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Deposition of aerosol particles from a subway microenvironment in the human respiratory tract

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ABSTRACT

Conventional subway systems are characterized by high particulate matter (PM) concentrations. To relate PM exposure to adverse health effects it is important to determine the dose of the inhaled particles in the human respiratory tract (HRT). Therefore, the total and regional doses of particles for a healthy adult male using the dosimetry model ExDoM in the subway system were estimated. The overall dose was determined using the average exposure $PM_{2.5}$ concentrations obtained from an extensive campaign in the Barcelona subway system, including measurements on the platforms and inside the trains. Despite the lower $PM_{2.5}$ concentrations inside the trains with respect to those on station platforms, the highest dose was observed inside the trains due to longer exposure time, evidencing the importance of the exposure period in the estimation of the particle dose. Overall, during a subway commuting travel, roughly 80% of the inhaled mass of subway $PM_{2.5}$ was deposited in the HRT. The highest amount of the inhaled particles was deposited in the extrathoracic region (68%), whereas the deposition was much smaller in the tracheobronchial tree (4%) and alveolar–interstitial region (10%). Individual's daily exposure to $PM_{2.5}$ and dose were estimated, considering a typical time-activity pattern of an adult male who lives in Barcelona and commutes by subway. While a subject typically spends approx. 3% of the day in the subway system, this microenvironment may account for up to 47% of the total $PM_{2.5}$ daily dose. These results might be similarly high for other commuting modes due to the reported high PM exposure levels. The dose is mainly dependent on the particle size and exposure concentrations.

1. Introduction

Urban population is daily exposed to air particulate pollution from a range of sources, including the ambient environment and three main microenvironments: home, workplace and commuting. In fact, the exposure to airborne particles depends on the lifestyle of each individual and the different microenvironments frequented (Buonanno, Fuoco & Stabile, 2011; Buonanno, Marks & Morawska, 2013). Epidemiological and toxicological studies have shown associations between

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particulate matter (PM) and adverse health effects (e.g. Dominici et al., 2006; Katsouyanni et al., 2001; Pope & Dockery, 2006; Russell & Brunekreef, 2009; Schikowski et al., 2007; Valavanidis, Fiotakis & Vlachogianni, 2008). Although the large majority of these studies relate health effects to PM exposure (the inhaled concentration), the negative outcomes are mainly caused by the subsequent deposition of PM in the respiratory tract during breathing (Salma, Balásházy, Winkler-Heil, Hofmann & Záray, 2002). Hence, in order to understand the mechanisms behind the health responses, it is crucial to determine the respiratory tract deposition fraction (DF) of aerosol particles, which is their probability to deposit, and the dose (amount of inhaled particles deposited) (Löndahl et al., 2009). For aerosols this dose can be given as number, surface area or mass of the deposited particles. The dose of particles in the human respiratory tract (HRT) depends on a number of factors, including PM exposure concentrations, physicochemical characteristics of PM, exposure duration, and exposed subject characteristics, such as age, gender, state of health, lung morphology, and breathing parameters (Broday & Agnon, 2007; Glytsos, Ondráček, Džumbová, Kopanakis & Lazaridis, 2010; Heyder, 2004; Hofmann, 2011; Lazaridis, Broday, Hov & Georgopoulos, 2001; Patterson, Zhang, Zheng & Zhu, 2014). However, most of the studies on the health impact of aerosol inhalation link the observed health effects with day-averaged concentrations from fixed ambient air quality monitoring stations, rather than personal exposure to particles at the indoor and outdoor places where the individual may be active (Aleksandropoulou, Mitsakou, Housiadas & Lazaridis, 2008).

Inhaled particles are carried with the tidal air through the respiratory system. However, when travelling along an airway, particles will be exposed to different physical mechanisms forcing them to displace off the streamlines of the inhaled air volume and eventually depositing on the surrounding airway surfaces. The most important mechanisms acting upon the inhaled particles are diffusion (Brownian motion), inertial impaction, electrostatic charging, and sedimentation (gravitational settling) (Hofmann, 2011; Hussain, Madl & Khan, 2011; Löndahl et al., 2014).

The deposited dose of atmospheric aerosols in the human respiratory tract is measured by monitoring the inhaled and exhaled particle concentrations (e.g. Löndahl et al., 2008; Montoya et al., 2004; Morawska, Hofmann, Hitchins-Loveday, Swanson & Mengersen, 2005). This provides an empirical estimation for the total deposition pattern of aerosol particles in the respiratory system. Due to experimental limitations, the regional dose in the respiratory system (extrathoracic, tracheobronchial, and alveolar–interstitial regions) cannot be determined experimentally and is typically estimated by means of mathematical models. Several dosimetry models have been developed over the years (Aleksandropoulou & Lazaridis, 2013; Asgharian, 2004; Georgopoulos & Lioy, 2006; Heyder & Rudolf, 1984; ICRP, 1994; Klepeis, 2006; Koblinger & Hofmann, 1990; Lazaridis et al., 2001; Mitsakou, Mitrakos, Neofytou & Housiadas, 2007; Rudolf, Köbrich & Stahlhofen, 1990; Sturm, 2007; Yeh & Schum, 1980). These models for both total and regional deposition have been compared to experimental studies, with a reasonable correlation being obtained between model predictions and experimental measurements (Asgharian & Price, 2007; Löndahl et al., 2008; Stuart, 1984).

Several studies on PM exposure in different commuting modes along complementary routes have stated that all commuting modes (passenger cars, bus, subway, motorbike, cycling and pedestrian) are characterized by high PM exposure levels due to the fact that commuters are close to mobile emission sources. Moreover, in this assessment studies it is very important to account for the breathing rates and journey times (e.g. de Nazelle et al., 2012; Gulliver & Briggs, 2004). Many people living in metropolitan areas worldwide commute using underground subway transportation. Several studies have investigated the air quality in underground subway systems (see Martins et al., 2015a and references therein), and most of them reported elevated pollution levels in terms of PM in comparison to the outdoor ambient air. The PM in this microenvironment is mostly generated internally by the motion of the trains and movement of passengers, but can also origin from the inflow of outside air through the ventilation system, which promotes the mixing and resuspension of PM (e.g. Querol et al., 2012). Despite the relatively short amount of time spent in the subway on a daily basis, or commuting in a general way, PM exposure levels in such microenvironment are of concern given the relatively high PM concentrations. Michaels and Kleinman (2000) reported that peak exposures of 1 h or less, just over the typical time spent in a transport environment, may be extremely relevant in terms of health effects. In addition, the exposure to PM in the subway system has been associated with adverse health effects (Bachoual et al., 2007; Bigert et al., 2008; Seaton et al., 2005). However there is an uncertain and limited nature of the evidence for subway metalliferous PM toxicity (Moreno et al., 2015 and references therein), as for example part of subway particles bioreactivity has been associated to the glass fibre filters in the extracted samples (Karlsson, Ljungman, Lindbom & Möller, 2006), and no increased lung cancer risk has been found amongst subway train drivers (Gustavsson, Bigert & Pollán, 2008).

To the authors' knowledge, there are no studies on the deposition of subway PM in the human respiratory tract. Therefore, the main objectives of this study were to (i) determine the $PM_{2.5}$ exposure of subway commuters, (ii) calculate the total and regional doses in the respiratory tract based on the $PM_{2.5}$ exposure during subway commutes, as a function of the time spent on the platforms and inside the trains, and (iii) estimate the overall daily $PM_{2.5}$ dose, considering a typical time-activity pattern. The exposure and dose assessment was performed using aerosol measurements in the Barcelona subway system and in the urban background of Barcelona (Spain), with limitations for the remaining indoor microenvironments considered, as explained later in results and conclusions. In this study, the $PM_{2.5}$ dose in the HRT was estimated applying the dosimetry model Exposure Dose Model (ExDoM) (Aleksandropoulou & Lazaridis, 2013).

2. Experimental method

2.1. Monitoring sites and measurements

The subway system of Barcelona is managed by the Transports Metropolitans de Barcelona (TMB) and it is one of the oldest underground transport systems in Europe, with its first line beginning operation in 1924. The measurement campaign was conducted in this subway system during two seasonal periods: warmer (2 April–30 July 2013) and colder (28 October 2013–10 March 2014). Different types of subway stations were considered in the experimental campaign in terms of architectural design, in order to investigate the effects on the exposure concentration levels and dose. Particle aerosol measurements were performed on the four stations described hereinafter:

– Joanic on the yellow line (L4): two platforms in the same tunnel with the two rail tracks in the centre, one for each direction, separated by a middle wall.
– Santa Coloma on the red line (L1): one wide tunnel with two rail tracks without middle wall.
– Tetuan on the purple line (L2): one platform in a single narrow tunnel with one rail track.
– Llefià on the new light blue line (L10): a single narrow tunnel with the platform separated from the rail track by a glass wall with mechanical doors that are opened simultaneously with the train doors (known as platform screen doors system – PSDs). The system is automatic, with computer controlled driving system that optimises speed, braking and stopping processes.

The techniques and procedures of the experimental study are described in detail elsewhere (Martins et al., 2015a), and will be only briefly summarised here. $PM_{2.5}$ samples were collected daily on quartz microfibre filters using a high volume sampler (HVS, Model CAV-A/MSb, MCV) over 19 h (from 5 a.m. to midnight, subway operating hours). The filters were gravimetrically analysed to determine the $PM_{2.5}$ mass concentrations. $PM_{2.5}$ mass concentrations ($\mu g\,m^{-3}$) were determined continuously by a light-scattering laser photometer (DustTrak, Model 8533, TSI) with a 5-minutes time resolution. These measurements were performed for each seasonal period during a month at each station. Furthermore, $PM_{2.5}$ mass concentrations were also determined inside the trains, from 6 subway lines (L1, L2, L3, L4, L5 and L10), with 5-seconds time resolution using a DustTrak. $PM_{2.5}$ concentrations provided by DustTrak monitor were corrected against the gravimetric $PM_{2.5}$. Additionally, in the current study the exposure concentrations represent the mean value of the measurements performed at each station and inside the trains of the different lines, in order to simulate the overall PM exposure and dose of a subway commuter. Air quality measurements were performed simultaneously at the urban background station of Palau Reial to obtain mean concentrations of the outdoor environment.

2.2. Respiratory tract deposition model

Aerosol deposition in human respiratory system was calculated by the dosimetry model ExDoM. A detailed description of this model has been reported by Aleksandropoulou and Lazaridis (2013) and Chalvatzaki and Lazaridis (2015). For modelling purposes, the respiratory tract is divided into different anatomical regions: an extrathoracic (ET) region – anterior nasal passages (ET1) and the posterior nasal passages, larynx, pharynx and mouth (ET2); a tracheobronchial (TB) tree – the bronchial region, including trachea and bronchi (BB) and the bronchiolar region consisting of bronchioles and terminal bronchioles (bb); and an alveolar–interstitial (AI) region, consisting of respiratory bronchioles, and alveolar ducts and sacs surrounded by alveoli. The exposure is adjusted by the inhalability, which is the fraction of aerosol particles that enters in the HRT during breathing. The PM deposition fractions for each region of the respiratory tract are calculated after accounting for the filtering effect of the preceding airways (Aleksandropoulou & Lazaridis, 2013).

In particular, the dose depends on the exposure concentration and physicochemical characteristics of the PM, time-activity pattern and the exposed subject characteristics, as the respiratory physiology parameters, physical activity level, breathing pattern, gender and age, among others (Aleksandropoulou & Lazaridis, 2013; ICRP, 1994).

2.3. Exposure scenario and dose calculation

For the application of the dosimetry model ExDoM the following aspects were considered: (i) selection of the exposed subject; (ii) identification of the microenvironments where the exposed subject spent time; (iii) estimation of the time spent in each microenvironment, (iv) determination of the $PM_{2.5}$ exposure concentrations, (v) election of the breathing mode; and (vi) selection of breathing rate (volume of air inhaled per unit of time) to be used as a function of the corresponding specific activity levels (classified as sleep, sitting/resting and light exercise).

Modelling of $PM_{2.5}$ deposition in the HRT was conducted for a healthy Caucasian adult male breathing through the nose. Lippmann, Yeates and Albert (1980) reported that the nasal passages are a more efficient particle filter than the oral ones, thus, persistent mouth breathers deposit more particles in their respiratory system than those breathing entirely through the nose. Additionally, Löndahl et al. (2007) conducted an intensive study determining the dose of particles in the HRT by gender (male and female) and they found that the amount of deposited particles varied remarkably between genders, increasing substantially for the male subjects, because of their higher breathing rate values. The lung of a child differs

significantly from that of adults in terms of airway dimensions and breathing rate (Ménache, Hofmann, Ashgarian & Miller, 2008). Due to the combination of smaller airway sizes, smaller tidal volumes, but higher breathing frequencies, the total deposition fraction in children is generally higher than in adults (Asgharian, Ménache & Miller, 2004). It is also worth noticing that the dose is, in general, higher in subjects with lung problems (such as asthma, obstructive lung diseases, etc.) than healthy subjects (e.g. Anderson, Wilson & Hiller, 1990; Kim & Kang, 1997; Chalupa, Morrow, Oberdörster, Utell & Frampton, 2004).

The size distribution of the subway $PM_{2.5}$ was considered monodisperse with a mass mean aerodynamic diameter (MMAD) of 2.1 μm and a geometric standard deviation (GSD) of 1.7. Although particles were assumed spherical (shape factor of 1) for the dose calculations it is known from scanning electron microscopy studies that a large fraction of subway PM is laminar (Moreno et al., 2015; Querol et al., 2012). Another important factor determining the deposition of particles is their density (Aleksandropoulou & Lazaridis, 2013). The density of the subway particles ranged from 2.2 to 3.1 g cm^{-3}, based on their chemical composition at each subway station and seasonal period (Martins et al., 2015b).

In the case of the subway microenvironment, the estimations were based on the assumption that the subject is under light exercise and consequently the breathing rate equals to 1.5 m^3 h^{-1} (reference values for adult Caucasian males; ICRP, 1994). Furthermore, the dose was determined for average exposure concentrations from the measurements on the platforms and inside the trains in order to represent the overall dose in the subway system. The exposure time assumed was based on the TMB information with an average subway commuting one-way travel of 5 min on the platform and 15 min inside the train.

The dose is the amount of particles deposited in the respiratory tract during breathing, and it can be expressed as

$$Dose = DF \times C \times t \times Q$$

where DF is the deposition fraction of aerosol particles in the respiratory system (dimensionless), C is the airborne particle concentration in units of μg m^{-3} (amount of particle inhaled per volume air), t is the exposure time in hours, and Q is the breathing rate in m^3 h^{-1}. From these parameters, DF is the least accessible factor, because it depends on the exposed subject characteristics, such as age, gender, health status, lungs morphology, respiratory parameters and activity, as well as on numerous other parameters including particle size, density, shape, and chemical composition (ICRP, 1994; Löndahl et al., 2007). DF increases with larger particle size leading to higher dose. Moreover, the DF is different for each region of the respiratory system (extrathoracic, tracheobronchial, and alveolar–interstitial). The breathing rate not only depends on the body size of the subject, but also of their activity and health status (Bennett & Zeman, 2004; ICRP, 1994). Furthermore, the deposition calculations used in the model are based upon the empirical equations proposed in the ICRP human respiratory tract model (Aleksandropoulou & Lazaridis, 2013).

To estimate the overall daily dose some activities were neglected, such as outdoor entertainment or indoor (at home and workplace) activity, therefore, assuming no indoor sources. The daily dose was determined for a healthy adult male living in Barcelona considering a typical time-activity pattern of a subject who has a sedentary job and commutes by subway. Time-activity pattern was based on information from the Spanish national statistical institute (http://www.ine.es/) and previous exposure studies carried out in Barcelona, which included Time-Microenvironment-Activity-Diaries (Schembari et al., 2013). The exposure concentrations at home and workplace were estimated just taking into account the infiltration of $PM_{2.5}$ from outdoor, for naturally ventilated buildings (Morawska & Salthammer, 2003). Therefore, these concentrations are underestimated due to the non-consideration of indoor sources, such as e.g. cooking at home or printer emissions in an office. Dosimetry calculations were performed using the aforementioned concentrations during exposure under variant physical activities. The additional physical activity levels considered were sitting/resting and sleeping with breathing rates of 0.54 and 0.45 m^3 h^{-1}, respectively. The aerosol density outside the subway system was assumed equal to 1.5 g cm^{-3}, which corresponds to the average density of typical ambient aerosols (Zhang, Canagaratna, Jayne, Worsnop & Jimenez, 2005). A monodispersed aerosol size distribution was considered with a MMAD of 0.21 μm and a GSD of 1.15.

Table 1
Average $PM_{2.5}$ mass concentrations (μg m^{-3}) on the subway platforms and at the urban background site (outdoor) for both measurement periods.

Warmer period			Colder period		
Measurement period	Subway station	Outdoor	Measurement period	Subway station	Outdoor
2 Apr–2 May 2013	32.3 (Joanic)	14.9	28 Oct–25 Nov 2013	69.7 (Joanic)	10.5
1 Jul–30 Jul 2013	51.1 (Santa Coloma)	16.6	10 Feb–10 Mar 2014	65.0 (Santa Coloma)	13.2
2 May–31 May 2013	39.6 (Tetuan)	14.3	25 Nov–20 Dec 2013	91.3 (Tetuan)	23.3
31 May–1 Jul 2013	20.2 (Llefià)	15.4	13 Jan–10 Feb 2014	40.5 (Llefià)	11.4

3. Results and discussion

3.1. Exposure concentrations

$PM_{2.5}$ concentrations in the Barcelona subway obtained from this experimental study have been reported by Martins et al. (2015a), and will only be summarised here.

Table 1 displays the average $PM_{2.5}$ concentrations measured at each subway station and in the outdoor environment. The outdoor concentrations were lower than those in the subway stations. Thus, the outdoor $PM_{2.5}$ concentrations do not seem to influence significantly the air quality in the subway stations, since most of the $PM_{2.5}$ load in the underground stations is generated within the subway system by the motion of the trains and the movement of the commuters (e.g. Querol et al., 2012). These results are in agreement with Nieuwenhuijsen, Gómez-Perale and Colvile (2007), who also found high PM concentrations in underground environments resulting from the generation or accumulation of PM in a confined space, particularly in old subway systems. In Barcelona subway, higher $PM_{2.5}$ concentrations were found in the stations during the colder period, mainly due to platform ventilation differences between seasons, being stronger during the warmer period. The new Llefià station showed on average lower $PM_{2.5}$ concentrations (around 50%) in comparison with old conventional stations (Joanic, Santa Coloma and Tetuan), which might be related to the design of the stations (with PSDs), but also due to the lower train frequency and more advanced ventilation setup.

Regarding the measurements inside the trains no seasonal pattern was found, thereby, in the current study average $PM_{2.5}$ concentrations under normal conditions inside the trains (with air conditioning) obtained in both measurement periods were used. These concentrations were 53.8, 37.1, 56.9, 46.9, 35.7 and 23.3 $\mu g\ m^{-3}$ in the lines L1, L2, L3, L4, L5 and L10, respectively. Again $PM_{2.5}$ concentrations inside the trains of older lines (L1-5) were higher than those in the trains of the new PSDs line (L10). On average, the $PM_{2.5}$ concentrations inside the trains were lower (around 15%) than those on station platforms.

Table 2 displays a typical time-activity pattern and the exposure concentrations for a 24-h period, including both indoor and outdoor environments considered for this study. The reported concentrations are the average values of all measurements during both seasonal periods to obtain overall exposure concentrations. For a subway commuting travel of 15 min inside the train and 5 min on the platform, the average $PM_{2.5}$ exposure would reach 44.5 $\mu g\ m^{-3}$ (Table 2), based on the exposure concentrations of 42.3 and 51.2 $\mu g\ m^{-3}$ inside trains and on platforms, respectively. Exposure concentrations at home and workplace were estimated taking into account the mean value of the indoor/outdoor ratio of 0.91 for $PM_{2.5}$, for naturally ventilated buildings in the absence of indoor sources (Morawska & Salthammer, 2003). Thus, the exposure concentrations at home and workplace were obtained based on the average $PM_{2.5}$ outdoor concentration of 15.0 $\mu g\ m^{-3}$ (Table 2). However, in addition to the particles from outdoor origin, aerosols are also generated in the presence of indoor activities (e.g. Abt, Suh, Allen & Koutrakis, 2000a; Abt, Suh, Allen, Catalano & Koutrakis, 2000b; Morawska & Salthammer, 2003; Afshari, Matson & Ekberg, 2005; Hussein, Hämeri, Heikkinen & Kulmala, 2005; Hussein et al., 2006; Lazaridis et al., 2006; Glytsos et al., 2010; Abdullahi, Delgado-Saborit & Harrison, 2013), and this should be taken into account when interpreting the dose results. Moreover, the natural ventilation does not provide a constant indoor/outdoor ratio (Hussein et al., 2006, 2005; Minguillón et al., 2012).

3.2. Particle dose in the subway system

The deposited $PM_{2.5}$ mass in the different regions of the HRT (ET, TB and AI) for a subway commuting travel, assuming the typical exposure time of 5 min on the platforms and 15 min inside the trains, is shown in Fig. 1. Particle dose in the HRT

Table 2
Daily time-activity pattern and $PM_{2.5}$ exposure concentrations.

| Time		Duration	Microenvironment	Activity level	$PM_{2.5}$ exposure
Start	End	(h)			($\mu g\ m^{-3}$)
00:00	07:00	7.0	Home	Sleeping	13.6
07:00	08:10	1.2	Home	Light exercise	13.6
08:10	08:20	0.2	Outdoor	Light exercise	15.0
08:20	08:40	0.3	Subway system	Light exercise	44.5
08:40	09:00	0.3	Outdoor	Light exercise	15.0
09:00	18:00	9.0	Workplace	Sitting	13.6
18:00	18:20	0.3	Outdoor	Light exercise	15.0
18:20	18:40	0.3	Subway system	Light exercise	44.5
18:40	18:50	0.2	Outdoor	Light exercise	15.0
18:50	21:00	2.2	Home	Light exercise	13.6
21:00	23:00	2.0	Home	Sitting/resting	13.6
23:00	00:00	1.0	Home	Sleeping	13.6
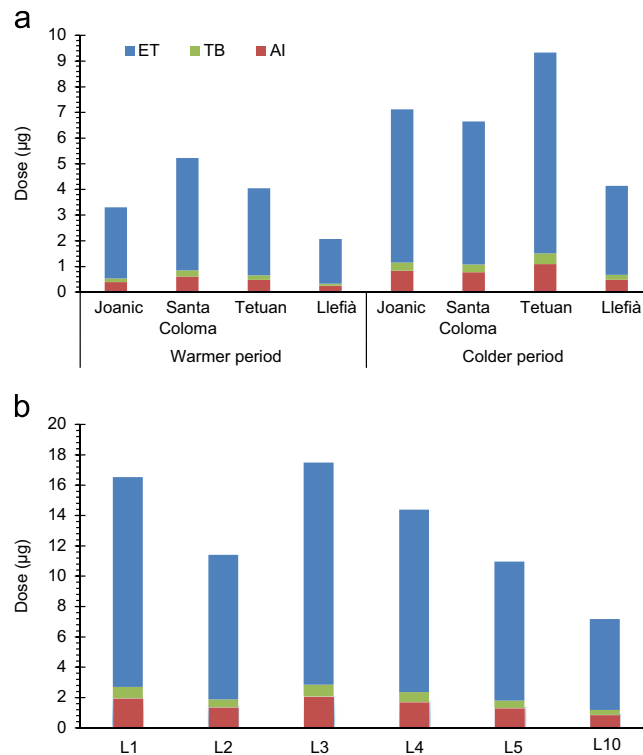
Fig. 1. PM$_{2.5}$ dose (µg) in the HRT for a subway commuting travel during (a) 5 min on the platforms and (b) 15 min inside the trains (regions of the HRT: ET – extrathoracic, TB – tracheobronchial and AI – alveolar–interstitial).

was proportional to the exposure concentrations. Thus, higher PM$_{2.5}$ dose occurred during the colder period in the subway stations (Fig. 1a), mainly due to platform ventilation differences between seasonal periods, as previously discussed.

Based on the intensive study performed in several subway stations of Barcelona by Martins et al. (2015a), different PM$_{2.5}$ concentrations were found among stations depending on the architectural design. The PM$_{2.5}$ dose for commuters in the new stations with PSDs (e.g. Llefià) was lower than in the older conventional stations. Among the latter, the stations with single narrow tunnel and one rail track (e.g. Tetuan) showed higher PM$_{2.5}$ doses than those observed in the rest of stations, taking into account that in the stations with one wide tunnel and two rail tracks without middle wall (e.g. Santa Coloma) a much more variable particle dose is expected, related to the large variability of PM$_{2.5}$ concentrations. Furthermore, the particle dose when a subject is in the subway stations may be affected by several influential parameters (Martins et al., 2015a). Experimentally, PM$_{2.5}$ concentrations showed clear differences during distinct times of the day and location on the platform, reflecting the influence of the ventilation settings, design of the stations and tunnels, train frequency and commuter density. Concentrations were lower during weekends, probably due to the lower frequency of trains. When travelling inside the trains in the oldest lines (L1-5), the PM$_{2.5}$ dose in the HRT was also higher than that in the new line 10 (Fig. 1b), as expected due to higher PM$_{2.5}$ concentrations.

During the subway commuting travel the highest dose was observed inside the trains as a result of the longer exposure time, evidencing that the exposure period is an important factor in the estimation of the particle dose, since the concentrations inside the trains were lower (around 15%) than those on station platforms. However, it is worth observing that the particle dose inside the trains was more than double (averagely 2.5 times higher) of those on the platforms.

Salma et al. (2002) reported that the biological response to airborne particles is assumed to be related to the amount of PM deposited in the different compartments of the HRT. With respect to deposition efficiencies, a large percentage (81.7%) of the inhaled mass of PM$_{2.5}$ was deposited in the whole human respiratory system and the remaining was exhaled. These total deposition follows the tendencies derived from the superposition of the regional depositions ($D_{total} = D_{ET} + D_{TB} + D_{AI}$). As shown in Fig. 1(a) and (b), the deposition fraction over the total inhaled mass showed substantial differences among the regions of the human respiratory tract (ET, TB and AI). The extrathoracic airways received the highest amount of the inhaled PM$_{2.5}$ mass deposited in the HRT (68.5%). This fraction reflects that the deposition of particles occurred mainly in the upper region of the respiratory tract, which does not penetrate into the lung, and is removed much more rapidly than the particles deposited in deeper regions of the respiratory system (e.g. Carvalho, Peters & Williams, 2011; Löndahl et al., 2014). In contrast, the lowest amount of inhaled PM$_{2.5}$ mass was deposited in the tracheobronchial region (3.7%) and the remaining mass in the alveolar–interstitial region (9.6%).

3.3. Estimated personal PM$_{2.5}$ daily dose

The current study deals with the individual's exposure to PM$_{2.5}$, in order to identify the activities and microenvironments that contribute most to an average daily dose. Given that short-term exposure may contribute significantly to average daily exposure to PM$_{2.5}$ mass concentration, the daily doses of total and regional deposited mass were calculated, considering a typical time-activity pattern (Table 2). Figure 2 shows the effect of PM$_{2.5}$ exposure concentrations on dose for total and regional respiratory tract. The deposition increases by increasing the exposure concentration.

In Table 3, dose, deposition rate and the contribution of the different activities/microenvironments for total PM$_{2.5}$ deposited mass are reported for a typical time-activity pattern (Table 2), considering no indoor sources at home and workplace. The minimum 24-h total dose of PM$_{2.5}$ in an adult male was around 78 μg. This value presents the lowest estimate of the particle dose and it is expected to be higher in real-life conditions after considering indoor sources of aerosol particles and spatial variability of outdoor aerosols. Abt et al. (2000a, 2000b) conducted an intensive study characterizing sources of indoor particles and they found that cooking activities, cleaning and the movement of people has a significant impact on indoor particle concentrations. Moreover, the difference in the absolute value is expected to yield higher personal exposure and dose depending on the residence and workplace of the subject being close to a road, in commute traffic, background environment, etc. (e.g. Buonanno et al., 2011; Knibbs, Cole-Hunter & Morawska, 2011; Minguillón et al., 2012; Salma et al., 2015; Wang, Morawska, Jayaratne, Mengersen & Heuff, 2011). In the current study, the outdoor concentrations were measured at a background station located in the urban area of Barcelona, despite the exposure and dose for people living and working nearby major roads and road junctions can be significantly higher, due to very high PM$_{2.5}$ concentrations recorded close to road traffic (Minguillón et al., 2014). Furthermore, as previously mentioned the particle dose is expected to increase while breathing through the mouth.

Regarding the total deposition fraction, 29.4% of the daily inhaled PM$_{2.5}$ was deposited in the HRT and the remaining was exhaled. The dose in the tracheobronchial and in the extrathoracic regions represented from 3.9% to 14.0% of the inhaled particles. The remaining particles were deposited in the alveolar–interstitial region (11.5%). Comparing the daily particle deposition with the particle deposition taking place only at the subway micro environment, the deposition fraction of the inhaled particles in the extrathoracic region and consequently the total deposition fraction were much lower for the daily deposition, due to the smaller particle size of the aerosol outside the subway system (see size distribution in Section 2.3). The decrease in deposition with decreasing particle size is in agreement with predictions of the ICRP model (ICRP, 1994). The deposition fraction in the alveolar–interstitial region was higher in the daily results since the smaller particles can penetrate into deep lung regions and can deposit there. Thus, these results showed that the regional distribution of deposited particles (i.e. the mass of particles that are deposited in each of the respiratory tract regions) is strongly dependent on particle size. However, for health effect purposes, the smaller particles depositing deeper in the lungs are less efficiently cleared compared to the larger particles that deposit preferentially in the upper airways where they are more easily cleared (Carvalho et al., 2011). The clearance mechanisms are a natural defence of the human body and operate in different regions of the lungs to eliminate the trapped foreign material (Hussain et al., 2011). Furthermore, comparing same mass deposits of large and small particles, the latter contains a much higher number of particles that need to be cleared (Carvalho et al., 2011).
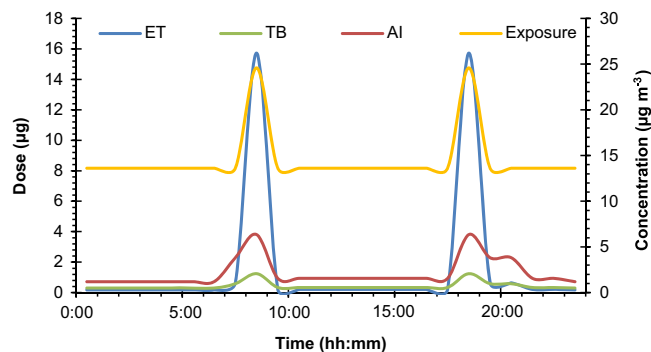

Fig. 2. Daily PM$_{2.5}$ exposure concentration and dose in the different regions of the HRT (ET – extrathoracic, TB – tracheobronchial and AI – alveolar–interstitial).

Table 3
Summary of PM$_{2.5}$ dose and time spend for each microenvironment.

Microenvironment	Dose (μg)	Daily dose fraction (%)	Time exposure (h)	Daily time fraction (%)	Deposition rate (μg h^{-1})
Home	24.4	31.2	13.3	55.6	1.8
Outdoor	3.9	5.0	1.0	4.2	3.9
Subway system	36.4	46.7	0.7	2.8	54.6
Workplace	13.3	17.1	9.0	37.5	1.5

There are considerable differences in the distribution of particle dose in the regions of the HRT along the day (Fig. 2) which are attributed to the different exposure concentrations and size distribution of PM$_{2.5}$ but also to the different physical activity levels. Brand et al. (1999) reported that for all particle sizes, the particle dose for each region of the HRT increases with increasing breathing rate. Increasing the breathing rate demonstrate the effect of inertial impaction in larger particle size, by increasing deposition in the upper respiratory tract and consequently increases the deposition fraction in the extrathoracic region. For smaller particle size, increasing the amount of aerosol inhaled due to high air velocity promotes an increase of particle deposition in the deeper lung.

An increase of physical activity from rest to exercise led to an increase of the dose, as also reported by Löndahl et al. (2007) and Daigle et al. (2003), because of the combined increase in deposition fraction and breathing rate. However, the effect of changing the breathing rate on the distribution of the daily particle dose in the HRT is lower compared to the particle size and the exposure concentrations.

Table 3 shows the contributions of the different microenvironments to the total particle daily dose. An important contribution arises from the commuting time spent in the subway ($\approx$ 3%), which accounted for a maximum of approximately 47% of the overall daily dose, corresponding to more than 36 μg per day. Therefore, commuting in the subway system represents the activity with the highest dose received per time unit (54.6 μg h^{-1}), due to the high particle concentrations and large particle size that contribute disproportionally to dose. Therefore, the high contribution of the subway exposure to the daily dose, despite the low time exposure, is mainly due to the higher PM concentrations and the larger particle size. Moreover, in relative terms, it becomes more relevant due to an underestimation of the dose received by people living/residing in microenvironments where higher particle concentrations are usually experienced, namely considering the indoor sources, as explained before.

Dose at home becomes less important (1.8 μg h^{-1}), partly because it includes night hours with lower PM concentrations and the exposed subject is under low activity level. However, this deposition rate was lower at home and workplace compared to background levels (outdoor), which is typical of an indoor microenvironment without any relevant particle mass sources as considered for this study (Table 3). It should be noted that a high PM dose is expected for all types of commuting means of transport due to the high PM exposure levels, as mentioned previously. Moreover, the particle size of the subway PM$_{2.5}$ is larger than the remaining means of transport, thus for a given concentration lead to a higher dose.

4. Conclusions

PM dosimetry models substantially improve personal dose assessment in replacing experimental investigations because they save time, efforts, and money in assessing the health effects arising from exposure to aerosol particles. However, modelling of the deposited dose require: a detailed description of the exposed subject characteristics and the time-activity pattern, a good estimate of the indoor and outdoor exposure levels to aerosol particle, physicochemical properties of inhaled particles, and a precise deposition fraction of aerosol particles in the different regions of the respiratory system. In this study, the above mentioned factors were taken into account and the dosimetry model ExDoM was applied for the estimation of the respiratory tract dose received by a healthy adult male exposed to PM$_{2.5}$ in the Barcelona environment, especially on subway system. Note that a monodispersed aerosol was considered for the dosimetry model, whereas it is possible that a finer mode is also present, and hence the reported results may underestimate the amount of particle mass that is deposited in the deeper regions of the respiratory system.

The dose of PM$_{2.5}$ during a subway commuting travel was calculated assuming the typical exposure time of 5 min on the platforms and 15 min inside the trains. Particle dose was proportional to the exposure concentrations both on the platforms and inside the trains. The highest dose was observed inside the trains due to the longer exposure time, evidencing that the exposure period is an important factor in the estimation of the particles deposited dose, despite concentrations inside the trains were lower than those on station platforms. Concerning the deposition fractions, a large percentage (82%) of the inhaled mass of PM$_{2.5}$ was deposited in the whole human respiratory system. The separation made for regional dose in the HRT for health effect purposes, shows that the highest amount of the inhaled particles deposited in the extrathoracic airways. However, the particles deposited in this region is removed much more rapidly than the ones in the deeper regions of the respiratory system.

Individual's typical daily exposure to PM$_{2.5}$ and dose were estimated for an adult male who lives in Barcelona and commutes by subway, considering no indoor sources at home and workplace. The distribution of deposited particles in the HRT showed considerable differences along the day, most dependent on the particle size and exposure concentrations. Changing the breathing rate had a minor effect on the distribution of deposited particles in the HRT.

The daily PM$_{2.5}$ dose obtained in this study represents the lowest estimate and it is expected to be higher in real-life conditions where aerosols are generated by indoor activities and outdoor exposure may take place at more polluted locations than the urban background environment, for example considering a traffic or a city-centre environment. Commuting by subway represented the highest dose received per time unit during the day, contributing to around 50% of the total particle dose, although the amount of time spent in the subway system accounts for only 3% of a day, owing mainly to the higher concentration and the larger particle size. However, the relative contribution to the total daily dose of PM$_{2.5}$ due to subway commuting is overestimated, and it should be interpreted as the maximum daily dose of particles received by a subject in this microenvironment. Given the relevance of the commuting dose with respect to the total daily dose, it is

important to mention that a high impact on $PM_{2.5}$ dose is expected regardless of the mean of transport, as reported in several studies, revealing that this result is not exclusive for subway commuting.

The results of this work show the importance of individual exposure and dose assessment, in order to provide information for the protection of public health. Personal exposure studies are an essential tool to identify health risks, set, and review air quality standards and evaluate effective policy interventions.

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