Spatial and temporal variation of particulate matter characteristics within office buildings — The OFFICAIR study

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HIGHLIGHTS

• PM characteristics varied markedly among the European office buildings.
• One order of magnitude difference was observed for the PNC and the OP metrics.
• Spatial variation within a building is less significant than temporal variation.
• Trace element concentrations showed the largest variation within a building.
• Differences within a building might be more relevant in long term than short term.

GRAPHICAL ABSTRACT

In the frame of the OFFICAIR project, office buildings were investigated across Europe to assess how the office workers are exposed to different particulate matter (PM) characteristics (i.e. PM2.5 mass concentration, particulate oxidative potential (OP) based on ascorbate and reduced glutathione depletion, trace element concentration and total particle number concentration (PNC)) within the buildings. Two offices per building were investigated during the working hours (5 consecutive days; 8 h per day) in two campaigns. Differences were observed for all parameters across the office buildings. Our results indicate that the monitoring of the PM2.5 mass concentration in different offices within a building might not reflect the spatial variation of the health relevant PM characteristics such as particulate OP or the concentration of certain trace elements (e.g., Cu, Fe), since larger differences were apparent within a building for these parameters compared to that obtained for the PM2.5 mass concentration in many cases. The temporal variation was larger for almost all PM characteristics
1. Introduction

There is an increasing concern on indoor air quality, as in our modern world, people spend about 80–90% of their time indoors. Modern office buildings became one of the most important workplaces in the past decades due to (i) the continuous growth and modernization of big cities and (ii) the shift in the economic sectors towards services (e.g., banking, information services). The exposure of office workers to health relevant air pollutants is still not fully characterized; however, some studies have recently been performed regarding indoor air quality in offices (e.g., Chatoutsidou et al., 2015; Nørgaard et al., 2014; Sangiorgi et al., 2013; Szigeti et al., 2014, 2016; Wolkoff, 2013). OFFICAIR (on the reduction of health effects from combined exposure to indoor air pollutants in modern offices) is the first large-scale international research project which is devoted, among others, to assess indoor air quality in modern office buildings across Europe and to answer numerous questions regarding the office environment (Blyussken et al., 2016; Mandin et al., 2017).

Particulate matter (PM) is one of the criteria air pollutants which serves as an indicator of both indoor and outdoor air quality. PM encompasses many different chemical components and physical characteristics, many of which are potential contributors to adverse health effects. The evidence is on the PM$_{2.5}$ size fraction (particles with aerodynamic diameter smaller than 2.5 μm) since several epidemiological and toxicological studies have already shown the association between the mass concentration of PM$_{2.5}$ and certain respiratory and cardiovascular diseases or mortality (e.g., Brook et al., 2010; Hoek et al., 2002; Laden et al., 2000). In the past years, oxidative potential (OP), which is a measure of the capacity of PM to oxidize target molecules (e.g., physiologically relevant antioxidants), has been proposed as an additional or alternative exposure metric alongside PM mass concentration (Boogaard et al., 2012; Borm et al., 2007; Szigeti et al., 2015; Yang et al., 2015). However, different methods have been developed for the assessment of particulate OP (Ayres et al., 2008) and there is still no consensus about which OP metrics are the most appropriate to predict PM-related health effects. Furthermore, only limited experimental data is available on the OP of different PM fractions especially for the indoor environment since it is a novel metric. The link between the OP metrics and different PM constituents has already been investigated and the results suggest that a certain panel of trace elements (e.g., Cu, Fe, Cr) is a major determinant of particulate OP (e.g., Godri et al., 2010a, 2011; Szigeti et al., 2015, 2016). Moreover, other PM constituents (e.g., quinones, humic-like substances, sulfate ion) may contribute to particulate OP (e.g., Godri et al., 2010a; Roginsky et al., 1999; Szigeti et al., 2015; Verma et al., 2012). Lastly, the particle number concentration (PNC) has also been considered as a relevant metric to characterize PM as ultrafine particles (UFPs; particles with aerodynamic diameter smaller than 100 nm), the dominant particles regarding PNC (Borsos et al., 2012; Crilley et al., 2012), represent a significant health risk (Delfino et al., 2006; Iahl-Muli et al., 2002). The evidence on health effects of long-term exposure to UFPs is still limited as they are usually not monitored routinely as well as the few epidemiological studies on UFPs and mortality reported inconsistent results (Lanzinger et al., 2016). Accordingly, the confidence on UFPs is still missing to allow introducing legislation specifically directed to this PM size fraction.

Since smoking is not permitted in the office buildings, resuspension of settled dust and particle emission from printers, photocopiers and multi-task devices are well-known indoor sources for larger particles (with aerodynamic diameter larger than 1 μm) and UFPs respectively (e.g., Chatoutsidou et al., 2015; Destaillats et al., 2008; He et al., 2007). However, it is likely that particles of outdoor origin are still the most important determinants of PM$_{2.5}$ mass concentration in these environments compared to the indoor generated particles (Szigeti et al., 2016). In contrast, UFP emission from office equipment can considerably increase the background PNC of these particles temporarily. The emission from these devices is well characterized by chamber experiments (e.g., He et al., 2007; Morawska et al., 2009). However, less data is available on the real exposure of office workers to PM mass and number concentration generated by office equipment (McGarry et al., 2011). Hanninen et al. (2010) concluded that the health risk associated with the daily uptake and lung deposition of particles emitted from printers is low and also substantially lower than the estimated risk linked to the exposure to ambient particles.

In our previous paper (Szigeti et al., 2016), we have tried to answer several questions regarding the office environment (i.e., “Which are the major determinants of indoor PM$_{2.5}$ mass concentration?”; “Indoor air vs. outdoor air: Where is it better to stay?”; “Which are the relevant PM constituents regarding OP?”) in the frame of the OFFICAIR project. We concluded that indoor air is generally characterized by lower oxidative potential values than the outdoor air. The results also suggested that office workers might be exposed to health relevant PM characteristics (e.g., trace elements, particulate OP) at a different extent within an office building; however, this conclusion was based only on limited experimental data. Thus one of the main goals of this study was to strengthen or reject this statement by simultaneous PM$_{2.5}$ sampling during the working hours at two indoor locations per building in two sampling campaigns. Furthermore, we aimed to investigate the spatial and temporal variation of the PNC in the offices and to study the influence of certain environmental parameters and human activities (e.g., windows opening, printing) on it.

2. Methods

2.1. Study design and sampling

In total, nine mechanically ventilated office buildings were investigated in France (n = 1), Greece (n = 2), Hungary (n = 1), Italy (n = 2), The Netherlands (n = 1) and Portugal (n = 2) between February 2013 and February 2014. The selection of the office buildings was based on some predefined criteria developed during the OFFICAIR project (Blyussken et al., 2016). All buildings were modern or retrofitted. The location of the sampling sites is depicted in Fig. 1 and some office characteristics are listed in Table 1.

PM$_{2.5}$ samples were collected at two indoor locations per office building during the working hours (5 consecutive days; 8 h per day; without filter replacement) in two sampling campaigns (2 × 1 week; 5 weeks difference between the two campaigns). Samples were collected by low-volume aerosol samplers operating at a flow rate of either 1.0 or 2.3 m$^3$ h$^{-1}$ onto quartz fiber filters (Ø 47 m, Pallflex® Tissuquartz) supplied by Pall Corporation (Port Washington, NY, USA). It must be
noted that two identical PM$_{2.5}$ samplers were used within the same office building. The sampling head was placed at 1.2 m height which approximately corresponds to the breathing zone of a sitting person. Field blank samples were also collected at each building. Before sampling, filters were 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. After sampling, loaded and field blank filters were shipped in cooled packages to the central laboratory to determine the collected PM$_{2.5}$ mass. Afterwards, one third of each loaded/blank filter was cut out by a ceramic lance and the concentration of seven trace elements (Al, V, Mn, Fe, Cu, Zn and Pb) was determined by inductively coupled sector field plasma mass spectrometry (ICP-SF-MS; ELEMENT2; ThermoFinnigan, Germany) after microwave-assisted aqua regia extraction by the method described elsewhere (Mihucz et al., 2015). Particulate OP was determined by antioxidant depletion using a synthetic respiratory tract lining fluid containing urate, ascorbate and reduced glutathione in a physiologically relevant concentration of 200 μmol L$^{-1}$ each. The whole procedure has been described in details previously (Szigeti et al., 2014, 2016). In the case of each loaded/blank filter, three filter cuts (Ø 5 mm) were subjected to the OP analysis. The OP was expressed in % antioxidant depletion/m$^3$ air and in % antioxidant depletion/µg PM. Field blank filters were also analyzed in the identical way and the results were used for blank corrections. An additional PM$_{2.5}$ sample was collected in one of the offices for method validation purposes. The loaded filter was cut into three equal strips and analyzed by the ICP-SF-MS method to investigate the uncertainty related to the determination of trace elements.

Total PNC values were also monitored with a time resolution of 1 min in the offices by different condensation particle counters (CPC; Model 3007, 8020 or 8525; TSI Incorporated, Shoreview, MN, USA) simultaneously with the PM$_{2.5}$ sampling (from 9:00 AM to 5:00 PM). The CPC Model 3007 counts the particles with diameter larger than 10 nm, while the other two CPCs (Model 8020 and 8525) only detect particles with diameter larger than 20 nm. However, the difference in the working range of the instruments does not cause considerable differences in the PNC values at low concentration levels ($<5 \times 10^4$ cm$^{-3}$; difference $<20$%; Cattaneo et al., 2009). Furthermore, the same type of device was used in both offices within an office building. Quality control check (i.e., zero reading by using HEPA filter) was carried out on the CPCs daily. The concentration of nitrogen dioxide (NO$_2$) and ozone (O$_3$) was also monitored in the offices with a time resolution of 1 min by Aeroqual™ S500 portable air quality sensors (Aeroqual Ltd., Auckland, New Zealand). The position of all equipment was selected carefully to avoid the direct influence of nearby occupants. A detailed time activity diary (i.e., number of people in the office, number of printed pages, windows opening) was filled in during the field campaigns for each office and was used to investigate the association among the different environmental factors and indoor air quality parameters.

2.2. Data analysis

Spearman’s rank correlation coefficients ($r_s$) with a two-tailed test of significance ($p$) were produced to show relationships among different PM characteristics by using the software package of IBM SPSS Statistics.
for Windows, version 21 (IBM Corp., Armonk, NY, USA). Significant correlation was defined as $p < 0.05$. The Shapiro-Wilk test was used to test the normality of the data distribution.

To assess the spatial variation of different PM characteristics within the office building, i.e., the difference between the results obtained for the two offices in the same building at the same time, we calculated the intra-class correlation (ICC) coefficients with a two-stage hierarchical linear regression model (office and building levels). The temporal variation was also characterized by the ICC coefficients.

The calculations were based on the following theory:

$$ y_{ijk} = \mu_i + \nu_{ijk} = \mu_i + \epsilon_{ijk} $$

(1)

where $i, j$ and $k$ represent the number of rooms ($i = 1, 2$); the number of buildings ($j = 1, \ldots, 9$) and number of field campaigns ($k = 1, 2$), respectively, $y_{ijk}$ is the measured value of a PM characteristic ($Y$) in room $i$ within building $j$ during campaign $k$, $\mu_i$ is the true mean value, $\nu_{ijk}$ is the deviation from $\mu_i$ due to the effect of campaign $k$, $\epsilon_{ijk}$ is the deviation from campaign $k$ mean due to the effect of building $j$, and $\epsilon_{ijk}$ is the deviation from building $j$ due to the effect of room $i$. The variance components are $\text{var (} \epsilon_{ijk} \text{)} = \sigma^2_\epsilon$, $\text{var (} \nu_{ijk} \text{)} = \sigma^2_v$ and $\text{var (} \mu_i \text{)} = \sigma^2_\mu$. $\sigma^2_\mu$ represents the variance obtained between rooms, $\sigma^2_v$ is for the level-2 (or inter-building) variance and $\sigma^2_\epsilon$ is related to the variance obtained between campaigns. The ICC coefficients were calculated as follows:

$$ \text{ICC (spatial variation)} = \frac{\sigma^2_\mu}{\sigma^2_\mu + \sigma^2_v + \sigma^2_\epsilon} $$

(2)

$$ \text{ICC (temporal variation)} = \frac{\sigma^2_v}{\sigma^2_\mu + \sigma^2_v + \sigma^2_\epsilon} $$

(3)

The lower the ICC coefficient is, the more variable the PM characteristic is. In the case of PNC, the median of the weekly data was used in the calculations. In the case of other PM characteristics, only the 5-day mean values were available.

In the case of trace elements and particulate OP, the uncertainty of the measurements was calculated to assess whether the uncertainty of the measurements is negligible compared to the spatial/temporal variation of the PM characteristics or not. Only one PM$_{2.5}$ sample was analyzed for this purpose in the case of trace elements; however, more data was available in the case of OP since three parallel filter cuts were subjected to the OP assessment in all cases. The total variance was calculated as the sum of the different variance components (spatial, temporal, residual and uncertainty). Then, the contribution of the variance component related to the uncertainty of the measurements to the total variance was expressed as their ratio.

3. Results and discussion

3.1. Concentration of primary indoor air quality indicators

Table 2 represents the PM$_{2.5}$ mass concentration, PNC and NO$_2$ concentration values obtained for the two sampling campaigns. The PM$_{2.5}$ mass concentration values ranged between 4.7 and 37.6 µg m$^{-3}$ with a median value of 11.7 µg m$^{-3}$. This difference is in line with our previous observations when the indoor PM$_{2.5}$ mass concentration values varied between 2.6 and 31.2 µg m$^{-3}$ in the 20 office buildings investigated across Europe (Szigeti et al., 2016). The lowest values were obtained for the Dutch offices which were the only unoccupied offices (meeting rooms) during the sampling campaigns. There is no dedicated guideline or toxicological reference value for the indoor PM$_{2.5}$ mass concentration, thus it is difficult to perform risk assessment in terms of potential adverse health effects. However, the PM$_{2.5}$ mass concentration values exceeded the ambient WHO guideline of 25 µg m$^{-3}$ (24-h mean; WHO, 2005) in 12.5% of the cases. It must be noted that the sampling was undertaken in different seasons and it is known that (i) the ambient PM$_{2.5}$ mass concentration values might show seasonal variation in Europe (e.g., Putaud et al., 2010; Szigeti et al., 2015) and (ii) the outdoor PM$_{2.5}$ mass concentration is one of the major determinants of the corresponding indoor value (Szigeti et al., 2016). Thus the comparison across the sites and seasons is difficult. Accordingly, the results represent a case...
study for each building rather than a representative picture. Temporal variation in the PM$_{2.5}$ mass concentration values could be observed for the sampling sites; however, the variation is low according to the results of the ICC analysis (ICC = 64%) (Table 3). The one order of magnitude difference between these values is not surprising since similar differences were reported in the literature for different days (from 9:00 AM to 5:00 PM in each building) even within a city (Salma et al., 2014). In contrast to the outdoor environment and the particles penetrate into the indoor environment and the particles penetrate into the indoor environment, high RSD values ranging from 12 to 46% (mean: 28%) were obtained which indicates that the PNC values between the two sampling campaigns (Table 3). The day to Friday) varied considerably among the buildings in Brisbane, Australia and slightly lower mean PNC values were obtained for the Greek offices where the PNC values occasionally reached the upper limit of the CPCs (10$^3$ particles cm$^{-3}$). In some cases (e.g., HU6), a broad and large peak with a maximum between 12:00 and 13:00 appeared (Fig. 2a) which is an indicator of particle formation event outdoors (Salma et al., 2011). The lack of the above-mentioned peak corresponds to a day without particle formation event (Fig. 2b) which is an indicator of particle formation event outdoors. Salma et al. (2011) monitored the PNC values at the same site investigated in this study in Hungary (HUF) and they observed similar curves for the outdoor environment than our results for the indoor environment. Since the monitoring was carried out during the working hours only, the morning and evening peaks related to traffic emissions cannot be clearly seen. It must be noted that the particle formation more likely takes place in the outdoor environment and the particles penetrate into the offices through the heating, ventilating and air conditioning (HVAC) system and/or opened windows and doors. Time series of the PNC values for other selected offices and days (from 9:00 AM to 5:00 PM in each case) are shown in Fig. A.1 as examples. In general, similar series of PNC values with some fluctuations could be observed between the two investigated offices in the same building (Fig. A.1 a and b) which suggests the domination of the influence of outdoor sources.

Regarding the gaseous air pollutants, the concentration of O$_3$ was below the limit of detection (2.0 µg m$^{-3}$) in most cases; the highest median concentration value was 42.3 µg m$^{-3}$ which is far below the WHO guideline of 100 µg m$^{-3}$ (maximum daily 8-h mean; WHO, 2005).

### Table 2

PM$_{2.5}$ mass concentration, particle number concentration (PNC) and nitrogen dioxide (NO$_2$) concentration values obtained for the investigated offices during the two campaigns.

<table>
<thead>
<tr>
<th>Building ID</th>
<th>Room ID</th>
<th>1st campaign</th>
<th>2nd campaign</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PM$_{2.5}$ mass concentration (µg m$^{-3}$)</td>
<td>PNC ($N \times 10^3$ cm$^{-3}$)</td>
<td>NO$_2$ (µg m$^{-3}$)</td>
</tr>
<tr>
<td></td>
<td>Median</td>
<td>Range</td>
<td>Median</td>
</tr>
<tr>
<td>FR1</td>
<td>A</td>
<td>37.6</td>
<td>15.8</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>31.1</td>
<td>12.5</td>
</tr>
<tr>
<td>GR4</td>
<td>A</td>
<td>n.a.</td>
<td>20.8</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
<tr>
<td>GR5</td>
<td>A</td>
<td>n.a.</td>
<td>13.1</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
<tr>
<td>HU6</td>
<td>A</td>
<td>9.3</td>
<td>14.6</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>9.4</td>
<td>n.a.</td>
</tr>
<tr>
<td>IT3</td>
<td>A</td>
<td>8.0</td>
<td>6.3</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>7.1</td>
<td>6.0</td>
</tr>
<tr>
<td>IT4</td>
<td>A</td>
<td>21.3</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>14.3</td>
<td>4.6</td>
</tr>
<tr>
<td>NL3</td>
<td>A</td>
<td>6.8</td>
<td>2.4</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>7.7</td>
<td>2.6</td>
</tr>
<tr>
<td>PT1</td>
<td>A</td>
<td>29.6</td>
<td>7.1</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>25.9</td>
<td>4.8</td>
</tr>
<tr>
<td>PT2</td>
<td>A</td>
<td>18.2</td>
<td>5.0</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>17.7</td>
<td>4.9</td>
</tr>
</tbody>
</table>

LOD = limit of detection.

n.a. = not available.

*Bold numbers: ICC < 40%, concentrations/OP levels are considered to be heterogeneous (i) between sampling campaigns (temporal variation) or (ii) within a building (spatial variation); grey numbers: ICC > 60%, concentrations/OP levels are considered to be homogeneous; italic numbers: 40% < ICC < 60%, no conclusion can be drawn.*

### Table 3

Intra-class correlation coefficients (ICCs) for the investigated PM characteristics in office buildings (two-stage model).

<table>
<thead>
<tr>
<th>PM characteristic</th>
<th>Temporal variation between campaigns</th>
<th>Spatial variation within buildings</th>
</tr>
</thead>
<tbody>
<tr>
<td>PM$_{2.5}$ mass concentration</td>
<td>64%</td>
<td>88%</td>
</tr>
<tr>
<td>O$_3$ &amp; NO$_2$ m$^{-3}$</td>
<td>5%</td>
<td>88%</td>
</tr>
<tr>
<td>O$_{3/NO}_2$ m$^{-3}$</td>
<td>21%</td>
<td>70%</td>
</tr>
<tr>
<td>Al</td>
<td>0%</td>
<td>36%</td>
</tr>
<tr>
<td>V</td>
<td>0%</td>
<td>79%</td>
</tr>
<tr>
<td>Mn</td>
<td>47%</td>
<td>39%</td>
</tr>
<tr>
<td>Fe</td>
<td>0%</td>
<td>27%</td>
</tr>
<tr>
<td>Cu</td>
<td>0%</td>
<td>39%</td>
</tr>
<tr>
<td>Zn</td>
<td>3%</td>
<td>62%</td>
</tr>
<tr>
<td>Pb</td>
<td>73%</td>
<td>75%</td>
</tr>
<tr>
<td>PNC</td>
<td>20%</td>
<td>92%</td>
</tr>
</tbody>
</table>

*Bold numbers: ICC < 40%, concentrations/OP levels are considered to be heterogeneous (i) between sampling campaigns (temporal variation) or (ii) within a building (spatial variation); grey numbers: ICC > 60%, concentrations/OP levels are considered to be homogeneous; italic numbers: 40% < ICC < 60%, no conclusion can be drawn.*

However, the NO2 concentration values varied markedly across the office buildings (Table 2). The lowest and the highest median values were 7 and 154 μg m⁻³ respectively. The hourly mean concentration of NO2 never exceeded the 200 μg m⁻³ WHO guideline (WHO, 2005); however, it is clear that the median values were higher than 40 μg m⁻³, the annual mean limit value set by the WHO (2005), in many cases.

Significant correlation ($r_s = 0.73; p < 0.01$) was observed between the PNC values (5-day median) and the concentration of NO2 (5-day median) after the removal of the outlier pairs obtained in the Netherlands (results obtained for the 1st campaign were removed only; unexpectedly high NO2 concentration values). The PNC (5-day median) and the PM2.5 mass concentration also correlated ($r_s = 0.52; p < 0.05$). In contrast, PM2.5 mass concentration and NO2 concentration (5-day median) were not correlated ($r = 0.17; p = 0.45$).

### 3.2. Particulate oxidative potential in office buildings: can we strengthen our previous findings and add another piece into the jigsaw?

The OP metrics, ascorbate and reduced glutathione depletion on a unit volume basis (OPAA m⁻³ and OPGSH m⁻³ respectively) and on a unit mass basis (OPAA μg⁻¹ and OPGSH μg⁻¹ respectively), varied markedly across the investigated offices. The OP data are depicted on box plots (Fig. 3) and are not classified further due to the low number of samples collected per country. Similar to previous observations, urate was not depleted by the particles (e.g., Künzli et al., 2006; Szigeti et al., 2014).

The indoor levels of OPAA and OPGSH (both on a unit volume and on a unit mass bases) were in the same order or magnitude compared to the results obtained in our previous study carried out also in offices (Szigeti et al., 2016). The maximum indoor OPAA m⁻³ and OPGSH m⁻³ values were about 20 and 60 times higher than the lowest ones respectively (Fig. 3). These differences are larger than that obtained for the PM2.5 mass concentration (factor of 8 difference between the two extreme values). Similar to the PNC, temporal differences could be observed between the two sampling campaigns at the same location for either the OPAA m⁻³ or the OPGSH m⁻³ values in many cases and it has been supported by the results of the ICC analysis (Table 3).

Indoor particles collected in Budapest, Hungary showed larger oxidative activity on a unit mass basis (especially OPAA μg⁻¹) than the other PM2.5 samples collected at different locations (the mean OPAA μg⁻¹ value was about 2.7 times higher for the Hungarian samples compared to the mean OPAA μg⁻¹ value determined for the other PM2.5 samples) which is in line with our previous observations (Szigeti et al., 2016). As a consequence, the contribution of oxidatively active PM constituents to the PM2.5 mass was higher in the case of PM2.5 samples collected in Budapest compared to the other indoor samples collected at different sites in Europe. However, it must be noted that the health relevant metrics are the OPAA m⁻³ and the OPGSH m⁻³, thus the higher OPAA μg⁻¹ and OPGSH μg⁻¹ values do not necessarily indicate that the office workers are exposed to larger amount of oxidatively active particles; the PM2.5 mass concentration has to be taken into account.

Significant correlation was observed between the PM2.5 mass concentration and both OPAA m⁻³ ($r_s = 0.69, p < 0.01$) and OPGSH m⁻³ ($r_s = 0.66, p < 0.01$). These findings suggest that PM2.5 mass concentration represents quite well the OP of the particles (i.e., OPAA m⁻³ and OPGSH m⁻³; potentially health relevant characteristics of PM); however, the link is uncertain since previous studies reported different correlation coefficients between OP metrics based on antioxidant depletion and PM2.5 mass concentration (e.g., Janssen et al., 2014; Künzli et al., 2006; Szigeti et al., 2014, 2015, 2016). Among the investigated PM constituents, only the concentration (μg g⁻¹) of Cu correlated with both
the PNC values showed less variation (92%) and the two OP metrics (OPAA m⁻³ and OPGSH m⁻³) exhibited a similar or slightly larger variation (88 and 70% respectively) than the PM_{2.5} mass concentration (88%). The ICC coefficient values calculated for the trace elements varied within a wide range; considerable spatial variation was apparent for Al, Mn, Fe and Cu (Table 3). Among them, Fe and Cu were linked to OP. These values indicate that the monitoring of the PM_{2.5} mass concentration in a limited number of offices within a building might not reflect the spatial variation of the health relevant PM characteristics such as particulate OP or the concentration of certain trace elements at a building scale.

If we compare the ICC coefficient values calculated to assess the temporal and the spatial variation (Table 3), the temporal variation was larger for almost all PM characteristics (except for Mn) than the spatial differences within the office buildings. This finding indicates that long-term monitoring campaigns are necessary to get an overview about the long-term variation of the investigated PM characteristics. However, the differences within an office building may cause different long-term individual exposure patterns, thus working in two different offices within an office building might be associated with different health risks. It must be noted that the uncertainty of the measurements has to be also taken into account since large uncertainty could cause false conclusions regarding the spatial/temporal variation of the investigated PM characteristics. In the case of trace elements the contribution of the uncertainty to the total variance was <5%, while the mean contribution was 11 and 21% for OPAA m⁻³ and OPGSH m⁻³ respectively. However, the largest contribution observed was 61 and 73% for OPAA m⁻³ and OPGSH m⁻³ respectively, which clearly indicates that the uncertainty of the measurements is not negligible in some cases and the outcomes of the ICC analysis must be handled carefully. The uncertainty of the measurements could be reduced by either increasing the sampling time or using larger filter cuts for the analyses.

3.4. Limitations of the study: what questions will remain unanswered?

The link between indoor activities, environmental parameters (both indoors and outdoors) and indoor air quality has still not clearly identified yet. We have already raised this issue through a question (“Geographical location, outdoor PM sources, infiltration and indoor PM sources: Which are the major determinants of indoor PM_{2.5} mass concentration?”) in our previous paper (Szigeti et al., 2016) and we concluded that, in general, the outdoor PM_{2.5} mass concentration is still the major factor affecting the corresponding indoor value and the physical position and the proper operation of the HVAC system play a role in the reduction of the PM_{2.5} mass concentration as well. In the case of PM_{2.5}, we cannot approach the problem from a different angle compared to our previous paper, i.e. we cannot identify the factors which might affect the actual PM_{2.5} mass concentration, since an integrated PM_{2.5} sampling (5 days × 8 h) was carried out in this study as well, and a long-term sampling period does not provide enough information. Continuous monitoring of the PM_{2.5} mass concentration is needed.

The comparison of the changes in the PNC values with the indoor activities recorded on the time activity diaries (i.e., number of occupants in the office, printing) did not show any clear associations. PNC is generally dominated by the UFPs which are basically non-resuspensible by physical activities such as walking (Fisk et al., 2000), thus it is not surprising that small changes in the number of occupants (i.e., in the occupancy density) did not affect PNC. No significant concentration peaks appeared during printing in contrast to the observations of McGarry et al. (2011). However, they also found that the PNC emitted from some printers (classified as low-emitting printers) was not distinguishable from the background PNC, even in a 1 m distance between the printer and the CPC. It must be noted that we did not test the particle emission from the printers and the distance between the printer and the CPC was generally more than 1 m. Some offices were not equipped with printers; the devices were placed on the corridors or in dedicated rooms. Moreover, the relatively high background PNC as well as the

3.3. Differences between offices in the same building: is it more significant than the temporal variation?

To demonstrate the spatial variation within the office buildings, the ICC coefficients for the PM_{2.5} mass concentration, OPAA m⁻³ and OPGSH m⁻³, trace element concentrations and the PNC were calculated and the results are listed in Table 3. Among the investigated PM characteristics,
influence of the ventilation conditions in the office might affect the identification of those periods when considerable PNC was emitted from the printer. Accordingly, the fluctuation in the indoor PNC values is more likely caused by the variation of the outdoor PNC. There are several emission sources and formation mechanisms of the UFPs in the ambient air of big cities and their intensities are complex in time and space and changes in the PNC might occur frequently (Salma et al., 2011). One of the limitations of the study is that we did not investigate the outdoor environment, thus the direct comparison is not available. We also checked those periods when the windows were opened and, again, no clear association was found between the PNC values and the windows opening activity. In some cases, the PNC values increased when the office workers opened the windows; however, it is difficult to distinguish whether the increase was caused by the opening of the windows or it was just caused by the natural fluctuation of the outdoor PNC values and the particles penetrating through the HVAC system into the indoor environment were more dominant than the particles entering into the indoor environment through the opened windows. Latter might happen in the case of large open space offices where the majority of the windows are closed and the air exchange mainly happens through the HVAC system. Of course, the proximity and the intensity of the particles penetrating from the outdoor sources (i.e., outdoor PNC level) have to be taken into account. It is clear that there are several factors which influence indoor air quality and, consequently, the spatial distribution of particulate OP characteristics within a building. It is very difficult to separate the contribution of each factor.

### 4. Conclusions

Recently, the modern design, easy access through major roads and public transport, low energy consumption and the number of comfort services (e.g., restaurants, gym) are probably the most important parameters that people take into consideration regarding modern office buildings; however, the healthy indoor air quality should not be overlooked. The spatial variation of the concentration of indoor air pollutants within office buildings has not been investigated extensively yet since it requires high amount of resources. Our results obtained by comparing certain indoor air quality parameters at two locations per building indicate that occupants might be exposed to different PM characteristics, especially particulate OP and trace elements, to a different extent within the same building in several cases. In the case of short-term exposure, the spatial variation within the building might not be as important as the temporal variation of the air quality parameters since significant differences are usually apparent for different seasons (e.g., warm summer vs. cold winter); however, long-term exposure to a slightly higher levels of air pollutants in one of the offices might lead to shorter life expectancy or increased risk of certain adverse health effects. Thus the simultaneous investigation of the air quality characteristics at different locations within the same office building at different seasons (at least summer and winter) is highly recommended when either regulation will be formed for the indoor environment or field campaigns are planned. It is also clear that integrated problem solving approaches (i.e. data collection by checklist and questionnaire; investigation of indoor air quality parameters/physical, chemical, biological/by different methods; evaluation of the data, etc.) are needed to assess occupants’ exposure to a variety of environmental pollutants and to find solutions to ensure human health and well-being. It is intended that the evidences reported in this study may contribute for the better understanding of the human’s exposure to PM in the office setting as well as for providing information for the design of further long-term exposure assessment studies. 

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