Children exposure assessment to particulate matter in urban environment

Inês da Cunha Lopes

Instituto Superior Técnico
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HIGHLIGHTS
- Children spend most of their time indoors indicating that risk assessment should focus on these microenvironments.
- Home and transport were responsible for high contributions to children's exposure to and inhaled dose of black carbon.
- Transport was the microenvironment in which children were exposed to the highest black carbon concentration.

ABSTRACT
The particulate matter (PM) has adverse impacts on human health. Children are considered a susceptible group since they breathe high volumes of air relative to their body weights and their tissues and organs are growing.

The main objective of this study is to quantify the children's daily exposure to PM and the respective inhaled dose. Nine children from Lisbon carried equipment along three days in order to quantify the PM2.5 and black carbon (BC) concentrations.

The children spent more than 80% of their time indoors, especially at home and in the classroom. The exposure depends on the microenvironment frequented and the activities performed. The PM2.5 concentrations varied between 12 - 28 μg/m³. Time series analysis of the BC concentrations showed high peaks in underground parking lots, when candles are burning and during charcoal grills. The mean daily exposure and the inhaled dose were equal to 1.3 μg/m³ and 15 μg, respectively. Home was the microenvironment that mostly contributed to the daily BC exposure (39%) and inhaled dose (28%) thanks to the large amount of time spent there (55%). Transportation accounted for 5.0% of daily time however, children received intense exposure (21%) and inhaled dose (23%) due to the high BC concentration (5.1 μg/m³) in which the children were exposed.

This study may be used to help prioritize targets for minimizing children's exposure to PM and to indicate outcomes of control strategies.

Keywords: Children, Time-activity pattern, PM2.5, Black carbon (BC), Personal exposure, Inhaled dose.

1. Introduction
The exposure to air pollution, in particular to particulate matter (PM) has several adverse effects on human health and may induce or aggravate vascular (Watson, 2006) and respiratory diseases, such as asthma (Kelly & Fussell, 2011) and lung cancer (Sax et al., 2013), especially in urban areas (EEA, 2017). Black carbon (BC) is a constituent of fine particles emitted from incomplete combustion such as mobile sources (ex.: motorized transports, mainly in diesel vehicles) and burning of biomass (EEA, 2013; Gelencsér, 2004; Seinfeld & Pandis, 2006). Human exposure was defined by Ott (1982) as ‘the event when a person comes into contact with a pollutant of a certain concentration during a certain period of time’. The exposure to air pollutants has traditionally been assessed based on data from fixed-site monitoring networks. This method usually provides a large quantity of data for a wide range of pollutants however, it provides for only one point in space (Steinle et al., 2013). The impacts on individual health are related not only to the concentrations of the pollutants but also to the personal exposure in the different microenvironments (ME), as it depends on the time that each person spend in a certain place, the activity profile, the characteristics of the microenvironment and its surroundings (Dimitroulopoulou & Ashmore, 2009; Dons, et al., 2011).

People spend around 90% of their time indoors (Klepeis, et al., 2001), so indoor air quality (IAQ) is a dominant factor for personal exposure (Sundell, 2004). Therefore, for an accurate personal exposure assessment, the different places in which time is spent (Dimitroulopoulou & Ashmore, 2009) should be considered, since it is more reliable and takes into account the time-activity patterns and it portrays the daily exposure to PM to which a person is exposed to (Steinle et al., 2013).

Children seem to be most vulnerable to the harmful effects of ambient air pollutants because their defense mechanisms are still evolving and they inhale a higher volume of air per body weight than adults (Mendell & Health, 2005; Salvi, 2007; Trasande & Thurston, 2005).

The assessment of personal exposure of vulnerable groups is essential to evaluate the exposure to air pollutants so that measures can be taken to control indoor and outdoor pollution and to ensure the protection of public health.

The main goal of this study is to quantify the children's daily exposure to airborne particles and respective inhaled dose through personal monitors since the exposure depends on a series of factors that vary according to the time-activity pattern and the microenvironments that they attend.
2. Materials and methods

2.1. Study design
Nine children (7-10 years old) living and studying in Lisbon metropolitan area (AML) (Figure 1) were selected to carry personal monitors during all their daily activities along three days. The personal measurements were conducted for 72 h each and took place from May 2 to June 22, 2018. Measurements were carried out on weekdays/school days.

2.2. Monitoring equipment used
Each child carried a trolley with three portable equipment. A GPS that registered the coordinates of the routes. A SKC Leland Legacy pump (9L/min) connected to a personal cascade impactor (PCIS) to collect the particles in different size ranges below 2.5 μm. The polytetrafluoroethylene (PTFE) filter loads were measured by gravimetric in a controlled clean room (class 10,000) with a semi-micro balance. The PM2.5 mass concentration in the 4 stages of PCIS was obtained through gravimetric analysis. Each filter was weighted three times (before and after sampling) and the differences between the masses were lower than 20μg. A micro-aethalometer (Model AE51) was used to determine BC concentrations by measuring changes in absorption of transmitted light at 880 nm with continuous collection of light-absorbing BC particles deposited on a small Teflon-coated borosilicate glass fiber filter. The filter strips were replaced to prevent the filter saturation and to maintain measurement integrity. The air was drawn into the device at 100 ml/min and logging on a 60 seconds basis. The data obtained was corrected using the “Optimized Noise-Reduction” (ONA) program to reduce the noise (Hagler et al., 2011).

A time-activity diary (TAD) was completed by the children in order to record all the information about their time-activity patterns, indicating children’s main indoor or outdoor activities, start and end times of each time-activity and the respective location. In addition, a questionnaire was developed in order to know some characteristics of the home and the classroom of each child.

2.3. Data analysis
The children’s inhaled dose was estimated by multiplying the arithmetic BC concentration in the ME (µg/m³) by the time spent in the ME and inhalation rate (IR, m³/h) (Table 1).

The contribution of different time-activities and MEs based on exposures for each time-activity and ME was examined (Eq. (1) and (2)).

The intensity of exposure and inhaled dose to BC (Eq. (3)) were also presented in order to compare the relative contributions of exposure and inhaled dose.

Table 1: Inhalation rate (m³/h) as a function of age group and activity performed. Based on Buonanno et al (2011).

<table>
<thead>
<tr>
<th>Activity</th>
<th>IR (m³/h) for 6-10 age group</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sleeping and rest</td>
<td>0.31</td>
</tr>
<tr>
<td>Sedentary activities</td>
<td>0.42</td>
</tr>
<tr>
<td>Studying</td>
<td>0.42</td>
</tr>
<tr>
<td>Transportation</td>
<td>Vehicle</td>
</tr>
<tr>
<td></td>
<td>Walking</td>
</tr>
<tr>
<td>Playing indoor</td>
<td>1.27</td>
</tr>
<tr>
<td>Non-sedentary activities</td>
<td>ex.; leisure</td>
</tr>
<tr>
<td>Sport indoor</td>
<td>1.27</td>
</tr>
<tr>
<td>Sport outdoor</td>
<td>1.44</td>
</tr>
</tbody>
</table>

Daily BC exposure contribution (%) = \( \frac{C_{ij} \times t_{ij}}{\sum_{j=1}^{m} C_{ij} \times t_{ij}} \)  (1)

Daily BC inhaled dose contribution (%) = \( \frac{C_{ij} \times t_{ij} \times IR_{ij}}{\sum_{j=1}^{m} C_{ij} \times t_{ij} \times IR_{ij}} \)  (2)

\( C_{ij} \) – Arithmetic mean BC concentration by ME (j) and individual (i) (µg/m³);
\( t_{ij} \) – Time spent by individual (i) in ME (j) (hours);
\( m \) – Total number of MEs, \( \sum_{j=1}^{m} t_{ij} = 24 \) h.

BC exposure (inhaled dose) intensity = Daily exposure (inhaled dose) contribution (%) / Daily time contribution (%)  (3)
2.4. Statistical analysis
Statistical calculations were performed using STATISTICA software. Mann–Whitney U test was used because samples were independent (differences between microenvironments and children). This test is non-parametric, hence it does not consider any assumptions related to the distribution, in that it compares between two medians to suggest whether both samples come from the same population or not. Statistical significance refers to $p < 0.05$.

3. Results and Discussion

3.1. Time–activity patterns
Daily time–activity patterns were obtained from the time–activity diary of each child. Table 2 shows the percentage of time spent by the participants on each ME/activity.

The overall subjects’ time–activity pattern was characterized by a substantial amount of time spent indoors (more than 80%). The largest amount of time was spent at home (55%) (sleeping and general activities), followed by school (classroom, 22% and playground, 8.3%) and transports (5.0%).

These results are similar to those found in previous studies. In the framework of the LIFE Index-Air (2017) project a questionnaire about time–activity patterns was developed targeting children (5 - 10 years old) in Lisbon. This study revealed that during the week children spend 89% of their time indoors – 55% in home, 27% in classrooms, 3.5% in vehicles and 2.7% practicing indoor physical activities. Furthermore, international studies revealed also similar results. Jeong & Park (2017a) assessed 10-12 years old Korean children and showed that the greatest amount of time was spent at home (59%) and on transport (7.1%).

Table 2: Percentage of time spent on each ME/activity.

<table>
<thead>
<tr>
<th>Activities/Microenvironments</th>
<th>% time in each MEs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Home</td>
<td>55</td>
</tr>
<tr>
<td>Sleeping</td>
<td>40</td>
</tr>
<tr>
<td>General activities</td>
<td>15</td>
</tr>
<tr>
<td>Vehicle</td>
<td>4.3</td>
</tr>
<tr>
<td>Transportation</td>
<td>22</td>
</tr>
<tr>
<td>Walking</td>
<td>0.69</td>
</tr>
<tr>
<td>Classroom</td>
<td>8.3</td>
</tr>
<tr>
<td>Playground at school</td>
<td>2.0</td>
</tr>
<tr>
<td>Sport indoor</td>
<td>0.63</td>
</tr>
<tr>
<td>Leisure</td>
<td>1.2</td>
</tr>
<tr>
<td>Extracurricular activities</td>
<td>1.0</td>
</tr>
<tr>
<td>Others</td>
<td>5.4</td>
</tr>
</tbody>
</table>

3.2. PM2.5 mass distribution and respective concentration
Figure 2 shows the PM2.5 concentration to which child was exposed and the respective particles contribution in each stage of PCIS.

No trend was found in the particles mass distribution among the 9 children. This may be because each child was exposed to different sources of PM2.5 throughout the day since each one had their own time–activity patterns in different locations. The PM2.5 concentration varied between 12 - 28 µg/m$^3$ (Figure 2), complying the World Health Organization (WHO) daily reference value (25 µg/m$^3$), except C2. The child C2 had been exposed to high PM2.5 concentration (28 µg/m$^3$), perhaps due to the sampling which was disturbed by school colleagues through the introduction of small objects into air intake of PCIS.

The stages with the greatest contribution to the PM2.5 concentrations are the A and D. For children C2, C4, C5, C6, C8, and C9 the particles with an aerodynamic diameter (AD) <0.25 µm (stage D) were those that presented a greater contribution to the PM2.5 concentration. This stage includes the inhaled and ultrafine particles, capable of depositing in the pulmonary alveoli, get into the bloodstream and reaching different organs (Guarieiro & Guarieiro, 2013), thus these children may be more likely to present greater health problems.

The child C7 was exposed to the lowest PM2.5 concentration. In addition, about 42% represent particles with an AD from 1.0 to 2.5 µm. Therefore, this child is probably less likely to have health problems associated with particle inhalation. However, the PM2.5 composition may better predict health effects than PM mass or size (Rohr & Wyzga, 2012; Stanek et al., 2011). In this way, it’s necessary to take into account the chemical analysis of the filters to identify possible adverse health effects.

Figure 2: PM2.5 concentration to which each child was exposed and the respective particles contribution from each stage.
### 3.3. BC concentrations

BC concentrations by time-activity and ME based on self-reported time-activity diary are shown in Table 3. Children were exposed to the lowest BC concentrations at home (0.89 µg/m³) and to the highest in transportation (5.1 µg/m³). In these MEs, the BC concentrations were significantly different (lower and higher, respectively) from the other MEs according to the statistical tests (p < 0.05). Rivas et al. (2015) assessed 45 Spanish children (7-10 years) obtaining the same result. They found that the highest BC concentration was in transport and the lowest was during the periods that the children were at home. This may be due to the fact that BC is a result of incomplete combustion of fossil fuels, with vehicles being a substantially relevant source of BC (Jeong & Park, 2017b; Buonanno et al., 2013; Climate and Clean Air Coalition, 2018).

The school location (Vieira, 2011) and the ventilation systems (Almeida, et al., 2011) are important parameters for a good indoor air quality (IAQ) in a classroom since the BC concentration is influenced by outdoor sources, as the transports and cooking (Jeong & Park, 2017b). At school, children were exposed to similar BC concentration in the classroom and in the playground (1.2 and 1.1 µg/m³, respectively). The playground frequented by children is outside, so it is influenced directly by the surrounding traffic. In this way, the classrooms have a high influence from outside BC sources. Rivas et al. (2015) analyzed the relationship among personal monitoring, fixed stations (school indoor and outdoor) and urban stations. They concluded that generally indoor and outdoor school levels followed the urban trends, confirming that there is infiltration and influence of traffic emissions in classrooms, as well as, the majority of BC concentration from personal measurements followed approximately the same levels and trends from indoor school monitoring stations.

Table 3: Black carbon (BC) concentrations by activity and microenvironment.

<table>
<thead>
<tr>
<th>Activities/Microenvironments</th>
<th>Black carbon (BC), µg/m³</th>
</tr>
</thead>
<tbody>
<tr>
<td>Home</td>
<td>0.89</td>
</tr>
<tr>
<td>Sleeping</td>
<td>0.81</td>
</tr>
<tr>
<td>General activities</td>
<td>1.1</td>
</tr>
<tr>
<td>Transportation</td>
<td></td>
</tr>
<tr>
<td>Vehicle</td>
<td>5.1</td>
</tr>
<tr>
<td>Walking</td>
<td>2.5</td>
</tr>
<tr>
<td>Classroom</td>
<td>1.2</td>
</tr>
<tr>
<td>Playground at school</td>
<td>1.1</td>
</tr>
<tr>
<td>Sport indoor</td>
<td>1.1</td>
</tr>
<tr>
<td>Sport outdoor</td>
<td>1.5</td>
</tr>
<tr>
<td>Leisure</td>
<td>2.2</td>
</tr>
<tr>
<td>Extracurricular activities</td>
<td>2.0</td>
</tr>
<tr>
<td>Others</td>
<td>1.4</td>
</tr>
</tbody>
</table>

### 3.4. BC time series

The analysis of BC concentration time series allows the identification of peak concentration events and the relation to specific activities, taking into account the TAD of each child. Figure 3 shows 24h BC time series of two children who were exposed to BC peak concentrations. In both graphs, it is verified that from 00h00 to 07h30 the concentrations are constant and low, being associated with the home environment, especially to the sleep period.

![BC time series](image)

Figure 3: BC time series corresponding to 2 children. A – Transport by car; A – Transport on foot; B – Underground parking lot; C – Birthday party; D – Party

The point A identified in Figure 3 corresponds to the transport during the rush hour. Rivas et al. (2015) verified in the children’s temporal analysis that BC peaks also coincided with the rush hour in which the children commute to school. The child C1 was exposed to a high BC concentration in the underground parking lot (point B). The underground parking lots are fully or partially enclosed, so the pollutants do not disperse as it does outside. The point C is relative to a birthday party and the peak concentration may be associated with the burning candles according to the TAD of this child. In accordance with Stabile et al. (2012) the combustion of candles essentially produces carbonaceous particles with the ratio BC/PM10 being greater than 80%. The child C8 had a peak BC concentration at night (point D) during an outdoor party with charcoal grills. Therefore, the reason of this BC peak may be due to the charcoal grills, since it releases fumes which contain a high rate of carbonaceous particles, being a substantial source of BC (Jeong & Park, 2017b; Buonanno et al., 2009).

### 3.5. BC exposure and respective inhaled doses

The children’s daily BC exposure and inhaled dose contribution are shown in Figure 4. Daily average BC exposure and inhaled dose measured was equal to 1.3 µg/m³ and 15 µg, respectively. These values were found to be lower than the results from Italy (5.1 µg/m³ for daily exposure and 39 µg for the daily inhaled dose) (Buonanno et al., 2013) and South Korea (1.9 µg/m³ for daily exposure 24 µg for the daily inhaled dose) (Jeong & Park, 2017b). These studies are also related to children (aged 7-12 years), so the inhalation rates used
to calculate the dose were the same as those used for this study.

First of all, it is necessary to consider the routines of each country, since exposure is influenced by the type of microenvironment and activity performed. These activities are related to an increase on BC concentration as Buonanno et al. (2013) and Jeong & Park (2017b) concluded and, according to Climate and Clean Air Coalition (2018), household cooking and heating account for 58% of global black carbon emissions. A possible reason for the higher values of Buonanno et al. (2013) can be because the study was carried out in the winter and according to the questionnaires, 83% of the houses had a fireplace and BC is formed from incomplete biomass combustion (Savolähti et al., 2016; Mousavi et al., 2018; Climate and Clean Air Coalition, 2018).

The key microenvironments who made a substantial contribution to the daily exposure to BC were home, transportation and classroom, whereas for the inhaled dose were home, transportation, classroom and also the playground due to the high IR in this ME. The microenvironment that contributed substantially to the children daily exposure (38%) and inhaled dose (28%) to BC was at home due to the greatest amount of time spent (55%). Similarly, home was the place where children received the greatest daily exposure in Korea and Italy (52% in both) (Jeong & Park, 2017b; Buonanno et al., 2013). Transportation was also crucial to the contribution to BC exposure and dose (21% and 23% respectively) in spite of involving only a small fraction of daily time (5.0%), the children were exposed to a high BC concentration (5.1 μg/m³). This result is in agreement with Jeong & Park (2017b) and Buonanno et al. (2013), which verified that the contribution to BC exposure in transportation was 15% (for a daily time 7.6%) and 11% (for a daily time of 4.0%), respectively. In this study, the contribution of the school was responsible for 21% of the exposure and 17% of the inhaled dose. Likewise, both studies also obtained very similar results in the contribution of this microenvironment, 20% in BC exposure for both studies (Buonanno et al., 2013; Jeong & Park, 2017b).

The children C4 and C9 presented the highest BC exposure, according to statistical test (p < 0.05). For C4, the ME that highly contributed to the BC exposure and inhaled dose was the transportation, since this child was exposed to the highest BC concentration in this ME (8.6 μg/m³) and spent more time in transport (10% of daily time). For C9, it was the home that contributed most to the daily exposure and dose. This child spent about 66% of the daily time in this ME and was exposed to the highest BC concentrations measured in homes (2.2 μg/m³). This may be due to the fact that this home has an open kitchen, so the BC emitted from cooking disperse easily to the living room. Furthermore, this child lives with a large family composed by nine members. Jeong & Park (2017b) concluded that children with large families faced higher BC exposures than those with small families, perhaps due to the fact that a greater number of family members generally require longer times spent on cooking.

The intensity was calculated in order to better understand the relative contribution of BC exposure and inhaled dose by MEs and daily time. Transportation incurred the most intense BC exposure (4.0) among the children, as it was observed in Jeong & Park (2017b) and Buonanno et al. (2013) (2.0 and 2.8, respectively). In this study, the intensity was higher since a smaller fraction of daily time was spent on transportation. Furthermore, there was a greater contribution in BC exposure due to a greater BC concentration that the children were exposed to in comparison to both studies mentioned.
4. Conclusions

The exposure of 9 children (aged 7 - 10) who lived and studied in AML was evaluated in terms of total PM2.5 concentration and BC concentration in real time. The TAD allowed to evaluate the activities/microenvironments that contributed to the daily exposure and to estimate the daily inhaled dose of each child in each ME.

The daily exposure depends on the ME frequented and activities performed. The results showed that children spend most of their time indoors (more than 80%), indicating that risk assessment should focus mainly on indoor MEs in order to protect children from adverse health effects that may be caused by BC exposure. Home and classroom were the MEs where the children spent more time. In terms to the particle size distribution of PM2.5, there was no trend in the particle mass distribution by the different stages of the PCIS. This may be due to the fact that each child has its time-activity pattern, so they are exposed to different sources of PM2.5. The mean of daily exposure to BC was 1.3 μg/m³ (range 0.88 - 2.0 μg/m³) and the mean of daily inhaled dose was 15 μg (range 12 - 27 μg). The children studied and lived in different places, so the individual time-space activity pattern leads to such variability. It was verified that underground parking lots, when candles are burning and during charcoal grills lead to high peaks in the BC concentration. The MEs that contributed most to the daily BC exposure were the home, the transportation, and the classroom. Although the home was the ME where the children were exposed to a lower concentration (0.89μg/m³), the greatest contribution to BC exposure and inhaled dose (39% and 28%, respectively) occurred at home due to the large amount of time spent there (55%). The BC concentration at home can be influenced by cooking and infiltration from traffic emissions. In contrast, transportation is the ME in which children were exposed to a higher BC concentration (5.1 μg/m³) and despite the small fraction of daily time (5.0%), this was the second ME that stands out in the contribution to the BC exposure in 21% and to the daily inhaled dose in 23%. Through the calculation of the intensity of the exposure and daily dose, it was evidenced that the transport was distinguished from the remaining MEs, and thus could represent a BC indicator.

In order to reduce children’s exposure to BC, it is necessary to take proactive measures based on precautionary principles. At home, measures should be taken such as not opening the windows at rush hour/heavy traffic and having an efficient exhaust fan in the kitchen in order to remove the BC emitted from cooking. For transportation, especially children’s transportation should choose routes with less traffic routes in order to help minimize children’s exposure. During weekdays, children spend a large amount of daily time at school. School zones should be built to ensure the protection from traffic-related pollutants or should have policies to reduce BC emissions around the school.

References


