



Mineralogical, chemical and toxicological characterization of urban air particles

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ABSTRACT

Systematic characterization of morphological, mineralogical, chemical and toxicological properties of various size fractions of the atmospheric particulate matter was a main focus of this study together with an assessment of the human health risks they pose. Even though near-ground atmospheric aerosols have been a subject of intensive research in recent years, data integrating chemical composition of particles and health risks are still scarce and the particle size aspect has not been properly addressed yet. Filling this gap, however, is necessary for reliable risk assessment. A high volume ambient air sampler equipped with a multi-stage cascade impactor was used for size specific particle collection, and all 6 fractions were a subject of detailed characterization of chemical (PAHs) and mineralogical composition of the particles, their mass size distribution and genotoxic potential of organic extracts. Finally, the risk level for inhalation exposure associated to the carcinogenic character of the studied PAHs has been assessed. The finest fraction (<0.45 μm) exhibited the highest mass, highest active surface, highest amount of associated PAHs and also highest direct and indirect genotoxic potentials in our model air sample. Risk assessment of inhalation scenario indicates the significant cancer risk values in PM 1.5 size fraction. This presented new approach proved to be a useful tool for human health risk assessment in the areas with significant levels of air dust concentration.

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1. Introduction

Polyaromatic hydrocarbons (PAHs) behaving like persistent organic pollutants (POPs) are of a concern for human health (Bartos et al., 2009; Delgado-Saborit et al., 2011; Shah and Balkhair, 2011; WHO, 2003) as well as for the ecosystems of remote areas (such as the polar regions) as they are bioaccumulative, resist degradation and cycle in the environment for a long time. The atmosphere is a main pathway for their long-range transport even though for most POPs the atmospheric burden is only a small fraction of the total environmental load. As POPs are semivolatile (saturation vapor pressures 10^{-6} – 10^{-2} Pa) they partition between the phases of the atmosphere according to temperature, particulate matter availability and chemical properties. The distribution among the various particle fractions is important as it controls the atmospheric fate. Gaseous molecules on one hand and molecules associated with aerosol particles on the other hand side undergo different degradation and physical removal processes (dry and wet deposition). Particles of different sizes and compositions have very different atmospheric lifetimes.

The processes on aerosol particles and partitioning between the gaseous and particulate phases are, however, insufficiently known (Lohmann and Lammel, 2004) and are to be better understood in

order to describe POPs cycling and assess related impacts to the ecosystems and human health.

The health hazard of ambient air is believed to be determined by conventional gaseous pollutants (ozone, nitrogen oxides) and particulate matter (PM). Numerous scientific studies have linked particle pollution exposure to a variety of health problems, including increased respiratory symptoms, such as irritation of the airways, coughing, difficulty breathing, decreased lung function, aggravated asthma, development of chronic bronchitis, irregular heartbeat, nonfatal heart attacks, and premature death in people with heart or lung disease (Atkinson et al., 2001, 2010; Kan et al., 2007; Li et al., 2011; Pope et al., 2009; Schwartz et al., 1996; Shi et al., 2003). A long-term exposition to elevated levels can cause higher mortality, shorter life, higher incidence of cardiovascular diseases, bronchitis and lung cancers (Parodi et al., 2005). PM is estimated to kill more than 500 000 people worldwide each year (UNEP, 1994). To prevent this loss of life we must understand the characteristics of PM and gain insight into how these characteristics are related to adverse health effects (Nel, 2005).

Particle size and morphology, disregarding its chemical properties, obviously represent one of the hazards. Respirable particles are usually divided into the coarse (diameter more than 2.5 μm), fine (0.1–2.5 μm in diameter) and ultrafine (less than 0.1 μm in diameter) size fractions. Currently, governments and quality monitoring agencies track and regulate 10 μm-diameter (PM₁₀) and 2.5 μm-diameter (PM_{2.5}) particles (European Parliament, 2008). Unfortunately, recent studies suggest that the unregulated ultrafine particles are potentially the most dangerous. Growing attention is given to the potential effects of the ultrafine

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dust particles in the human health because they can penetrate deeper into the respiration tract than fine or coarse particles do (Oberdorster et al., 2002), and cause the respiratory problems (Boldo et al., 2011). Pulmonary effects of PM include the triggering of inflammation in the smaller airways, which can lead to the exacerbation of asthma and chronic bronchitis, airway obstruction, and decreased gas exchange. PM can also interfere with the clearance and inactivation of bacteria in lung tissue.

The chemical composition of the particles themselves, as well as the variety and amount of compounds sorbed on their surfaces, are other factors expectedly responsible for the health effects. In the ambient air, coarse particles are mostly derived from soil and sea salt. Fine and ultrafine particles are predominantly derived from combustion of fossil fuels and transformations of biogenic emissions (Manoli et al., 2002; Ruusunen et al., 2011; Valavanidis et al., 2006). Ultrafine particles are a major component emitted from vehicles propelled by fossil fuel combustion engines. The high temperature processes are considered to be the most significant contribution to air pollution in urban areas (Oberdorster et al., 2002). A very large fraction of the total surface area of PM is associated with this size range. Combustion particles commonly have a core of elemental carbon or fly ash coated with a layer of chemicals including organic hydrocarbons and oxygenated hydrocarbons, metal ions, nitrates, and sulfates. It has been estimated for the urban environment in Europe that 42% of these compounds originate from traffic, 22% from industry, 11% from refineries and energetic sources, and 9% from the local incineration, and all these components may play a role in particle toxicity (Donaldson et al., 2002). There are a lot of mineralogy studies (Ebert et al., 2004; Vester et al., 2007) however connection with other particle characterization (such as toxicology) is missed.

In spite of growing attention is devoted to the effects of PM on human health, there are no complex studies linking the toxicological effects of air particles to the PM size dependent chemical composition and the most important chemical compounds sorbed on their surfaces. Filling this gap is vital for future evaluation of the health effects of various fractions of the atmospheric PM. We have to determine whether in addition to monitoring the mass and the number of particles in the air we also need to consider the chemical composition of the particles as well as a number of toxic chemicals associated with their surfaces when evaluating the effects of emission ultrafine particles. This study also characterizes the human health risk for inhalation exposure to main pollutant PAHs sorbed on air particles. The contribution of individual PAHs congeners to total risks is determined.

In our study, a new combination of methods was used to describe the morphology and sorption potential of various size fractions of the PM, to determine the main chemicals associated with their surfaces and to link it to main toxicological effect (genotoxicity) and related human health risks.

2. Materials and methods

2.1. Air particle sampling procedures

For the purpose of this study, samples of airborne particulate matter (<PM₁₀) were collected in Brno, Czech Republic, in August, 2006. The sampling location was in the industrially influenced area affected also by road traffic and domestic emissions in the city of Brno (WGS 84 coordinates: X: 49.17725, Y: 16.573556, Z: 272).

A high volume ambient air sampler PM-10 (Graseby-Andersen, USA, flow 1.13 m³ min⁻¹, volume 1620 m³ per 24 h) equipped with a multi-stage cascade impactor (Andersen Instruments Incorporated, USA, series 230, model 235) for particle-size fractionation was used for particle sampling. This impactor fractionates suspended particulates into six size fractions (below 10, 7.2, 3.0, 1.5, 0.95, and 0.45 μm). Particles were sampled at quartz slotted collection substrates (Tisch

Environmental, Inc, USA) and quartz filters (Whatman, UK), sampling duration was 7 days.

Each filter has been stabilized for 48 h in a chamber with constant temperature and humidity before weighting. Weighted filters were loaded in to the cartridge, wrapped in two layers of aluminum foil and transported to the sampling site. After exposition the sampling cartridges were wrapped in the new aluminum foil, returned to the laboratory and weighted (Mettler-Toledo GmbH, Switzerland) after another 48 h of stabilization.

2.2. Mineralogical analysis

Scanning electron microscope (SEM) CamScan CS 3200 equipped with microanalytical system Link ISIS 300 (Oxford Instruments) with energy-dispersive SiLi spectrometer (EDS) was applied to characterize morphology and semiquantitative chemical composition. The filters with particulate matter (PM) were coated by carbon and point spectra were measured in spot mode of selected grains, flakes or droplets at magnification 1000×. For bulk composition the 200× magnification was used and an area of 0.2×0.2 mm was measured. The compositional spectra were compared with the EDS library of clay and rock forming minerals (Reed, 1996). After the chemical analysis was done, the samples were coated by gold to reach better morphology resolution. Photomicrographs were taken in secondary electron image (SEI) and backscattered electron image modes (BEI). X-ray diffraction analysis (XRD) was performed using powder diffractometer Philips X'pert MPD system with Bragg–Brentano reflecting geometry and vertical goniometer PW 3020. From each filter a piece ca 10×10 cm was treated by ethanol in an ultrasonic bath and the particles were transferred to a suspension. Ethanol was partly evaporated, the denser suspension was placed on a silicon monocrystal wafer with zero diffraction background and air-dried.

2.3. Chemical analysis

Each particulate fraction was a subject of the chemical analysis with the goal of qualitative and quantitative determination of bound organic compounds with the special focus on PAHs. Our previous experiments indicate that other pollutants like HM or OCPs do not have significant levels on this site. All samples were extracted with 120 ml dichloromethane (DCM) in a Büchi System B-811 automatic extractor (Büchi, Switzerland). Surrogate recovery standards (D8-naphthalene, D10-phenanthrene, D12-perylene) were spiked on each filter prior to extraction. The solvent was evaporated and then condensed back to sample over 60 min. Volume was reduced after extraction under a gentle nitrogen stream at ambient temperature, and fractionation achieved on silica gel column (30 cm length, 1 cm i.d.). Sample was added to activated silica gel in column (activation: 12 hod, 150 °C). Column was rinsed with n-hexane from aliphatic fraction. Dichloromethane was used for elution of PAHs. Terphenyl was used as an internal standard. Samples were analyzed using a GC–MS instrument (HP 6890–HP 5973, Agilent Technologies, Germany) supplied with a J&W Scientific fused silica column DB-5MS (60 m×0.25 mm i.d.×0.25 μm of stationary phase). Helium was used as an inert carrier gas. Injection of sample was automatic in splitless mode (1 μl, 280 °C). Details of temperature program: 80 °C (1 min), 15 °C min⁻¹ to 180 °C, 5 °C min⁻¹ to 310 °C (10 min). Temperature of transfer line was 280 °C and of ion source was 200–300 °C. The 29 polyaromatic hydrocarbons (naphthalene, biphenyl, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzonaphtho-thiophene, benzo[b]fluorene, benzo[g,h,i]fluoranthene, cyclopenta[c,d]pyrene, benz[a]anthracene, triphenylene, chrysene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, perylene, indeno[1,2,3-c,d] pyrene, dibenz[a,h]anthracene, dibenz[a,c]anthracene, benzo[g,h,i]perylene, athanthrene, coronene) were analyzed (more details see (Dvorska et al., 2012)).

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