

## Seasonal variation in exposure to particulate matter among children attending different levels of education: Comparison of two dosimetry models

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dosimetry

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- 31

#### 32 Abstract

Exposure to particulate matter (PM) has been associated with several adverse health 33 outcomes. Studies indicate that children may be exposed to much higher concentrations 34 of PM at school than in other environments. There exists very little data on the deposited 35 36 dose of PM while children attend classes. This study was carried out in a school located near an industrial complex in Portugal and attended by children aged 3-12 years. Indoor 37 PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> were measured over two seasons in classrooms representing 38 different school year groups. Particle deposition fractions in the respiratory tract, as well 39 40 as the deposited doses, were calculated using the Multiple-Path Particle Dosimetry (MPPD) and the Exposure Dose Model (ExDoM2). Both models were implemented 41 assuming an 8-hour exposure scenario to represent the school day. In general, differences 42 in PM concentrations were observed depending on room occupancy periods and season. 43 The highest mean PM<sub>2.5</sub> concentration was recorded in winter when the classroom was 44 vacant  $(23.7 \pm 20.5 \ \mu g \ m^{-3})$ , while the highest mean PM<sub>10</sub> level was observed in spring 45 during school hours ( $61.7 \pm 24.2 \ \mu g \ m^{-3}$ ). Regardless of the dosimetry model, the highest 46 deposition of PM<sub>10</sub> and PM<sub>2.5</sub> was in the upper region, while the lowest was in the 47 tracheobronchial (TB) region. The results indicate that deposited dose and deposition 48 49 fraction in spring may be more harmful to pupils' health than in winter. PM<sub>10</sub> presented the highest doses, ranging from 54.2 to 128 μg and from 83.9 to 185 μg, according to
MPPD and ExDoM2 estimates, respectively.

#### 52 Keywords

53 Schoolchildren; Particulate matter; Dose; PM deposition; Dosimetry models.

#### 54 1. Introduction

According to a recent report on air pollution and child health, nearly 600,000 children 55 aged 5-15 years died from exposure to unhealthy levels of ambient and household air 56 57 pollution in 2016 (WHO, 2018). Only in Europe, air pollution causes more than 1,200 58 deaths in people under the age of 18 every year (EEA, 2023). PM is the principal component of indoor and outdoor air pollution and includes a range of particle sizes (Lee 59 60 et al., 2021). Several studies have suggested that the primary exposure mechanism of PM is inhalation, which exacerbates respiratory symptoms in patients with chronic airway 61 62 diseases (Leikauf et al., 2020). PM is mostly absorbed through the respiratory tract, where it can infiltrate the lung alveoli and reach the bloodstream. In the respiratory system, PM 63 induces the activation of alveolar macrophages and neutrophils that release reactive 64 oxygen or nitrogen species. Oxidative stress stimulates the production of mediators of 65 pulmonary inflammation and begin or foster numerous illnesses (Thangavel et al., 2022). 66 Short-term exposure to particulate matter with aerodynamic diameter lower than 2.5 µm 67 (PM<sub>2.5</sub>) and its respiratory tract depositions are dose-responsive and related to higher 68 blood pressure, prevalence of prehypertension and hypertension among children (Liu et 69 70 al., 2021). There is also evidence that prenatal exposure to  $PM_{2.5}$  and its components 71 increases the risk of preterm birth (Shi et al., 2024). In addition, poor air quality in school buildings also contributes to health problems and can affect children's concentration and 72 73 cognitive development (Sunyer et al., 2015). According to the study by Martins et al. (2020), concentrations of PM were often higher inside than outside 74 the microenvironments evaluated in Lisbon (Portugal), with children being exposed to much 75 76 higher concentrations of PM at school than at home. Faria et al. (2020) showed that although children spend more time at home than at school during weekdays, the 77 78 classroom was the microenvironment that most contributed to the daily inhaled dose of 79 PM<sub>2.5</sub> and PM<sub>10</sub> in Lisbon. Madureira et al. (2015) found that indoor air pollutants in primary schools located in Porto (Portugal) were related to greater odds of wheezing in 80 children. Branco et al. (2020) also reported that high exposure to PM<sub>2.5</sub> and O<sub>3</sub> in nurseries 81 and primary schools in urban and rural areas in northern Portugal was associated with a 82 83 reduction in lung function among children.

84

In order to evaluate pupils' exposure to air pollutants in the school environment, many studies have been carried out on the assessment of indoor and outdoor air in primary schools or nurseries in both urban and rural areas, focusing mainly on investigating the

effects related to ventilation in classrooms, evaluating comfort parameters, and carrying 88 out the chemical characterisation of indoor and outdoor atmospheric pollutants (e.g., 89 Abhijith et al., 2022; Almeida Sousa et al., 2021; Mainka et al., 2015; Mumovic et al., 90 2009). Especially in Portugal in the last four years, some studies have investigated 91 potential sources of particulate matter in classrooms (Madureira et al., 2016), the 92 93 relationship between allergy symptoms and indoor air quality in schools (Branco et al., 94 2020; Szabados et al., 2022), and inhaled doses of particulate matter (Faria et al., 2020), but only a few aimed to calculate PM deposition among children (Chalvatzaki et al., 95 2020a; Madureira et al., 2020). Although the regional deposition and dose of particles 96 deposited in the respiratory tract are an important factor in understanding the health 97 98 effects of aerosol particles (Goel et al., 2018; Linell et al., 2023), it is still unknown how 99 size segregated PM is deposited in regions of the respiratory system and the dose deposited by children while attending classes, even when previous studies reported 100 adverse effects related to particle deposition in the airways. For example, it was 101 documented that the deposition of PM2.5 increases the risk of severity of pulmonary 102 tuberculosis in the upper and middle lobes (Makrufardi et al., 2023). In the study carried 103 104 out by Kesavachandran et al. (2015), the deposition of fine particles in the airways resulted in a decline in forced expiratory volume and peak expiratory flow among outdoor 105 106 exercisers. Therefore, the development of dosimetry models is an important step in 107 understanding exposure-dose-response relationships for PM and can help in evaluating the human health effects of inhaling toxic substances in different environments 108 (Chalvatzaki and Lazaridis, 2015). Different dosimetry modelling approaches have been 109 broadly used to predict particle deposition and dose in human airways, such as the MPPD 110 111 and the ExDoM2, for which estimates can be made for monodisperse and polydisperse aerosols, in a user-interactive environment. For example, Chalvatzaki et al. (2021) carried 112 out simulations with ExDoM2 considering three study cases with seasonal and diurnal 113 variations in Greek cities. Overall, a higher daily deposited dose was obtained in the cold 114 period compared to the warm periods for all sites. This finding was associated with 115 increased deposition rate in the cold period during the afternoon/evening, because of 116 significant heating emissions. Recently, Khan et al. (2022)carried out MPPD simulations 117 for a city in India and found that the mass deposited in winter was significantly greater 118 119 than in monsoons for  $PM_1$ ,  $PM_{2.5}$  and  $PM_{10}$ , mainly due to the higher PM levels in winter. 120

As deposition fractions and doses may vary from season to season and there are very few studies on the deposited dose of PM while children attend classes, the current work aimed to assess winter and spring indoor levels of PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> during school hours to investigate: i) temporal variability of mass concentrations and, ii) hourly and seasonal variations in the deposition of total and regional fractions and doses of PM in the children's respiratory system using two dosimetry models.

- 128129 2. Material and methods
- 130
- 130 131 2.1. Study area

Estarreja is a small town of about 7,000 inhabitants located in northern Portugal. The 132 town has an important chemical complex, comprising 30 companies in an area of 290 ha, 133 which is home to various economic sectors, such as heavy industry, retail, warehousing, 134 and services (Alves et al., 2023). This study was carried out at a school in the north side 135 of the town, close to the industrial complex. In addition to kindergarten, this school 136 integrates the first two cycles of basic education. In Portugal, basic education comprises 137 3 stages: 1<sup>st</sup> cycle (1<sup>st</sup> to 4<sup>th</sup> grades, ages 6-9 years), 2<sup>nd</sup> cycle (5<sup>th</sup> and 6<sup>th</sup> grades, 10-11 138 years) and  $3^{rd}$  cycle ( $7^{th}$  to  $9^{th}$  grades, 12-14 years). 139

140 The school of this study is surrounded by low-rise buildings, a road with little traffic, and a railway line to the west. It consists of 3 main buildings, each one dedicated to a different 141 level of education: 1) preschool, 3-5 years, 2)  $1^{st}$  cycle, 6-10 years, and 3)  $2^{nd}$  cycle, 142 143 10 – 12 years. Buildings 1, 2 and 3 are composed of 6, 12 and 7 classrooms, respectively. 144 To cover the different age groups and the various types of school activities, a classroom was selected in each of the buildings. By imposition of the school principal, classrooms 145 with children with special educational needs or disabilities were considered ineligible. 146 147 The general characteristics of the classrooms were: concrete walls, wooden windows, and 148 floor, white painted and whiteboard with markers. All of them depend only on natural 149 ventilation with windows and exterior doors.

The school is open from 7:30 to 18.30. However, the school day starts at 8:30 and finishes at 15:30. Every child is entitled to a 60-minute school lunch. In general, cleaning takes place between 16:00 and 18:00. In this study, class time corresponds to the school day considering the lunch break.

154 2.2. Experimental set-up and instrumentation

155 Measurements took place over two seasons. The winter campaign was performed between November and December 2022, and the spring campaign between April and May 2023. 156 157 However, for this study, fifteen days of monitoring during each season were considered after checking the amount of complete data for each working day and in every classroom. 158 The monitoring equipment was installed at a height of around 1 m above the floor, and at 159 160 least 1 m away from any doors, windows, and walls. The indoor instruments were removed from the room and transferred to another in the next building at the end of every 161 Friday afternoon (Table 1). PM<sub>10</sub> simultaneous concentrations were rectified against in-162 163 situ gravimetric PM<sub>10</sub> measurements.

164 **Table 1** 

Location	Description	Measurement periods
Indoor, ground	Kindergarten; room attended by the same	Nov. 14 – 18, 2022
floor	class (20 pupils); age group between 3 and 5	April 13 – 21, 2023
	years.	
Indoor, ground	1 <sup>st</sup> cycle, 1 <sup>st</sup> grade; room attended by the	Nov. 18 – 25, 2022
floor	same class (17 kids); age group between 6	April 21 – 28, 2023
	and 7 years.	
Indoor, 1 <sup>st</sup> floor	2 <sup>nd</sup> cycle, 5 <sup>th</sup> and 6 <sup>th</sup> grades; room attended by	Nov. 25 – Dec. 02, 2022
	different classes (on average 22 children per	April 28 – May 05, 2023
	class); age group between 10 and 11 years.	
Indoor, 1 <sup>st</sup> floor	2 <sup>nd</sup> cycle, 5 <sup>th</sup> and 6 <sup>th</sup> grades; room attended by different classes (on average 22 children per class); age group between 10 and 11 years.	Nov. 25 – Dec. 02, 2022 April 28 – May 05, 2023

165 Sampling campaign characteristics.

Simultaneous measurements of  $PM_{10}$ ,  $PM_{2.5}$ , and  $PM_1$  were obtained using an aerosol spectrometer (Grimm Model EDM 164) operated at a 1-minute time resolution. In this spectrometer, each particle is detected in the optical measuring cell and based on the intensity of the scattered light signal it is assigned a size. This model measures particle size distributions within the range from 0.25 to 32.0 µm. In addition, gravimetric  $PM_{10}$ samples were collected on 15 cm quartz microfibre filters using a high-volume sampler at an airflow rate of 500 l/min (HVS, Model CAV-A/MSb, MCV S.A).

174 The high-volume sampler was intercompared with real-time measurements. The 175 regression slopes for the results obtained from parallel measurements during the two 176 monitoring campaigns were 0.95 and 0.75, with correlation coefficients ( $\mathbb{R}^2$ ) of 0.91 and 0.95 for winter and spring, respectively. However, as there was no gravimetric equipment 177 for fine particles, PM1 and PM2.5 data were corrected from the ratios obtained between 178 the initial concentrations of  $PM_1/PM_{10}$  and  $PM_{2.5}/PM_{10}$  and multiplied by the 179 180 concentrations of corrected PM<sub>10</sub>, in accordance with the method applied by Cipoli et al. (2022).181

182 2.3. Estimation of particle deposition and deposition dose in the respiratory system of183 pupils using two models

184

• The MPPD model (V3.04, ARA Inc.)

The MPPD model calculates breathing from transport, deposition, and clearance in the respiratory tract of rats and humans based upon a multiple-path method (Asgharian and Anjilvel, 1998; Asgharian et al., 2001). In general, this model can predict both deposition in a typical path per airway generation (single-path) and particle deposition in all ways of the lung (multiple-path). The model provides specific human lung geometry for 10 different ages. It has three major applications: risk assessment, drug delivery and threat

assessment. In this study, MPPD was used for risk assessment. Lung geometries for ages 192 3, 8 and 9 were selected because of the age of children attending the school. The model 193 input parameters are presented in four categories: inhalant properties (aerosol), airway 194 morphometry, exposure condition and deposition/clearance settings. For aerosol 195 properties, the density and distribution were assumed to be 1 g  $cm^{-3}$  and single, 196 respectively, while equations 1 and 2 (Hinds and Zhu, 2022) were employed to calculate 197 198 the Mass Median Aerodynamic Diameter (MMAD) and the Geometric Standard Deviation (GSD) using the mass size distribution of PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> obtained in 199 200 both seasons to derive the hourly values for all classrooms (Table S1, Supplementary 201 Material):

202 
$$MMAD = e^{\frac{\sum c_i(\ln(d_i))}{\sum c_i}}$$
 (1)

203 
$$GSD = e^{\sqrt{\frac{\sum c_i (\ln(d_i) - \ln(MMAD))^2}{\sum c_i}}}$$
(2)

where  $c_i$  is the mass fraction in GRIMM channel *i* and  $d_i$  is the cut-off diameter of GRIMM channel *i*.

206 For airway morphometry, the age-specific 5-lobe was chosen and the default values for 207 Upper Respiratory Tract (URT) and Functional Residual Capacity (FRC) were obtained from the model. Exposure was considered constant and the parameters for this category 208 included the inspiratory fraction (value of 0.5), the Breathing Frequency (BF) and Tidal 209 210 Volume (TV) that were also provided by the model, while concentrations were calculated 211 for each of the three selected classrooms and each season considering weekdays only. In addition, since the aim of the study was to evaluate particle deposition while children 212 213 were in their classrooms, only the sitting activity pattern was contemplated, and the associated breathing pattern was nasal (Patterson et al., 2014). The selection of the nasal 214 route assumed that the study population (pupils) was sitting most of the time (low-215 intensity activity) and breathing spontaneously through the nose. The input parameters 216 for each age are shown in Table 2, and seasonal averages of PM concentration, MMADs 217 218 and GSDs are presented in Table S1, Supplementary Material.

MPPD calculates the deposition fraction based on mathematical equations described in detail by Asgharian et al. (2001). Based on the estimated concentration of DF and PM, the dose rate is calculated for each region of the respiratory tract and a report with the results is provided to the user. In this study, the period between 8 and 16 h represents the school day, therefore, dose rates were calculated for 1-hour exposure intervals and the sum of hourly doses is equivalent to the school day.

225

• The ExDoM2 model

ExDoM2 was also used for the calculation of deposition fraction and dose profile for the 227 school day. The model calculates the deposition dose, clearance, and retention of aerosol 228 particles in the human respiratory tract based on the International Commission on 229 Radiological Protection (ICRP) human respiratory tract model. ExDoM2 allows the user 230 to set variable or static exposure conditions, such as exposure concentration, physical 231 232 exertion levels, and different environments (Chalvatzaki and Lazaridis, 2015). 233 Furthermore, the model works with monodisperse or polydisperse aerosol size distributions where the user has the flexibility to introduce the aerosol size parameters as 234 235 median aerodynamic diameter and geometric standard deviation or to quantify these 236 values directly from the model using as input the measurement data. For a detailed 237 description of the model see Chalvatzaki and Lazaridis (2015) and Lazaridis (2023). Table 2 displays the values of the physiological variables used in the deposition 238 239 calculations in the models.

Input parameters according to the study group and model, applicable at normal conditions ofsitting breathing.

Age	URT (ml)		FRC (ml)		BF (min	-1)	TV (ml)	
(years)	MPPD	ExDoM2	MPPD	ExDoM2	MPPD	ExDoM2	MPPD	ExDoM2
3	9.47	nd	48.2	nd	24	nd	121.3	nd
5	nd	13.3	nd	767	nd	25	nd	213
8	21.0	nd	501.3	nd	17	nd	278.2	nd
9	22.4	nd	683.0	nd	17	nd	295.8	nd
10	nd	25	nd	1484	nd	19	nd	333

<sup>243</sup> <sup>nd</sup> indicates missing values when that age is not considered in the model.

244

In general, the input parameters required in each model are similar. However, while 245 ExDoM2 considers wind speed and automatically selects physiological parameters based 246 247 on the reference values provided in the ICRP66 model, MPPD offers the user the option 248 of directly insert these values or select the model's default values that change according 249 to the morphology of the lung and the person's age (Table 3). Both models consider age 250 as an input parameter, but the selection options differ from one model to another. In 251 ExDoM2 the user can choose between 5 options that consider gender (1 year, 5 years, 10 252 years, 15 years, or adults), while with MPPD, the user has the option to choose one of the following ages: 3 months, 21 months, 23 months, 28 months, 3 years, 8 years, 9 years, 14 253 years, 18 years, or 21 years. Additionally, different from MPPD, in ExDoM2 the user 254 must specify the gender of the individual. The respective ages used in this study are 255 presented according to the model in Table S2, Supplementary material. 256

257 **Table 3** 

258 Model specifications according to input options.

Â			
Parameter	Entered by user	Select from a list of	Automatically set
		options	by the model

PM concentration	Both models		
Density	Both models		
MMAD	Both models		
GSD	Both models		
Wind speed	ExDoM2 only		
Exposure scenario		Both models	
Activity level		Both models	
URT	MPPD only		ExDoM2 only
FRC	MPPD only		ExDoM2 only
BF	MPPD only		ExDoM2 only
TV	MPPD only		ExDoM2 only
Total lung capacity			Both models
Age		Both models	
Gender		ExDoM2 only	

The same input data for the implementation of MPPD for each group was used as input in the ExDoM2: (1) mean hourly PM concentration, (2) breathing pattern, (3) particle density, (4) hourly MMADs and GSDs, and (5) shape factor. The hourly wind speed was only considered in ExDoM2, and it was calculated for each classroom from data obtained during the winter monitoring campaign (Table S3, Supplementary material). In addition, in ExDoM2 model, the simulations were also implemented for each gender, female and male.

In addition to using a very different approach, another difference between the models is related to the presentation of the results. While MPPD provides an output report, the ExDoM2 model estimates the dose by formula (3), and generates an output file that includes hourly doses, making it necessary to isolate the DF in equation 3:

271 
$$Dose(\mu g h^{-1}) = PM_i * DF_{i,j} * VT$$
 (3)

where  $PM_i$  is the mean exposure concentration of PM in the size fraction i (µg m<sup>-3</sup>),  $DF_{i,j}$ is the deposition fraction in the region j of the respiratory tract for PM in the size fraction i, and VT is the ventilation per hour (m<sup>3</sup> h<sup>-1</sup>) of the exposed pupil, depending on age. To represent the school day, dose rates were calculated for a total of 8 h exposure duration in 1 h exposure time intervals.

277

#### 278 2.4. Data analysis

For the treatment and analysis of the PM concentrations, a minimum data availability of 75% per school day was considered. After checking the normality of the data, PM concentrations were examined with typical descriptive statistics including measures of central tendency and dispersion, diurnal cycles, highlighting the school period and significant differences between seasons and between the time of occupancy versus nonoccupancy with the non-parametric Wilcoxon test, where a p-value <0.05 was considered</li>
 statistically significant.

Subsequently, the data from the dosimetry models were analysed separately and then the
results of the two models were combined, considering the size of the PM and the groups
evaluated to extract the general behaviour of the deposited fractions and doses.
Association, agreement, and comparison analyses were performed using linear
regression, Bland-Altman method, and t-test for paired samples.

291

#### 292 **3.** Results and discussion

293

#### 294 3.1. General overview of PM concentrations

Median concentrations of  $PM_{10}$  varied from 6.7 to 24.4 µg m<sup>-3</sup> in winter, and between 13 and 18 µg m<sup>-3</sup> in spring. As shown in **Table 4**, the median concentrations of  $PM_{10}$ ,  $PM_{2.5}$ and  $PM_1$  in the room attended by 2<sup>nd</sup> cycle students were 1.3, 1.2 and 1.4 times higher in winter. PM levels were higher in winter in the 2<sup>nd</sup> cycle room, while PM concentrations were higher in spring in kindergarten and 1<sup>st</sup> cycle room. In general, PM concentrations were statistically different between the two seasons in all classrooms.

#### 301 Table 4

302 Median of PM concentrations (µg m<sup>-3</sup>) for weekdays in each classroom in winter and spring.

		Winter			Spring	
Classroom	$PM_{10}$	PM <sub>2.5</sub>	$\mathbf{PM}_1$	$PM_{10}$	PM <sub>2.5</sub>	$PM_1$
Kindergarten						
Median	6.67	6.05	4.42	14.0	12.5	10.0
<b>IQR</b> <sub>a</sub>	15.6	3.07	1.10	15.8	1.93	5.15
1 <sup>st</sup> cycle						
Median	18.1	9.96	6.31	18.1	15.0	11.4
IQR	21.6	5.76	3.06	39.6	3.99	2.62
2 <sup>nd</sup> cycle						
Median	24.5	15.7	13.8	18.7	13.0	9.21
IQR	24.9	14.8	16.1	8.79	4.17	5.82

303 <sup>a</sup>Interquartil range.

To compare PM concentrations in the school environment, the hourly average 304 concentration for PM is shown in **Fig.1**. The highest and lowest  $PM_{10}$  hourly 305 concentration was recorded in two different seasons. While the maximum value was 306 observed in the  $1^{st}$  cycle room (128 µg m<sup>-3</sup>; 16:00) in spring, the lowest concentration 307 was recorded in the kindergarten (4.9  $\mu$ g m<sup>-3</sup>; 7:00) in winter. PM<sub>10</sub> concentrations in 308 both seasons showed the main peaks during class time and periodic cleaning hour, which 309 can be explained because the rooms are normally swept, in addition to the resuspension 310 311 of soil material brought in on the soles of shoes, emissions associated with student 10

activities (e.g., paper and clothing fibres, skin peeling) and infiltration from outside to the
classroom environment, as reported in other studies (Alves et al., 2013; Madureira et al.,
2016). PM<sub>2.5</sub> represented, on average, 35% and 31% of the PM<sub>10</sub> concentrations during
class time compared to 91% and 90% during unoccupied periods in winter and spring,
respectively.

The Fig. 1 also revealed that PM<sub>2.5</sub> and PM<sub>1</sub> concentrations in the 1<sup>st</sup> and 2<sup>nd</sup> cycle rooms 317 318 in winter began to increase almost at the end of the school day and, depending on the 319 classroom, reached their peak between 18:00 and 22:00, which may suggest the 320 penetration of particles from residential biomass combustion. A different pattern was 321 observed in the kindergarten room, where the concentration of fine particles did not increase at night. It is believed that this behaviour is because the door always remains 322 closed after cleaning hours, making it difficult for particles to penetrate the environment, 323 as reported by other authors (Long et al., 2001). In a sampling campaign carried out in a 324 nearby school between September and November, it was concluded, based on the 325 326 chemical speciation of PM<sub>2.5</sub>, that biomass combustion represented 9% of the mass concentrations (Alves et al., 2023). In the present study, it is expected that the contribution 327 from this source will be greater, since it was carried out in winter, with lower 328 temperatures. In spring, concentrations for both particle sizes remained almost constant 329 throughout the day, with mean values below 15 and 11  $\mu$ g m<sup>-3</sup> for PM<sub>2.5</sub> and PM<sub>1</sub>, 330 331 respectively.

Although the daily mean concentrations were not higher than the values recommended 332 by the World Health Organisation ( $PM_{10}$ : 45 and  $PM_{2.5}$ :15 µgm<sup>-3</sup>), in winter, hourly  $PM_{10}$ 333 334 concentrations above 45  $\mu$ g m<sup>-3</sup> were observed in the kindergarten and the 1<sup>st</sup> cycle room, with a maximum of 62.7  $\mu$ g m<sup>-3</sup> during the school day. In spring, the peaks were registered 335 in the kindergarten and the 1<sup>st</sup> cycle room between 11:00 and 15:00, while the 2<sup>nd</sup> cycle 336 room did not record concentrations above 45  $\mu$ g m<sup>-3</sup> at any time of the day. Regarding 337 PM<sub>2.5</sub>, hourly concentrations above 15  $\mu$ g m<sup>-3</sup> were recorded in both the 1<sup>st</sup> and 2<sup>nd</sup> cycle 338 rooms, with levels in the 1<sup>st</sup> cycle room varying from 15.5 to 23.5  $\mu$ g m<sup>-3</sup> between 6:00 339 and 16:00 in spring. 340



341 Time
342 Figure 1. Variation in hourly average PM concentrations during weekdays in a) winter and b) spring. The highlighted band represents the class time.

According to the difference between the 5<sup>th</sup> and 95<sup>th</sup> percentiles (**Fig. 2**), there was greater 345 variability in PM<sub>2.5</sub> and PM<sub>1</sub> concentrations in winter than in spring, mainly when the 346 classrooms were not occupied. On the other hand, PM<sub>10</sub> concentrations varied more in 347 348 class time than unoccupancy periods in the two seasons. In general, much higher  $PM_{10}$ concentrations were observed during the school day compared to concentrations 349 350 measured during the period when classrooms were vacant. The maximum difference was recorded in spring when the average concentration in the kindergarten classroom was 17.2 351  $\mu$ g m<sup>-3</sup> during school hours and rose up to 51.7  $\mu$ g m<sup>-3</sup> when the classroom was vacant. 352 Unlike PM<sub>10</sub>, PM<sub>1</sub> concentrations were higher during free periods than class hours in both 353 winter and spring, except in the 1<sup>st</sup> cycle room. PM<sub>2.5</sub> concentrations between class time 354 and unoccupancy periods in the kindergarten were similar in winter and spring, while in 355 the 1<sup>st</sup> cycle room, class time levels were higher in spring. 356



357

Figure 2. Box plots showing the concentrations of particulate matter in class time and rest of the day in
 each room for a) winter and b) spring. Lower and upper boundaries of box plots represent the 25<sup>th</sup> and 75<sup>th</sup>
 percentiles, respectively; the line represents median values. The highlighted orange frames indicate when
 concentrations were not statistically different between school day and unoccupancy periods.

In this study, the average concentrations of  $PM_{10}$  and  $PM_{2.5}$  considering the two seasons 363 and all classrooms were 3.82 and 3.16 times lower than those reported by Branco et al. 364 (2019) in nurseries and primary schools in urban and rural locations in the north of 365 Portugal. Some studies have shown that student activities increase PM generation 366 367 indoors. Martins et al. (2020) reported that children in Lisbon were exposed to significantly higher concentrations of PM at school than at home, and that indoor 368 concentrations of PM<sub>10</sub> and PM<sub>2.5</sub> were often higher than outdoors. According to 369 researchers, human activity and external infiltration are the main sources associated with 370 internal PM. However, the results of the Lazović et al. (2022) study indicate that the 371 average concentrations of PM<sub>10</sub> and PM<sub>2.5</sub> in the outdoor environment in primary schools 372 in Serbia were 20% and 32% higher than the concentrations observed in classrooms. 373 374 Therefore, PM concentrations can be affected by several factors, such as classroom activities, ventilation, occupancy rate and external sources (Alameddine et al., 2022;
Tippayawong et al., 2009).

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- 378 379

#### 3.2. Deposition fraction of PM in two seasons and using two deposition models

Beposition fractions were obtained for the school day using two models (**Table 5**). In general, for all groups and in both seasons, the deposition fractions of  $PM_{2.5}$  and  $PM_{10}$  in the various regions of the respiratory tract showed the following trend: tracheobronchial < pulmonary < head airway. This is in line with the trends reported in previous studies (Jia et al., 2021; Khan et al., 2022).

385

Overall, there were small differences between spring and winter DFs, as DFs do not 386 387 depend on PM concentrations (Sánchez-Soberón et al., 2015), as previously reported in studies using MPPD (Li et al., 2015). In this study, the total DF of PM<sub>10</sub> calculated with 388 MPPD and ExDoM2 was up to 4% and 7% higher in spring than in winter, respectively. 389 This behaviour was slightly different for the finer particles (PM<sub>1</sub> and PM<sub>2.5</sub>), being mainly 390 larger in winter, except for children from the 2<sup>nd</sup> cycle, for whom DF were 4% and 10% 391 392 higher in spring according to MPPD and ExDoM2, respectively (Table 5). The seasonal 393 DF of PM<sub>10</sub> and PM<sub>2.5</sub> was identified to be associated with the mass mean aerodynamic diameter, as reported by Manojkumar and Srimuruganandam (2022) in the city of Vellore, 394 395 India. Based on the data from this study and using both models, it was possible to observe 396 that larger MMADs led to greater DF, regardless of the children's age. This indicates that the DF varies with the particle properties and, therefore, is normally not directly 397 proportional to the mass concentration, as reported in the results of the study by Kumar 398 399 et al. (2017).

400

**Table 5**. Average deposition fractions for the different PM sizes during a school day.

	MPPD				ExDoN	12		
Winter	Head	ТВ	Р	Total	Head	ТВ	Р	Total
$PM_1$								
Kindergarten	0.22	0.03	0.13	0.39	0.12	0.02	0.09	0.23
1 <sup>st</sup> cycle	0.23	0.04	0.17	0.44	0.07	0.02	0.08	0.17
2 <sup>nd</sup> cycle	0.21	0.04	0.18	0.43	0.04	0.03	0.08	0.15
PM <sub>2.5</sub>								
Kindergarten	0.26	0.03	0.16	0.45	0.37	0.02	0.13	0.52
1 <sup>st</sup> cycle	0.26	0.04	0.23	0.52	0.25	0.02	0.14	0.41
2 <sup>nd</sup> cycle	0.24	0.04	0.17	0.44	0.09	0.02	0.09	0.21
$PM_{10}$								
Kindergarten	0.33	0.09	0.24	0.66	0.73	0.03	0.08	0.84

1 <sup>st</sup> cycle	0.32	0.12	0.30	0.74		0.63	0.04	0.11	0.78
2 <sup>nd</sup> cycle	0.34	0.16	0.18	0.68		0.56	0.03	0.10	0.69
Spring									
$PM_1$					_				
Kindergarten	0.21	0.04	0.14	0.39		0.07	0.03	0.09	0.18
1 <sup>st</sup> cycle	0.21	0.04	0.19	0.43		0.04	0.03	0.08	0.15
2 <sup>nd</sup> cycle	0.21	0.04	0.17	0.43		0.05	0.03	0.08	0.15
$PM_{2.5}$									
Kindergarten	0.25	0.03	0.15	0.43		0.30	0.02	0.12	0.44
1 <sup>st</sup> cycle	0.25	0.03	0.19	0.47		0.15	0.02	0.11	0.28
2 <sup>nd</sup> cycle	0.25	0.04	0.19	0.48		0.17	0.02	0.12	0.31
$PM_{10}$									
Kindergarten	0.35	0.10	0.24	0.69		0.79	0.03	0.07	0.89
1 <sup>st</sup> cycle	0.35	0.14	0.26	0.75		0.69	0.04	0.10	0.82
2 <sup>nd</sup> cycle	0.34	0.14	0.24	0.72		0.63	0.03	0.10	0.77

402 TB: tracheobronchial; P: Pulmonary.

403

404 Still according to **Table 5**, the fraction in the head was the highest of the total airway 405 deposition, representing up to 35% with MPPD and 79% with ExDoM2. However, this 406 pattern was not observed for PM<sub>1</sub>, when using the ExDoM2 model, especially in spring, since in all ages the highest value was observed in the lung region and not in the head. 407 408 This can be because particles with a larger mass median aerodynamic diameter deposit more in the head region, while smaller aerodynamic diameters deposit preferably in the 409 410 alveolar region, considering that deposition by Brownian diffusion occurs mainly in the acinar region of the lung (Darquenne, 2020), Although with MPPD the greatest 411 contribution was not from the head, the fraction of PM<sub>1</sub> deposition in the alveolar region 412 was higher when compared to the fractions of  $PM_{10}$  and  $PM_{2.5}$ . 413

414

415 In general, the main difference between the models occurs for smaller particles. ExDoM2 calculates higher total DF of PM<sub>10</sub> but lower values for PM<sub>2.5</sub> and PM<sub>1</sub>. MPPD calculates 416 higher DF in TB and pulmonary regions, while ExDoM2 estimates higher values for the 417 418 head airway region. This is mainly due to discrepancies in the morphometry data such as 419 airway lengths, diameters, and branching angles, which in turn depend on the choice of the lung model and the input values of the respiratory variables, such as URT and FRC. 420 421 For example, ExDoM2 uses average values from different models (Weibel, 1963; Yeh and Schum, 1980; Phalen et al., 1985; Hansen and Ampaya, 1975) that vary depending 422 on the respiratory region (Aleksandropoulou and Lazaridis, 2013), but MPPD considers 423 8 geometries of the human lung, with the user being responsible for choosing one of the 424 options. In addition, the models use different approaches for deposition calculations. 425 426 While ExDoM2 is mainly based on the empirical equations proposed in ICRP, MPPD is 427 based on that proposed by Asgharian and Anjilvel. (1998), also using a Monte Carlo428 approach.

429

430 The smallest variation between the total deposition results of the models was observed for the 2<sup>nd</sup> cycle, whereas the largest difference between the models was identified in 431 kindergarten. The total DF of PM<sub>10</sub> estimated with the ExDoM2 was up to 22% higher 432 than the results from MPPD. However, the total DF of PM<sub>2.5</sub> and PM<sub>1</sub> derived from 433 MPPD was higher (up to 66% for PM<sub>1</sub>) than the value estimated with ExDoM2 in both 434 435 seasons (Table S4, Supplementary material). By way of example, Fig. 3 displays the diurnal cycles of deposition fraction of particulate matter in the group of 9- and 10-year-436 437 old children, split into sizes and model. It is possible to observe little difference from one model to another in the total DF of PM<sub>10</sub>, although there are large differences in the DF 438 439 by regions.

440

441 It is noticeable in Fig. 3 that a larger fraction of PM<sub>1</sub> and PM<sub>2.5</sub> is deposited in the lung region, while PM<sub>10</sub> is deposited in the head region. ExDoM2 predictions show DFs of 442 443  $PM_{2.5}$  of 0.09 in the head and lung, while the fraction of  $PM_{10}$  deposited in the upper airway and pulmonary region averaged 0.56 and 0.10, respectively. MPPD follows the 444 445 same trend but with different values. While the fraction of PM<sub>2.5</sub> deposited in the head 446 and P was, on average, 0.24 and 0.17, respectively, the fraction of  $PM_{10}$  deposited in head and P was, on average, 0.34 and 0.19, respectively. This trend was also observed by Gao 447 448 et al. (2022) in a study carried out in China with 10-year-old children during commuting trips to school, which showed that for the three transport modes evaluated and the three 449 450 particle sizes, the proportion of DFs in the head was the highest, especially for  $PM_{10}$ , representing up to more than 85% of the total fraction deposited. For PM<sub>1</sub> and PM<sub>2.5</sub>, the 451 DFs of the P significantly exceeded those of the TB part. This is especially important 452 since children with asthma are more sensitive to air pollution because they breathe at 453 454 higher tidal volumes, which can increase the efficiency of PM<sub>2.5</sub> deposition in the lung (Afshar-Mohajer et al., 2022). 455 456



Figure 3. Hourly deposition fraction for different particle sizes in children aged 10 years (ExDoM2) and 9
 years (MPPD) during the school day in winter. DFs were calculated from average values during a week of
 measurements in December.

461

After the comparison of both models, it was observed that the deposition of particles 462 463 larger than 1.17 µm was greater than that of smaller particles. Location wise, it was greater in the head region due to their higher sedimentation and impaction rates, while the 464 deposition of particles with smaller sizes was greater in the P region due to the diffusion 465 effect (Guo et al., 2019). Overall, this trend was most noticeable with the ExDoM2 results, 466 467 which can be explained because the respiratory tract model (RTM) used in ExDoM2 is an updated version of the RTM (ICRP, 2012), in which particles deposited in the 468 extrathoracic region (ET) are partitioned 65% to ET1 and 35% to ET2 (Chalvatzaki and 469 Lazaridis, 2015). 470

472 3.3. Deposition dose during the school day

Fig. 4 shows the deposited dose during the school day of the three sizes of PM in different 473 474 parts of the respiratory system for all children in the two periods evaluated. Important differences can be observed between the two seasons and the two models, with values 475 476 being higher in spring. While seasonal variations showed only a slight influence on the 477 total deposition fraction, they had a large effect on the school day deposited dose, 478 particularly during spring as a direct consequence of higher PM concentrations (Table S5, Supplementary material). On the other hand, after calculating the deposited dose and the 479 deposition fraction considering the gender of the children, it was observed that regardless 480 of their age, the results were the same for both girls and 5-year-old boys, and likewise for 481 girls and boys aged 10 years. This may be because ExDoM2 uses as input the same values 482 for physiological parameters without differentiating by gender. For this reason, the results 483 in this study were always reported and discussed for children according to age, and not 484 by gender. 485

486 The highest accumulated dose of PM<sub>2.5</sub> and PM<sub>10</sub> in the two seasons occurred for children attending the 1<sup>st</sup> cycle and coincides with the highest environmental concentration of PM<sub>1</sub> 487 and  $PM_{2.5}$  observed in this room. The overall results show that while the total  $PM_{10}$ 488 deposition dose estimated with ExDoM2 was up to 58% higher than the values derived 489 from the MPPD model, the dose for PM<sub>1</sub> obtained with the MPPD was between 15% and 490 491 69% higher than the value calculated with ExDoM2. The differences can be attributed to the specific values used in the two models for the tidal volume and respiratory frequency 492 parameters and, consequently, the value of the ventilation rate. Furthermore, the increase 493 and decrease in dose estimated in any of the models coincides with the observed pattern 494 495 in the deposition fraction (Fig. 4).



**498 Figure 4.** Regional deposited dose of a)  $PM_{1,}$  b)  $PM_{2.5}$  e c)  $PM_{10}$  for each group during the school day in 499 winter and spring. Comparison between the two models. Head: upper airway; P: pulmonary; TB: 500 tracheobronchial. Different scales on the y-axis were purposely chosen to highlight the values and aid 501 visualisation.

The PM<sub>10</sub> doses for the school day calculated in the present study were lower than the 502 503 results reported in previous studies. Chalvatzaki et al. (2020) applied ExDoM2 to predict 504 the  $PM_{10}$  dose received by students at five primary schools in Lisbon, Portugal, assuming that the exposed children were 10-year-old nasal breathers. The researchers found that 505 506 the total deposited dose ranged from 72 to 239 µg in indoor school environments, while in the current study it was, on average, 84.4 µg with the same model (10 years old) and 507 508 66 μg with MPPD (9 years old). Faria et al. (2020) quantified the exposure of children 509 between 5 and 10 years old to PM and the respective daily inhaled dose considering various microenvironments in Lisbon. The researchers observed that during the week 510 children inhale, on average, 96 and 177  $\mu$ g of PM<sub>2.5</sub> and PM<sub>10</sub>, respectively, with the 511 classroom contributing more than the residential environment. In their study, the inhaled 512 doses were higher than the doses calculated with both models in this study, as the inhaled 513 dose does not consider the deposition fraction and, therefore, the estimated value will 514 515 always be higher. In any case, these results showed that children inhale more PM in 516 classrooms than in residential environments, despite spending more time at home.

Regarding the amount of PM deposited per region, higher cumulative doses occurred 517 mainly in the head region and lower ones in the TB region, except for PM<sub>1</sub> calculated 518 with ExDoM2 (Fig. 4), coinciding with the pattern of deposition fractions. However, 519 unlike the DFs of PM<sub>1</sub> (ExDoM2) from kindergarten in winter, the DFs of PM<sub>1</sub> with 520 ExDoM2 were always greater in the lung region than in the upper airways. This can be 521 522 associated with the aerodynamic diameter of the particles, since when compared by classrooms and seasons, the largest MMADs of PM1 obtained was in kindergarten in 523 winter (Table S1, Supplementary Material). However, even though the DFs of PM<sub>1</sub> 524 525 (ExDoM2) were higher in the lung region, they were lower than the values estimated with the MPPD. 526

In general, a comparison between the two models revealed that higher deposition doses 527 in the lung and TB region were always estimated with MPPD for 8- and 9-year-old 528 children in both seasons. Nevertheless, the same did not happen in kindergarten due to 529 the ventilation rate in ExDoM2 being 0.095  $\text{m}^3 \text{h}^{-1}$  higher than the value used by MPPD, 530 531 despite the DFs calculated with MMPD for these two regions being always higher than those estimated with ExDoM2. In both seasons, the majority of the PM<sub>10</sub> dose was 532 deposited in the head region, while PM<sub>1</sub> and PM<sub>2.5</sub> were predominantly deposited in the 533 lung region. It is noteworthy that different mechanisms govern PM deposition in the TB 534 535 and lung regions, such as diffusion, impaction, and sedimentation (Deng et al., 2018). 536 Regardless of the model, special attention should be paid to these results, particularly 537 those related to finer particles, since PM<sub>2.5</sub> depositions in the respiratory tract, including P and TB regions, are associated with elevated blood pressure and greater risk of 538 539 prehypertension and hypertension in children aged 4 to 12 years (Liu et al., 2021).

540 Fig. 5 shows a statistical summary of the hourly deposited doses of  $PM_1$ ,  $PM_{2.5}$  and  $PM_{10}$ for all groups between 8 h and 16 h. With MPPD, a 3-year-old child has a lower mean 541 dose deposited per hour compared to children aged 8 and 9 years for the three PM sizes 542 543 evaluated in both seasons, while with ExDoM2, the lowest values were observed in children aged 10 years of the 2<sup>nd</sup> cycle. The differences observed in deposited doses can 544 be explained by the DFs predicted by the two models. In the case of MPPD, deposition 545 546 efficiency is lower in the respiratory tract of 3-year-old children compared to 8- and 9-547 year-old children, whatever the particle size and season, while ExDoM2 predictions showed the highest DF values for 5-year-old children. As shown by Poorbahrami et al. 548 (2021), the differences in total and regional deposition calculations are probably due to 549 the use of different lung models, which in turn suggest volumetric and structural 550 551 differences in lung morphologies according to the ages of the individuals.



553

Figure 5. Boxplots with hourly doses of different sizes of PM for the three groups evaluated, split into model and a) winter and b) spring.

Although both models describe the same hourly profile pattern, with peaks at different 556 times depending on classroom activities and particle size, the deposited dose values show 557 clear differences from model to model. In the 1<sup>st</sup> cycle and 2<sup>nd</sup> cycle classrooms, the PM<sub>2.5</sub> 558 and PM<sub>10</sub> peaks coincided at the same time, different from the PM<sub>1</sub> pattern, with more 559 stable values being observed throughout the day. The hourly profile for winter reveals 560 that both particle doses calculated with ExDoM2 and those estimated from MPPD showed 561 562 variations linked to PM sources (Fig. 6). Normally, the deposited dose increased between 8 h and 10 h (which is related to the start of the school day) and decreased during rest and 563 564 lunch times. On the other hand, high doses, generally from 14 h onwards, are associated with students leaving, as well as emissions from cleaning classrooms. 565

The difference between the  $PM_{10}$  doses predicted by ExDoM2 were 58%, 31% and 24% higher than those calculated with the MPPD for kindergarten, 1<sup>st</sup> cycle and 2<sup>nd</sup> cycle, respectively, while for PM<sub>1</sub> the MPPD calculated doses 46% and 57% higher for children in the 1<sup>st</sup> cycle and 2<sup>nd</sup> cycle, respectively. This is in accordance with the pattern of the total deposition fraction of these PM sizes and their respective concentrations. However, despite the MPPD calculating higher DFs for PM<sub>1</sub> and PM<sub>2.5</sub> in kindergarten and 1<sup>st</sup> cycle compared to ExDoM2, the doses did not follow this behaviour. This is due to the ventilation rate used by ExDoM2 being higher, compared to the values used by MPPD for children aged 3 and 8 years. Similar relationships were also observed in spring.



575

576 Figure 6. Hourly cycle of PM deposition doses during school hours in winter according to group and the577 dosimetry model.

Furthermore, high R values (R > 0.89) were observed between the MPPD and ExDoM2 578 hourly doses in both seasons (Table S6, Supplementary Material). However, there were 579 some exceptions, as in the case of the 2<sup>nd</sup> cycle classroom. The low correlations occur 580 581 because some dose peaks calculated with MPPD were not present in the ExDoM2 dataset or because one of the models showed lower doses compared to the other. Coincidentally, 582 583 weak correlations between the hourly doses of the two models were observed only for R values below 0.63 between hourly deposited dose and hourly exposure (Table S5, 584 585 Supplementary Material).

In addition to linear regression, the difference and agreement between the hourly doses of the two models were also evaluated. While the paired t-test applied at a significance level of 5% revealed that there are statistically significant differences in the hourly deposited dose of all PM sizes in the two seasons, the Bland-Altman test confirmed that
even though there were differences in the hourly values, good agreement was found
between the predictions of the two models for deposited particle doses (Table S7 and
Figure S1, Supplementary Material.

Overall, this study highlights the importance of evaluating the personal exposure to PM 593 594 of children of different ages who attend a large school that integrates several levels of 595 education (~700 students), and which is located less than 1 km away from the largest industrial complex in the country. In Portugal, schools offering pre-school, 1<sup>st</sup> cycle and 596 597 2<sup>nd</sup> cycle education are abundant (10,991 establishments) and represent 71% of the population of children attending these levels of education (~1,189,353 students from pre-598 school up to the 3<sup>rd</sup> cycle of basic education). Thus, the results of this study may be valid 599 in other establishments with these levels of education, and serve to encourage 600 601 measurements in school environments considering a larger group of classes to calculate in more detail the exposure of schoolchildren to PM. However, the results should be 602 603 interpreted with caution because they are based on a short data set that may have been affected by specific conditions of the sampling period or by sporadic events. The 604 impossibility of monitoring and collecting samples simultaneously in different 605 606 classrooms and other school environments constitutes another limitation of the study. In 607 addition, variability in the activities carried out in the classrooms was not considered. 608 Therefore, the children's respiratory patterns were constant, and the same exposure 609 scenario was evaluated (nasal breathing under sitting activity level). Finally, whenever possible, monitoring in classrooms for longer periods (more than 1 week) should be taken 610 611 into account, as well as carrying out at least one sampling campaign in each season with the aim of improving the representation of variations in weather conditions and school 612 613 activities throughout the academic year.

614

#### 615 4. Conclusions

This study suggests that PM concentrations are statistically different from one season to 616 another, and that the variability of concentrations is also influenced by room occupancy 617 618 periods, with higher levels of PM<sub>10</sub> during the school day compared to periods when the classrooms are empty. Although PM levels were low compared to other studies, 619 dosimetry models indicate that a significant amount of PM was deposited in pupils' 620 respiratory tracts during school hours. The patterns indicate larger deposited doses in 621 622 spring compared to winter, with children aged 9 and 10 attending the 1<sup>st</sup> cycle being the 623 most affected. Furthermore, the variations observed in the deposition fractions and deposited doses for the models are the result of differences in the physiological 624 parameters of the respiratory system (e.g., URT, FRC, BF and TV) used as input, which 625 626 are age dependent. None of the models covers all ages of the children targeted in the 627 present study. In general, a good agreement was obtained between both models, although 628 some differences, especially for regional deposition, could be observed. The dosimetry 629 models applied in this research may be useful in other educational settings to identify and 630 mitigate the impact of local and regional sources of air pollution in the school 631 environment.

632

#### 633 Declaration of Competing Interest

634 The authors declare that they have no known competing financial interests or personal635 relationships that could have influenced the work documented in this manuscript.

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Appendix A. Supplementary material. Additional material related to this paper isavailable online at:

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#### SUPPLEMENTARY MATERIAL

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847 **Table S6.** Mean concentration ( $\mu$ g m<sup>-3</sup>), MMADs ( $\mu$ m) and GSDs ( $\mu$ m) of PM<sub>10</sub> used as input 848 data in the two models.

Kindergarten		Winter			Spring	
Time	PM <sub>10</sub>	MMAD	GSD	PM <sub>10</sub>	MMAD	GSD
8:00	5.53	1.24	2.95	14.6	4.05	2.18
9:00	30.7	3.80	1.97	49.7	3.74	2.31
10:00	41.2	2.84	2.04	47.0	4.05	1.79
11:00	48.4	3.73	1.76	71.3	3.28	1.96
12:00	18.4	2.55	2.09	45.7	3.61	2.15
13:00	28.7	3.60	2.00	44.6	4.31	1.70
14:00	61.7	4.45	1.56	71.9	4.39	1.65
15:00	44.4	4.08	1.62	67.4	3.62	1.98
16:00	17.5	2.51	2.21	34.3	2.89	2.45
1 <sup>st</sup> cycle	Winter			Spring		
Time	$PM_{10}$	MMAD	GSD	$PM_{10}$	MMAD	GSD
8:00	9.50	1.01	2.21	19.1	3.22	3.03
9:00	27.3	1.32	2.47	54.2	3.54	2.65
10:00	52.9	3.18	2.41	69.1	3.66	2.26
11:00	62.7	4.17	1.80	68.0	3.91	1.97
12:00	34.9	4.21	1.63	66.6	3.10	2.82
13:00	48.2	3.48	1.93	44.7	3.86	2.13
14:00	32.4	3.70	1.80	71.0	4.15	1.82
15:00	56.1	3.15	2.05	81.4	4.51	1.57
16:00	41.6	4.02	1.76	128.7	2.42	3.32
2 <sup>nd</sup> cycle	Winter			Spring		
Time	$PM_{10}$	MMAD	GSD	$PM_{10}$	MMAD	GSD
8:00	19.2	0.98	5.41	24.9	3.19	2.81
9:00	27.7	1.70	5.82	38.1	3.49	2.37
10:00	44.6	2.78	3.86	40.2	3.28	2.47
11:00	46.0	3.08	3.25	33.4	3.53	2.34
12:00	48.2	3.40	2.73	37.3	3.29	2.32
13:00	31.1	2.81	3.20	29.9	2.53	2.45
14:00	19.1	2.00	3.99	21.5	3.64	2.13
15:00	46.4	3.90	2.25	36.5	2.64	2.89
16:00	27.5	2.85	3.06	25.1	2.17	3.22

849

The equations used by ExDoM2 to calculate the particle deposition for the upper respiratory tract region (DF<sub>H</sub>), which includes the extrathoracic airways ET1 (anterior nose) and ET2 (posterior nasal passages, larynx, pharynx and mouth); the tracheobronchial region (DF<sub>TB</sub>) which includes the bronchial (BB) and the bronchiolar (bb); the alveolar region (DF<sub>AL</sub>) and the total deposition (DF) are those formulated by the International Commission on Radiological Protection (ICRP) and are presented below. In the equations, d<sub>p</sub> is the particle diameter in  $\mu$ m, and IF is the inhalable fraction (in equations 1 and 4) and is calculated according to equation 5.

857 
$$DF_{H} = IF\left[\frac{1}{1 + e^{(6.84 + 1.183 \ln d_{p})}} + \frac{1}{1 + e^{(0.924 - 1.885 \ln d_{p})}}\right]$$
(1)

858

859 
$$DF_{TB} = \left(\frac{0.00352}{d_p}\right) \left[ e^{(-0.234(\ln d_p + 3.40)^2)} + 63.9e^{(-0.819(\ln d_p - 1.61)^2)} \right]$$
(2)

861 
$$DF_{AL} = \left(\frac{0.0155}{d_p}\right) \left[ e^{(-0.416(\ln d_p + 2.84)^2)} + 19.11e^{(-0.482(\ln d_p - 1.362)^2)} \right]$$
(3)

863 
$$DF = IF \left[ 0.0587 + \frac{0.911}{1 + e^{(4.77 + 1.485 \ln d_p)}} + \frac{0.943}{1 + e^{(0.508 - 2.58 \ln d_p)}} \right]$$
(4)

865 
$$IF = 1 - 0.5 \left( 1 - \frac{1}{7.6x10^{-4}d_p^2 + 1} \right) + 10^{-5}U^{2.75}e^{(0.055d_p)}$$
(5)

<b>Table S7.</b> Age considered according to the r	nodel.
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	MPPD	ExDoM2
Kindergarten	3 years	5 years
1 <sup>st</sup> cycle	8 years	10 years
2 <sup>nd</sup> cycle	9 years	10 years

Thermal conditions, including wind direction, were measured with a portable monitoring
equipment (Delta Ohm 32.1 and 32.3), which included the following sensors: AP3203 Omnidirectional Hot Wire Probe, TP3275 - Globe Temperature Probe, and HP3217R - Combined
Temperature and RH Probe. The equipment was installed at a height of 1 m above the floor and
operated at a 1-minute time resolution. Each classroom was monitored for a week in winter 2022
(Table S3).

#### **Table S8**. Average wind speed (m $s^{-1}$ ).

Time	Kindergarten	1 <sup>st</sup> cycle	2 <sup>nd</sup> cycle
8:00 - 8:59	0.000	0.001	0.001
9:00 - 9:59	0.000	0.002	0.001
10:00 - 10:59	0.005	0.005	0.002
11:00 - 11:59	0.003	0.002	0.002
12:00 - 12:59	0.000	0.002	0.002
13:00 - 13:59	0.000	0.003	0.001
14:00 - 14:59	0.001	0.005	0.001
15:00 - 15:59	0.008	0.002	0.001
16:00 - 16:59	0.002	0.002	0.002

877 The discrepancies between the dosimetry models for different particle sizes in the two seasons

878 are presented in Table S4. For  $PM_{10}$ , the total DF is up to 22%, showing that the MPPD

underestimates the total DF compared to the ExDoM2 counterparts. However, ExDoM2 appearsto underestimate the total DF for finer particles compared to the other model.

Table S9. Discrepancy of total DFs derived from the models. Bold indicates that ExDoM2 gave
 values higher than MPPD.

	Winter			Spring			
	$PM_{10}$	PM <sub>2.5</sub>	$\mathbf{PM}_1$	$PM_{10}$	PM <sub>2.5</sub>	$PM_1$	
Kindergarten	22%	13%	41%	22%	3%	53%	
1 <sup>st</sup> cycle	5%	20%	60%	8%	40%	66%	
2 <sup>nd</sup> cycle	2%	53%	66%	6%	36%	64%	

Table S10. Linear correlation coefficients between hourly exposure and hourly deposited dose
 according to the model (R).

	Winte	er					Spring					
	MPPD			ExDoM2			MPPD			ExDoM2		
	$PM_{10}$	PM <sub>2.5</sub>	$PM_1$	$PM_{10}$	PM <sub>2.5</sub>	$PM_1$	PM10	PM <sub>2.5</sub>	$PM_1$	$PM_{10}$	PM <sub>2.5</sub>	$PM_1$
Kindergarte												
n	0.99	0.99	0.99	0.99	0.96	0.99	0.99	0.98	1	0.99	0.78	0.99
1 <sup>st</sup> cycle	0.98	0.99	0.99	0.95	0.96	0.90	0.95	0.87	0.99	0.91	0.09*	0.99
2 <sup>nd</sup> cycle	0.98	0.98	1	0.97	0.09*	0.99	0.63*	0.12*	0.41*	0.92	0.06*	0.99

\*Values indicate weak correlations.

# 889 Table S11. Linear correlation coefficients between the hourly deposited doses by the two890 models (R).

		Winter		Spring			
	$PM_{10}$	PM <sub>2.5</sub>	$\mathbf{PM}_1$	$PM_{10}$	PM <sub>2.5</sub>	$\mathbf{P}\mathbf{M}_1$	
Kindergarten	0.99	0.98	0.98	0.99	0.89	0.99	
1 <sup>st</sup> cycle	0.99	0.98	0.92	0.99	0.40*	0.99	
2 <sup>nd</sup> cycle	0.99	0.20*	0.99	0.58*	0.45*	0.40*	

891 \*Values indicate weak correlation.

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**Table S12**. Average hourly deposited dose of PM in each monitoring campaign and p-values

894	from the	paired sat	mple t-test.	Dose	values are	presented	in µg	h-1 fc	r both	models
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	$PM_{10}$			PM <sub>2.5</sub>			$PM_1$			
Campaign	MPPD	ExDoM2	p-value	MPPD	ExDoM2	p-value	MPPD	ExDoM2	p-value	
Kindergarten -wr	3.96	9.40	1.0E-03	0.76	1.64	2.8E-04	0.28	0.31	1.11e-02	
1 <sup>st</sup> cycle - wr	8.77	12.6	7.2E-04	1.61	1.75	0.046	0.60	0.32	5.8E -10	
2nd cycle - wr	7.20	9.44	1.2E-03	1.81	1.05	1.0E-05	1.29	0.55	2.0E-06	
Kindergarten - sg	6.20	14.2	6.1E-05	1.01	1.92	1.6E-05	0.49	0.42	2.0E-04	
1 <sup>st</sup> cycle - sg	14.3	20.7	4.6E-05	2.40	1.93	2.8E -03	1.26	0.57	1.8E-07	
2 <sup>nd</sup> cycle - sg	7.49	9.32	1.3E-03	1.95	1.36	9.4E -05	1.29	0.40	7.3E-06	

895 Wr: winter; sp: spring.

The Bland-Altman graph was generated to assess the agreement between the two models. The differences between doses are displayed on the y axis; on the x-axis, dose averages are plotted. This method has two limits of agreement at  $\pm 1.96$  times the standard deviation. Ideally, all points should fall between these two limits. After making the graphs for all rooms and the three particle sizes of the two seasons, the 1<sup>st</sup> cycle room presented a point outside the limits of agreement for PM<sub>10</sub> and PM<sub>1</sub> and, the 2<sup>nd</sup> cycle room for PM<sub>10</sub> in spring. The other cases were always within this limit, as shown in **Figure S1**.



903Average hourly deposited dose904Figure S7. Bland-Altman plot generated for the hourly deposited doses of PM1 calculated for 1st cycle in<br/>winter.905winter.

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