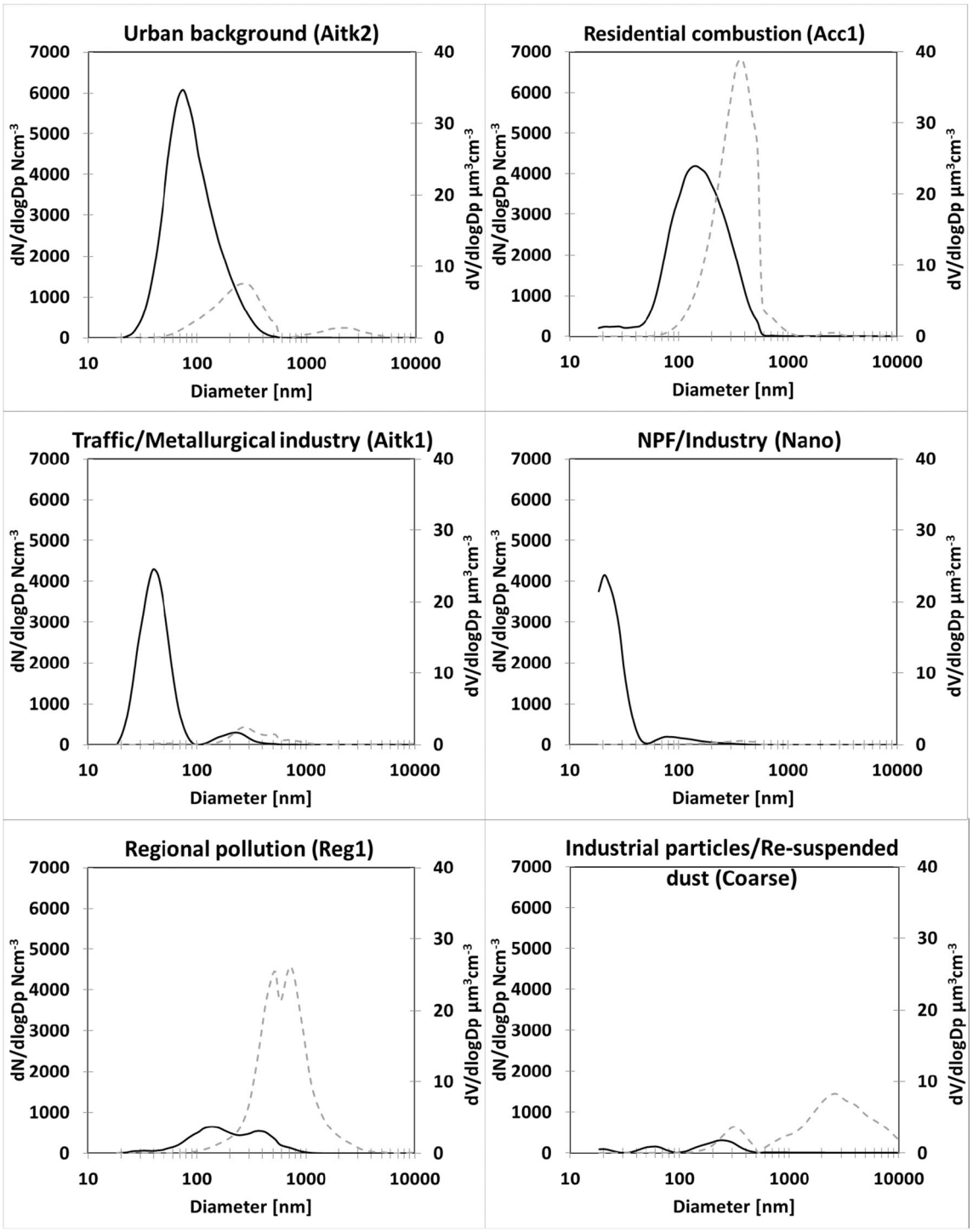

Fig. 5 PNSD PMF factors for Plesná. The volume size distribution (dashed lines) was re-calculated from the PNSD assuming spherical particles

with the factors resolved based on the $PM_{0.15–1.15}$ chemical composition in Plesná, this factor has a moderate correlation with local residential coal combustion ($r = 0.50$), local residential biomass burning ($r = 0.48$) and industrial coal combustion ($r = 0.44$). This factor represents the residential combustion sources of local origin. In Radvanice, a coal combustion factor with NMD at 160 nm and VMD at 500 nm contributed by 14% to PNC and by 42% to PVC (Leoni et al. 2018).

Factor 3 contributed 12% to PNC and 1% to PVC, having a prominent mode at 39 nm (Aitk1, Fig. 5). It shows an average concentration of 7.6×10^3 particle cm^{-3} with a peak at 9.7×10^3 particle cm^{-3}. The polar plot shows high PNC from the north-northeast to south-southeast for $W > 2$ m s^{-1} (Fig. 6). There was

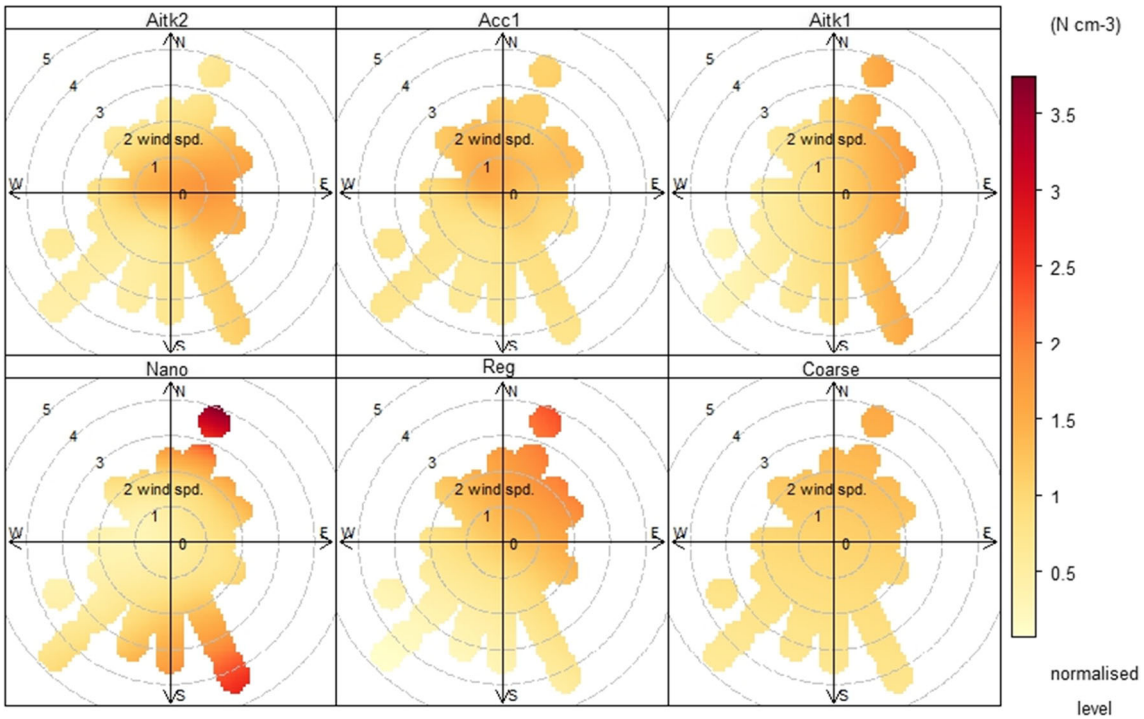

Fig. 6 Polar plots for Plesná NSD factors

no significant correlation with the factors resolved based on the PM chemical composition due to the size range of the impactor. These particles are likely to be from traffic and industrial origins given the prevailing wind direction and the industrial source identification by Leoni et al. (2018) with NMD at 45 nm in Radvanice. Other studies reported a high concentration of ultrafine particles emitted by steel and iron production (Weitkamp et al. 2005; Cheng et al. 2008; Riffault et al. 2015).

Factor 4. This factor includes particles in the nucleation mode (Nano) with a mode at 21 nm (Fig. 5) and a contribution of 8% to PNC. The average particle concentration was 6.0×10^2 particle cm^{-3} with a maximum of 3.7×10^3 particle cm^{-3}. The factor shows a diel pattern with the maxima between 1 PM and 2 PM (Fig. SI4) pointing to photochemical nucleation by the highest solar radiation and particle growth recorded on 26.2, beginning between 1 PM and 2 PM (Fig. SI5). The highest PNC was recorded with WS > 3 m s^{-1} from the north-northeast and south-southeast. According to the polar plots, new particle formation (NPF) events were most probably accompanied by industrial emissions of local and regional origin, from transboundary transport of pollution from the Polish part of Silesia (Fig. 6). The metallurgical industry has been identified as a source of nanoparticles (Dall'Osto et al. 2008; Riffault et al. 2015; Leoni et al., 2016 and 2018); however, no statistically significant correlation with the factors resolved based on the PM chemical composition was found.

Factor 5. It is characterized by accumulation mode particles (Reg) with two NMD (144 and 379 nm) and VMD (524 and 724 nm). This factor is a major contributor to volume concentrations (33%) and a minor contributor to PNC (7%) with an average concentration of 4.9×10^2 particle cm^{-3} and a maximum at 2.3×10^3 particle cm^{-3}. The factor shows the dial course linked to the heating pattern and/or the stagnant atmospheric conditions during winter nights. The highest PNC occurred for northeasterly winds with a WS of > 2 m s^{-1}. The factor correlates moderately with the $PM_{0.15–1.15}$ factors of industrial coal combustion ($r = 0.65$), local residential biomass burning ($r = 0.47$) and secondary inorganic aerosol along with biomass burning.

The last factor, *Factor 6* is characterized by supermicrometre mode particles (Coarse) with appreciable VMD at 3 μm (small VMD at 306 nm, Fig. 5). It contributes 16% to PVC and 2% to PNC. The factor average concentration was 1.4×10^2 particle cm^{-3} with a peak value of 8.8×10^2 particle cm^{-3}. The polar plots showed the highest PNC for northeasterly winds of > 2 m s^{-1} (Fig. 6). The factor does not have a clear diel pattern (Fig. SI4). The factor shows moderate correlation with raw iron production ($r = 0.53$). However, the expected correlation with the re-suspended dust factor is very weak ($r = 0.16$), probably due to the upper cut point of the impactor being 1.15 μm. In comparison with Radvanice, Leoni et al. (2018) identified a mixed industrial coarse particle and road dust factor.

Radvanice

Since the source apportionment of aerosol particles based on PNSD in Radvanice is the subject of the study by Leoni et al.

(2018), only the PNSD sources apportionment in Plesná is fully presented in this study. Briefly, Leoni identified six sources. Mixed source industrial and fresh road traffic, industrial ultrafine particles and urban background contributed most to the PNC (26%, 28%, 24%, respectively). The highest contribution to the particle volume concentration (PVC) was coal combustion along with mixed source industrial coarse particles and road dust (42% and 20%).

For the purpose of this study, the Pearson correlation coefficients (r) between the Radvanice PNSD and $PM_{0.15–1.15}$ resolved factors were computed. Mixed source industrial and fresh road traffic nucleation (*Factor 1, Radvanice*; Leoni et al. 2018) shows moderate to strong correlation with sinter (*Factor 4*) and raw iron production (*Factor 3*) $r = 0.47$ and $0 = 0.74$, respectively. Industrial ultrafine particles (*Factor 2*, *Radvanice*; Leoni et al. 2018) strongly correlated ($r = 0.74$) with sinter production (*Factor 4*). Coal combustion (*Factor 4, Radvanice*; Leoni et al. 2018) shows moderate correlations with mixed sources residential heating and industrial coal combustion (*Factor 2*, $r = 0.64$), and secondary inorganic and coal combustion (*Factor 1*, $r = 0.57$). Regional pollution (*Factor 5, Radvanice*; Leoni et al. 2018) correlated moderately with mixed sources residential heating and industrial coal combustion (*Factor 2*, $r = 0.53$). Mixed source industrial coarse particles and road dust (*Factor 6, Radvanice*; Leoni et al. 2018) shows moderate correlations with raw iron and steel production (*Factor 3 and 4*) $r = 0.49$ and $r = 0.64$, respectively.

Plesná and Radvanice source comparisons

To assess the relationship between the resolved PNSD factors at the two sites, the Pearson correlation coefficients (r) were computed to compare the shared variance. The factor coal combustion (*Factor 4, Radvanice*; Leoni et al. 2018) shows moderate correlation with the Plesná factors of residential combustion of local origin (*Factor 2*) $r = 0.68$ and regional pollution (*Factor 5*) $r = 0.52$. Additionally, the factor regional pollution (*Factor 5*, *Radvanice*; Leoni et al. 2018) correlated moderately with the factor regional pollution in Plesná (*Factor 5*) $r = 0.53$.

Conclusions

This study accessed the spatial and temporal variability of the winter air particulate pollution sources in an urban settlement influenced by metallurgical industry using atmospheric aerosol chemical composition and PNSD. The modelling results confirmed the relevance and importance of the complementary datasets enabling the identification of sources contributing to both particle mass and particle number. While in Radvanice, the PNSD and $PM_{0.15–1.15}$ sources show moderate to strong correlation, in Plesná, the two mixed PNSD sources show weak correlation (traffic and industry—Aitken mode at 39 nm and nanoparticles with a mode at 21 nm) with the $PM_{0.15–1.15}$ sources.

The study results elucidated the importance of the local meteorology given by the region's orography. Similar sources according to the factor chemical profiles resulted from different physical sources. In Plesná, residential combustion sources of local origin followed by regional combustion sources primarily contributed to $PM_{0.15–1.15}$ (Tab. SI4). Surprisingly, the influence of the local industrial emissions was not very important at the site. In Radvanice, local residential combustion and the metallurgical industry were the most important sources (Tab. SI5), which agrees with our previous studies (Pokorná et al. 2015, 2016). Aitken and accumulation mode particles emitted by the urban background (residential heating, industry and traffic) along with local residential combustion sources were the major contributors to PNC in Plesná (Tab. SI4). Additionally, NPF events contributing 8% to PNC were probably mixed with local industrial nanoparticles similar to what was reported in the previous study by Leoni et al. (2018). In contrast, at the Radvanice site, the local industrial sources were the major contributor to PNC (Leoni et al. 2018). However, for a NE prevailing wind with speeds of > 2 m s^{-1}, air pollution transboundary transport and regional sources were evident at both receptor sites in agreement with the results presented by Kozáková et al. (2019). Last but, not least, since the measurement campaign was conducted in winter and the sampling sites are moderately influenced by the traffic density, the contribution of road traffic sources to PM and PNSD seems to be reasonable. However, the measurement of additional road traffic tracers and/or the annual measurement will enable more precise source apportionment of road traffic at both sites.

To conclude, the presented results are representative for winter seasons by the same meteorological conditions, since the meteorology plays a crucial role in the air pollution not only in the Ostrava city, but also in the whole Moravian-Silesian region. Therefore, the long-term complex measurement (highly time- and size-resolved chemical composition and PNSD) at least at three measurement sites well distributed in the city will enable precise source apportionment, and consequent planning and implementation of effective abatement strategies to improve air quality.

The methodology presented in this study can be replicated at any receptor to elucidate the spatial and temporal variability of the air particulate pollution sources contributing to particle mass as well as particle number concentrations.

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References

Almeida AM, Pio CA, Freitas MC, Reis MA, Trancoso MA (2005) Source apportionment of fine and coarse particulate matter in sub-urban area at the Western European Coast. Atmos Environ 39:3127–3138

Ancelet T, Davy PK, Mitschell T, Trompetter WJ, Markwitz A, Weatherburn DC (2012) Source of particulate matter pollution in a small New Zealand city. Environ. Sci. Technol. 46:4767–4774

Ancelet T, Davy PK, Trompetter WJ, Markwitz A, Weatherburn DC (2014) Sources and transport of particulate matter on an hourly time-scale during the winter in a New Zealand urban valley. Urban Clim 10:644–655

Beddows DCS, Harrison RM, Green DC, Fuller GW (2015) Receptor modeling of both particle composition and size distribution. Atmos Chem Phys 15:10107–10125

Carslaw DC, Ropinks K (2012) Openair e an R package for air quality data analysis. Environ Model Soft 27-28:52–61

Caseiro A, Bauer H, Schmidl C, Pio CA, Puxbaum H (2009) Wood burning impact on PM10 in three Austrian regions. Atmos Environ 43:2186–2195

Černikovský L, Krejčí B, Blažek Z, Volná V (2016) Transboundary air pollution transport in the Czech-polish border region between the cities of Ostrava and Katovice. Cent Eur J Public Health 24:45–50

Cheng YH, Chao YC, Wu CH, Tsai CJ, Uang SN, Shih TS (2008) Measurement of ultrafine particles concentrations and size distribution in an iron foundry. J Hazard Mater 158:124–130

CHMI, Czech Hydrometeorological Institute (2018) Air pollution in the Czech Republic in:2018 http://portal.chmi.cz/files/portal/docs/uoco/isko/grafroc/18groc/gr18en/KO_rocenka_2018.pdf

Clarke LB (1993) The fate of trace elements during coal combustion and gasification: an overview. Fuel 72(6):731–736

Cohen DD, Crawford J, Stelcer E, Bac VT (2010) Characterisation and source apportionment of fine particulate matter at Hanoi 2001 to 2008. Atmos Environ 44:230–328

Cusack M, Pérez N, Pey J, Alastuey A, Querol X (2013) Source apportionment of fine PM and sub-micron particle number concentrations at a regional background site in the western Mediterranean: a 2.5 year study. Atmos Chem Phys 13:5173–5187

Dall'Osto M, Booth MJ, Smith W, Fisherb R, Harrison RM (2008) A study of the size distributions and the chemical characterization of airborne particles in the vicinity of a large integrated steelworks. Aerosol Sci Technol 42:981–991

DeGaetano AT, Doherty OM (2004) Temporal, spatial and meteorological variations in hourly PM2.5 concentration extremes in New York City. Atmos Environ 38:1547–1558

Dostál, M., Pastorková, A., Rychlík, S., Rychlíková, E., Švecová, V., Schallerová, E., Šrám, R.J., 2013. Comparison of child morbidity in regions of Ostrava, Czech Republic, with different degrees of pollution: a retrospective cohort study. Environ Health 12, 1–11

Duan M, Zhuang G, Li X, Tao H, Zhuang Y (2004) The characteristics of carbonaceous species and their sources in PM2.5 in Beijing. Atmos Environ 38:3443–3452

Finkelman R, Trace B, minor elements in coal. In Organic Geochemistry; Engel, M. H., Macko, S. A. (eds) (1993) Plenum Press: New York: 593–607

Guan Q, Cai A, Wang F, Yang L, Xu C, Liu Z (2017) Spatio-temporal variability of particulate matter in the key part of Gansu Province, Western China. Environ Pollut 230:189–198

Guerreiro CBB, Foltescu V, de Leeuw F (2014) Air quality status and trends in Europe. Atmos Environ 98:376–384

Harrison RM, Beddows DCS, Dall'Osto M (2011) PMF analysis of wide range particle size spectra collected on a major highway. Environ. Sci. Technol. 45:5522–5528

Heal MR, Kumar P, Harrison R (2012) Particles, air quality, policy and health. Chem Soc Rev 41:6606–6630

Hopke PK (2016) A review of receptor modeling methods for source apportionment. Journal of Air and Waste Management Association 66:237–259

Hovorka, J., Pokorná, P., Hopke, P.K., Křůmal, K., Mikuška, P., Píšová, M., 2015. Wood combustion, a dominant source of winter aerosol in residential district in proximity to a large

Kasumba J, Hopke PK, Chalupa DC, Utell MJ (2009) Comparison of sources of submicron particle number concentrations measured at two sites in Rochester. NY Sci of the Total Environ 407:5071–5084

Kim, E., Hopke P.K., Larson, T.V., Covert, D.S., 2004. Analysis of ambient particle size distributions using Unmix and positive matrix factorization Environ. Sci & Technol 38, 202–209

Kozáková J, Pokorná P, Vodička P, Ondráčková L, Ondráček J, Křůmal K, Mikuška P, Hovorka J, Moravec P, Schwarz J (2019) Influence of regional air pollution transport at a European air pollution hotspot. Environ Sci Pollut Res 26:1675–1692

Kumar P, Hopke PK, Raja S, Casuccio G, Lersch TL, West RR (2012) Characterization and heterogeneity of coarse particles across an urban area. Atmospheric Environ 46:339–359

Lagudu, U.R.K., Raja, S., Hopke, P.K., Chalupa, D.C., Utell, M.J., Casuccio, G.; Lersch, T. L, West, R.R., 2011. Environ. Sci Technol 45,8, 3288–3296

Leoni C, Hovorka J, Docekalova V, Cajthaml T, Marvanova S (2016) Source impact determination using airborne and ground measurements of industrial plumes. Environ. Sci. Technol. 50:9881–9888

Leoni C, Pokorná P, Hovorka J, Masiol M, Topinka J, Zhao Y, Křůmal K, Cliff S, Mikuška P, Hopke PK (2018) Source apportionment of number size distributions and mass chemical composition in a European air pollution hot spot. Environ Pollut 234:145–154

Li XH, Yang K, Han JZ, Ying Q, Hopke PK (2019) Sources of humic-like substances (HULIS) in PM2.5 in Beijing: receptor modeling approach. Sci Total Environ 671:765–775

Masiol M, Vu TV, Beddows DCS, Harrison RM (2016) Source apportionment of wide range particle size spectra and black carbon collected at the airport of Venice (Italy). Atmos Environ 139:56–74

Masiol M, Hopke PK, Felton HD, Frank BP, Rattigan OV, Wurth MJ, LaDuke GH (2017) Source apportionment of PM2.5 chemically speciated mass and particle number concentrations in New York City. Atmos Environ 148:215–229

Mkoma SL, Kawamura K, Fu PQ (2013) Contributions of biomass/biofuel burning to organic aerosols and particulate matter in Tanzania, East Africa, based on analyses of ionic species, organic and elemental carbon, levoglucosan and mannosan. Atmos Chem Phys 13:10325–10338

Moon C (2001) Exposure assessment of air pollutants: a review on spatial heterogeneity and indoor/outdoor/personal exposure to suspended particulate matter, nitrogen dioxide and ozone. Atmos Environ 35:1–32

Moreno T, Kojima T, Amato A, Lucarelli F, de la Rosa J, Calzolai G, Nava S, Chiari M, Alastuey A, Querol X, Gibbons W (2013) Daily and hourly chemical impact of springtime transboundary aerosols on Japanese air quality. Atmos Chem Phys 13:1411–1424

Ogulei D, Hopke PK, Chalupa DC, Utell MJ (2007a) Modeling source contributions to submicron particle number concentrations measured in Rochester. New York Aerosol Sci and Technol 41:179–2001

Ogulei D, Hoppke PK, Ferro AR, Jaques PA (2007b) Factor analysis of submicron particle size distributions near a major United States-Canada trade bridge. Journal of the Air & Waste Management Assoc 57:190–203

Pancras JP, Landis MS, Norris GA, Vedantham R, Dvonch JT (2013) Source apportionment of ambient fine particulate matter in Dearborn, Michigan, using hourly resolved PM chemical composition data. Sci Total Environ 448:2–13

Petit JE, Amodeo T, Meleux F, Bessagnet B, Menut L, Grenier D, Pellan Y, Ockler A, Rocq B, Gros V, Sciare J, Favez O (2017) Characterising an intense PM pollution episode in March 2015 in France from multi-site approach and near real time data: climatology, variabilities, geographical origins and model evaluation. Atmos Environ 155:68–84

Pey J, Querol X, Alastuey A, Rodríguez S, Putaud JP, Van Dingenen R (2009) Source apportionment of urban fine and ultrafine particle number concentration in a Western Mediterranean city. Atmos Environ 43:4407–4415

Pokorná P, Hovorka J, Kroužek J, Hopke PK (2013) Particulate matter source apportionment in a village situated in industrial region of Central Europe. J Air Waste Manag Assoc 63:1412–1421

Pokorná P, Hovorka J, Klán M, Hopke PK (2015) Source apportionment of size resolved particulate matter in European air pollution hot spot. Sci Total Environ 502:172–183

Pokorná P, Hovorka J, Hopke PK (2016) Elemental composition and source identification of very fine aerosol particles in a European air pollution hot-spot. Atmospheric Pollution Research 7:671–679

Polissar AV, Hopke PK, Poirot RL (2001) Atmospheric aerosol over Vermont: chemical composition and sources. Environmental Science & Technology 35:4604–4621

Querol X, Viana M, Alastuey A, Amato F, Moreno T, Castillo S, Pey J, de la Rosa J, Sánchez de la Campa A, Artíñano B, Salvador P, García Dos Santos S, Fernández-Patier R, Moreno-Grau S, Negral L, Minguillón MC, Monfort E, Gil JI, Inza A, Ortega LA, Santamaría JM, Zabalza J (2007) Source origin of trace elements in PM from regional background, urban and industrial sites of Spain. Atmos Environ 41:7219–7231

Riffault V, Arnds J, Marris H, Mbengue S, Setyan A, Alleman LY, Deboudt K, Flament P, Augustin P, Delbarre H, Wenger J (2015) Fine and ultrafine particles in the vicinity of industrial activities: a review. Environ Sci Technol 45(21):2305–2356

Rogula-Kozłowska W, Klejnowski K (2013) Submicrometer aerosol in rural and urban backgrounds in southern Poland: primary and secondary components of PM1. Bull Environ Contam Toxicol 90:103–109

Rogula-Kozłowska W, Klejnowski K, Rogula-Kopiec P, Mathews B, Szopa S (2012) A study on the seasonal mass closure of ambient fine and coarse dusts in Zabrze, Poland. Bull Environ Contam Toxicol 88:722–729

Rogula-Kozłowska W, Błaszczak B, Szopa S, Klejnowski K, Sówka I, Zwoździak A, Jabłońska M, Mathews B (2013a) PM2.5 in the central part of Upper Silesia, Poland: concentrations, elemental composition, andmobility of components. EnvironMonit Assess 185:581–601

Rogula-Kozłowska W, Kozielska B, Klejnowski K, Szopa S (2013b) Hazardous compounds in urban PM in the central part of Upper Silesia (Poland) in winter. Arch Environ Prot 39:53–65

Shen S, Jaques PA, Zhu Y, Geller MD, Sioutas C (2002) Evaluation of the SMPS–APS system as a continuous monitor for measuring PM2.5, PM10 and coarse (PM2.5–10) concentrations. Atmos Environ 36:3939–3950

Sowlat MH, Hasheminassab S, Sioutas C (2016) Source apportionment of ambient particle number concentrations in central Los Angeles using positive matrix factorization (PMF). Atmos Chem Phys 16: 4849–4866

Squizzato S, Masiol M, Emami F, Chalupa DC, Utell MJ, Rich DQ, Hopke PK (2019) Long-term changes of source apportioned particle number concentrations in a metropolitan area of the northeastern United States. Atmosphere 10(27):1–17

Šram RJ, Binková B, Dostál M, Merkerová-Dostalová M, Líbalová H, Milcová A, Rossner P, Rossernová A, Schmuczerová J, Švecová V, Topinka J, Votavová H (2013) Health impact of air pollution to children. Int J Hyg Environ Health 216:533–540

Topinka J, Rossner P, Milcová A, Schmuczerová J, Penčíková K, Rossnerová A, Ambrož A, Štolcpartová J, Bendl J, Hovorka J, Machala M (2015) Day-to-day variability of toxic events induced by organic compounds bound to size segregated atmospheric aerosol. Environ Pollut 202:135–145

Vejahati F, Xu Z, Gupta R (2010) Trace elements in coal: associations with coal and minerals and their behavior during coal utilization − a review. Fuel 89(4):904–911

Viana M, Kuhlbusch TAJ, Querol X, Alastuey A, Harrison RM, Hopke PK, Winiwarter W, Vallius M, Szidat S, Prévôt ASH, Hueglin C, Bloemen H, Wåhlin P, Vecchi R, Miranda AI, Kasper-Giebl A, Maenhaut W, Hitzenberger R (2008) Source apportionment of particulate matter in Europe: a review of methods and results. J Aerosol Sci 39:827–849

Vu T, Delgado Saborit JM, Harrison RM (2015) Review: particle number size distribution from seven major sources and implications for source apportionment studies. Atmos Environ 122:144–132

Vu, T.V., Beddows, D.C.S., Delgado-Saborit, J.M., Harrison, R.M., 2016. Source apportionment of the lung dose of ambient submicrometre particulate matter. Aerosol Air Qual. Res. 16 (7), 1548–1557

Waked A, Bourin A, Michoud V, Perdrix E, Alleman LY, Sauvage S, Delaunay T, Vermeesch S, Petit JE, Riffault V (2018) Investigation of the geographical origins of PM10 based on long, medium and short-range air mass back-trajectories impacting Northern France during the period 2009-2013. Atmos Environ 193:143–152

Wang Q, Shao M, Liu Y, William K, Paul G, Li X, Liu Y, Lu S (2007) Impact of biomass burning on urban air quality estimated by organic tracers: Guangzhou and Beijing as cases. Atmos Environ 41:8380–8390

Wang YG, Hopke PK, Utell MJ (2012) Urban-scale seasonal and spatial variability of ultrafine particle number concentrations. Water. Air and Soil Pollution 223(5):2223–2235

Weitkamp EA, Lipsky EM, Pancras PJ, Ondov JM, Polidori A, Turpin BJ, Robinson AL (2005) Fine particle emission profile for a large coke production facility based on highly time-resolved fence line measurements. Atmos Environ 39(36):6719–6733

Zhou L, Hopke PK, Paatero P, Ondov JM, Pancras JP, Pekney NJ, Davidson CI (2004) Advanced factor analysis for multiple time resolution aerosol composition data. Atmos Environ 38:4909–4920

Zhou, L., Kim, E., Hopke, PK., 2005a. Mining airborne particulate size distribution data by positive matrix factorization. J. of Geophysical Research 110, D07S19

Zhou, L., Hopke, P.K., Stanier, C.O., Pandis, S.N., Ondov, J.M., Pancras, J.P., 2005b. Investigation of the relationship between chemical composition and size distribution of airborne particles by partial least squares and positive matrix factorization J. of Geophysical Research 110, D07S18

Zíková N, Wang YG, Yang FM, Li XH, Tian M, Hopke PK (2016) On the source contribution to Beijing PM2.5 concentrations. Atmos Environ 134:84–95