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Lung-deposited dose of particulate matter from residential exposure to smoke from wood burning

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Abstract

Residential settings are of utmost importance for human exposure, as it is where people spend most of their time. Residential wood combustion is a widespread practice known as a source indoor particulate matter (PM). Nevertheless, research on the risks of exposure associated with this source is scarce and a better understanding of respiratory deposition of smoke particles is needed. The dosimetry model ExDoM2 was applied to determine the deposited dose of inhalable particulate matter (PM_{10}) from residential biomass combustion in the human respiratory tract (HRT) of adults and children. The dose was estimated using PM_{10} exposure concentrations obtained from a field campaign carried out in two households during the operation of an open fireplace and a woodstove. Simultaneously, PM_{10} levels were monitored outside to investigate the outdoor dose in a rural area strongly impacted by biomass burning emissions. Indoors, the 8-h average PM_{10} concentrations ranged from 88.3 to 489 µg m^{-3} and from 69.4 to 122 µg m^{-3} for the operation of the fireplace and the woodstove, respectively, while outdoor average PM_{10} concentrations ranged from 17.3 to 94.2 µg m^{-3}. The highest amount of the deposited particles was recorded in the extrathoracic region (68-79%), whereas the deposition was much lower in the tracheobronchial tree (5-6%) and alveolar-interstitial region (16-21%). The total dose received while using the fireplace was more than twofold the one received in the room with a woodstove and more than 10 times higher than in the absence of the source. Overall, indoor doses were higher than the ones received by a subject exposed outdoors, especially at the alveolar–interstitial region. After 24 h of exposure, it was estimated that approximately 35 to 37% of the particles deposited in the HRT were transferred to the gastrointestinal tract, while approximately 2.0-2.5% were absorbed into the blood. The results from exposure and dose of indoor particles gathered in this work suggest that homeowners should be encouraged to upgrade the wood burning technology to reduce the PM levels inside

their residences. This study also provides biologically relevant results on the lung deposition of particles from residential biomass burning that can be used as a reference for future research.

Keywords: Dose, Human respiratory tract, Lung deposition, Indoor air pollution, PM_{10}, Residential wood combustion.

Abbreviations: AI: alveolar–interstitial, B: ventilation rate, BB: trachea, bb: bronchiolar, C: exposure concentration, COPD: chronic obstructive pulmonary disease, DF: deposition fraction, ET: extrathoracic, ExDoM2: exposure dose model, GI: gastrointestinal, GSD: geometric standard deviation, HRT: human respiratory tract, ICRP: International Commission on Radiological Protection, MMAD: mass mean aerodynamic diameter, PM_{10}: Particulate matter with equivalent aerodynamic diameters below 10 µm, t: exposure time.

1. Introduction

The term aerosol refers to solid and/or liquid particles in suspension in the air with different origins, composition and granulometric distribution (Calvo et al. 2013). Ambient particulate matter (PM) is regarded as the leading risk factor among all environmental and occupational risks. Long-term exposure to ambient PM pollution contributed to 4.14 million deaths in 2019 (Health Effects Institute 2020). Exposure to PM has been associated with an array of adverse health outcomes including both acute (e.g. pulmonary inflammation, exacerbation of chronic diseases, changes in blood pressure, heart rate variability) and chronic effects (e.g. lung cancer, pneumonia, cardiovascular mortality, myocardial infarction, hypertension, premature death, stroke) (Pope 2000; Anderson et al. 2012; Kim et al. 2015; Darquenne et al. 2020).

Residential biomass combustion is well-known as a major source of particulate matter below 10 and 2.5 µm (PM_{10} and $PM_{2.5}$) worldwide (Vicente and Alves 2018; Olsen et al. 2020). In addition to its contribution to ambient PM levels, this source also greatly affects household air quality (Guo et al. 2008; McNamara et al. 2013; Salthammer et al. 2014; de Gennaro et al. 2015; Saraga et al. 2015; Parajuli et al. 2016; Bartington et al. 2017; Castro et al. 2018). Studies conducted in the United States found evidence of respiratory symptoms in children living in wood burning households (reviewed by Naeher et al. 2007). Moreover, the use of a fireplace for 4 h was associated with increased risk of respiratory symptoms by about 16-20% of women living in tobacco-free homes (Naeher et al. 2007, and references therein). The inhalation and particle deposition in the human respiratory tract (HRT) are behind the PM-related health effects. However, the actual dose is seldom considered in epidemiological and toxicological studies, and frequently exposure is used as a measure for dose (Schlesinger et al. 2006; Paur et al. 2011; Schmid and Cassee 2017).

In previous studies, the total lung dose of biomass combustion-generated aerosols was measured directly *in vivo*, monitoring the inhaled and exhaled particle concentrations (Löndahl et al. 2008; Muala et al. 2015). Despite the valuable information provided by total dose estimations, knowledge of regional deposition in the HRT is crucial to assess the potential hazard of inhaled particles (Hinds 1999). The regional dose in the respiratory system is difficult to be determined experimentally, although some methods are available (Kim 2009; Löndahl et al. 2014). Therefore, the regional dose is typically estimated by means of mathematical models (ICRP 1994; Hussain et al. 2011; Hofmann 2011; Aleksandropoulou and Lazaridis 2013).

Few research studies have been conducted to characterise the exposure and lung burden arising from biomass combustion in indoor microenvironments. In Italian households, Stabile et al. (2018) carried out on-site measurements to evaluate the exposure and dose of particles received by the population living in dwellings where biomass-burning systems were used for heating. The researchers estimated the alveolar and tracheobronchial dose considering the measured exposure concentrations, the exposed individual's inhalation rate and assuming a constant value of 0.2 for the PM_{10} deposition fraction in the lungs. Recently, Nicolaou et al. (2020) characterised the exposure of household biomass-related pollution in the Peru Andean region and determined the lung-deposited dose and regional deposition fractions of inhaled PM through modelling.

Considering the importance of i) dosimetry to assess the health risks posed by exposure to PM (Schmid and Cassee 2017), ii) residential environments for human exposure (Tham 2016), and iii) the role of specific indoor sources (such as residential biomass combustion for heating) on indoor air quality (e.g. Salthammer et al. 2014; Stabile et al. 2018; Vicente et al. 2020), the goal of the present study was to estimate the total and regional doses in the HRT based on the indoor exposure to PM_{10} when using common biomass wood burning appliances in many European countries. In this work it was hypothesised that the combustion appliance selected for household heating might play a crucial role on the lung dose received by the subject exposed indoors. Additionally, through modelling, this study aimed to compare the doses obtained indoors with those associated with exposure to outdoor particles in a rural area highly impacted by residential biomass burning.

2. Methodology

2.1. Site description and PM_{10} measurements

A winter sampling campaign was carried out in January 2017 in a small village in central Portugal. The weather was typical for the season, with mean diurnal temperatures between 7 and 14 °C. Wood burning for residential heating is common in this area. There are no major industries nearby or major roads close to the village, where traffic is limited.

To assess the indoor exposure to PM from residential wood burning two detached houses of similar characteristics (age, construction materials, exposure to wind, etc.) equipped with aluminium window frames, double glazed casement windows, and outdoor blinds, were selected. One household was equipped with an open fireplace in the kitchen (about 38 m^3) and the other with a woodstove, also installed in the kitchen (about 67 m^3) (Figure 1). The monitoring programme was carried out under controlled conditions, meaning that during the weeks of experiments no other activities took place in the houses and only the person responsible for the measurements was allowed in the residences. The experiments were conducted under minimum ventilation conditions (doors and windows closed) with an average air exchange of 0.78 ± 0.12 and 0.72 ± 0.13 h^{-1} in the rooms equipped with fireplace and woodstove, respectively. Three (woodstove) to four (fireplace) experiments of 8-h each were performed in different days, mimicking the rural resident's behaviour. During the burning period, parallel outdoor sampling was carried out. To start the combustion experiments, pinecones were ignited and used to lit pine and eucalyptus split logs, two abundant tree species in the region. Throughout the burning period, the combustion appliance was refuelled several times: three and five times for the fireplace and woodstove, respectively. The duration of the experiments and number of batches to refuel the combustion chambers tried to mimic common European burning practices (Gustafson et al. 2008; Wöhler et al. 2016; Reichert et al. 2016). Additionally, background measurements, in the absence of indoor sources of PM, were carried out in each residence.

Figure 1. Wood combustion appliances of the present study: open fireplace (left) and woodstove (right) both located in the kitchen of the households.

PM_{10} mass concentrations were continuously measured by a light-scattering laser photometer (DustTrak DRX 8533, TSI,) with a 1-minute resolution, in the indoor and outdoor environments, simultaneously. Additionally, concurrent indoor and outdoor PM_{10} samples were collected on quartz filters using two high volume air samplers (CAV-A/mb, MCV). Indoors, the samplers were placed in the middle of the room and the height of the air uptake inlet was positioned at about 1.2 m above the floor, to simulate the human sitting breathing height. After gravimetric quantification of PM_{10} mass concentrations (XPE105 DeltaRange®, Mettler Toledo), the chemical composition was determined (organic and elemental carbon, water soluble ions, speciated organic compounds, metals). The detailed description of the analytical techniques and the PM_{10} chemical composition can be found in a previous work (Vicente et al. 2020). The concentrations recorded by the DustTrak monitor were corrected using the gravimetric measurements.

2.2. Particle dosimetry model

The particle deposition in the HRT was estimated by the dosimetry model ExDoM2, a revised version of ExDoM (Aleksandropoulou and Lazaridis 2013), which is based on the International Commission on Radiological Protection model (ICRP 1994, 2015). A full description of the model has been reported by Aleksandropoulou and Lazaridis (2013) and Chalvatzaki and Lazaridis (2015). The ExDoM2 model simulates the dynamics of inhaled particulate matter in human airways and estimates the dose, based upon empirical equations (ICRP 1994), in the five regions of the HRT: extrathoracic (ET1: anterior nose and ET2: posterior nasal passages), tracheobronchial (BB: trachea and bb: bronchiolar), and alveolar–interstitial (AI). To model particle deposition, the regions were treated as a series of filters during both inhalation and exhalation. The two sub-compartments of the extrathoracic compartment (ET), ET1 and ET2, receive approximately 65% and 35% of the ET deposits of inhaled aerosols, respectively (ICRP 2015).

The model takes into account the particle's inhalability, fraction of particles that effectively enter the human body, considering the aerodynamic diameter of the particles and the air velocity at the exposure site (Aleksandropoulou and Lazaridis 2013). The deposition pattern of particles in the HRT is closely related to the particle size and to the breathing pattern and the anatomical and physiological characteristics of the exposed subject. The dose (µg) is estimated as the product of exposure concentration (C, $\mu g\ m^{-3}$), ventilation rate of the exposed subject (B, $m^3\ h^{-1}$), the exposure time (t, h) and deposition fraction (DF) of particles in the respiratory system (equation 1).

$$\text{Dose} = C \times B \times t \times DF \qquad (1)$$

The ventilation rate depends on the activity level of the exposed subject, age and gender (ICRP 1994). Age- and gender-specific standardised values for different physical activity levels (sleeping, sitting awake, light exercise, and heavy exercise) are listed in the ICRP (1994) report.

The model allows to estimate the retention of particles in the HRT and the mass transferred to the gastrointestinal (GI) tract, lymph nodes and absorbed into blood during exposure and post-exposure times (taken as 24-h in the present work). Additionally, particles deposited in the ET region are also eliminated by extrinsic means (e.g. nose blowing). The mechanical clearance of particles is calculated by the ICRP compartment model (ICRP 2015). The model uses Caucasians reference values for particle residence times and clearance rates for mechanical transport. In the present work, the absorption of PM_{10} into blood was assumed to be moderate and to occur at the same rate in all regions (except in ET1 for which it was assumed that no absorption takes place) (ICRP 2015). Absorption is treated as a two stage process consisting of dissociation and absorption (ICRP 2015). A fully description of the clearance model can be found elsewhere (Chalvatzaki and Lazaridis 2015).

2.3. Exposure scenario

Input parameters of the model cover the exposed subject (age and gender), PM exposure concentrations (hourly average), breathing mode (nose or mouth breathing), activity level (sleep, sitting/resting and light activity, heavy activity), wind speed and particle size distributions (Table 1).

Table 1. Input data for ExDoM2 model

	Indoor				Outdoor	
	Fireplace	Background	Woodstove	Background	Fireplace	Woodstove
Particle properties						
PM_{10} Concentration (μg m^{-3}) [a]	88.3 - 489	14 - 17	69.4 - 122	21 - 24	49.4 - 94.2	17.3 - 72.2
PM_1/PM_{10}	0.97	0.97	0.84	0.97	0.89	
Density (g cm^{-3})	1.5	1.5	2.0	1.5	1.9	
MMAD (μm)	0.7	1.0	0.7	1.0	0.87	
GSD (μm)	1.8	2.4	1.8	2.4	4.02	
Exposure scenario						
Breathing scenario	Nose					
Exposure duration (h)	8					
PM_{10} concentration	Hourly average					
Activity level	Light exercise					

[a]8-h average (minimum-maximum concentrations); MMAD: Mass mean aerodynamic diameter; GSD: Geometric standard deviation

In the present study, PM_{10} deposition in the HRT was modelled for three different healthy subjects, male, female and 10 years male child exposed to biomass burning particles indoors 8 hours per day. The burning period defined in the present study is similar to the daily average time reported previously for residential heating in Europe (Gustafson et al. 2008; Stabile et al. 2018). It was assumed that the subjects were under light physical activity and breathing through the nose. For comparison purposes, the same assumption was made to assess exposure to outdoor PM_{10} and indoor PM_{10} in the absence of sources (background). Particles were considered spherical (shape factor of 1) (Martins et al. 2015; Sánchez-Soberón et al. 2015; Mammi-Galani et al. 2017). The particle density was calculated based on their chemical composition at each sampling site (indoor fireplace, indoor woodstove and outdoors) (Vicente et al. 2020) (Table 1). A mass mean aerodynamic diameter (MMAD) of 0.87 µm (Castro et al. 2018) and 0.66 µm (Bari et al. 2011a) was considered indoors and outdoors, respectively (Table 1). The density of the particles indoors in the absence of activity (background) was considered to be 1.5 g cm^{-3} and the MMAD of the particles equal to 1.0 µm (Castro et al. 2018).

3. Results

3.1. Exposure concentrations

The range and average PM_{10} concentrations indoors and outdoors, as well as the daily profiles, during the operation of the woodstove and fireplace, have been reported in detail in a previous manuscript (Vicente et al. 2020). Regarding the daily profiles (Figure 2A), the lighting and refuelling were found to be the main polluting phases. In general, during the wood burning periods, indoor concentrations were higher than those outdoors. Figure 2B displays the average exposure concentrations obtained with the DustTrak for an 8-h period for each measurement day, which include four monitoring periods with the fireplace in use, three periods with the woodstove in operation, and the respective outdoor data. Additionally, measurements of background levels in the rooms for an equivalent period (8-h) were also included. The results showed a 16- (fireplace) and 4-fold (woodstove) increase, on average, in exposure concentrations during the operation of wood burning appliances in comparison with levels in the absence of indoor activity (background measurements). During the operation of the fireplace, indoor PM_{10} levels (8-h average) were in the range from 88.3 to 489 µg m^{-3}. In the room equipped with woodstove, PM_{10} concentrations (8-h average) were lower but still high, in the range from 69.4 to 122 µg m^{-3}. The door in the woodstove allows sealing off the combustion chamber from the room, however, it is periodically open to refuel, which might lead to smoke leakage into the room. The impact of the refuelling operations on the indoor PM levels was also highlighted in a recent study conducted in twenty English households using low cost air quality monitors (Chakraborty et al. 2020).

In the present study, the outdoor PM_{10} concentrations during the indoor burning periods ranged from 49.4 ± 19.9 to 94.2 ± 76.5 µg m^{-3} and from 17.3 ± 6.44 to 72.3 ± 27.0 µg m^{-3} for the operation of the fireplace and the woodstove, respectively. In the winter of 2006, the daily average $PM_{2.5}$ concentrations in a residential area of Kurkimäki (Finland), where there are no major roads or other emission sources, ranged from 5 µg m^{-3} to over 40 µg m^{-3}. In this area, the researchers recorded short-time concentration peaks up to 1000 µg m^{-3} (minute averages), which were ascribed to local wood combustion (Hellén et al. 2008). In a Danish small rural town with widespread use of wood combustion for heating, Glasius et al. (2006) measured $PM_{2.5}$ concentrations about 4 µg m^{-3} higher than at a nearby background site. The average $PM_{2.5}$ concentration in the residential area during the intensive measuring period was 16.0 µg m^{-3}. In

Germany, at a residential site in Dettenhausen, Bari et al. (2011b) reported that at the beginning of winter months (November, December), the average PM_{10} concentrations varied from 10 to 40 μg m^{-3}, while the highest peak concentrations were observed from middle of January to the early February, which the researchers attributed to the limited dispersion of air pollutants caused by surface inversions.

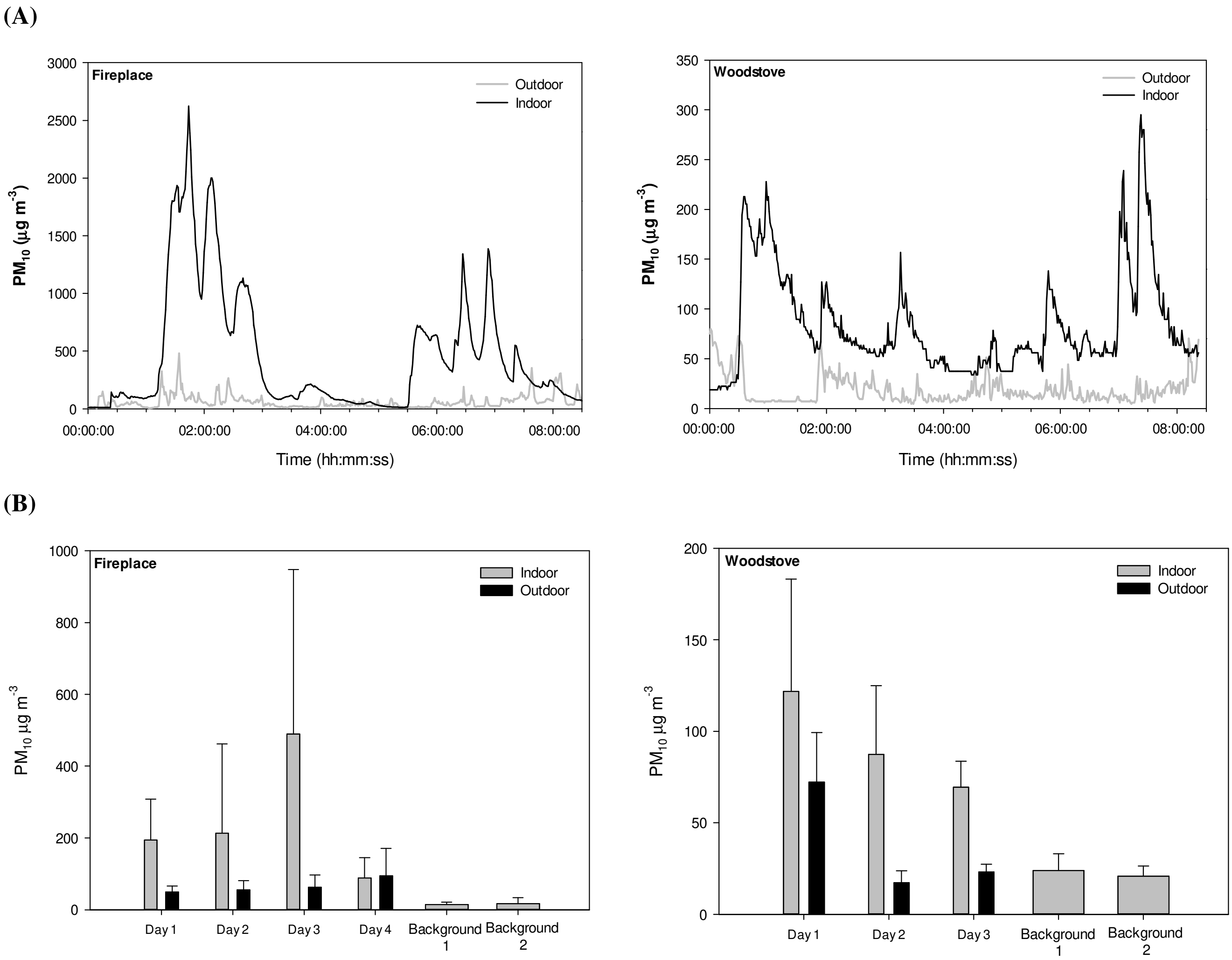

Figure 2. Example of PM_{10} profiles (μg m^{-3}) (A) and 8-h average concentrations (B) for wood burning experiments and background measurements.

3.2. Total and regional doses

The 8-h PM_{10} doses in the regions of the HRT for each subject are shown in Figure 3 for indoor exposure during the operation of the fireplace and woodstove and the corresponding outdoor values. Indoors, the highest and lowest total dose were obtained for males and females, respectively, whereas outdoors, the highest total dose for PM_{10} was obtained in the HRT of males and the lowest total dose was observed in 10-year old male children. Nevertheless, children inhale more air per unit of body weight than adults and are more susceptible to respiratory risks than adults due to their immature immune system. Respiratory disease is a leading cause of childhood mortality globally (Xi et al. 2015).

Indoors, the highest deposited dose was received by subjects exposed to particles produced during wood combustion in the open fireplace with an average 8-h cumulative dose of 954 ± 660 µg, 1119 ± 773 µg and 974 ± 673 µg for females, males and 10-year male children, respectively. The higher deposited dose obtained is directly linked (R^2 = 0.977) to the higher PM_{10} concentrations measured while the fireplace was operating compared to the woodstove (Table 1). The corresponding values for a subject in the room equipped with a woodstove were 391 ± 123 µg, 459 ± 144 µg and 398 ± 125 µg for females, males and 10-year male children, respectively. The total dose received by a subject in the room where the fireplace was in operation was more than twice the one received in the room with a woodstove and 11 to 12-fold higher than the total dose received by a subject in the room without indoor pollution sources. A lower increase (3-fold) in the total dose received by a subject exposed to particles from the woodstove operation in comparison with the one received in the absence of indoor sources was recorded. As displayed in Figure 4, high variability in the hourly dose was recorded during the 8-h measurement period, especially when the open fireplace was in use. As explored in Vicente et al. (2020), the daily profiles revealed high PM_{10} peak concentrations during the start-up phase, as well as during refuelling periods. In the periods when the stove was active, the dose was lower but still noticeable. Outdoors, the received dose for a male subject ranged from 77 to 413 µg and from 111 to 535 µg for 8-h exposure during the campaign with the fireplace and the woodstove, respectively. The variability found in outdoor doses might be ascribed to the distinct weather conditions in different monitoring days. A linear increase of the dose rate with the exposure concentration was also observed outdoors (R^2 = 0.893).

Regarding the regional deposition of inhaled particles, the results showed that the ET airways received the highest amount of the particulate mass deposited in the HRT (from 206 – 1520 µg and from 209 – 426 µg during the fireplace and woodstove operation, respectively) whilst the lowest was recorded in the TB region (from 14 – 120 µg and from 15 – 36 µg during the fireplace and woodstove operation, respectively) (Table S1). The nose has an important role as an air conditioner and a defender of the lower HRT since it is responsible for filtering, humidifying, and heating the inhaled air, as well as for trapping inhaled particles, protecting the gas-exchange regions of the lung (Hinds 1999; Harkema et al. 2013).

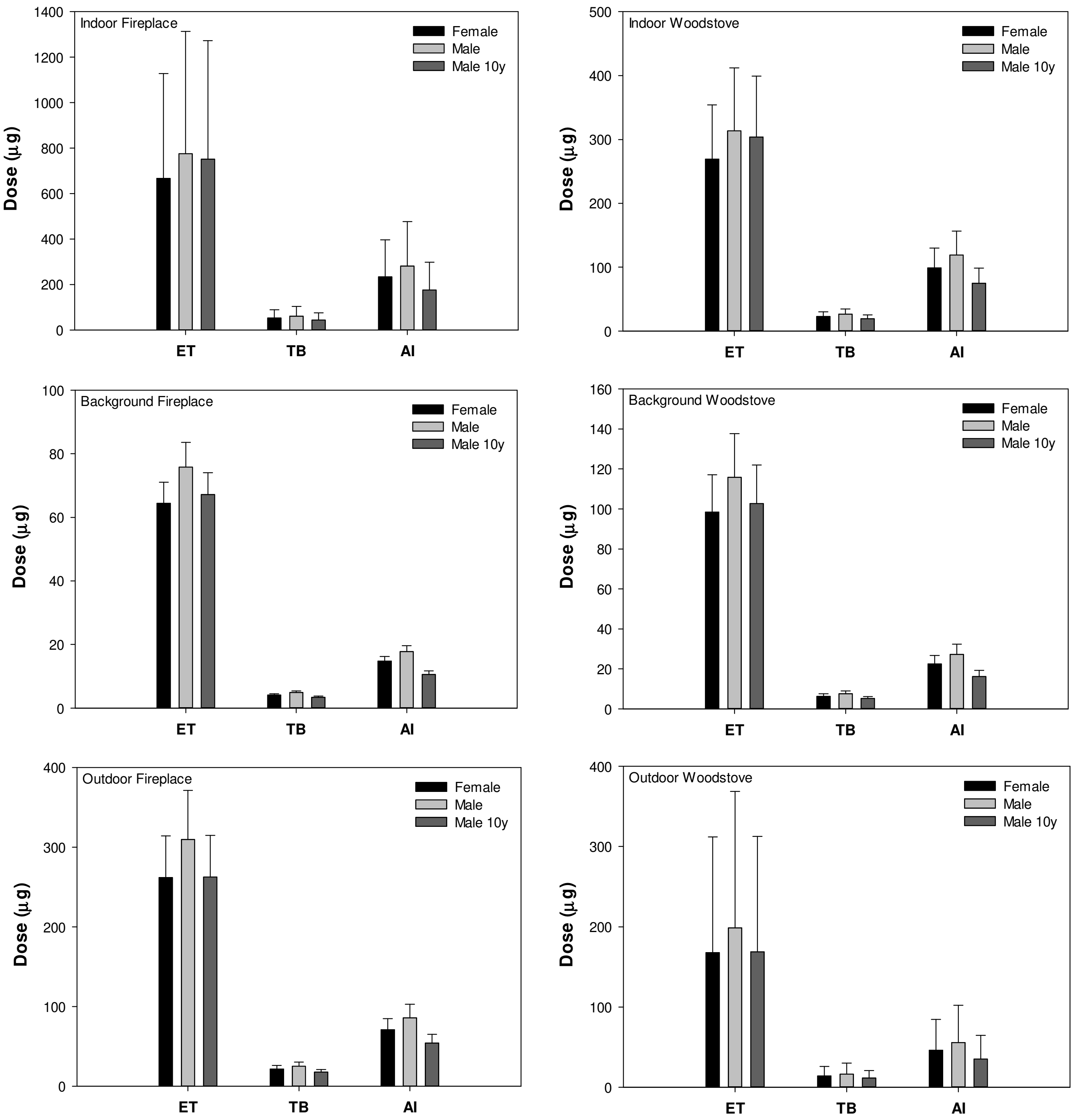

Figure 3. Particulate matter 8-h dose in the different regions of the HRT tract (ET – extrathoracic, TB – tracheobronchial and AI – alveolar–interstitial) for different subjects.

The AI region received from 55 to 522 µg and from 58 to 162 µg of PM_{10} during the fireplace and woodstove operation, respectively. Globally, indoor doses were higher than the ones received by a subject exposed outdoors, especially at the AI region (Figure 3).

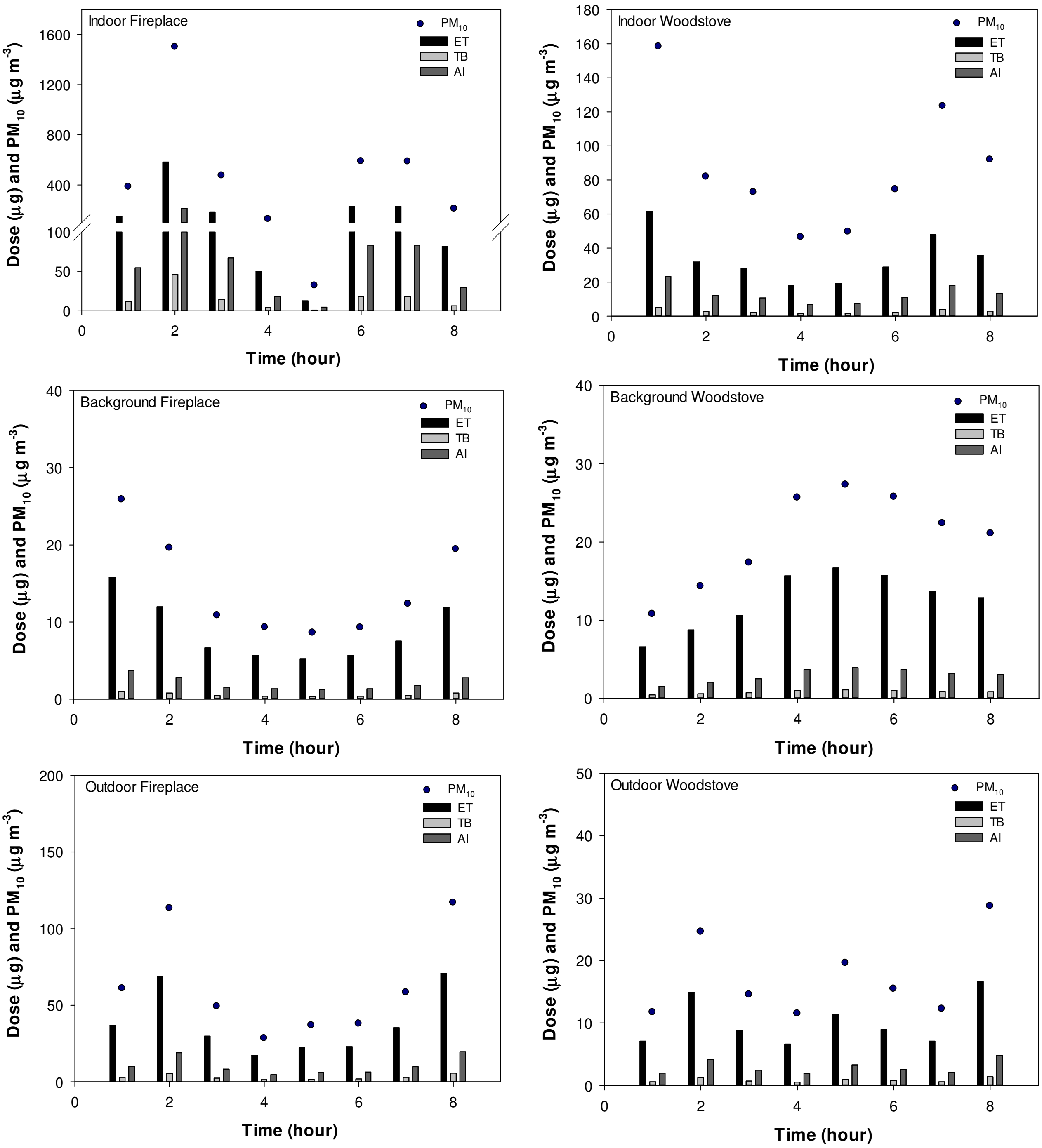

Figure 4. Example of hourly PM_{10} exposure concentration and dose in the different regions of the HRT (ET – extrathoracic, TB – tracheobronchial and AI – alveolar–interstitial) estimated for an adult male.

The doses at the AI region for a subject exposed to indoor particles from woodstove operation were, on average, 2.8 times higher than those received outdoors. Indoors, the operation of the fireplace led to doses at the AI region 3.5 times higher, on average, than outdoors. The dose received by a subject at the AI region in the absence of indoor sources of PM was 16-17 and 3-4 times lower than during the operation of the fireplace and woodstove, respectively.

The normalised delivered dose (dose per surface area or mass of lung/tissue) plays a crucial role in a toxicological dose-response analysis with significance for human risk assessment (Schmid and Cassee 2017). Considering the age and gender specific superficial area of the HRT regions (ET, TB, AI) reported by Sarangapani et al. (2003), it was observed that, although the mass received at the AI region was greater than the one recorded in the TB region, the deposited mass of particles per square centimetre of tissue surface area was higher at the latter region (Figure 5).

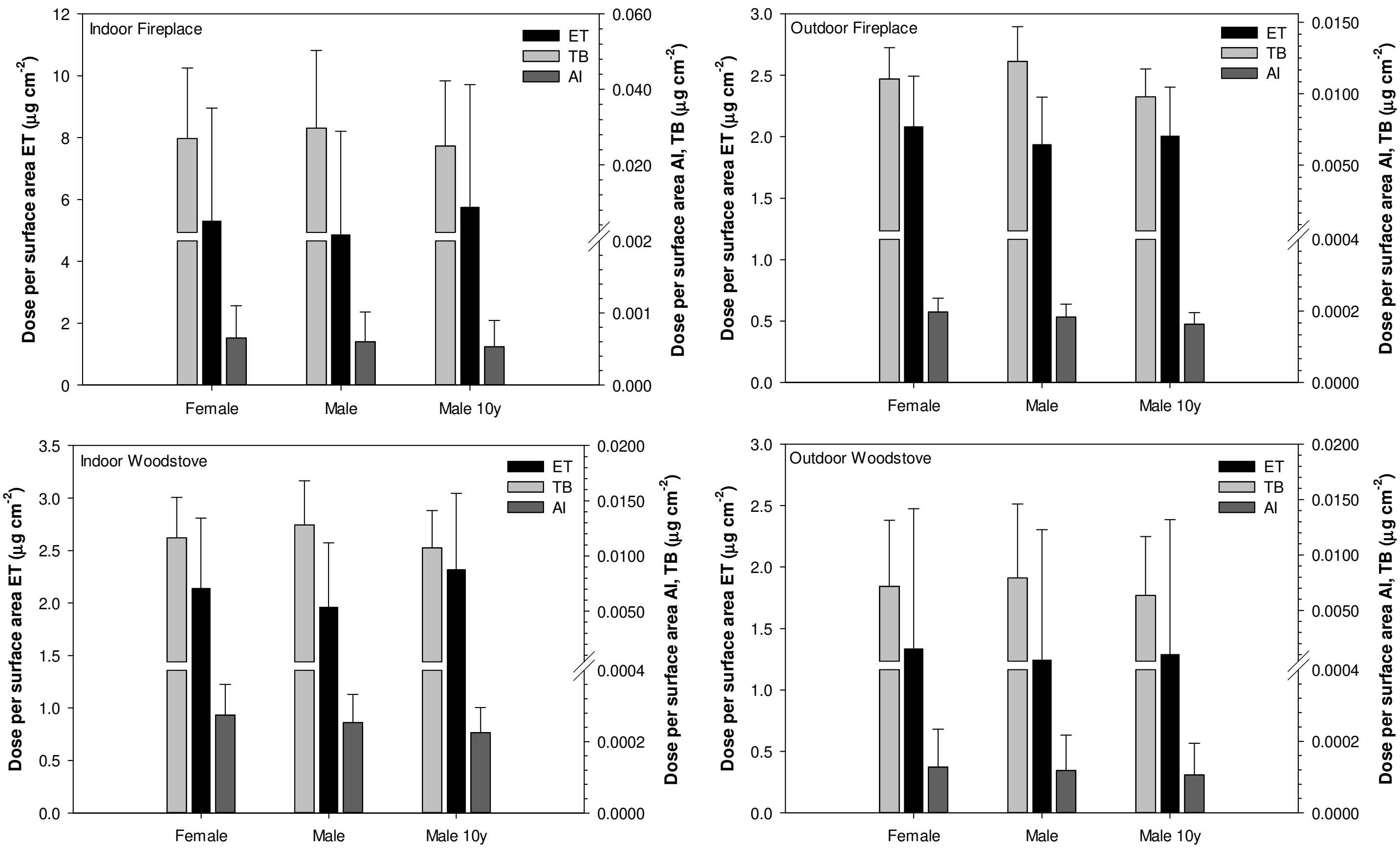

Figure 5. Dose per surface area of the target tissue considering 8-h exposure (ET – extrathoracic, TB – tracheobronchial and AI – alveolar–interstitial).

In fact, the alveoli account for more than 90% of the lung surface area. The alveolar region, where the air-blood barrier is thinner, represents the potentially most vulnerable site of deposition due to the easier access to the blood stream. Additionally, considering that clearance mechanisms are slower in the lower RT, the probability of adverse health effects due to particle–cell/tissue interactions is higher in this region of the HRT (Paur et al. 2011).

For *in vitro* toxicological studies, the target tissue/site dose reflect more accurately the amount of material coming in contact with the cells than measures of exposure (Paur et al. 2011; Schmid and Cassee 2017). Thus, this metric yields important information about the dosage to be tested in *in vitro* assays. Considering the exposure scenario evaluated in the present study, a realistic alveolar dose ranging from $(6.5 \pm 4.5) \times 10^{-4}$ µg cm^{-2} to $(5.3 \pm 3.7) \times 10^{-4}$ µg cm^{-2} and from $(2.7 \pm 0.862) \times 10^{-4}$ µg cm^{-2} to $(2.3 \pm 0.707) \times 10^{-4}$ µg cm^{-2} could be considered for indoor exposure to particles from fireplace and woodstove operation, respectively. Outdoors, lower doses at the AI region were observed ($1.1 \times 10^{-4} – 2.0 \times 10^{-4}$ µg cm^{-2}) (Figure 5).

3.3. PM retention and clearance

The PM_{10} retention in the HRT and the mass transferred to the gastro-intestinal tract (oesophagus), lymph nodes and blood (absorption into the blood) 24-h after exposure are displayed in Table 2. Indoors, 49 to 67% and 53 to 61% of the particles deposited in the HRT remained in the RT of a subject exposed to wood smoke from the fireplace and woodstove, respectively. Outdoors, 49 to 60% of the deposited particles were retained in the HRT.

After 24-h of exposure, the highest dose of particles was recorded in the oesophagus (Table 2), which derives from the higher deposited dose in the ET region. Particles deposited in the ET2 region, or transferred to this region from the anterior nasal passage and trachea, are cleared rapidly by mucociliary action to the throat and swallowed, transferring the particles to the GI tract (ICRP 2015). The particulate fraction that deposits in the tracheobronchial region, consisting of trachea, bronchi and terminal bronchioles, can be trapped in the mucus produced by the bronchial epithelial cells and cleared by mucociliary transport into the throat, and then swallowed to the GI tract. The ICRP (2015) assumes that a fraction of particles deposited in the bronchial tree clears slowly, with mucus velocities generally increasing towards the trachea. The association between the exposure to biomass burning smoke and the development of gastrointestinal cancers has been reported in previous studies (Kayamba et al. 2017; Sheikh et al. 2020). Of the particles deposited in the HRT, about 2% were absorbed into the blood (assuming moderate blood absorption) after 24-h of exposure. More than 90% of the particles deposited in the AI region remained deposited after 24-h of exposure.

Table 2. Retention of particles in the HRT, mass of PM_{10} (µg) transferred to the gastrointestinal tract, lymph nodes and absorbed into the blood after 24-h exposure (ET – extrathoracic, TB – tracheobronchial and AI – alveolar–interstitial).

	ET	TB	AI	Oesophagus	Lymph nodes	Blood Absorption
Fireplace						
Indoor						
Female	318 ± 205	35.1 ± 23.1	217 ± 148	326 ± 239	$(9.02 \pm 7.61) \times 10^{-5}$	23.1 ± 18.8
Male	370 ± 239	39.7 ± 26.0	260 ± 177	380 ± 279	$(10.5 \pm 8.84) \times 10^{-5}$	27.5 ± 22.3
Male 10y	358 ± 232	27.7 ± 18.1	163 ± 111	366 ± 269	$(9.70 \pm 8.18) \times 10^{-5}$	18.3 ± 14.8
Background						
Female	28.0 ± 0.771	2.28 ± 0.0560	13.4 ± 1.10	33.6 ± 6.45	$(1.04 \pm 0.328) \times 10^{-5}$	1.84 ± 0.524
Male	32.9 ± 0.908	2.66 ± 0.0813	16.2 ± 1.33	39.6 ± 7.62	$(1.23 \pm 0.386) \times 10^{-5}$	2.21 ± 0.630
Male 10y	29.2 ± 0.801	1.74 ± 0.0798	9.62 ± 0.794	34.8 ± 6.68	$(1.05 \pm 0.330) \times 10^{-5}$	1.41 ± 0.394
Outdoor						
Female	124 ± 21.7	14.5 ± 2.48	65.3 ± 12.7	128 ± 28.9	$(3.66 \pm 0.877) \times 10^{-5}$	7.71 ± 1.94
Male	147 ± 25.7	16.5 ± 2.80	79.0 ± 15.3	152 ± 34.3	$(4.32 \pm 1.03) \times 10^{-5}$	9.22 ± 2.33
Male 10y	124 ± 21.8	11.4 ± 1.92	50.0 ± 9.71	128 ± 28.8	$(3.51 \pm 0.877) \times 10^{-5}$	6.07 ± 1.57
Woodstove						
Indoor						
Female	122 ± 33.5	14.9 ± 4.22	90.5 ± 27.8	136 ± 47.1	$(4.23 \pm 1.71) \times 10^{-5}$	10.8 ± 4.25
Male	142 ± 39.0	16.7 ± 4.72	109 ± 33.3	159 ± 54.9	$(4.91 \pm 1.99) \times 10^{-5}$	12.9 ± 5.05
Male 10y	138 ± 37.8	11.7 ± 3.25	68.5 ± 21.0	153 ± 52.8	$(4.53 \pm 1.84) \times 10^{-5}$	8.58 ± 3.35
Background						
Female	47.3 ± 0.842	3.84 ± 0.126	21.8 ± 1.89	52.7 ± 8.38	$(1.54 \pm 0.434) \times 10^{-5}$	2.76 ± 0.650
Male	55.6 ± 0.990	4.49 ± 0.135	26.3 ± 2.28	62.1 ± 9.89	$(1.82 \pm 0.512) \times 10^{-5}$	3.32 ± 0.780
Male 10y	49.2 ± 0.879	2.95 ± 0.0678	15.7 ± 1.36	54.7 ± 8.68	$(1.56 \pm 0.438) \times 10^{-5}$	2.12 ±0.489
Outdoor						
Female	81.4 ± 69.6	9.62 ± 8.06	42.5 ± 35.5	80.6 ± 69.1	$(2.30 \pm 1.99) \times 10^{-5}$	4.81 ± 4.05
Male	96.2 ± 82.3	11.0 ± 9.21	51.4 ± 42.9	95.4 ± 81.9	$(2.71 \pm 2.34) \times 10^{-5}$	5.76 ± 4.85
Male 10y	81.7 ± 69.6	7.58 ± 6.35	32.5 ± 27.1	80.5 ± 68.9	$(2.22 \pm 1.92) \times 10^{-5}$	3.82 ± 3.22

4. Discussion

In the present study, PM_{10} exposure concentrations were obtained in two households during the operation an open fireplace and a woodstove. Parallel outdoor measurements were conducted to investigate the PM_{10} levels in an area strongly impacted by biomass burning emissions. Indoors, the 8-h average PM_{10} concentrations were 246 ± 171 (range from 88.3 to 489 µg m^{-3}) and 92.9 ± 26.6 µg m^{-3} (range from 69.4 to 122 µg m^{-3}) during the operation of the fireplace and the woodstove, respectively, exceeding the WHO guideline (50 µg m^{-3} 24-h mean). These concentrations fall within the range reported in previous studies carried out in Southern Europe during the operation of similar (open versus closed) wood combustion appliances (Canha et al. 2018; Castro et al. 2018; Stabile et al. 2018). Under real life conditions, Stabile et al. (2018) investigated the indoor exposure to particles emitted by biomass-burning heating systems in private Italian households. During the combustion periods, the researchers found particle concentrations in the range from 24-552 µg m^{-3}, 29-227 µg m^{-3} and 16-70 µg m^{-3} for open fireplaces, woodstoves and pellet stoves, respectively. As observed in the present study, the greatest rise in particle concentrations was recorded for wood combustion in the open fireplace while a smaller, but still clear increase was observed for the woodstove. The woodstove door allows to seal off the combustion chamber from the room, meaning that the release of pollutants into the indoor environment occurs mainly when the stove door is opened for refuelling. On the other hand, the open fireplace continuously releases pollutants into the air, increasing the levels of indoor particles more drastically. Moreover, the combustion conditions (e.g. lower combustion temperatures), achieved in open fireplaces also enhance the release of incomplete combustion products. Salthammer et al. (2014) investigated on-site the effects of wood-burning appliances on indoor air quality, in private German households. The study comprised seven households, six with closed combustion appliances and one with an open device. The 24-h average $PM_{2.5}$ concentrations were lower than the ones recorded in the present study (6 to 55 µg m^{-3}). The variations in the results of several studies can be attributed to differences in sampling duration and conditions, design of combustion appliances and fuels burned, operation of the combustion appliances, building characteristics, among other factors. Additionally, the chimney draft can also affect the pollutant concentrations indoors.

Outdoors, the 8-h average PM_{10} concentrations ranged from 17.3 ± 6.44 to 94.2 ± 76.5 µg m^{-3}. The widespread range of concentrations found outdoors is in agreement with the results of previous studies (e.g. Hellén et al. 2008; Bari et al. 2011b), reflecting the variability in weather conditions (e.g. wind velocity and direction and occurrence of rain), and possibly also the usage patterns of the combustion appliances by the village residents.

The total dose received by the exposed subjects was directly correlated with the exposure concentrations. During wood combustion in the open fireplace, the 8-h cumulative dose ranged from 295-1870 µg, 346-2192 µg and 301-1908 µg for females, males and 10-year male children, respectively. When the woodstove was in use, the corresponding total doses were in the range from 303 to 532 µg, 35 to 623 µg and 308 to 541 µg for females, males and 10-year male children, respectively. Similarly, Stabile et al. (2018) reported larger doses from exposure to particles from wood combustion in open fireplaces in comparison with woodstoves and automatically fed appliances (pellet stove). The researchers reported that the hourly extra-dose

(in relation to background), in terms of lung deposited PM_{10}, received by people exposed to particles released during the operation of open fireplaces was 5 μg h^{-1}. For closed combustion appliances, operated in batch mode, the derived dose was 4 μg h^{-1}, while for automatically fed appliances (pellet stove) the value was 1 μg h^{-1} (Stabile et al. 2018). In the present study, the hourly dose ranged from 37 to 274 μg h^{-1} and from 38 to 78 μg h^{-1} when using the fireplace and the woodstove, respectively. The lower doses found in the study of Stabile et al. (2018) may result from the calculation method employed. Firstly, the researchers subtracted the background concentrations to the levels measured during the operation of the biomass combustion appliances. Additionally, the authors estimated the dose assuming a constant value of 0.2 for the PM_{10} deposition fraction in the lungs. Nicolaou et al. (2020) characterised the exposure to $PM_{2.5}$ during biomass cooking (burning of wood, animal dung, and crop residue in open fires) in a rural area of Pruno. The estimated daily deposited doses of particles from biomass smoke based on personal exposures showed high variability (751 ± 1092 μg day^{-1}). The differences observed between genders and ages are related to the anatomy and physiology of the HRT, which determine the deposition of particles in its different regions. For example, when it comes to physiological parameters, an adult inhales more air than a child, whereas the breathing frequency is decreased. In the present study, the representative values for physiological parameters of Caucasian subjects under different activities provided by the ICRP were used. In addition, the main anatomical parameters, which are used for the calculations of particle deposition in the HRT, are also distinct for male, female and children (ICRP 1994).
Regarding the deposition of inhaled particles in each region of the HRT, the results revealed that the ET airways received 68–79% of the inhaled PM_{10}, whilst the lowest deposition was recorded in the TB region (5-6%) (Figure 2). The AI region received from 18 to 26% and from 16 to 21% of the total particulate mass deposited in the HRT indoors and outdoors, respectively. The fractional particle deposition in each region of the respiratory tract is determined by the particle parameters (size, shape and density). It is also affected by anatomical and physiological parameters such as, for example, the airways dimensions and flow rates (ICRP 1994). Lazaridis et al. (2001) applied the ICRP model to study the particle deposition at different parts of the HRT for different particle granulometries in man and woman. The authors reported that at the ET regions the deposition of particles with a diameter smaller than 0.2 μm was higher for males compared to females, which was attributed to higher volumetric flow rates. Similar deposition fractions for both genders was recorded for larger particles. In the BB region, while coarse particles presented similar deposition characteristics, particles smaller than 0.002 μm displayed higher deposition in the HRT of females, whereas particles with diameter in the range between 0.002 - 0.2 μm deposited with higher probability in the HRT of man. A similar behaviour was observed in the bb and AI regions. The distinct results were due to anatomical differences between women and men.

Concerning the target tissue dose, it was observed that realistic doses ranging from 1.1 to 6.5 × 10^{-4} μg PM_{10} cm^{-2} could be used to evaluate the toxicological potential on confluent alveolar epithelial cell cultures *in vitro*. It should be borne in mind that the doses obtained in the present study, for healthy subjects, may be higher in subjects with pre-existing respiratory diseases (Bennett et al. 1997; Kim and Kang 1997; Brown et al. 2002; Chalupa et al. 2004; Löndahl et al. 2012). The differences in doses have been ascribed to increased deposition efficiency, less even distribution of inhaled air, and decreased particle clearance rates in individuals with pre-existing lung diseases (Phalen et al. 2006). Studies performed to assess the dose received by individuals with COPD found an increased particle deposition rate compared to healthy subjects as a result of higher minute ventilation (Bennett et al. 1997; Kim and Kang 1997; Brown et al. 2002; Löndahl et al. 2012). For example, Bennett et al. (1997) reported deposition rates 2.5 higher in COPD patients compared to healthy subjects. Pre-existing lung disease along with other factors, such as the effects of exercise, oral breathing and unusual anatomy, can produce doses that exceed those of the average resting person by factors of about 33–67 (Phalen et al. 2006). Additionally, considering spatially non-uniform deposition regions and clearance, Paur et al. (2011) assumed a factor of 10 to account for high-dose regions, or hot spots. Taking into account these factors, particle doses ranging from about 0.07 to 0.44 μg PM_{10} cm^{-2} could be considered to expose alveolar epithelial cell cultures *in vitro* for the worst-case exposure scenario.

The evaluation of the retention and clearance of particles from the HRT revealed that although the higher deposited dose was recorded in the ET region, particles are cleared rapidly to the GI tract. On the other hand, after 24-h exposure, the percentage of cleared particles from the AI region was reduced (less than 10%). Thus, particles deposited deeper in the lung take longer to be cleared, increasing the probability of adverse health effects in this region of the HRT (Paur et al. 2011). Furthermore, the direct translocation of particles from the respiratory epithelium towards circulation can provoke adverse effects on different extra pulmonary sites (Schwarze et al. 2006; Nemmar et al. 2013; Du et al. 2016; Fiordelisi et al. 2017; Corsini et al. 2019).

The main limitation of the present work is the small sample size. Future research should be conducted to examine differences in lung deposition between different types of combustion appliances and designs and distinct biomass fuels burned, as well as the effect of the building envelope on the results. In the present study, a simplified exposure scenario was considered (nasal breathing under light physical exertion level) not accounting for the variability in breathing patterns of individuals (nasal, oral and mixed) and inhalation rates, which are closely related to subjects's activity, body position and health status. The modelling estimations obtained in the current study could be improved with more refinement of the assumptions and with the inclusion of country specific physiological parameters and time activity patterns.

Regarding the inter-subject variability in the particle doses, Löndahl et al. (2008) experimentally determined the deposition fraction of aerosol from efficient and low temperature biomass combustion in 10 healthy subjects (4 men and 6 women) aged 21–31. A difference of a factor greater than 2 was reported between the subjects with the highest deposition fraction and those with the lowest (Löndahl et al. 2008). Finally, the particle size increase of hygroscopic particles due to exposure to near-saturated surfaces, which can be of significant for biomass burning derived particles (Löndahl et al. 2008), was not accounted for in the present study. Future work should also evaluate the penetration of combustion related PM in other rooms of the house, including the estimate of the dose inhaled during sleep.

Despite the limitations mentioned above, to the authors knowledge, this is the first study in Europe encompassing a field campaign under controlled conditions (no concurrent sources in the households) and mimicking the households burning practices (duration of the burning experiments and number of batches to refuel the combustion chambers) to evaluate the deposited dose of inhalable particulate matter in the HRT of adults and children. Thus, this study provides an innovative approach and novel data regarding PM deposition in lungs from exposure to an important indoor PM source.

Conclusions

The dose received by different subjects (male, female and 10-year male) indoors, during the operation of wood heating systems (fireplace and woodstove) was evaluated, using the dosimetry model ExDoM2, by means of the concentration levels measured during an experimental campaign. Measurements were performed during the periods of use of the wood combustion appliances (8 hours) simultaneously inside and outside. Measurements in the absence of indoor PM sources were also conducted.

Higher deposited PM_{10} doses in the HRT were registered indoors during the operation of the open fireplace (up to twofold) in relation to those obtained for the woodstove. The doses received by a subject exposed indoors to particles emitted during the use of wood heating equipment were estimated to be 3- (woodstove) up to 10 times (fireplace) higher compared to those in the absence of activity. Indoor doses were in general higher than those received by a subject exposed outside the home. At the AI region, indoor doses were, on average, 2.8 and 3.5 times higher than the ones received outdoors during the operation of the woodstove and fireplace, respectively.

The results indicated that the highest mass of particles was deposited in the extrathoracic airways. However, the particles deposited in this region are removed much more rapidly to the gastrointestinal tract than those in the deeper regions of the respiratory system. On the contrary, it was observed that more than 90% of the particles deposited in the alveolar-interstitial region remained deposited after 24 h of exposure.

Given the main findings of the present study, the replacement of old-type wood combustion appliances should be encouraged in order to reduce the particle doses in the human respiratory tract. Additionally, considering that the deposition of inhaled particles in the HRT is one of the key factors for assessing their toxic effects, the results of this work provide novel data on PM regional deposition, which can be employed in future research on toxicological assessment of biomass burning particles.

Ethics approval and consent participate

Not applicable

Consent for publication

Not applicable

Availability of data and materials

The datasets used and/or generated during the current study are available from corresponding author on reasonable request

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

Conceptualisation: Estela D. Vicente, Célia A. Alves; **Formal analysis and investigation:** Estela D. Vicente; **Writing - original draft preparation:** Estela D. Vicente; **Writing - review and editing:** Célia A. Alves, Vânia Martins, Susana M. Almeida, Mihalis Lazaridis; **Funding acquisition:** Célia A. Alves; **Resources:** Mihalis Lazaridis; **Supervision:** Célia A. Alves, Mihalis Lazaridis; **Validation:** Mihalis Lazaridis; **Project administration:** Célia A. Alves.

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SUPLEMENTARY MATERIAL

Lung-deposited dose of particulate matter from residential exposure to smoke from wood burning

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Table S1. Total and regional (ET – extrathoracic, TB – tracheobronchial and AI – alveolar–interstitial) PM_{10} 8-h dose (µg) in the HRT tract for different subjects.

	ET	TB	AI	Total
Fireplace				
Indoor				
Female	667 ± 461 (206 - 1307)	53 ± 37 (16 - 104)	235 ± 162 (73 - 460)	954 ± 660 (295 - 1870)
Male	776 ± 536 (240 - 1520)	61 ± 42 (19 - 120)	282 ± 195 (87 - 552)	1119 ± 773 (346 - 2192)
Male 10y	752 ± 520 (233 - 1474)	45 ± 31 (14 - 88)	177 ± 122 (55 - 346)	974 ± 673 (301 - 1908)
Background				
Female	64 ± 6.6 (6.0 - 6.9)	4.1 ± 0.42 (3.8 - 4.4.)	15 ± 1.5 (14 - 16)	83 ± 8.6 (77 - 89)
Male	76 ± 7.8 (70 - 81)	4.9 ± 0.51 (4.6 - 5.3)	18 ± 1.8 (17 - 19)	99 ± 10 (91 - 106)
Male 10y	67 ± 6.9 (62 - 72)	3.4 ± 0.35 (3.2 - 3.7)	11 ± 1.1 (10 - 11)	81 ± 8.3 (75 - 87)
Outdoor				
Female	262 ± 52 (226 - 337)	22 ± 4.3 (19 - 28)	71 ± 14 (61 - 91)	355 ± 70 (306 - 456)
Male	310 ± 62 (267 - 399)	25 ± 5.0 (22 - 33)	86 ± 17 (74 - 110)	421 ± 84 (364 - 541)
Male 10y	263 ± 52 (227 - 338)	18 ± 3.5 (15 - 23)	54 ± 11 (47 - 70)	355 ± 66 (289 - 430)
Woodstove				
Indoor				
Female	269 ± 85 (209 - 366)	23 ± 7.2 (18 - 31)	99 ± 31 (77 - 135)	391 ± 123 (303 - 532)
Male	313 ± 98 (243 - 426)	26 ± 8.3 (20 - 36)	119 ± 37 (92 - 162)	459 ± 144 (355 - 623)
Male 10y	304 ± 95 (235 - 413)	19 ± 6.1 (15 - 26)	75 ± 24 (58 - 102)	398 ± 125 (308 - 541)
Background				
Female	99 ± 19 (85 - 112)	6.3 ± 1.2 (5.5 - 7.2)	23 ± 4.2 (20 - 26)	127 ± 24 (110 - 144)
Male	116 ± 22 (100 - 131)	7.5 ± 1.4 (6.5 - 8.6)	27 ± 5.1 (24 - 31)	151 ± 28 (131 - 171)
Male 10y	103 ± 19 (89 - 116)	5.2 ± 1.0 (4.5 - 5.9)	16 ± 3.1 (14 - 18)	124 ± 23 (108 - 141)
Outdoor				
Female	168 ± 144 (69 - 333)	14 ± 12 (6 - 28)	46 ± 38 (19 - 90)	194 ± 57 (94 - 451)
Male	199 ± 170 (81 - 394)	16 ± 14 (7 - 32)	56 ± 46 (23 - 109)	230 ± 58 (111 - 535)
Male 10y	169 ± 144 (69 - 334)	11 ± 10 (5 - 22)	35 ± 29 (15 - 69)	183 ± 63 (89 - 425)