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1 **Indoor air quality in urban nurseries at Porto City: particulate matter assessment**

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12

13 **Abstract**

14 Indoor air quality in nurseries is an interesting case of study mainly due to children's high vulnerability to
15 exposure to air pollution (with special attention to younger ones), and because nursery is the public
16 environment where young children spend most of their time. Particulate matter (PM) constitutes one of
17 the air pollutants with greater interest. In fact, it can cause acute effects on children's health, as well as
18 may contribute to the prevalence of chronic respiratory diseases like asthma. Thus, the main objectives of
19 this study were: i) to evaluate indoor concentrations of particulate matter (PM_{10} , $PM_{2.5}$, PM_{10} and PM_{Total})
20 on different indoor microenvironments in urban nurseries of Porto city; and ii) to analyse those
21 concentrations according to guidelines and references for indoor air quality and children's health. Indoor
22 PM measurements were performed in several class and lunch rooms in three nurseries on weekdays and
23 weekends. Outdoor PM_{10} concentrations were also obtained to determine I/O ratios. PM concentrations
24 were often found high in the studied classrooms, especially for the finer fractions, reaching maxima
25 hourly mean concentrations of $145 \mu\text{g m}^{-3}$ for PM_{10} and $158 \mu\text{g m}^{-3}$ $PM_{2.5}$, being often above the limits
26 recommended by WHO, reaching 80% of exceedances for $PM_{2.5}$, which is concerning in terms of
27 exposure effects on children's health. Mean I/O ratios were always above 1 and most times above 2
28 showing that indoor sources (re-suspension phenomena due to children's activities, cleaning and cooking)
29 were clearly the main contributors to indoor PM concentrations when compared with the outdoor
30 influence. Though, poor ventilation to outdoors in classrooms affected indoor air quality by increasing the
31 PM accumulation. So, enhancing air renovation rate and performing cleaning activities after the
32 occupancy period could be good practices to reduce PM indoor air concentrations in nurseries and,
33 consequently, to improve children's health and welfare.

34

35 **Keywords**

36 Indoor air, nursery, particulate matter, children, exposure

37

38 **1. Introduction**

39 Public health awareness on indoor air pollution has lagged behind that on outdoor air pollution. However,
40 air quality inside public and private buildings where people spend a large part of their life is an essential
41 determinant of healthy life and people's welfare. Evidence has been made that people, especially children,
42 spend most of their time in indoor environments and therefore are more exposed to indoor air pollution
43 (Almeida et al., 2011). Whilst this does not per se mean that indoor exposures will produce more harmful
44 effects, the evidence is that indoor concentrations of many air pollutants are often higher than those
45 typically encountered outside (Jones, 1999).

46 In this particular field, nurseries could be a very interesting case study (Sousa et al., 2012a) for two main
47 reasons. Firstly, because of children's not fully developed immune system and lungs, their relative higher
48 amount of air inhalation (the air intake per weight unit of a resting infant is twice that of an adult) and
49 their growing tissue and organs (Mendell and Heath, 2005), which together raise the possibility of higher
50 exposures than seen in adults (Schwartz, 2004). Secondly, because children spend more time in schools
51 (or preschools and nurseries in the case of younger children) than in any other indoor environments
52 besides home, and there is a correlation between pollutant concentrations and the onset of health
53 problems in schoolchildren (Cartieaux et al., 2011). Indoor air quality in nurseries and pre-schools is
54 different from primary or higher schools (Yoon et al., 2011), although this has been largely ignored
55 (Ashmore and Dimitroulopoulou, 2009).

56 Several pollutants are present in nurseries' indoor air, but particulate matter (PM) is of great interest
57 mainly because of its public health significance (Harrison et al., 2002). PM comprises material in solid or
58 liquid phase suspended in the air and may have very diverse chemical compositions that are highly
59 dependent on their source. On the other hand, it has been demonstrated that the finer PM fractions are the
60 ones with the most acute effects on human health (Schwartz and Neas, 2000). This is why recently
61 measurements of total suspended PM (PM_{Total}) have been replaced by total thoracic particles (particles
62 with an aerodynamic diameter smaller than $10\ \mu m$, PM_{10}) and also, more recently, by finer particles
63 (particles with an aerodynamic diameter smaller than $2.5\ \mu m$, $PM_{2.5}$, and smaller than $1\ \mu m$, PM_1) (Monn,
64 2001).

65 PM concentrations on nurseries can be influenced by several factors and can arise from both indoor and
66 outdoor sources. Physical activities of the pupils lead to the re-suspension of mainly indoor coarse
67 particles and greatly contribute for increasing PM_{10} in classrooms (Fromme et al., 2008). Furthermore,
68 Lim et al. (2012) suggested that the impact of the activity pattern on personal exposure of PM is
69 significant. Cleaning activities and ventilation are also major factors that determine indoor air PM
70 concentrations in classrooms (Heudorf et al., 2009). Cooking is also an important source of indoor PM
71 (Monn et al., 1997). Dust coming from outside of the buildings can be a major source of PM
72 concentration and it can be responsible for the existence of very adverse compounds in particles, as the
73 example of heavy metals mainly due to automobile emissions (Darus et al., 2012). Sousa et al. (2012b)
74 recently reviewed the available studies that have been done concerning PM_{10} and $PM_{2.5}$ concentrations in
75 nurseries and primary schools from 2008 to 2012, and found that: i) PM concentrations observed
76 worldwide exceeded several times national legislations and WHO guidelines; ii) indoor/outdoor ratios
77 were several times higher than 1; and iii) PM concentrations were reported as mainly due to constant re-
78 suspension of particles. Added to it, there is spatial and temporal heterogeneity in the distribution of air
79 quality within school environments, which is affected by the penetration of outdoor pollutants, wall
80 absorption, emissions from furniture and other materials, level and length of occupancy, and quality of
81 ventilation (Mejía et al., 2011).

82 Indoor air quality problems often cause non-specific symptoms rather than clearly defined illness,
83 especially regarding the respiratory system (Jones, 1999). However, there are evidences that pollutants

84 such as PM may cause acute effects as irritation in the skin, eyes, nose and throat and upper airways, as
85 well as may contribute to the prevalence of chronic respiratory diseases, like asthma (Sousa et al., 2012a).

86 In addition to higher health concerns, classroom air quality also affects the performance of school
87 activities by children, so it is important to understand cost-effective good practices and measures to
88 improve indoor air quality in nurseries (Wargocki and Wyon, 2013). In order to protect human health
89 from PM indoor air pollution exposure, national and international authorities set up standards and
90 guidelines. Some of these are for industrial or occupational purposes, like the example of the U.S.
91 Department of Labor, Occupational Safety and Health Administration (OSHA) that sets the limits of 5
92 000 and 15 000 $\mu\text{g m}^{-3}$ (8-hour time weighted average) for $\text{PM}_{2.5}$ fraction and PM_{Total} , respectively. Other
93 example is set by the Institute of Environmental Epidemiology, Ministry of the Environment of Singapore
94 (Singapore, 1996), which recommended the maximum concentration of 150 $\mu\text{g m}^{-3}$ for PM_{10} as the limit
95 for acceptable indoor air quality. On the other hand, the Indoor Air Quality Management Group from the
96 Government of the Hong Kong Special Administrative Region (Hong Kong, 2003) established, for 8-hour
97 average in offices and public spaces, the PM_{10} limits of 180 and 20 $\mu\text{g m}^{-3}$ for good (represents the IAQ
98 that provides protection to the public at large including the young and the aged) and excellent (represents
99 an excellent IAQ that a high-class and comfortable building should have) classes respectively, the latter
100 accordingly to the Finnish Society of Indoor Air Quality and Climate. The World Health Organization
101 (WHO, 2010) recommended to apply to indoor spaces the same PM guidelines as for ambient air,
102 presented on the 2005 global update, which are 25 and 50 $\mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$ and PM_{10} , respectively (over
103 24 hours). These WHO guidelines are adopted by other authorities, like ANSES - French Agency for
104 Food, Environmental and Occupational Health & Safety. The Federal Department of Health Canada
105 recommended that $\text{PM}_{2.5}$ indoor concentrations should be kept as low as possible in all indoor
106 environments (Health Canada, 2012). The Portuguese national legislation (*Decreto-Lei n° 79/2006*)
107 established a maximum limit of 150 $\mu\text{g m}^{-3}$ for PM_{10} , specifically in school indoor environments.

108 In the recent decades many studies have been carried out in children's dwellings to study indoor air
109 quality, but children's dwelling is not, however, their only microenvironment; the most important indoor
110 environment for children and their primary place of social activity is the nursery, and up till now indoor
111 environment quality in this place has been poorly documented (Roda et al., 2011). In fact, and as far as
112 known, there are only a few studies published concerning the indoor air quality in nurseries, particularly
113 regarding PM measurements. Fromme et al. (2005) analysed respirable PM and elemental carbon levels
114 in the indoor air of apartments and nursery schools in the urban area of Berlin (Germany), and found that
115 the outdoor motorway traffic was correlated with the indoor air in the studied nurseries. However, only 1-
116 day measurements were performed (sampling time from 7 to 8 hours) and the samples occurred merely in
117 one place per nursery. Yang et al. (2009) characterized the concentrations of different indoor air
118 pollutants, including PM_{10} , within Korean schools and nurseries and concluded that, in average, children
119 were more exposed to PM inside nurseries than outdoors and suggested that increasing ventilation rate
120 could play a key role to improve indoor air quality in nurseries. Although measurement campaigns were
121 performed during summer, autumn and winter, and it has had into account the building age, this study did
122 not performed measurements in the lunch rooms neither in different floors inside each studied building,
123 and only considered the PM_{10} fraction. Wichmann et al. (2010) studied the extent of infiltration of $\text{PM}_{2.5}$
124 (as well as soot and NO_2) from outdoor to indoor in the major indoor environments occupied by children
125 (10 pre-schools, 6 schools and 18 homes) in different locations (city centre, suburban area and
126 background), and found that, despite outdoor infiltrations, $\text{PM}_{2.5}$ concentrations in these indoor
127 environments were mainly due to indoor sources. However, this study was limited to places occupied by
128 children over 6 years old and measurements were only made for $\text{PM}_{2.5}$ fraction and in one classroom per
129 pre-school. More recently, Yoon et al. (2011) studied 71 classrooms in 17 nurseries (preschools) and
130 searched for indoor air quality differences (several pollutants including PM_{Total} and respirable
131 particulates) between urban and rural ones, and confirmed that the PM concentrations indoors were higher
132 than those in outdoor, and also that those in urban areas were higher than in rural areas. Lack of
133 comparative analysis between different classrooms and other environments inside the same nursery and a
134 limited analysis to the coarser PM fractions were the major limitations of this study.

135 In Portugal, as far as it is known there are no studies focusing on PM in nurseries' indoor air; there are
136 only few studies focusing on the indoor air of primary schools (Almeida et al., 2011; Pegas et al., 2012).

137 To reduce the above referred lacks, the main objectives of this study were: i) to evaluate indoor
138 concentrations of particulate matter (PM_1 , $PM_{2.5}$, PM_{10} and PM_{Total}) on different indoor
139 microenvironments in urban nurseries in Porto city; and ii) to analyse those concentrations according to
140 guidelines and references for indoor air quality and children's health.

141 2. Methods

142 In Portugal, there are a considerable number of children attending nurseries. In fact, there are 276,125
143 children (2.26% of the Portuguese population) attending a total of 6,812 nurseries, 64.3% of which are
144 public. In the urban area of Porto city there are 161 nurseries of which 32.3% are public (PORDATA,
145 2013). This study was carried out on three different nurseries (N_URB1, N_URB2 and N_URB3), all
146 located at urban sites influenced by traffic emissions in Porto city, Portugal (Figure 1). N_URB1 and
147 N_URB2 buildings were located in the same traffic busy street and their front facades were directly
148 facing this street. N_URB3 building was located in the same area, although its front facade was not facing
149 directly the street. These three nurseries had different management models: i) N_URB1 was a full private
150 for-profit nursery; ii) N_URB2 was managed by a private institution of social solidarity, non-profit and
151 with a mix of public and private funds; and iii) N_URB3 was a public nursery, entirely managed with
152 public funds by the municipality authorities and the Ministry of Education.

153 N_URB1 nursery had children until 5 years old, separated by age into 6 different classrooms, divided into
154 3 floors. During the period of measurements, the use of oil and/or electric heaters or air conditioners was
155 common to heat the rooms in this nursery. To prevent heat loss to the outside, windows were usually
156 closed, and the only natural ventilation in the rooms was done with the doors opened to the inside
157 corridors. Very young children, infants (< 1 year old) and toddlers (1-3 years old) spent all the period in
158 this nursery inside the same classroom, including sleeping and eating. On the opposite, older children (3-5
159 years old) used to have different daily patterns in this nursery, especially because they went to the lunch
160 room to eat. There was a small outdoor playground, although rarely used during the measurements
161 campaign.

162 N_URB2 nursery also had children until 5 years old, divided by age into 7 different classrooms. Although
163 the building had 2 floors, all the nursery rooms were located in the ground floor. All the classrooms had
164 direct access to small outdoor playgrounds and the access doors were usually opened. N_URB2 building
165 had no air conditioning system and the electric heaters were rarely used during the periods of
166 measurements.

167 N_URB3 nursery had children from 3 to 5 years old (pre-school children), mixed in 4 different
168 classrooms, all located in the ground floor, although it was a building with 2 floors. There were also
169 classrooms in the first floor, but they were occasionally used.

170 All the nurseries had a lunch room in the ground floor with a kitchen using gas stoves, except for
171 N_URB3 where there were no cooking activities, as the food were brought to the nursery already cooked.

172 Cleaning activities' patterns were also different in all the three studied nurseries. In N_URB1 younger
173 children classrooms (< 3 years old), daily cleaning activities were made during the sleeping time (after
174 lunch), with children sleeping in the classroom; in the other classrooms, cleaning used to be made during
175 lunch time (when children are not in the classroom) or at the end of the afternoon, after classes. On the
176 opposite, daily cleaning activities in N_URB2 in all the building were made after the occupancy period,
177 and the deep cleaning was made on weekends. In N_URB3 almost the same happened, except that some
178 daily cleaning in corridors and common spaces were made during occupancy and the deep cleaning was
179 made on weekdays after the occupancy period.

180 Measurements were performed in 4 classrooms in nursery N_URB1, 3 classrooms in nursery N_URB2,
181 and 2 classrooms in nursery N_URB3, as well as in the lunch rooms of all nurseries. Table 1 summarizes
182 some of the main important characteristics for indoor air quality in each studied room.

183 Indoor concentrations of the different fractions of PM (PM_1 , $PM_{2.5}$, PM_{10} and PM_{Total}) were continuously
184 measured using a TSI DustTrak DRX 8534 particle monitor using light-scattering laser method. The
185 minimum and maximum limit detections for this equipment are, respectively, 0.001 mg m^{-3} and 150 mg
186 m^{-3} . The equipment was submitted to a standard zero calibration (available in the equipment) and data
187 were validated prior to each new measurement (in each new room). Indoor measurements were performed
188 from 2 to 9 days in each considered room, and, in some cases, both in weekdays and weekends, between
189 February and June 2013. Hourly averages were calculated from a set of four measurements per hour (each
190 15 minutes) per day of measurement.

191 Simultaneously, hourly PM_{10} concentrations were obtained from the nearest air quality station classified
192 as urban traffic. These measurements were conducted by the Air Quality Monitoring Network of Porto
193 Metropolitan Area, managed by the Regional Commission of Coordination and Development of Northern
194 Portugal (*Comissão de Coordenação e Desenvolvimento Regional do Norte*) under the responsibility of
195 the Ministry of Environment.

196

197 3. Results

198 3.1 PM concentrations

199 As previously stated and according to Table 1, samplings were performed for more than one day in each
200 studied room of the three nurseries and hourly averages were calculated. Figure 2 shows as an example
201 (a) PM_{total} measured during five days on weekdays at N_URB1 and (b) $PM_{2.5}$ measured on weekend at
202 N_URB2. Assuming that there are no significant differences on indoor air pollution between different
203 weekdays, and as the daily patterns during the different sampling weekdays in each room were very
204 similar, average daily weekdays profiles were performed to represent an average indoor air quality
205 scenario. The same was performed for the weekends.

206 Figures 3 to 7 show the average daily profiles of PM_1 , $PM_{2.5}$, PM_{10} and PM_{Total} , respectively (a) to (d), for
207 N_URB1 and N_URB2 during weekdays and weekends (respectively Figures 3 to 6) and N_URB3
208 during weekdays and weekends (Figure 7). Table 2 summarizes the statistical parameters (minimum,
209 maximum, mean and median) of the hourly means for each room studied in the three nurseries.

210 Figures 3, 5 and 7 showed that PM concentrations in the classrooms started to rise up at the beginning of
211 the occupancy period and started decreasing after the end of the occupancy period (time variable,
212 depending on the room). Figures 4, 6 and 7 showed that the concentrations during weekends and non-
213 occupancy periods did not seem to have high fluctuations neither peaks, thus being considered
214 background concentrations for each respective room. The highest PM_1 and $PM_{2.5}$ concentrations were
215 registered in N_URB2 (classroom C), while the highest PM_{10} and PM_{Total} concentrations were found in
216 N_URB1 (classroom C). The minimum concentrations of all PM fractions were observed in the LR of
217 N_URB1. Likewise, the minima concentrations in N_URB3 were observed in the LR; nevertheless, in
218 N_URB2, the LR had higher concentrations than the other measured rooms. Minima concentrations were
219 always found during weekends or periods of non-occupancy and maxima concentrations were always
220 registered during occupancy periods, as can be observed on Figures 3 to 7. Table 2 showed that median
221 values were very close to mean values, so there was not great scattering in the measurements in each
222 room. The only exception was registered in PM_{Total} , in which mean concentrations were in general higher
223 than median values.

224

225 3.2 PM size distribution

226 PM size ratios allowed to understand the size distribution on the PM measured concentrations. Three
227 different ratios were used here: i) $PM_1/PM_{2.5}$; ii) $PM_{2.5}/PM_{10}$; and iii) PM_{10}/PM_{Total} . These ratios were
228 calculated per microenvironment (room) and per nursery, with the calculated hourly mean concentrations,
229 in three different conditions: (i) occupancy; (ii) non-occupancy (according to data on Table 1); and (iii)
230 weekends (when applicable). These ratio results are represented in Table 3.

231 In N_URB1 during occupancy on weekdays, $PM_1/PM_{2.5}$ ratio varied from 0.91 to 0.95, $PM_{2.5}/PM_{10}$ ratio
232 from 0.50 to 0.75, and PM_{10}/PM_{Total} ratio from 0.42 to 0.59. During non-occupancy periods on weekdays,
233 $PM_1/PM_{2.5}$ ratio varied from 0.94 to 0.98, $PM_{2.5}/PM_{10}$ ratio from 0.95 to 0.97, and PM_{10}/PM_{Total} ratio from
234 0.97 to 0.99. On weekends, $PM_1/PM_{2.5}$ ratio varied from 0.87 to 0.98, $PM_{2.5}/PM_{10}$ ratio from 0.88 to 0.98,
235 and PM_{10}/PM_{Total} ratio from 0.95 to 1.

236 On weekdays during occupancy in N_URB2, $PM_1/PM_{2.5}$ ratio varied from 0.94 to 0.97, $PM_{2.5}/PM_{10}$ ratio
237 varied from 0.60 to 0.76, and PM_{10}/PM_{Total} ratio varied from 0.41 to 0.61. During non-occupancy periods
238 on weekdays, $PM_1/PM_{2.5}$ ratio varied from 0.96 to 0.98, $PM_{2.5}/PM_{10}$ ratio from 0.93 to 0.95, and
239 PM_{10}/PM_{Total} ratio from 0.97 to 0.98. On weekends, ratios were very close to 1 ($PM_1/PM_{2.5}$ and
240 $PM_{2.5}/PM_{10}$ ratios were 0.97, and PM_{10}/PM_{Total} ratio was 0.99).

241 In N_URB3 on weekdays during occupancy, $PM_1/PM_{2.5}$ ratio varied from 0.95 to 0.98, $PM_{2.5}/PM_{10}$ ratio
242 from 0.64 to 0.89, and PM_{10}/PM_{Total} ratio from 0.50 to 0.89. During non-occupancy periods on weekdays,
243 $PM_1/PM_{2.5}$ ratio varied from 0.95 to 0.99, $PM_{2.5}/PM_{10}$ ratio from 0.93 to 0.98, and PM_{10}/PM_{Total} ratio from
244 0.97 to 0.99. On weekends, ratios were also very close to 1 ($PM_1/PM_{2.5}$ and PM_{10}/PM_{Total} ratios were 0.99,
245 and $PM_{2.5}/PM_{10}$ ratio was 0.97).

246

247 3.3 Comparison with standards and guidelines

248 PM concentrations were compared with WHO guidelines and with the Portuguese legislation (*Decreto-*
249 *Lei n° 79/2006*). Table 4 summarizes the exceedances per room and per nursery to the WHO guidelines,
250 as no exceedances were observed to the Portuguese standards.

251 In nursery N_URB1, the worst scenario was found in classroom C, where the WHO guidelines were
252 exceeded 80% and 40% of times, for $PM_{2.5}$ and PM_{10} respectively. On the opposite, in classroom A the
253 WHO guidelines were not exceeded. In N_URB2, it was possible to found the worst scenario in LR,
254 where the WHO guideline for $PM_{2.5}$ was always exceeded and for PM_{10} was exceeded half of the times. It
255 is also important to point out that in classroom C the WHO guidelines were exceeded 40% and 20% of
256 times for $PM_{2.5}$ and PM_{10} , respectively. Lastly, in the case of N_URB3, WHO guideline for $PM_{2.5}$ was
257 mostly exceeded (60%, 50% and 100% of times respectively in rooms A, B and LR). On the other hand,
258 the WHO guideline for PM_{10} was never exceeded in this nursery.

259

260 3.4 Indoor/Outdoor ratios

261 Collected outdoor PM_{10} concentrations allowed obtaining an average daily profile of PM_{10} , represented in
262 Figure 8. It was possible to observe an increase throughout the morning, a decrease in the early afternoon
263 (12h-14h), and an increase throughout the rest of afternoon and evening, decreasing along the dawn.
264 According to the obtained results, PM_{10} concentration profiles were found similar on weekdays and
265 weekends.

266 Indoor measured concentrations were compared with outdoors using the indoor/outdoor ratio (I/O ratio).
267 Mean I/O ratios were obtained for each studied room in the three nurseries (Table 5). Generically, I/O
268 mean ratios were always higher than 1. On a closer look, in N_URB1, the highest I/O mean ratio was

269 found in LR and the lowest was found in classroom A, both for weekdays and weekends. Unfortunately,
270 there was not enough outdoor data available to determine I/O ratio for classroom C (considering the
271 period of measurement in this classroom). In N_URB2, it was possible to find the highest I/O ratio of all
272 the studied nurseries on weekdays (in classroom C). It is also important to point out the high I/O ratio
273 observed in classroom A. In N_URB3, the worst scenario was found in LR. On weekends, I/O mean
274 ratios were never higher 2.65 (N_URB3, classroom A).

275 4. Discussion

276 In nursery N_URB1, classroom C had the highest PM concentrations which could have been the result of
277 the cumulative effect of three major conditions: i) poor ventilation (there were no open direct access to
278 the outdoor and the door to the inner corridor was almost always closed); ii) high occupancy, with a total
279 of 25 persons, despite being the room with the higher volume; and iii) intense activity, characteristic of 5
280 years old children. Additionally, it was possible to notice three peaks in the PM profiles for all the studied
281 classrooms, which represented the three main occupancy periods (morning and afternoon before and after
282 the break). In nursery N_URB1, classroom B revealed the lower PM concentrations during occupancy,
283 most probably due to the lower occupancy on this classroom (only 7 people) when comparing to the
284 others. The lower concentrations observed in the LR on this nursery were possibly due to its size and the
285 existence of a small hall that creates a discontinuity between the kitchen and the lunch room, which
286 possibly diminishes kitchen PM penetration into the lunch room. On weekends the concentrations were
287 lower than on weekdays, and the behaviour for the different rooms was similar, with the exception of
288 classroom C where they were higher on the first hours of the day. As this was clearly the room with the
289 highest concentrations during weekdays, this was the result of the decrease of PM concentrations in the
290 beginning of the weekend (Saturday dawn) – settlement phenomenon.

291 In nursery N_URB2, LR showed the highest PM concentrations for the finer fractions (PM_1 and $PM_{2.5}$)
292 during the occupancy period and during the dawn and morning. Cooking activities are also one of the
293 major indoor sources of PM (Monn, 2001) and might explain the higher concentrations observed as these
294 activities started very early in the morning (8h) and ended late at the afternoon (19h). In this nursery it
295 was also possible to observe that classroom C had the maximum PM concentrations (peaks) in all
296 fractions, but especially higher for PM_{Total} , which can be attributed to three major synergetic factors: i)
297 a higher occupancy in this classroom when compared with others in this nursery with similar areas (Table
298 1); ii) poor ventilation (doors to outdoor were always closed and to the inner corridor were almost always
299 closed); and iii) normal activities characteristic of 4 years old children (occupants of this classroom). Also
300 in classrooms C and B in this nursery, it was possible to observe the three peaks in the concentrations on
301 weekdays, also in the three main occupancy periods (morning and afternoon before and after the break),
302 and for the same reasons than in N_URB1. On the other hand, classroom A (baby nursery) showed a
303 different pattern, with the highest concentrations being registered between 13-15h. This was the period of
304 sleeping for the babies in the cribs room (next to and opened to classroom A) and teachers took the
305 chance to do some tidying. On weekends, PM concentrations were lower and the profiles were similar
306 and almost constant for the two measured classrooms (A and C).

307 In nursery N_URB3, PM concentrations in classroom A had a typical behaviour throughout the
308 weekdays, with clear peaks matching the occupancy periods. On the opposite, classroom B had a peculiar
309 PM profile, due to its occupancy (a wide space that was only used late at the afternoon, from 16 to 19h).
310 In the lunch room of this nursery, the PM concentrations profile was slightly different from the other two
311 lunch rooms (in N_URB1 and N_URB2). As there were no cooking activities in the kitchen attached to
312 the lunch room and the cleaning activities were made immediately after lunch time, PM concentrations
313 were lower and the maximum was observed after the lunch time (early afternoon). On weekends,
314 concentrations were found much lower, and there was an expected almost constant PM behaviour during
315 this period.

316 There was occasionally an increase of PM concentrations at the end of the afternoon, which was kept
317 even after the end of classroom occupancy, mainly due to cleaning activities. Fromme et al. (2005) also

318 reported that cleaning activities could contribute to the increase of PM in the indoor air. To minimize this
319 contribution, cleaning activities in nurseries should be performed when children go home and with high
320 ventilation rates to outdoor.

321 $PM_1/PM_{2.5}$ ratios were, in all situations, equal or higher than 0.90, i.e., very close to 1, meaning that the
322 majority of the $PM_{2.5}$ was less than $1 \mu m$. On weekends and non-occupancy periods, PM concentrations
323 were mainly due to the finer fraction, with $PM_{2.5}/PM_{10}$ ratios close to 1, on the opposite to the periods of
324 occupancy when $PM_{2.5}/PM_{10}$ (as well as PM_{10}/PM_{Total}) ratios were in average half of those in weekends
325 and non-occupancy periods.

326 Overall, PM concentrations on nurseries were found to be much higher during occupancy periods than
327 during non-occupancy periods and weekends and almost constant on the latter ones, which was consistent
328 with the presence of children and their activities, even in lunch rooms. However, PM_{10} mean levels in all
329 studied rooms were below mean level obtained by Yang et al. (2009) in Korean nurseries ($94.94 \mu g m^{-3}$).
330 This means that the presence of children and their activities in nurseries' microenvironments potentiated,
331 in general, the suspension and/or re-suspension phenomena of PM indoors, mainly coarser fractions,
332 which was also found by Parker et al. (2008) for school buildings. In general, occupancy increases PM
333 concentrations indoors (Sousa et al., 2012b).

334 The PM concentrations found in all the studied nurseries were high, often above WHO guidelines, which
335 is concerning, especially for the finer fractions. Those were often found in the classrooms of older
336 children (4-5 years old). These have greater freedom and ability to move when compared with younger
337 ones, which is reflected in their usual daily activities on nurseries increasing PM concentrations in indoor
338 air, as reported by Fromme et al. (2005). Lunch rooms also exceeded WHO guidelines, especially in
339 N_URB2 and N_URB3, mainly due to cooking activities and children movements. Of concern were also
340 the exceedances in 50% of the measurement days to WHO $PM_{2.5}$ guideline in N_URB2 classroom 1,
341 which is a baby nursery, and these younger children are most vulnerable to adverse health effects of PM
342 suspended in the air.

343 I/O ratios were always higher than 1, meaning that PM_{10} indoor concentrations were, in average, higher
344 than ambient levels, which is consistent with the findings from Yoon et al. (2011) in urban preschools in
345 Korea and from Almeida et al. (2011) in Portuguese primary schools. On weekdays, indoor
346 concentrations were always at least 2 times higher than those found outdoors. Even on weekends indoor
347 concentrations were found to be until 2.65 times (in average) higher than those found outdoors. This
348 suggested that outdoor influence on PM indoor concentrations was not significant when compared with
349 indoor sources and re-suspension phenomena. In fact, the highest I/O ratios in N_URB1 and N_URB3
350 were found in lunch rooms, which is consistent with indoor sources already stated (cooking activities and
351 children drives). The higher I/O ratio found in classroom C in N_URB2, as well as the high ratio found in
352 classroom A in the same nursery, were also due to indoor sources and poor ventilation to outdoors. In
353 fact, poor ventilation to the outdoor turned indoor sources as the major increasing factor of indoor PM
354 concentrations, which was also stated by Yang et al. (2009).

355

356 5. Conclusions

357 PM concentrations were often found high in the studied classrooms, mainly in the finer fractions (PM_1
358 and $PM_{2.5}$), and often above the limits recommended by WHO, which is concerning in terms of exposure
359 effects on children's health. The classrooms occupied by older children were found to be those with the
360 highest PM concentrations, due to their higher mobility when compared with younger ones, thus
361 increasing PM re-suspension. Results allowed concluding that indoor sources were clearly the main
362 contributors to indoor PM concentrations when compared with outdoor influence. Due to that, the poor
363 ventilation to outdoors in classrooms affected indoor air quality by increasing the PM accumulation.
364 Results also confirmed that cleaning activities increased PM concentrations in indoor air and suggested

365 that cooking activities could increase PM concentrations in lunch rooms. To improve the air renovation
366 rate (higher and better ventilation), as well as to do the cleaning activities after the occupancy period
367 could be good practices to reduce PM indoor air concentrations in nurseries and, consequently, improve
368 children's health and welfare.

369 For the future, it could be important to study other nurseries to help supporting these findings, not only in
370 urban traffic influence context, but also in other contexts, like urban background and rural. In next studies
371 it could be important to determine the particulate matter composition (heavy metals, PAH's).
372 Measurements of the air flow rates could also be important to refine the analysis on the occupancy and air
373 renovation rates. It could also be important to study the association of PM air pollution in these nurseries
374 with children's daily exposure. Further investigations at home and in other microenvironments occupied
375 by children are needed to understand if there is, or not, an increased risk of adverse health effects on
376 children attending nurseries when compared with those cared at home.

377

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467

468 **Figure captions**

469 Figure 1. Location of the three studied urban nurseries in Porto city: a) N_URB1, b) N_URB3 and c)
470 N_URB2.

471 Figure 2. Distribution of PM hourly average concentrations of a) N_URB1 Room A weekdays, and b)
472 N_URB2 Room A weekend.

473 Figure 3 – PM average concentrations on weekdays in N_URB1: a) PM_1 , b) $PM_{2.5}$, c) PM_{10} and d)
474 PM_{Total} .

475 Figure 4 – PM average concentrations on weekends in N_URB1: a) PM_1 , b) $PM_{2.5}$, c) PM_{10} and d)
476 PM_{Total} .

477 Figure 5 – PM average concentrations on weekdays in N_URB2: a) PM_1 , b) $PM_{2.5}$, c) PM_{10} and d)
478 PM_{Total} .

479 Figure 6 – PM average concentrations on weekends in N_URB2: a) PM_1 , b) $PM_{2.5}$, c) PM_{10} and d)
480 PM_{Total} .

481 Figure 7 – PM average concentrations in N_URB3: a) PM_1 , b) $PM_{2.5}$, c) PM_{10} and d) PM_{Total} .

482 Figure 8 – Distribution of PM_{10} outdoor hourly average concentrations in weekdays and weekend.

Table 1 – Summary of the main characteristics for indoor air quality analysis in each studied microenvironment.

Nursery	Room	Type of use	Children's age (years)	Floor	Volume (m ³)	Occupancy	Period of occupation	Ventilation	Sampling time (weekdays + weekend days)
N_URB1	A	Classroom	1	Ground floor (back)	115	19	07h30 – 19h30	Windows to outdoor closed. Door to inner corridor almost always closed. A/C on.	5 + 2
	B	Classroom	3	1 st floor (front)	63	7	09h – 11h30 15h – 15h30	Windows to outdoor closed. Door to inner corridor almost always closed. No A/C. Electric/oil heater on.	3 + 2
	C	Classroom	5	2 nd floor (front)	176	25	08h – 11h30 15h30 – 17h30	Windows to outdoor closed. Door to inner corridor almost always closed. No A/C. Electric/oil heater on.	3 + 2
	LR	Lunch room	3-5	Ground floor (back)	115	21-74	11h30 – 13h30	Open to kitchen and to inner corridor. No direct connection to outdoor.	7 + 2
N_URB2	A	Classroom	<1	Ground floor (front)	51	12	09h – 12h00 15h30 – 18h	Windows directly to outdoor (traffic street) closed – opened only after occupancy. Door to inner corridor always open. Open passage to cribs room and a small lunch room.	2 + 2
	B	Classroom	2	Ground floor	120	20	09h30-12h 14h-16h30	Door to inner corridor almost always closed. Direct access to outdoor playground often opened. No A/C and heating off.	3 + 0
	C	Classroom	4	Ground floor (back)	151	27	09h30-12h 14h-16h30	Door to inner corridor almost always opened. Direct access to outdoor playground often closed. No A/C and heating off.	3 + 2
	LR	Lunch room	1-5	Ground floor (back)		17-68	11h – 12h30	Open to kitchen, to inner corridor, and to outdoor (during occupancy).	2 + 0
N_URB3	A	Classroom	3-5	Ground floor	133,5	23	09h – 11h30 13h30 – 16h	Door to inner corridor often closed. Passage to outdoor playground usually opened. No A/C and heater.	3 + 2
	B	Classroom	3-5	1 st floor	108	35	16h – 19h	Door to inner corridor often opened. Window to outdoor open during occupancy. No A/C and heater.	2 + 0
	LR	Lunch room	3-5	Ground floor	168	17-45	11h30 – 13h30	Open to inner corridor and kitchen. Windows to outdoor closed.	2 + 0

Table 2 – Statistical parameters of the hourly mean data for each room studied in all the three nurseries (values in $\mu\text{g m}^{-3}$).

PM	Nursery Room	N_URB1				N_URB2				N_URB3		
		A	B	C	LR	A	B	C	LR	A	B	LR
PM ₁	Min	8.60	7.25	6.67	2.75	13.75	4.00	7.00	16.25	13.00	11.75	7.00
	Max	46.29	45.25	120.25	70.25	74.25	54.75	145.00	125.25	71.25	62.00	82.00
	Mean	18.38	21.97	33.08	16.79	27.84	19.95	25.42	47.85	27.84	32.29	26.74
	Median	15.42	19.38	29.25	14.00	23.13	21.00	16.63	42.50	24.75	33.25	19.25
PM _{2.5}	Min	8.95	8.00	8.00	3.25	14.00	4.25	7.00	17.00	13.25	12.00	7.25
	Max	47.77	46.00	135.75	74.25	77.75	58.75	158.00	126.75	74.75	62.75	86.50
	Mean	19.70	22.75	34.69	18.17	28.69	21.09	26.65	48.94	28.50	32.63	28.01
	Median	17.04	20.00	30.00	15.25	23.75	21.50	17.38	43.25	25.00	33.25	20.75
PM ₁₀	Min	9.42	8.00	10.00	3.25	14.75	5.00	7.00	19.25	14.00	13.25	7.75
	Max	71.72	71.00	318.00	84.00	129.50	104.50	197.25	139.00	134.50	73.50	166.00
	Mean	26.11	25.56	50.94	22.31	34.82	31.62	28.88	56.77	34.15	34.86	40.15
	Median	19.53	21.75	32.00	17.29	24.25	23.63	18.13	46.75	26.00	35.00	23.00
PM _{Total}	Min	9.42	8.00	10.33	3.25	15.00	5.00	7.25	19.75	14.00	14.25	8.00
	Max	208.34	190.50	605.00	202.00	368.75	248.25	427.25	224.75	336.00	86.00	401.25
	Mean	48.89	32.61	85.81	32.97	63.74	66.09	40.18	77.69	50.55	37.04	70.55
	Median	20.23	23.00	32.50	18.88	25.38	23.63	19.50	55.50	26.25	36.25	23.25

Table 3 - PM size ratios in each studied microenvironment: average values according to the occupancy patterns.

Nursery	Room	Week						Weekend		
		During occupancy			During non-occupancy			PM ₁ /PM _{2.5}	PM _{2.5} /PM ₁₀	PM ₁₀ /PM _{Total}
		PM ₁ /PM _{2.5}	PM _{2.5} /PM ₁₀	PM ₁₀ /PM _{Total}	PM ₁ /PM _{2.5}	PM _{2.5} /PM ₁₀	PM ₁₀ /PM _{Total}			
N_URB1	A	0.93	0.63	0.42	0.96	0.95	0.97	0.90	0.94	0.99
	B	0.95	0.70	0.50	0.98	0.97	0.99	0.98	0.98	1.00
	C	0.91	0.50	0.51	0.98	0.95	0.98	0.98	0.96	0.98
	LR	0.92	0.75	0.59	0.94	0.95	0.98	0.87	0.88	0.95
N_URB2	A	0.96	0.69	0.41	0.97	0.95	0.98	0.97	0.97	0.99
	B	0.94	0.61	0.45	0.96	0.93	0.97	-	-	-
	C	0.94	0.60	0.49	0.97	0.94	0.98	0.97	0.97	0.99
	LR	0.97	0.76	0.61	0.98	0.95	0.97	-	-	-
N_URB3	A	0.95	0.64	0.50	0.97	0.97	0.97	0.99	0.97	0.99
	B	0.98	0.89	0.89	0.99	0.98	0.99	-	-	-
	LR	0.96	0.64	0.54	0.95	0.93	0.98	-	-	-

Table 4 – Exceedances of 24-hour mean PM concentrations to the WHO guidelines ($PM_{2.5}$ - $25 \mu g m^{-3}$ and PM_{10} - $25 \mu g m^{-3}$).

Nursery	Room	24h exceedances (%)	
		WHO ($PM_{2.5}$)	WHO (PM_{10})
N_URB1	A	0	0
	B	40	0
	C	80	40
	LR	11	0
N_URB2	A	50	0
	B	33	0
	C	40	20
	LR	100	50
N_URB3	A	60	0
	B	50	0
	LR	100	0

Table 5 – PM₁₀ I/O ratios: mean values observed in each studied site for weekdays and weekends, and respective minima (min) and maxima (max) values.

Nursery	Room	Weekday	Weekend
N_URB1	A	2.17 (min-max: 0.46-18.32)	1.06 (min-max: 0.34-9.42)
	B	2.23 (min-max: 0.42-12.75)	1.35 (min-max: 0.55-3.80)
	C	*	*
	LR	3.05 (min-max: 0.41-37.50)	1.54 (min-max: 0.35-11.50)
N_URB2	A	5.31 (min-max: 0.56-129.50)	2.02 (min-max: 0.40-20.00)
	B	1.96 (min-max: 0.23-11.00)	-
	C	13.96 (min-max: 0.57-213.63)	2.02 (min-max: 0.39-7.00)
	LR	2.41 (min-max: 0.60-9.35)	-
N_URB3	A	2.67 (min-max: 0.48-10.44)	2.65 (min-max: 0.83-15.00)
	B	2.12 (min-max: 0.42-21.00)	-
	LR	4.57 (min-max: 0.43-25.44)	-

* For room C in N_URB1 nursery, outdoor PM₁₀ concentrations data available were only for less than 50% of the study period, which was not statistically relevant.



Figure 1.

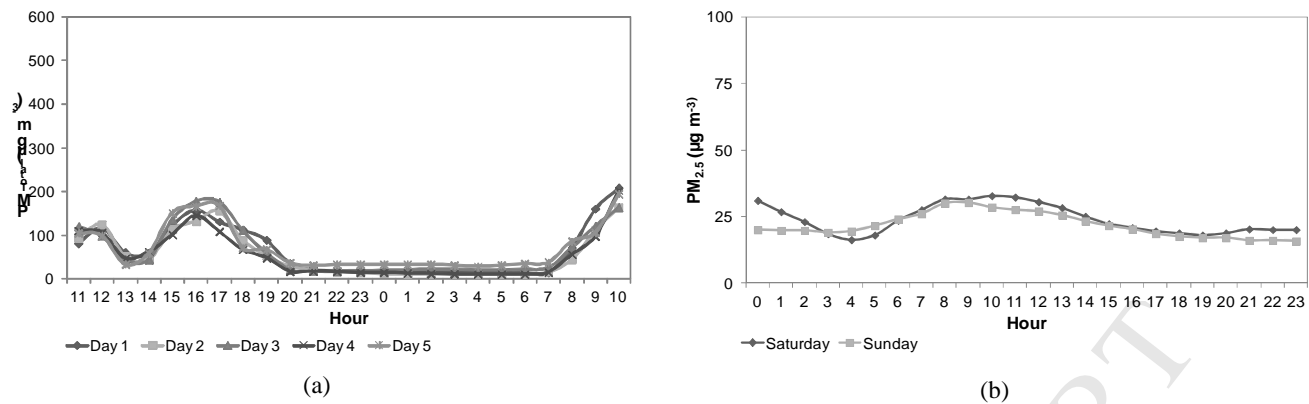


Figure 2.

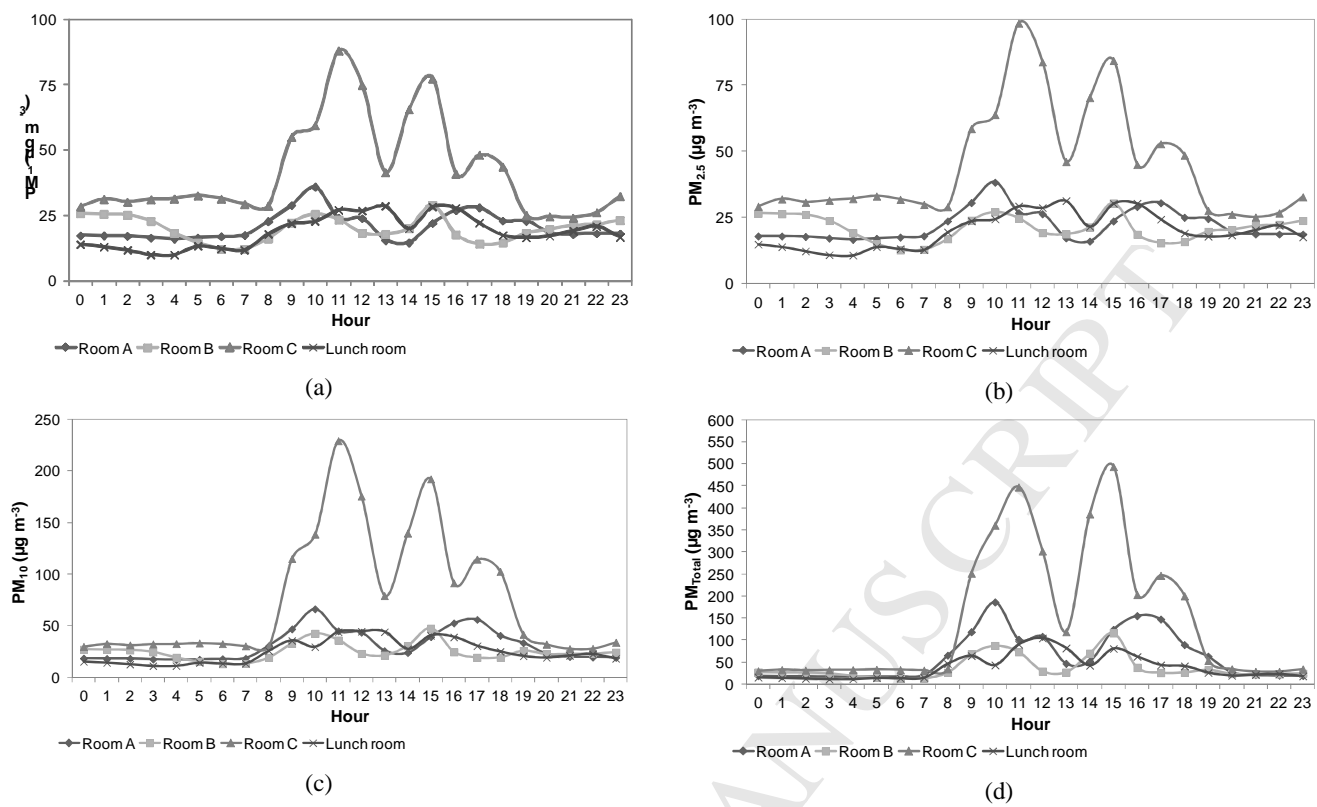


Figure 3.

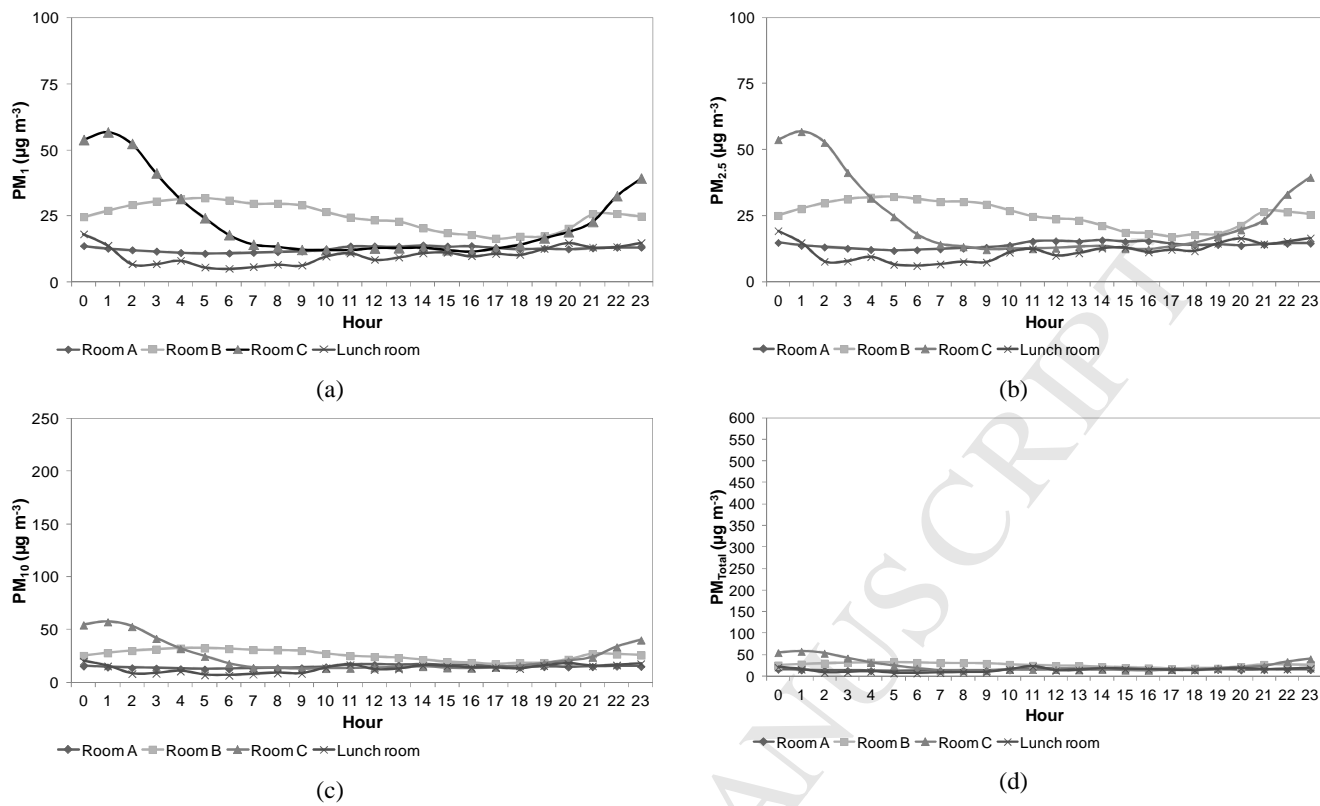


Figure 4.

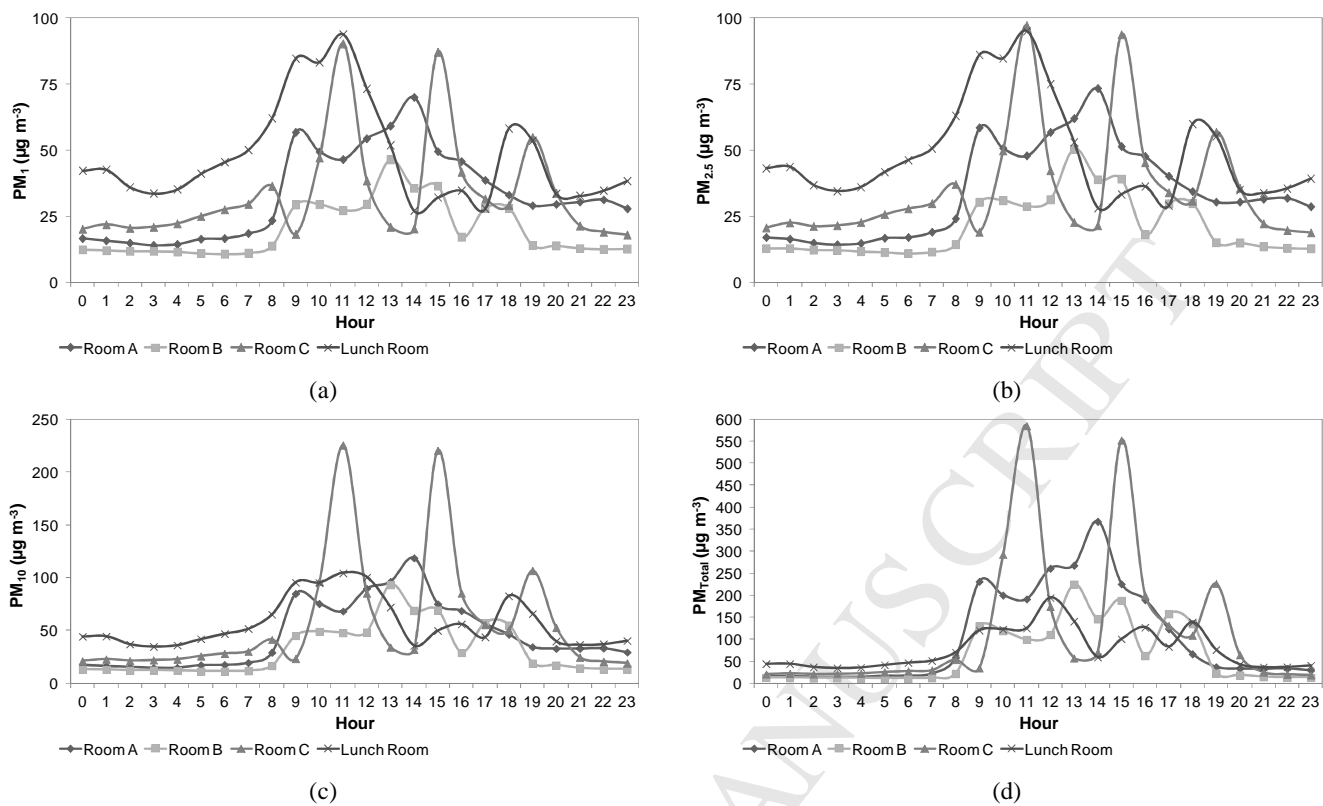


Figure 5.

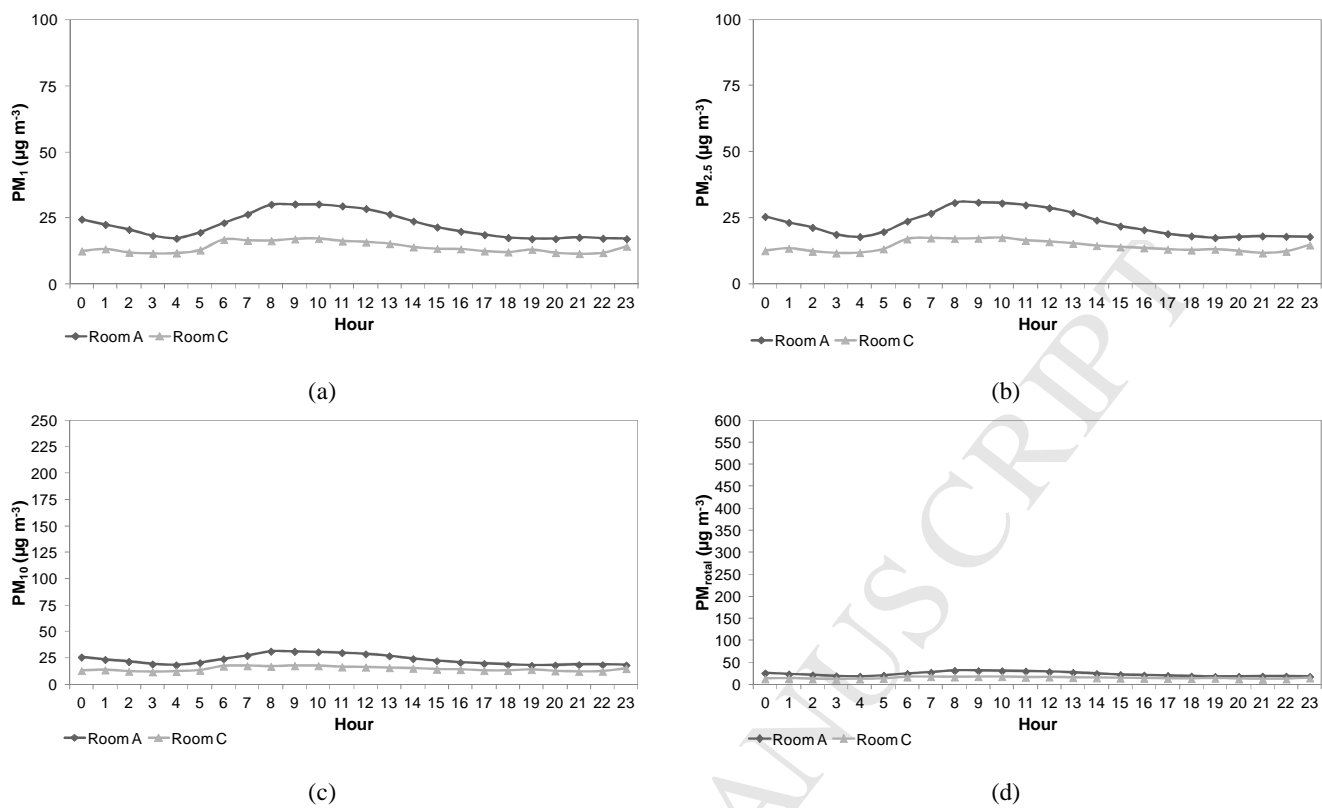


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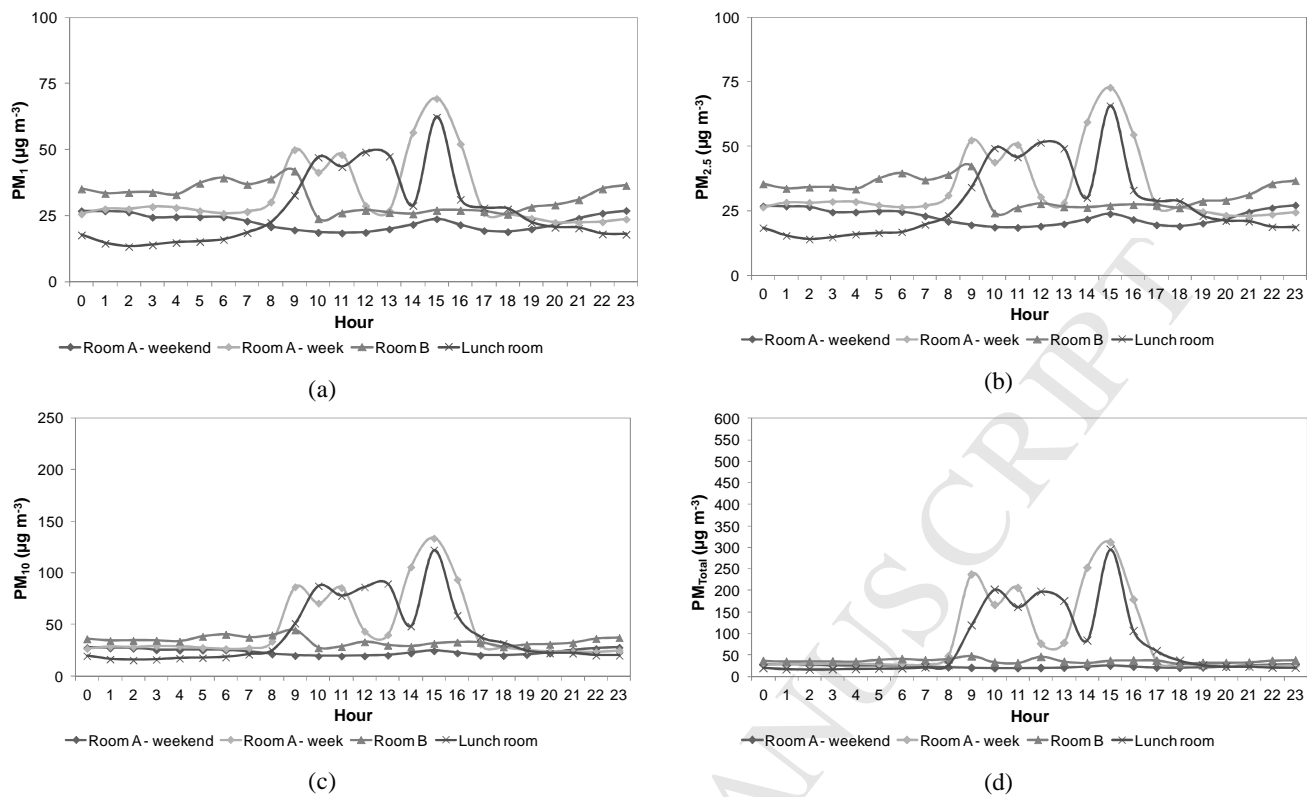


Figure 7.

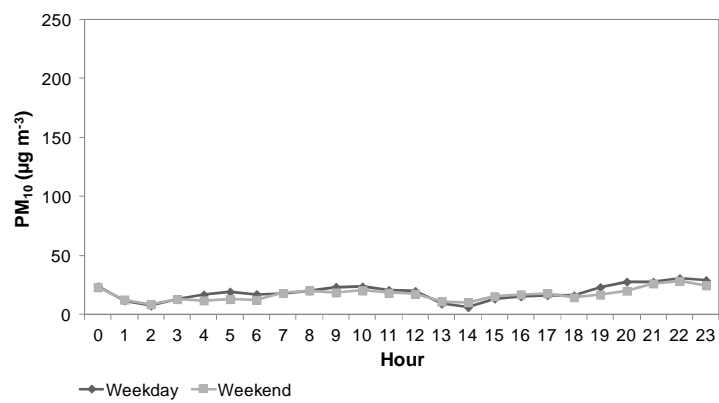


Figure 8.



Figure 1.

Highlights:

- PM concentrations were often found high in the studied classrooms
- Indoor sources were clearly the main contributors to indoor PM
- Poor ventilation to outdoors affected IAQ by increasing the PM accumulation