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Oxidative potential and chemical composition of $PM_{2.5}$ in office buildings across Europe – The OFFICAIR study

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ABSTRACT

In the frame of the OFFICAIR project, indoor and outdoor $PM_{2.5}$ samples were collected in office buildings across Europe in two sampling campaigns (summer and winter). The ability of the particles to deplete physiologically relevant antioxidants (ascorbic acid (AA), reduced glutathione (GSH)) in a synthetic respiratory tract lining fluid, i.e., oxidative potential (OP), was assessed. Furthermore, the link between particulate OP and the concentration of the PM constituents was investigated.

The mean indoor $PM_{2.5}$ mass concentration values were substantially lower than the related outdoor values with a mean indoor/outdoor $PM_{2.5}$ mass concentration ratio of 0.62 and 0.61 for the summer and winter campaigns respectively. The OP of $PM_{2.5}$ varied markedly across Europe with the highest outdoor OP^{AA} m^{-3} and OP^{GSH} m^{-3} (% antioxidant depletion/m^3 air) values obtained for Hungary, while $PM_{2.5}$ collected in Finland exhibited the lowest values. Seasonal variation could be observed for both indoor and outdoor OP^{AA} m^{-3} and OP^{GSH} m^{-3} with higher mean values during winter. The indoor/outdoor OP^{AA} m^{-3} and OP^{GSH} m^{-3} ratios were less than one with 4 and 17 exceptions out of the 40 cases respectively. These results indicate that indoor air is generally less oxidatively challenging than outdoors. Correlation analysis revealed that trace elements play an important role in determining OP, in particular, the Cu content. Indoor air chemistry might affect OP since weaker correlations were obtained for indoor $PM_{2.5}$. Our findings also suggest that office workers may be exposed to health relevant PM constituents to a different extent within the same building.

1. Introduction

Numerous epidemiological and toxicological studies have shown a relationship between ambient particulate matter (PM) exposure and adverse health effects in humans (Brook et al., 2010; Hoek et al., 2002; Laden et al., 2000; Schwartz et al., 2002). Different pathophysiological mechanisms have been proposed to explain PM's contribution to respiratory and cardiovascular diseases; however, there are still remaining questions to be answered. Many of the air pollution related health outcomes are thought to derive from oxidative stress initiated by certain gaseous air pollutants (i.e., nitrogen dioxide, ozone) or PM constituents in the lung (Ayres et al., 2008; Kelly, 2003; Nel, 2005). The pulmonary epithelial cells are protected against undue oxidation by the respiratory tract lining fluid (RTLF) which contains low-molecular weight antioxidants (i.e., reduced glutathione, ascorbate, urate) and antioxidant enzymes. Either the increased exposure to oxidants or the presence of decreased antioxidant defenses could lead to oxidative stress which in turn can activate a number of redox sensitive signaling pathways (Anseth et al., 2005; Kelly, 2003; Kelly and Fussell, 2012). However,

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the identification of the oxidatively active components of PM is still a challenging task since particles vary in size, mass, number, shape, aggregation status, surface area as well as chemical composition.

Oxidative potential (OP) is a novel metric which is defined as a measure of the capacity of PM to oxidize target compounds. In vitro acellular methods based on different principles (i.e., antioxidant depletion, hydroxyl radical formation in the presence of H_2O_2, consumption of dithiotreitol, fluorescent probes) have been developed and used for the assessment of the OP of aerosol particles. Previous studies suggest that various chemical compounds such as certain transition metals (e.g., Fe, Cu, Cr), aromatic organic compounds (e.g., quinones) and some major PM constituents (e.g., humic-like substances) contribute to particulate OP (e.g., Godri et al., 2010; Roginsky et al., 1999; Szigeti et al., 2015; Verma et al., 2012). The importance of the water-soluble part of the trace elements has also been proposed since higher correlations may occur between them and the OP metrics compared to the total trace element concentrations (Szigeti et al., 2015). Although, several studies have linked increased $PM_{2.5}$ mass concentration to adverse health effects, the use of OP in epidemiology has also been suggested as a promising metric since it may better represent the health impact of the aerosol particles (Borm et al., 2007; Boogaard et al., 2012; Szigeti et al., 2015; Yang et al., 2015). However, there is still no consensus about which OP metrics are the most appropriate to predict PM-related health effects.

Outdoor locations (i.e., urban, rural, industrial sites) are well characterized (in terms of PM mass concentration and chemical composition) over almost the entire world due to the existing regulations on the mass concentration of PM_{10} and $PM_{2.5}$; however, less information is available about PM in indoor microenvironments, particularly in offices (Chatoutsidou et al., 2015; Sangiorgi et al., 2013; Saraga et al., 2011; Szigeti et al., 2014). Office buildings are generally located in big cities near traffic intersections and busy roads in order to make them easily accessible for the employees. These buildings are generally equipped with heating, ventilation and air conditioning (HVAC) system to improve the air quality and create an acceptable feeling of comfort for the office workers. The inlet of the HVAC system is generally located on the roof of the buildings where the air is thought to be less polluted due to the distance from the traffic-related sources compared to the ground level. The filtering effect of the mechanical ventilation system, indoor sources and sinks of aerosol particles as well as the different microclimatic conditions (i.e., temperature, relative humidity) all have an influence on the size and chemical composition of the indoor particles as well as on the indoor/outdoor PM mass concentration ratio (Meng et al., 2007). However, it is still a challenge to determine the indoor generated part of the concentration of indoor $PM_{2.5}$ mass and the PM constituents. Substantial fraction of outdoor particulate matter pollution infiltrates indoors (Hänninen et al., 2004). Building occupants have some influence on the infiltration by operating windows and doors and many studies have shown that during the summer time infiltration levels are higher than in winter (Hänninen et al., 2011).

Many indoor activities (e.g., cooking, smoking, incense and candle burning) as well as re-suspension of settled dust may contribute to indoor $PM_{2.5}$ mass concentration in residential homes (Urso et al., 2015); however, fewer indoor sources are apparent for offices. Particle emission from printers, photocopiers and multi-task devices are well-known sources for ultra-fine particles, but these have almost no impact on the $PM_{2.5}$ mass concentration (e.g., Destaillats et al., 2008; He et al., 2007). Re-suspension of settled particles (with aerodynamic diameter larger than 1 μm) from indoor surfaces are common particle sources in offices (Chatoutsidou et al., 2015). Some specific activities (i.e., enveloping) could also contribute to the concentration of certain PM constituents (Szigeti et al., 2014). Furthermore, in-situ ozone-initiated chemistry with reactive volatile organic compounds could lead to the formation of secondary organic aerosol (Weschler and Shields, 1999). However, it is likely that particles of outdoor origin are still the most important determinants of $PM_{2.5}$ in these environments compared to the indoor generated particles.

The spatial and temporal variation of particulate OP is not as well characterized as the mass concentration and chemical composition of PM. Moreover, only limited information is available about the OP of indoor PM. In the frame of the European Union project OFFICAIR (on the reduction of health effects from combined exposure to indoor air pollutants in modern offices), indoor and outdoor $PM_{2.5}$ samples were collected in office buildings to facilitate (i) the OP assessment and chemical characterization of the particles and (ii) the investigation of the relationship between particulate OP and PM constituents. Furthermore, the aim of this study was to fill gaps and answer questions regarding the indoor and outdoor environment.

2. Experimental

2.1. Description of sampling sites and instrumentation

The selection of the office buildings was made according to predefined criteria developed during the OFFICAIR project (Bluyssen et al., 2015). For highlighting regional similarities and differences in aerosol characteristics, sampling sites were selected in different regions across Europe. $PM_{2.5}$ samples were collected in Finland (FI), Greece (GR), Hungary (HU), Italy (IT) and The Netherlands (NL) in a total of 20 office buildings ($n = 5$ for GR and HU, $n = 4$ for IT and $n = 3$ for FI and NL). The location of the sampling sites is depicted in Fig. A.1. $PM_{2.5}$ was sampled at one indoor and one outdoor location per building for approximately 100 h (from Monday 9 AM until Friday 5 PM) during summer 2012. The sampling campaign was repeated in winter 2012/2013. In order to investigate spatial variability within an office building, parallel indoor sampling ($n = 2$) was performed in one Dutch building (NL3) in April and May 2013. The indoor and concomitant outdoor $PM_{2.5}$ samples were collected onto quartz fiber filters (Ø 47 mm and Ø 37 mm, Whatman QM-A) supplied by GE Healthcare (Little Chalfont, Buckinghamshire, UK). Field blank samples were also collected. Before sampling, filters were wrapped in aluminium foil and pre-treated at 550 °C in an electric oven for 8 h in order to eliminate any possible organic contaminants. Thereafter filters were conditioned in an acclimatized room for 48 h at 20 ± 1 °C and 50 ± 5% relative humidity, and then weighed on a Mettler Toledo XP26DR balance with a readability of 2 μg. Low-volume aerosol samplers equipped with a $PM_{2.5}$ head operating at a constant flow rate of 0.24–2.3 $m^3\ h^{-1}$ were used. We performed our PM sampling method as well as the gravimetric analysis in accordance with the BS EN 12341:2014 standard. The sampling head was placed at 1.2 m height (sitting height) in the indoor locations. Two sampling strategies were applied for the outdoor $PM_{2.5}$; at the air inlet of HVAC systems or at the same height as that which the indoor sampling took place since, in some cases, it was not possible to perform the outdoor sampling at the inlet of the HVAC system. In the case of Greece and Hungary, all buildings were selected in the corresponding capital cities: Athens and its metropolitan area and Budapest. Two of the Dutch monitored buildings were located in Delft, while one was in the resort settlement of Noordwijk. Two of the Italian buildings chosen were in Florence, one in the center of Milan and another one in the metropolitan area of Milan. Furthermore, office buildings located in a small (Varkaus) and in a medium sized city (Kuopio) were selected in Finland. The main characteristics of the monitored buildings are compiled in Table 1. The windows of the offices were generally kept closed throughout the study; however, episodes of opened windows were registered during the summer sampling campaign in some office buildings. During the sampling period, indoor and outdoor air temperature and relative humidity were monitored close to the PM samplers with a time resolution of 5 min.

2.2. Chemical characterization of $PM_{2.5}$

Loaded as well as field blank filters were subjected to different chemical analyses in order to determine the concentration of trace