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Respiratory deposition dose of $PM_{2.5}$ and PM_{10} during night and day periods at an urban environment

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Abstract
Inhalation of particulate matter (PM) has been extensively associated with the worsening and onset of cardiorespiratory diseases, being responsible for millions of deaths annually. Assessment of PM deposition in the human respiratory tract is critical to better understand the health risks from environmental exposure of vulnerable age groups. In this study, $PM_{2.5}$ and PM_{10} day-night monitoring campaigns during the cold season were carried out in Bragança, Portugal. The multiple-path particle dosimetry (MPPD) model was used to quantify total and regional depositions in the human respiratory tract for four different age groups: infant (3 months), child (9 years), adult (21 years) and elderly (65 years). The results showed that concentrations for both PM fractions were higher during the night, a period marked by the burning of biomass for residential heating. Regional deposition fractions (DF) for $PM_{2.5}$ were in the ranges 17–38% (head), 4–14% (tracheobronchial) and 20–28% (pulmonary), while for PM_{10} were 24–67% (head), 4–27% (tracheobronchial) and 12–22% (pulmonary). Children and the elderly were found to be the most vulnerable groups to PM deposition, especially for the TB and H regions, respectively. The lifetime cancer and non-cancer risks associated with exposure to $PM_{2.5}$ exceeded the recommended limits, especially for children <10 years old. These findings provide useful information to alert authorities to the need to take action to reduce the pollution burden and protect the health, in particular of those most susceptible.

Keywords PM_{10} · $PM_{2.5}$ · MPPD · Inhaled dose · Exposure assessment · Risk assessment

Highlights
- MPPD was applied to $PM_{2.5}$ and PM_{10} for night/day periods and different age groups
- Regional doses of $PM_{2.5}$ showed significant differences between day and night periods
- $PM_{2.5}$ deposits mainly in the lung and PM_{10} in the head and tracheobronchial regions
- Children and infants presented higher percentages of deposition in the lung regions
- Non-carcinogenic and carcinogenic risks were found to be higher at night

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Introduction

Urban air pollution has grown attention over the past decades due to the clear deleterious human health effects resulting from exposure to various contaminants, among which particulate matter (PM) with aerodynamic diameters below 10 and 2.5 µm (PM_{10} and $PM_{2.5}$, respectively) stands out as the most dangerous (Alemayehu et al. 2020; Duan et al. 2021; Huang et al. 2021), especially due to its significant relationship with cardiorespiratory diseases (Bateson and Schwartz 2007; Khosravipour et al. 2022; Zhang et al. 2018). Plenty of studies support the hypothesis that the health effects of inhaling air particles are governed by more than just particle mass, since the chemical composition and the size fraction are highly variable in space and time (Alves et al. 2021; Cesari et al. 2018; Lucarelli et al. 2018; Phuc and Oanh 2022; Pio et al. 2020; Stojić et al. 2016). The complexity of urban aerosols comes from the different emission sources, which can produce different size-segregated PM as function of the strength of the sources in specific periods (Cipoli et al. 2023; Li et al. 2023;

Rogula-Kozłowska 2016; Rönkkö et al. 2018; Theodosi et al. 2011; Yang et al. 2023).

Apart from particle size, the deposition of PM in the human respiratory tract (HRT) also depends on lung morphometry and respiratory physiology. Ultrafine particles are prone to penetrate into the lung alveoli and bloodstream, while coarse particles are mainly settled in the upper respiratory tract (Hofmann 2011; Rissler et al. 2012; Sá et al. 2015; Deng et al. 2019), demonstrating the pivotal role of size in PM deposition. It has been documented that PM deposition depends on age, with children and the elderly identified as the groups most susceptible to exposure to air pollution (Sacks et al. 2011). The amounts deposited in the HRT are a relevant potential causative factor for investigating the health effects of PM pollution (Voliotis et al. 2021). Different models have been developed to estimate the total and regional deposition fractions: the ExDoM (Exposure Dose Model) (Aleksandropoulou and Lazaridis 2013), the ICRP model (International Commission on Radiological Protection), the NCRP model (National Council on Radiation Protection and Measurement) and the MPPD model (Multiple-Path Particle Dosimetry) (Asgharian et al. 2001; ICRP and Protection 1994; Koblinger and Hofmann 1990; National Council on Radiation Protection and Measurements 1997). Due to its user-friendly interface, the MPPD model has had a wide application in the literature (Behera et al. 2015; Oller and Oberdörster 2016; Avino et al. 2016; Lv et al. 2021; Amoatey et al. 2022; Khan et al. 2022). Most previous studies on PM exposure in Portugal are based on daily data (Almeida et al. 2014; Alves et al. 2023; Garrett and Casimiro 2011; Vicente et al. 2017). However, temporal variation in air pollutant concentrations and human activities may be crucial factors in health outcomes (Cheng et al. 2021; Lin et al. 2018). The concentration of pollutants in ambient air shows a temporary pattern, with higher levels at night compared to daytime, especially in winter (Barmpadimos et al. 2012; Gama et al. 2018). On the other hand, human behaviour, PM sources and meteorological conditions in urban areas show significant variations throughout the day, affecting the formation, removal and transport of atmospheric aerosols (Freiman et al. 2006; Galindo and Yubero 2017). In addition, day-night variations end up being reflected in different exposure patterns and, consequently, in distinctive health outcomes. Recent studies have pointed out that exposure to air pollution can vary dramatically between periods, with significant differences in the toxicological properties of PM between day and night periods (Galindo and Yubero 2017; Jalava et al. 2015).

Adverse health effects from exposure to PM result from its inhalation through the mouth or nose and subsequent deposition in the HRT. Particle deposition in HRT is governed and influenced especially by respiratory parameters, characteristics of lung morphology, and particle properties (Heyder et al. 1986; Heyder et al. 1980). Depositions in specific regions, including the head (H), tracheobronchial (TB) and pulmonary (P), of the respiratory system, are important to determine the adverse effects to human health (Goel et al. 2018). Currently, there are few studies focused on PM deposition for different periods of the day (Cipoli et al. 2022; Lv et al. 2021). In addition, most of the existing PM data of lung deposition target adult males (Subramaniam et al. 2003; Kameda et al. 2005; Avino et al. 2016; Menon and Nagendra 2018; Rajput et al. 2019). Only a few works have focused on PM deposition in children (Protano et al. 2017; Rissler et al. 2017; Patterson et al. 2014), and less attention has been paid to elderly groups (Kim and Jaques 2005; Manojkumar et al. 2019a; Manojkumar and Srimuruganandam 2022). As far as we know, there are no previous studies addressing $PM_{2.5}$ and PM_{10} deposition for different age groups using daytime and nighttime measurements in urban environments in Portugal. Furthermore, there is a lack of data on deposition in the HRT of vulnerable groups such as children and the elderly. For a clearer understanding of the $PM_{2.5}$ and PM_{10} deposition in the HRT, this study considers the concentrations obtained from different periods in an urban background site, aiming at (1) investigating the diurnal and nocturnal variability of $PM_{2.5}$ and PM_{10} concentrations during the cold season, (2) identifying potential risks, both carcinogenic and non-carcinogenic, due to PM exposure and (3) evaluating the total and regional deposition fractions, mass rate depositions and inhaled doses in the HRT of four age groups, by applying the widely accepted MPPD model. To establish a comparative scenario between day and night for different age groups, doses per minute were estimated assuming very light activities. The estimates produced by the model for the night and day periods constitute essential information to define the doses to be tested in vitro in PM toxicity assays to be carried out in the near future.

Material and methods

Sampling site

The small urban area of Bragança (10 km^2) has approximately 35,000 inhabitants (PORDATA 2021). The city is located in the northeast of Portugal in a mountainous region (700 m altitude). Bragança is in a Csa climate region (in the Köppen-Geiger classification, Peel et al. 2007), with rainy periods centred in the cold season (December to February), while the summer and spring are drier. Deterioration of air quality is more evident during the cold season because of vehicular traffic emissions and residential heating activities associated with more stable atmospheric conditions. In addition, as other regions of the Iberian Peninsula, Bragança is sometimes affected by

Saharan dust intrusions, which are more persistent in the warm period and with some less frequent occurrences in the cold period (Pereira et al. 2008; Russo et al. 2020). The city has been the subject of just a few climate and air quality studies (Dantas et al. 2019; Cipoli et al. 2022; Cipoli et al. 2023).

Particulate matter data ($PM_{2.5}$ and PM_{10}) were collected on the roof of the Agricultural High School, Polytechnic University of Bragança (41°48′20″ N, 6°45′42″ W) at about 15 m above ground level (Fig. 1). The selected site can be considered representative of the average altitude of the city. The school is located in the southwest part of the city and is surrounded by cafes, bars, supermarkets, small companies and residential buildings. In addition, the school is in the vicinity of busy avenues, which are adjacent to bike path often used by the population that runs around the entire outer perimeter of the Polytechnic (3 km long). Adults with infants and the elderly generally use this pathway for short walks during the midday, especially on sunny days in the cold season. However, it is observed that most adults who practice outdoor activities in this time of the year chose the beginning of the night period after the workday.

Particulate matter monitoring

The monitoring campaign took place over 55 days in the 2021 cold season (January 14th to March 17th), covering weekdays and weekends. Continuous monitoring of $PM_{2.5}$ was performed with a VEREWA Beta Attenuation Monitor (BAM model F701-20, Durag group, Germany). The BAM was operated with a flow rate of 1 $m^3\ h^{-1}$ and a temporal resolution of 2 h. Simultaneously, collection of PM_{10} was performed by a low-volume sampler (Echo PM TECORA, Italy) operated at a flow rate of 2.3 $m^3\ h^{-1}$, equipped with pre-weighed 47 mm quartz filter, covering two different periods (08:00 a.m – 06:00 p.m and 06:30 p.m – 07:30 a.m, local time) in order to determine intra-day concentrations. The selection of sampling periods was based on the hours of sunlight and the predominant activities in each period. The daytime period was monitored during the hours of sunlight, during which there is a high probability of the elderly and children frequenting the outdoor environment. The night period covered the return home after the working day, during which there is an intensification of biomass burning. Due to partial or complete loss of data caused by power outages, equipment failures and/or heavy rains, only 40 days with valid data for both size fractions were selected for model application.

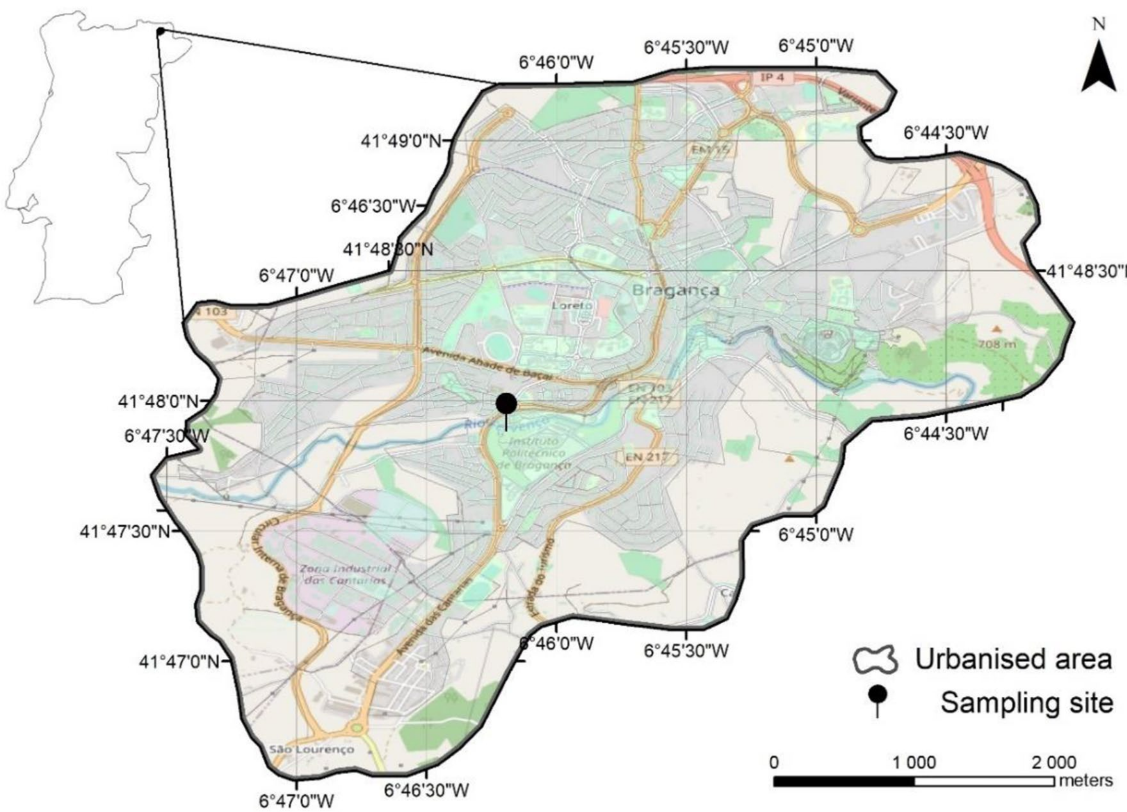

Fig. 1 Location of the monitoring site in the city of Bragança

Both the Beta monitor and the gravimetric sampler were factory calibrated before the monitoring campaign, and flow/zero checks are made during the whole period of experiments. Additionally, weather variables (air temperature, relative humidity, precipitation, wind direction and speed) were obtained at 1-h resolution from a portable weather station (Campbell Scientific Inc., USA), installed next to the other equipment on the roof.

As gravimetric measurements of $PM_{2.5}$ were not performed due to the unavailability of a gravimetric equipment with this cutoff size, and since BAM values were used in the model without any correction, some discrepancies between estimated and actual $PM_{2.5}$ doses may occur. However, several intercomparisons between gravimetric and BAM measurements have shown very good correlations with slopes close to 1, although in winter conditions the continuous monitor tends to slightly underestimate the concentrations of the gravimetric reference method (Chung et al. 2001; Hauck et al. 2004; Shukla and Aggarwal 2022; Takahashi et al. 2008).

Multiple-path particle dosimetry model

The mass rate and deposition fractions of $PM_{2.5}$ and PM_{10} were estimated for each region of the human respiratory tract (Head (H), Tracheobronquial (TB) and Pulmonary (P)) using the Multiple-path particle dosimetry model (MPPD). The Age-Specific 5-Lobe model was selected to simulate the deposition of lung particles, which has 10 different geometric configurations of the lung structure for different age groups from 3 months to 21 years. Four different age groups were considered in this study: infants (3 months), children (9 years), adults (21 years) and the elderly (>65 years). The lung structure for the elderly was not available, so the lung geometry of adults was used, and specific respiratory parameters were changed to reflect the characteristics of the elderly population. The physiological parameters of specific age groups (functional residual capacity, upper respiratory tract volume, breathing frequency and tidal volume) used in the software are given in Table 1, according to the recommendations of the International Radiological Protection Commission (ICRP 1994) and the model itself. For the infant, child and adult groups, the default values provided by the model were used. As the values for the elderly were not available, these parameters were adopted from the literature (Manojkumar and Srimuruganandam 2022). In addition, to establish a comparative scenario of short-term outdoor exposure between age groups, the level of activity was considered to be very light.

The properties of inhaled PM play an essential role in lung deposition. Due to the unavailability of specific information during the study period, the values found by Cipoli et al. (2022) for the mass median aerodynamic diameter (MMAD) and for the geometric standard deviation (GSD) during the cold season for the city of Bragança were used. These values can be consulted in more detail in Table S1.

In order to estimate the deposition fraction (DF) in the lung airways, several assumptions were made. Particles have

Table 1 Input parameters used in the MPPD model

Parameters		Options/values			
		Infant	Child	Adult	Elderly
Airway Morphometry	Species	Human			
	Model	Age-specific 5-lobe			
	FRC (ml)	17.97	683	2123.75	3301
	URT volume (ml)	2.45	22.44	42.27	50
Particle properties	Density (g cm^{-3})	1			
	Aspect ratio	1			
	Diameter (µm)	MMAD			
	Concentration (µg m^{-3})	Mean of PM			
	Particle distribution	Single			
	MMAD	Based on Cipoli et al. (2022)			
	GSD	Based on Cipoli et al. (2022)			
Exposure scenario	Exposure condition	Constant exposure			
	Aerosol concentration (µg m^{-3})	$PM_{2.5}$ and PM_{10}			
	Breathing frequency (min^{-1})	39	17	14	12
	Tidal volume (ml)	30.44	278.2	477.2	750
	Inspiratory fraction	0.5	0.5	0.5	0.5
	Breathing scenario	Nasal			

a density of 1 g cm^{-3}, all particles enter the respiratory tract through the nose and body orientation is assumed to be vertical, except for infants, who were considered to be lying on their backs. Additional information about the model and its capabilities can be found at Anjilvel (1995) and Miller et al. (2016). Version 3.04 of the model can also be downloaded for free at https://www.ara.com/products/multiple-path-particle-dosimetry-model-mppd-v-304.

Inhalation dose calculations

Based on the mean concentrations obtained for both $PM_{2.5}$ and PM_{10} and the deposition fractions calculated using MPPD, the individual short-term exposure dose was estimated for each period and age group using the following equation (Goel et al. 2018):

$$Dose\left(\mu g\ \min^{-1}\right) = PM_i * DF * VE \quad (1)$$

where PM_i is the mean PM concentration of fraction i (µg m^{-3}), DF is the deposition fraction and VE represents ventilation per minute (m^3 min^{-1}). VE values for infant, child, adult and the elderly were 2.4×10^{-3}, 8.4×10^{-3}, 10.9×10^{-3} and 9.9×10^{-3} USEPA (2011) respectively.

Health risk assessment

The methodology proposed by the United States Environmental Protection Agency (USEPA 2009) was applied for estimating the health risk from inhalation for all age groups. This methodology is commonly used to estimate the risks of specific PM-bound compounds such as metals and polycyclic aromatic hydrocarbons (e.g. Alves et al. 2021; Alves et al. 2020; Liu et al. 2019; Slezakova et al. 2014) but has also been successfully applied to PM concentrations in other studies (Heydari et al. 2019; Pavel et al. 2020; Yunesian et al. 2019). The lifetime excess cancer risk (ELCR) and the non-cancer risk were calculated for PM inhalation based on the mean concentrations obtained in each period. It has been considered that ambient levels are representative of the total population exposure given the lack of time-activity patterns for distinct microenvironments (Kazakos et al. 2020). According to the World Health Organisation (WHO), acceptable ELCR limit values range from 1×10^{-5} to 1×10^{-6} (USEPA 2007). The ELCR was calculated using the following equation:

$$ELCR = LADD\ \left(\mu g.kg^{-1}.day^{-1}\right) \times SF\ \left(kg.day\ \mu g^{-1}\right) \quad (2)$$

where $LADD$ refers to the lifetime average daily dose and SF is the slope carcinogenic potency factor. $LADD$ was calculated as follows:

$$LADD = \frac{C\ \left(\mu g.m^{-3}\right) \times IR\ \left(m^3.day^{-1}\right) \times ET \times EF\left(days.year^{-1}\right) \times ED(years)}{AT\ (days) \times BW(kg)} \quad (3)$$

where C is the PM concentration, IR is the inhalation rate, ET refers to the exposure time (hours/day), EF is the exposure frequency, ED is the duration of exposure, BW is body weight for each age group and AT is the average lifetime. Given that the risk analysis was based on a winter campaign, a period during which concentrations in Bragança are higher than those recorded in summer (Cipoli et al. 2022), the results obtained must be seen as the 'worst' scenario. All input parameters segregated by age and period are described in detail in Table S2. To determine the SF value, Eq. (4) provided by the USEPA was used:

$$SF = \frac{BW}{UR \times IR} \quad (4)$$

where UR is the unit risk (µg m^{-3}), BW is body weight (kg) and IR is the inhalation rate (m^3 day^{-1}). No studies were found that reported UR for PM_{10}. The UR value for $PM_{2.5}$ (0.008 µg m^{-3}) was obtained from the study by Greene and Morris (2006). Thus, ELCR was estimated only for $PM_{2.5}$. Since there are no government or health agencies (e.g. USEPA and WHO) reporting unit risks for PM_{10} and $PM_{2.5}$, the carcinogenic risk results should be regarded as indicative. The non-carcinogenic risk of $PM_{2.5}$ and PM_{10} was assessed using Eq. (5), based on the ratio between the LADD and the reference dose (RfD):

$$HQ\ (hazard\ quotient) = \frac{LADD\ (lifetime\ average\ daily\ dose)}{RfD\ (reference\ dose\ \left(\mu g.kg^{-1}.day^{-1}\right)} \quad (5)$$

To calculate the RfD (Eq. 6), the inhalation reference concentration (RfC) of 5 µg m^{-3} for $PM_{2.5}$ and 50 µg m^{-3} for PM_{10} were used (De Oliveira et al. 2012; Li et al. 2017), in order to assess the harmful impacts on human health.

$$RfD = \frac{RfC\ \left(inhalation\ reference\ concentration\ \mu g.m^{-3}\right) \times IR}{BW} \quad (6)$$

In general, if HQ is greater than 1, the potential non¯carcinogenic chronic risks are considered to be of concern and require action to be taken, while values lower than 1 are considered as acceptable risk (USEPA 2007).

General statistical analysis

PM concentration data were statistically analysed using SPSS software (IBM Statistics software v. 24). The non-parametric method of Mann-Whitney was used to evaluate the differences between daytime and nighttime. Pearson's correlation test was utilised to analyse linear correlations between $PM_{2.5}$ and PM_{10} and obtain significant differences at a confidence level of 95% ($p < 0.05$).

Results and discussion

General overview

During the sampling campaign, the mean air temperature and relative humidity varied between 1.2 and 15.2 °C and 39.8 and 98.3% respectively. Accumulated precipitation was 12.8 mm, falling on 15 days out of the 55 days of sampling. The average wind speed was 2.7 m s^{-1} with a predominantly southeasterly direction.

The in situ intercomparison between the gravimetric sampler and the BAM produced a high degree of agreement with significant correlations ($r^2 = 0.93$, slope of 1.72 and y-intercept of 12.3 µg m^{-3} for daytime with a $p < 0.05$ and $r^2 = 0.91$, slope of 1.76 and y-intercept of 9.74 µg m^{-3} for nighttime with a $p < 0.05$) for the entire period. Overall, the mean concentrations of $PM_{2.5}$ and PM_{10} (Fig. 2) were 38% and 17% higher at night than during the day, respectively. In the daytime, mean concentrations of $PM_{2.5}$ ranged between 4.78 and 46.9 µg m^{-3}, while PM_{10} varied from 13.6 to 92.3 µg m^{-3}. At night, values from 3.77 to 44.9 µg m^{-3} were found for $PM_{2.5}$ and from 12.4 to 96.7 µg m^{-3} for PM_{10}.

The mean $PM_{2.5}/PM_{10}$ ratio exhibited significant differences between nighttime (0.46) and daytime (0.37), which can be explained by the presence of sources that contribute to the emission of fine particles in the nocturnal period (e.g. biomass burning for residential heating) and lower dispersion conditions. Results of a previous source apportionment study in the city for the same monitoring campaign (Cipoli et al. 2023) indicated that, during the nocturnal period, the main contributions to the PM_{10} levels were as follows: biomass burning (30%) > traffic (26%) > dust (22%) > secondary inorganic aerosol (12%) > sea salt (10%). It has been shown that the $PM_{2.5}/PM_{10}$ ratio varies widely between regions in Europe. Ratios in the range 0.6–0.8 have been reported for regional background sites, while at urban background sites the values varied from 0.4 to 0.5 (Southern Spain and the Canary Islands) to 0.8 in Germany, The Netherlands and Northern and Central regions of Spain. At kerbside sites, the ratios are typically 0.6–0.7, but lower values (0.4) have been obtained in Sweden and in the Canary Islands, pointing to stronger inputs from road dust emissions and intrusions of PM_{10} plumes from the Sahara desert, respectively (Querol et al. 2004).

The difference between daytime and nighttime periods in the present study may, at least in part, be related to the possible underestimation of $PM_{2.5}$ concentrations by the Beta monitor under typical winter meteorological conditions (Le et al. 2020). The mean daily concentrations of $PM_{2.5}$ exceeded the value of 15 µg m^{-3} recommended by the World Health Organisation in 45% of the time, while for PM_{10} the exceedances of the daily value of 50 µg m^{-3} (Directive 2008/50/EC) corresponded to 30% of the monitoring period. In the previous source apportionment study it was demonstrated that biomass burning during nighttime is the main responsible for those exceedances (Cipoli et al. 2023).

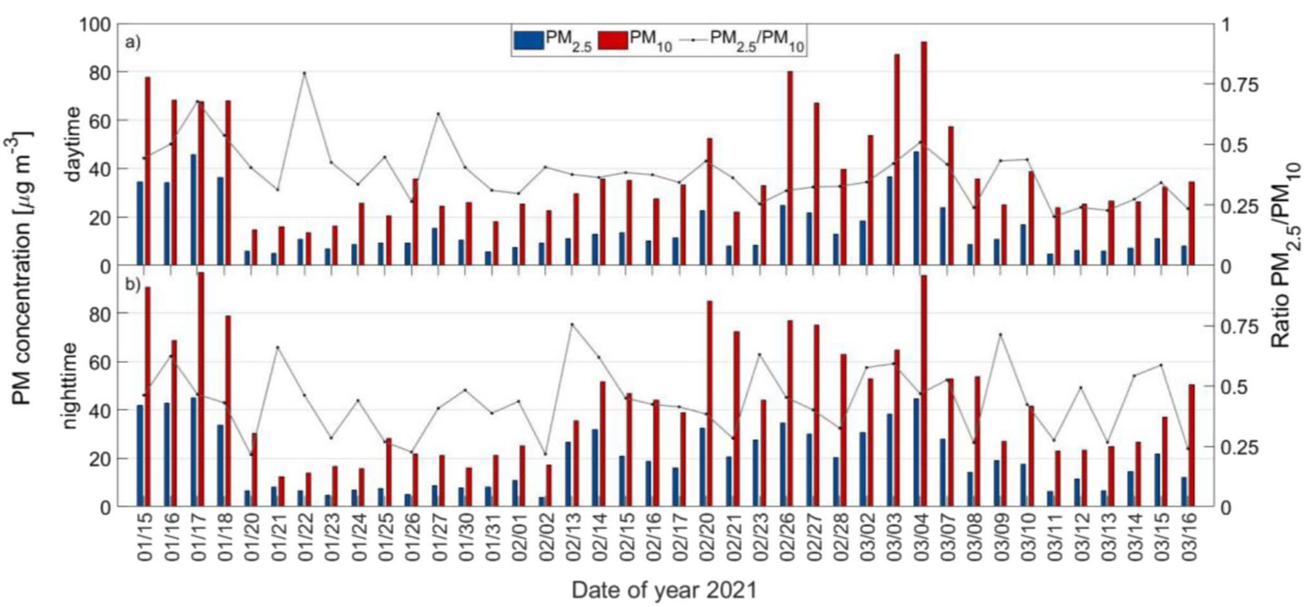

Fig. 2 Mean concentrations of $PM_{2.5}$ and PM_{10} during **a** daytime and **b** nighttime and ratios of $PM_{2.5}/PM_{10}$ for the entire sampling period

Total and regional PM deposition fractions

The deposition fractions segregated by PM size, age (Infant (I), Child (C), Adult (A) and Elderly (E)) and period of the day are shown in Table 2. PM_{10} showed the highest DF (0.58 – 0.86) in the respiratory tract, followed by $PM_{2.5}$ (0.43 – 0.64). Furthermore, for both fractions, higher DFs were found during the day than at night, with more pronounced increments (1.38 times for $PM_{2.5}$ and 1.46 times for PM_{10}) for the elderly category. When considering the $PM_{2.5}$ fraction, the following order of vulnerability was found for the daytime period: child > elderly > infant > adult. For the night period, the order was as follows: child > infant > elderly > adult. For PM_{10}, the next sequence was observed for the daytime period: elderly > adults > child > infant. The corresponding order for the night period was: child > infant > elderly > adult. It is worth noting that the estimated DF for the daytime period for both fractions pointed to the elderly as the most vulnerable age group, while for the nighttime period, the child category proved to be the most susceptible. Similar results for total DFs have been reported elsewhere (Madureira et al. 2020; Manojkumar et al. 2019; Manojkumar and Srimuruganandam 2022).

In relation to the regional DF, the deposition in the H, TB and P regions varied in a considerable range. During the day, PM_{10} and $PM_{2.5}$ showed deposition percentages in the H region for both size fractions (PM_{10}: I (33%), C (40%), A (63%), E (67%) and $PM_{2.5:}$ I (24%), C (29%), A (34%) E (38%)) higher than those of the night period. However, during the night, the deposition of $PM_{2.5}$ was dominant in the P region ($PM_{2.5}$: I (24%), C(28%), A (21%), E (21%)). Head and pulmonary depositions for specific periods agree with the results of other studies (Cipoli et al. 2022; Lv et al. 2021). The deposition percentages in the TB region for both PM fractions were generally lower than in the other regions of the respiratory tract, except for PM_{10} in the infant category, where TB (27% daytime and 23% nighttime) exceeded the percentages found in the P region (15% daytime and 13% nighttime).

In general, considering both periods, the percentages of $PM_{2.5}$ deposition in the P region were more pronounced for children, followed by infants, while for the H region, the most affected groups were the elderly, followed by adults. For the TB region, the infant category had the highest DFs. PM_{10} deposition was dominant in the H region for all age categories, with substantial differences in the regional deposition in P. For PM_{10}, the more persistent deposition in the upper respiratory tract may be related to impaction and sedimentation mechanisms, while $PM_{2.5}$ is governed by the Brownian diffusion mechanism, penetrating deeper into the airways. It should be noted that it was estimated that, during the day, the contribution of dust represented up to 25% of PM_{10} concentrations, while traffic (exhaust and non-exhaust emissions) accounted for 38% (Cipoli et al. 2023). In summary, the main factors that induce these variations are linked to the properties of the particles (MMAD and GSD), which depend on the emission sources, secondary reactions in the atmosphere and deposition mechanisms in the HRT (as a function of age).

Deposited mass rate

The deposited mass rate by age, period and particle size (Fig. 3) were also estimated for this study. $PM_{2.5}$ showed more evident deposition in the lung region compared to PM_{10}, especially for infants. The highest deposition rates of $PM_{2.5}$ were found in the elderly category (daytime 2.54×10^{-5} µg min^{-1} and nighttime 4.66×10^{-5} µg min^{-1}), while for PM_{10}, children were subject to a higher deposition rate (daytime 2.70×10^{-3} µg min^{-1} and nighttime 1.80×10^{-3} µg min^{-1}). In general, mass deposition for $PM_{2.5}$ decreased with age, being lower in

Table 2 Total and regional deposition fractions of $PM_{2.5}$ and PM_{10} for age-specific groups.

Daytime								
	$PM_{2.5}$				PM_{10}			
	Infant	Child	Adult	Elderly	Infant	Child	Adult	Elderly
Total	0.59	0.64	0.59	0.62	0.75	0.79	0.83	0.86
H	0.24	0.29	0.34	0.38	0.33	0.40	0.63	0.67
TB	0.14	0.07	0.04	0.04	0.27	0.17	0.05	0.04
P	0.21	0.25	0.21	0.20	0.15	0.22	0.15	0.14
Nighttime								
	$PM_{2.5}$				PM_{10}			
	Infant	Child	Adult	Elderly	Infant	Child	Adult	Elderly
Total	0.52	0.55	0.43	0.45	0.61	0.64	0.58	0.59
H	0.20	0.23	0.17	0.19	0.24	0.30	0.40	0.43
TB	0.08	0.06	0.05	0.05	0.23	0.14	0.05	0.04
P	0.24	0.28	0.22	0.21	0.13	0.19	0.13	0.12

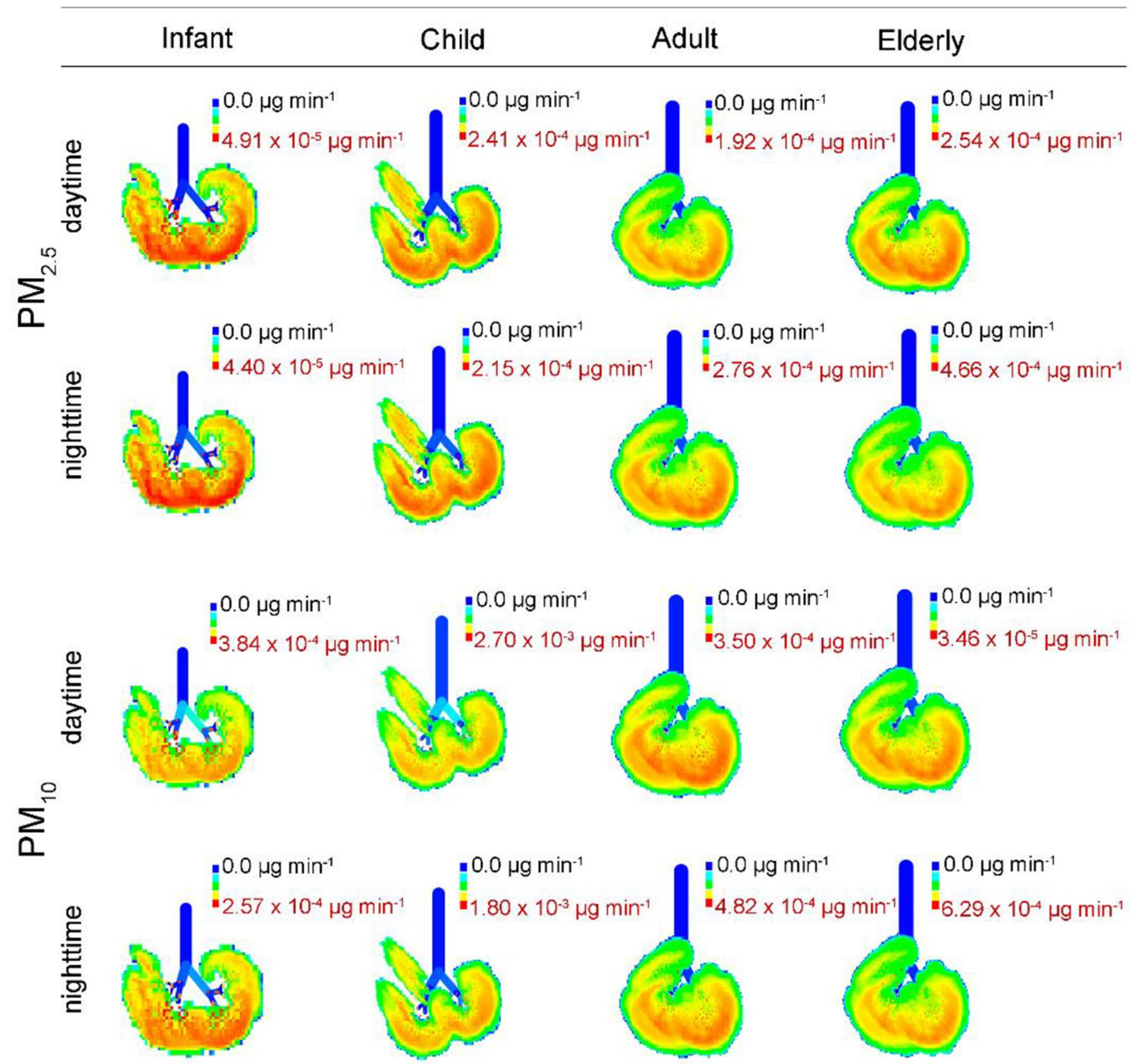

Fig. 3 Visualisation of deposited mass rates of $PM_{2.5}$ and PM_{10} in lungs for specific age groups and periods of the day

infants (4.65×10^{-5} μg min^{-1}) and higher in the elderly (3.60×10^{-4} μg min^{-1}). These results are in line with those reported by Madureira et al. (2020) and Manojkumar and Srimuruganandam (2022), who found similar relationships for fine particles as a function of age.

Considering all age groups, the deposited mass rate of $PM_{2.5}$ and PM_{10} was higher for the night period. Although this can be explained by the higher concentrations and specific particle properties in this period (e.g. smaller aerodynamic diameter due to biomass combustion for residential heating), the mass deposition rates are greatly influenced by the GSD values. In fact, changes in particle properties and distribution can affect the region and deposition rate in the respiratory tract.

Lobar deposition

The deposition fractions in the lobar region were estimated up to the 23^{rd} generation and can be seen in Fig. 4. The first generation begins in the bronchi, and the last corresponds to the alveolar sacs. In addition, deposition was evaluated for the entire lung (EL), and for the 5 lobes: right upper (RU), right middle (RM), right lower (RL), left upper (LU) and left lower (LL). Both PM fractions have high deposition in the LL ($PM_{2.5}$: 31–37% and PM_{10}: 30–37%) and RL ($PM_{2.5}$: 26–39% and PM_{10}:26–38%) regions, whereas the smallest depositions are found in RM ($PM_{2.5}$: 5–8% and PM_{10}: 5–8%). Infants and children presented lobar depositions according to the following order: RL > LL > LU> RU

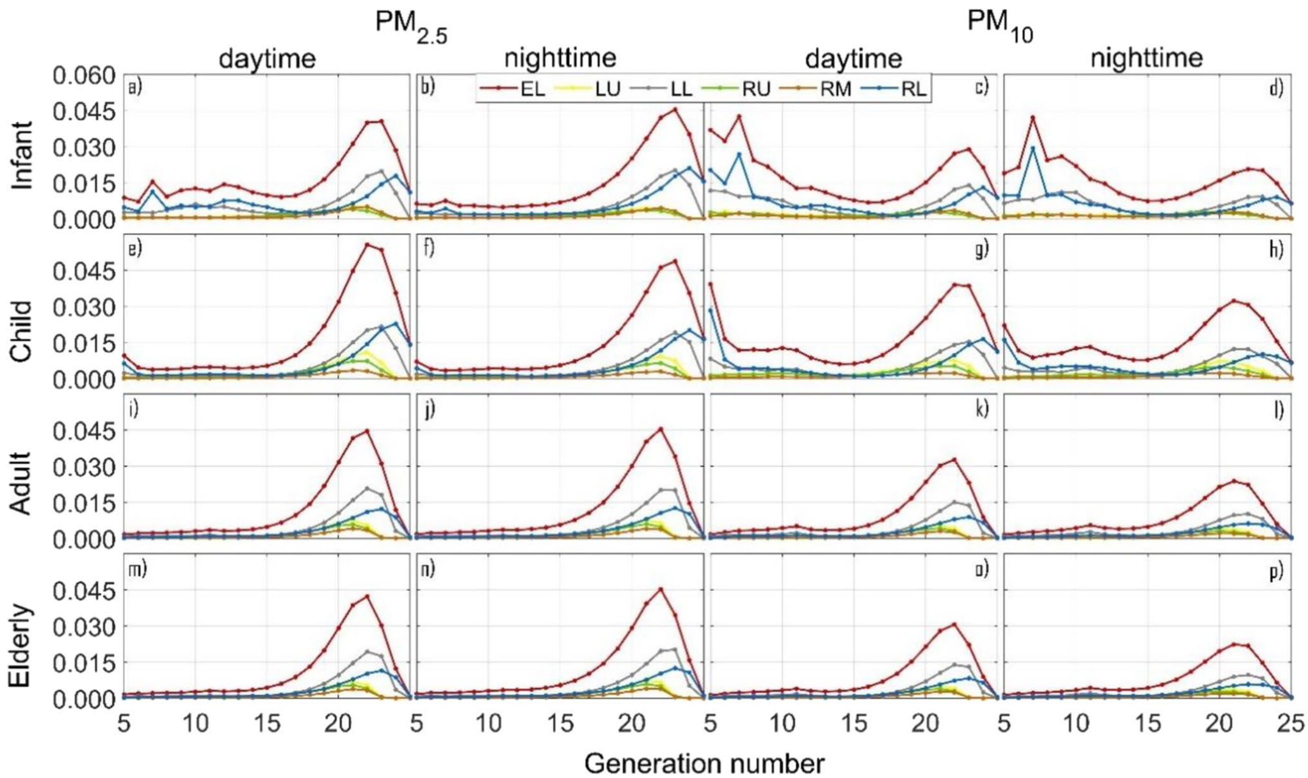

Fig. 4 Lobar deposition fractions of $PM_{2.5}$ and PM_{10} per generation number by age-specific groups for different day periods

> RM, while for adults and the elderly the sequence was as follows: LL > RL > LU > RU > RM. Regardless of age groups, the lowest deposition rates were found for the upper and middle lobes. This difference can be attributed to lobar lengths and volumes, since LU, RU and RM have about 3 to 4 times less volumes than the lower lobes, and therefore, lower lobar deposition (Islam et al. 2017).

The child and infant age groups presented the highest total lobar deposition fractions for $PM_{2.5}$ and PM_{10} in both periods, respectively. For exposure to PM_{10}, infants showed a bimodal behaviour, with PD peaks related to the 8th and 23rd generations. For the other age groups, the highest PDs were centred on generations 21–23. The 23rd generation (alveolar) received about 4–19 and 1–14 times more deposition of $PM_{2.5}$ and PM_{10} than the bronchial region (5th generation), except for the PM_{10} exposure of the infant age group, whose deposition in the 5th generation was 1.3 times higher than that of the 23rd generation. The peaks of PM_{10} lobar deposition in infants can be explained by the smaller diameter of the airways and higher respiratory rate in this age group (book toxicology of the lung). Although the deposition fraction between the number of generations varied in all age groups, the trend towards an increase in the deposition fraction after the 20th generation was similar.

In general, lung anatomical structure, inhalation rates and particle inertia play a key role in understanding deposition in specific regions of the lung (Islam et al. 2017). Deposition in lobar regions is linked to decreased lung function and an increase in chronic diseases (Guo et al. 2018; Zhao et al. 2017), as well as to the development of carcinomas in specific regions (Hofmann 2011).

Inhaled PM doses

The inhalation doses associated with specific deposition by region of the HRT can be seen in Fig. 5. Regardless of the period analysed, the elderly age group experienced the highest average PM_{10} inhaled doses by region: H (267.6×10^{-3} µg min^{-1}), TB (15.2×10^{-3} µg min^{-1}) and P (57.5×10^{-3} µg min^{-1}). Detailed information on inhaled doses by sampling period and day period can be found in the supplementary material (Tables S3-S6). The doses found for the daytime period by $PM_{2.5}$ inhalation were only higher than those for the nighttime period for the TB region of the infant age group (5.4×10^{-3} µg min^{-1} in daytime and 3.7×10^{-3} µg min^{-1} in nighttime) and for the H region of adults and the elderly (57.2×10^{-3} µg min^{-1} in daytime and 36.4×10^{-3} µg min^{-1} in nighttime, and 57.9×10^{-3} µg min^{-1} in daytime and 37.6×10^{-3} µg min^{-1} in nighttime, respectively). The higher nighttime dose of fine particles is in agreement with the results reported by Lv et al. (2021). These findings show that the deposited

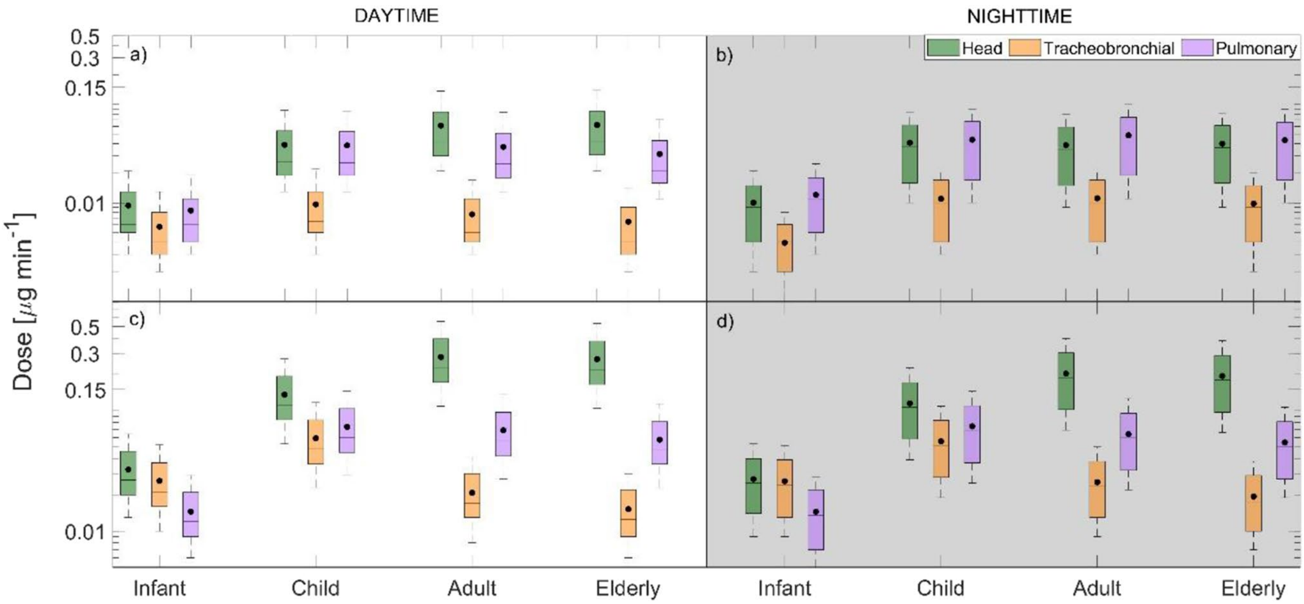

Fig. 5 Boxplot of: **a** $PM_{2.5}$ daytime, **b** $PM_{2.5}$ nighttime, **c** PM_{10} daytime and **d** PM_{10} nighttime inhaled dose (μg min^{-1}) by age-specific group for each region of the human respiratory tract

dose is dependent not only on the ambient concentration but also on the particle size. Day–night regional doses of $PM_{2.5}$ showed statistically significant differences ($p <$ 0.05) for all groups, due to specific sources in different periods. In summary, the vulnerability of the age groups followed the following order: Head—E > A > C > I, Tracheobronchial—E > C > A > I and Pulmonary—E > A > C > I.

Inhaled doses of PM_{10}, regardless of age groups, were high for all regions during the day compared to the night. Children revealed the highest doses of PM_{10} (57.4×10^{-3} μg min^{-1} in daytime and 53.4×10^{-3} μg min^{-1} in nighttime) for the TB region, being higher than those estimated for infants, adults and the elderly by about 2.5, 2.3 and 2.1 times, respectively. The H and P regions presented the highest deposited doses of PM_{10} for the elderly age group, being up to 15 and 7 times higher than those of infants. With regard to PM_{10} inhalation, in general, the following order was observed for each region: Head—E > A > C > I, Tracheobronchial—C > E > I > A and Pulmonary—E > C > A > I. For both PM fractions significant differences were found in the H region for adults and the elderly depending on the period analysed ($p < 0.05$). This suggests that specific sources act in certain periods, contributing to the emission of different PM sizes. According to the results obtained, some kind of long-term health effect is to be expected, especially for the elderly and children, who are the most vulnerable groups to PM exposure.

Health risk assessment of PM

Carcinogenic (ELCR) and non-carcinogenic (HQ) risks were calculated for all age groups and are shown in Table 3. All the ELCR values exceeded the recommended limits (1×10^{-5} to 1×10^{-6}) for all age groups. The higher levels for children aged 9 and under indicate that this age group is at risk due to exposure to $PM_{2.5}$, and therefore measures to mitigate and control the levels of this pollutant must be applied to reduce potential carcinogenic risks. In agreement with the present results, similar findings were previously reported by Yunesian et al. (2019), who

Table 3 Cancer risk (ELCR) related to $PM_{2.5}$ concentrations and hazard quotient (HQ) related to $PM_{2.5}$ and PM_{10} concentrations for all age groups

Period	ELCR of $PM_{2.5}$ (10^{-4})			
	Infant	Child	Adult	Elderly
Daytime	224	146	1.93	1.93
Nighttime	377	244	3.22	3.22
	HQ of $PM_{2.5}$			
Daytime	30.9			
Nighttime	51.6			
	HQ of PM_{10}			
Daytime	7.78			
Nighttime	11.6			

also observed that children and young people in Teheran are at high risk of adverse health effects.

The non-carcinogenic risk pointed out that exposure to both PM fractions by all age groups can trigger or aggravate the development of non-malignant chronic diseases (HQ > 1). Higher ELCR and HQ values for $PM_{2.5}$ exposure in the night period reinforce the strength of specific sources, such as biomass burning, contributing to fine particles that are potentially harmful to human health.

Implications and limitations

Exposure to $PM_{2.5}$ and PM_{10} in the cold season was associated with high risks of chronic effects on human health, with the elderly and children being especially susceptible. In the city of Bragança, 30% of the population is elderly (PORDATA 2021), which reinforces the importance of adopting mitigation measures by the authorities. As biomass burning for residential heating has been shown to be the main source of PM in winter, greater attention should be given to the heating systems, since most devices are old and out of date (unpublished results from the group), not complying with the emissions of the eco-design directive (EU Regulation 2015/1185). Because of the usual lack of time-activity patterns for different microenvironments, inhalation exposure assessments are generally based on ambient concentrations, which as considered representative of the total population exposure (Kazakos et al. 2020). However, outdoor levels may not always be surrogates of inhaled doses in the indoor air. In fact, outdoor measurements may underestimate concentrations in dwellings, as reported by studies that simultaneously evaluated the indoor and outdoor air quality in residences equipped with fireplaces and woodstoves (e.g. Vicente et al. 2021).

Some additional limitations can be identified in the present study. The monitoring campaign did not cover other seasons, and the results were considered representative of the worst exposure scenario. Furthermore, as there was no equipment available to monitor the particle size distribution, specific values of MMAD and GSD obtained in a previous campaign to characterise the properties of the local aerosol were assumed. However, these properties can change depending on the local emission sources, atmospheric conditions, monitoring distance from the sources and secondary reactions. Changes in particle properties can lead to different depositions and inhaled doses. Therefore, the results are important indicators for the identification of potentially susceptible groups, but should be analysed with caution, requiring the determination of specific factors for understanding the properties of the particles in future studies. In addition, to more accurately estimate exposure of different age groups to PM, monitoring in different seasonal periods should be carried out.

Conclusions

$PM_{2.5}$ and PM_{10} showed higher concentrations at night, which can be explained by greater atmospheric stability, accumulation of pollutants due to lower wind speed (when compared to daytime) and specific emission sources, such as residential wood combustion for heating during winter. Furthermore, the highest $PM_{2.5}/PM_{10}$ ratios were found at night, reinforcing the presence of fine particles in the atmosphere due to incomplete combustion processes. The highest deposition values in the upper respiratory tract (head and tracheobronchial regions) were mostly related to PM_{10} for all age groups, while depositions in the pulmonary region were dominated by $PM_{2.5}$. In addition, higher deposited doses for both PM fractions were found as a function of age: elderly > adults > child > infant. Regarding the deposited dose, the elderly group was identified as the most vulnerable. However, the assessment of potential carcinogenic and non-carcinogenic risks indicated values above the safety limits for the infant and child groups, highlighting the adverse impact on the health of these vulnerable groups from exposure to fine particles resulting from intense biomass burning. The results of this study show the urgency and importance of controlling air pollution, reinforcing the need to update the systems used for residential heating in order to reduce personal exposure (particularly of risk groups) to anthropogenic emissions. Given that exposure varies depending on PM concentrations, which in turn may show seasonal variations, to complement the present study, a new experimental campaign should be designed with samplings during the summer.

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Author contribution All authors contributed to the study conception and design. YC: investigation, methodology, conceptualisation, writing original draft and editing; CA: writing original draft and review, conceptualisation, methodology, supervision, funding acquisition; LF: investigation, methodology; MF: supervision, conceptualisation, funding acquisition. All authors read and approved the final manuscript.

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Data availability The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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