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Source apportionment of aerosol particles at a European air pollution hot spot using particle number size distributions and chemical composition☆

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A B S T R A C T

Ostrava in the Moravian-Silesian region (Czech Republic) is a European air pollution *hot spot* for airborne particulate matter (PM), polycyclic aromatic hydrocarbons (PAHs), and ultrafine particles (UFPs). Air pollution source apportionment is essential for implementation of successful abatement strategies. UFPs or nanoparticles of diameter <100 nm exhibit the highest deposition efficiency in human lungs. To permit apportionment of PM sources at the hot-spot including nanoparticles, Positive Matrix Factorization (PMF) was applied to highly time resolved particle number size distributions (NSD, 14 nm-10 μm) and $PM_{0.09-1.15}$ chemical composition. Diurnal patterns, meteorological variables, gaseous pollutants, organic markers, and associations between the NSD factors and chemical composition factors were used to identify the pollution sources. The PMF on the NSD reveals two factors in the ultrafine size range: industrial UFPs (28%, number mode diameter - NMD 45 nm), industrial/fresh road traffic nanoparticles (26%, NMD 26 nm); three factors in the accumulation size range: urban background (24%, NMD 93 nm), coal burning (14%, volume mode diameter - VMD 0.5 μm), regional pollution (3%, VMD 0.8 μm) and one factor in the coarse size range: industrial coarse particles/road dust (2%, VMD 5 μm). The PMF analysis of $PM_{0.09-1.15}$ revealed four factors: SIA/CC/BB (52%), road dust (18%), sinter/steel (16%), iron production (16%). The factors in the ultrafine size range resolved with NSD have a positive correlation with sinter/steel production and iron production factors resolved with chemical composition. Coal combustion factor resolved with NSD has moderate correlation with SIA/CC/BB factor. The organic markers homohopanes correlate with coal combustion and the levoglucosan correlates with urban background. The PMF applications to NSD and chemical composition datasets are complementary. PAHs in PM_1 were found to be associated with coal combustion factor.

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1. Introduction

During the last 15 years, the 75% of the air quality monitoring stations in European Union (EU) registered a drop in the concentrations of atmospheric particulate matter (PM, EEA Report 2016). However, the concentrations of PM_{10} and $PM_{2.5}$ still exceed the EU limit values in some regions (EEA, 2016). Recently, ultrafine particles (UFPs, diameter <100 nm) have received great attention because they are particularly hazardous for human health: (i) they can reach the alveolar region of lung; (ii) they have high deposition efficiency (Venkatamaraan, 1999); and (iii) they have orders of magnitude higher surface area to mass ratios compared to larger particles.

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The Moravian-Silesian region (Fig. 1), in the north-eastern part of the Czech Republic, is an EU *hot spot* for air pollution. Epidemiological studies have demonstrated that the air pollution in Ostrava, the major city of the region, significantly affects the health of the population, with an increased rate of respiratory illnesses compared to other regions of the Czech Republic (Šrám et al., 2013a; Topinka et al., 2015). High anthropogenic emissions, due to the steel industry, coke plants, domestic heating, vehicular traffic, and the transport of polluted air masses from Poland, contribute to the worsening of air quality especially in the winter (Mikuška et al., 2015; Pokorná et al., 2015, 2016). The 24-h PM_{10} limit (50 $\mu g\ m^{-3}$) is frequently exceeded (CHMI, 2017) along with elevated concentrations of polycyclic aromatic hydrocarbons (PAHs) (Mikuška et al., 2015). High concentrations of UFPs (up to $1.4 \times 10^5\ cm^{-3}$) highly enriched with PAHs (2.9 mg/g) were observed in the winter 2014 (Leoni et al., 2016). PAHs are formed by the incomplete combustion of fossil fuels and wood, and they have carcinogenic and mutagenic properties (Ravindra et al., 2008).

The planning and the application of abatement strategies to improve air quality are only possible when the pollution sources are identified and apportioned. This is challenging in this location, due to the presence of several sources, some of them situated near urban settlements. The source apportionment is possible through the application of Positive Matrix Factorization (PMF) bilinear model (Polissar et al., 2001; Hopke, 2016), where input data are composed of two matrices of temporal variability of aerosol chemical composition and mass. Highly time resolved aerosol data provide information at temporal resolution capable of identifying not only the main PM sources, but also sources that may have too short duration impact to be observed in 24 h integrated samples (e.g. Elsasser et al., 2012; Ancelet et al., 2012, 2014; Pancras et al., 2013; Moreno et al., 2013; Hovorka et al., 2015). Recently, source apportionment studies focus not only on particle mass, but also NSD (Harrison et al., 2011; Beddows et al., 2015; Masiol et al., 2017; Sowlat et al., 2016). The analysis of the mass chemical composition data distinguish sources contributing mainly to particle mass, while the analysis of the particle NSD identifies sources contributing principally to particle number, enabling the source apportionment down to nanoparticles (Beddows et al., 2015; Masiol et al., 2017).

Recent pollution source apportionment studies in this EU air pollution *hot spot,* based on size segregated aerosol chemical composition (Pokorná et al., 2015, 2016) revealed coal combustion, raw iron production, steel production and traffic being sources of $PM_{0.34\text{-}1.15}$ and road dust source of coarse particles ($PM_{1.15\text{-}10}$). The use of specific molecular markers (Mikuška et al., 2015) revealed combustion of wood and coal, vehicular emissions and industrial production of coke and iron the main $PM_{2.5}$ sources. Despite these studies, detailed pollution source identification down to nanoparticles was not performed yet.

The aim of this study is to give further insights on sources of winter air pollution including nanoparticles at the district Ostrava-Radvanice and Bartovice in, the example of industry impacted residential receptor site. Gaseous pollutants, organic markers and meteorological variables are used to help the source identification. The factors resolved with NSD are compared with mass chemical composition factors and associations are disclosed. Lastly, since PAHs are in high concentration in Ostrava and they are strongly harmful to human health, their association with PMF factors is discussed.

2. Methods

2.1. Experimental

An intensive sampling campaign was performed from 4th February to 7th March 2014 in the residential district of Ostrava – Radvanice and Bartovice, at the same site as in 2012 (Pokorná et al., 2015, 2016; Mikuška et al., 2015). The site was proven being representative at least for the district by $PM_{2.5}$ network measurements (Pokorná et al., 2015). A large metallurgy complex is located 1.5 km southwest of the sampling station (Fig. 1). The instruments were placed in an air-conditioned container. Five-minute integrated particle NSD were measured with a Scanning Mobility Particle Sizer (14–730 nm, SMPS-3936L25, TSI Inc.) and an Aerodynamic Particle Sizer (0.523–10 µm, APS-3321, TSI Inc.). Size segregated PM was collected with a Davis Rotating-drum Uniform-size-cut Monitor – 8DRUM (DELTA Group UC-Davis), from the 10th to 28th of February and is used to provide 2-h resolved PM compositions. Particles were collected on Mylar substrates lightly greased with Apiezon™. The 8DRUM collects particles in 8 size ranges from 0.09 µm to 10 µm. The five smallest size range samples, from 0.09 µm to 1.15 µm, were analyzed for 24 elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Pb)

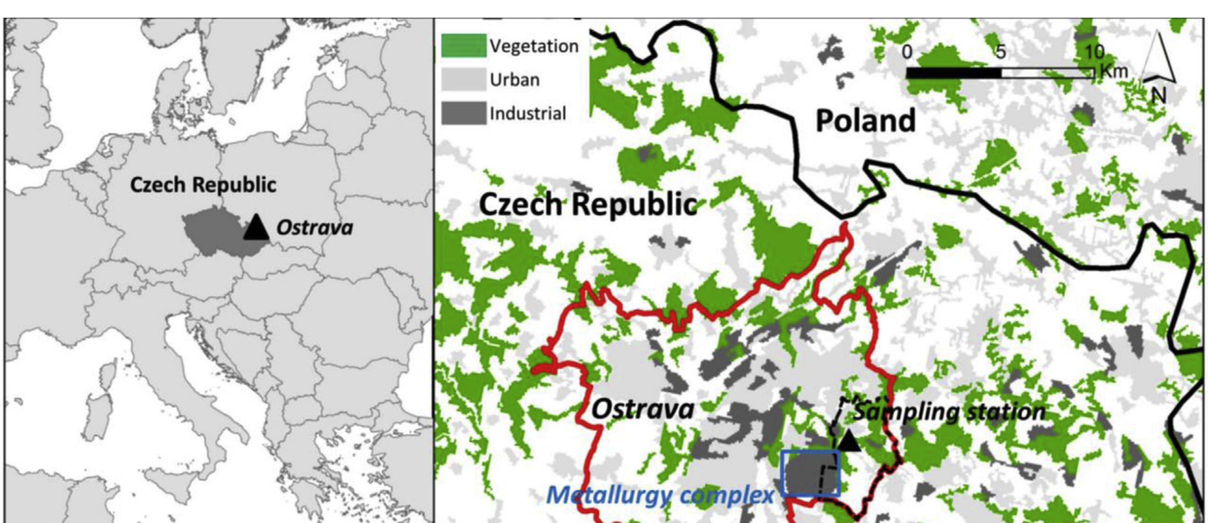

Fig. 1. Left: Czech Republic and Ostrava. Right: Sampling station (triangle), close to the metallurgy complex. Background map: Corine Land Cover 2012 version v.18.5.1.

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