- 1 Gaseous pollutants on rural and urban nursery schools in Northern Portugal
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#### 14 Abstract

15 Indoor air quality in nursery schools is different from other schools and this has been 16 largely ignored, particularly in rural areas. Urban and rural nursery schools have different environmental characteristics whose knowledge needs improvement. Thus, this study 17 aimed to evaluate continuously the concentrations of CO<sub>2</sub>, CO, NO<sub>2</sub>, O<sub>3</sub>, CH<sub>2</sub>O and total 18 VOC in three rural nursery schools and one urban, being the only one comparing urban 19 and rural nurseries with continuous measurements, thus considering occupation and non-20 occupation periods. Regarding CO<sub>2</sub>, urban nursery recorded higher concentrations (739-21 2328 mg m<sup>-3</sup>) than rural nurseries (653-1078 mg m<sup>-3</sup>). The influence of outdoor air was 22 23 the main source of CO, NO<sub>2</sub> and O<sub>3</sub> indoor concentrations. CO and NO<sub>2</sub> concentrations were higher in the urban nursery and O<sub>3</sub> concentrations were higher in rural ones. CH<sub>2</sub>O 24 25 and TVOC concentrations seemed to be related to internal sources, such as furniture and flooring finishing and cleaning products. 26

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Capsule: Gaseous pollutant levels were higher in the urban nursery than in rural ones,
except for O<sub>3</sub>. High concentrations were due to lack of ventilation, outdoor air and internal
sources.

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# 32 Keywords

33 Indoor air, nursery, children, rural, urban, gaseous

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#### 35 **1. Introduction**

In recent years, numerous scientific studies highlighted that citizens spend most of their 36 time in indoor environments (Jenkins et al., 1992; Silvers et al., 1994; Klepeis et al., 2001; 37 Schweizer et al., 2007; de Gennaro et al., 2014; Wu et al., 2015). The largest part of 38 human exposure to air pollution occurs in indoor environments, commonly considered 39 40 non-polluted such as homes, offices and schools (WHO 2006; de Gennaro et al., 2014; 41 Branco et al., 2014). Nevertheless, it is actually known that indoor air pollution has equal 42 or even greater impact on human health than outdoor pollution. This occurs because time spent indoor is usually higher than time spent outdoor; also, there is a great variety of 43 indoor sources, that include outdoor and specific indoor sources associated with 44 45 formaldehyde and volatile organic compounds (VOC) emissions, leading frequently to higher concentration than outdoor (Franklin 2007; Faustman et al. 2000; Sofuoglu et al. 46 2011). Furthermore, children are more vulnerable to air pollution exposure than adults, 47 48 being considered a risk group (Sousa et al., 2012). Exposure to indoor air pollution has been related to long and short-term health problems. Respiratory, cardiovascular and 49 50 central nervous systems are the most affected, leading also to adverse effects on children's 51 productivity and academic performance (Jones 1999; Wang et al., 2015; Annesi-Maesano et al., 2013, Mohai et al., 2011). 52

Indoor pollutant sources are related to structural conditions of buildings (interior finishes, coverings and furniture), occupants' activities (heating, cooling and cooking habits, metabolism, hygiene, cleaning and disinfection products) and outdoor pollution (Jones, 1999). The control and analysis of indoor air quality (IAQ) assume an extremely important role because indoor pollutants' concentrations may vary significantly with location and time (de Gennaro et al., 2014). Studies of IAQ in schools have been performed mainly in primary or secondary schools. Nevertheless, IAQ in nursery schools

is different from other schools and this has been largely ignored, particularly in rural areas 60 61 (Ashmore and Dimitroulopoulu, 2009). IAQ studies comparing urban and rural contexts are relevant because there are evident environmental and social differences. On the 62 environmental level this idea is supported essentially by the influence of traffic emissions. 63 On the social level, habits and life styles in these two contexts are significantly different. 64 65 Studies already made in nursery schools were essentially of three types: i) only focusing 66 on comfort parameters (Gladyszewska-Fiedoruk 2013), and/or on CO<sub>2</sub> concentration as global IAQ indicator (Theodosiou and Ordoumpozanis, 2008; Carreiro-Martins, 2014); 67 or ii) focusing on the study of one specific pollutant such as PM, allergens or phthalates 68 69 (Arbes, et al., 2005; Fromme, et al., 2013). As far as known there are only five studies focussing on various gaseous pollutants in nursery schools' indoor air, from which one 70 was in rural areas. Zuraimi and Tham (2008) investigated comfort parameters, air velocity 71 72 and air exchange rates, as well as concentrations of several pollutants in nursery schools 73 of Singapore, concluding that outdoor concentrations and occupant density were the main 74 determinants for CO<sub>2</sub> concentrations. For indoor CO and O<sub>3</sub> levels, outdoor concentrations were the main precursors. Yang et al. (2008) characterized the 75 concentrations of different indoor air pollutants in Korean nursery schools and compared 76 77 them according to age and characteristics of buildings. The main problems reported in that study were caused by chemicals emitted from building materials or furnishing, and 78 insufficient ventilation rates. Yoon et al. (2011) measured IAQ in rural and urban 79 80 preschools in Korea, by investigating the indoor air concentrations of PM and several chemical compounds, and they found evidences that pollutant concentrations were in 81 82 general higher in urban context and indoors than in rural context and outdoors. However, indoor/outdoor (I/O) ratios of  $CH_2O$ , CO and total volatile organic compounds (TVOC) 83 were higher in rural schools. 84

Cano et al. (2012) studied IAQ in nursery schools in Lisbon and Porto (Portugal) considering various chemical pollutants, comfort parameters and microbiological parameters. The results of that study demonstrated an association between CO<sub>2</sub> concentrations and the number of children present in classrooms, as well as the need to improve ventilation and comfort of the spaces to promote healthier indoor environments.

Despite considering a large number of nursery schools, gaseous compounds, comfort 90 91 parameters and comparisons between rural and urban nursery schools, in the above mentioned studies, samplings were only conducted during weekdays and during 92 93 occupation periods. That did not allow understanding differences in IAQ between occupation and non-occupation periods (including nights and weekends), which permit 94 95 to better understand sources of indoor air pollution, as well as the baseline room scenario. Additionally, some chemical compounds were measured by passive sampling and not 96 97 continuously. Moreover, some important compounds were missing as for example NO<sub>2</sub>, 98 which is an important traffic marker.

99 Following Nunes et al. (2015) study that focused on the PM assessment, and in the scope of INAIRCHILD project (Sousa et al., 2012), the present study is the only one comparing 100 101 urban and rural nurseries with continuous measurements considering the comparison of 102 occupation and non-occupation periods, thus aiming to reduce the above referred gaps. 103 Therefore, the continuous evaluation of the indoor concentrations of  $CO_2$ , CO, ozone 104 (O<sub>3</sub>), NO<sub>2</sub>, TVOC and formaldehyde (CH<sub>2</sub>O) on different indoor microenvironments, 105 namely classrooms and lunch rooms was performed. Furthermore, gaseous concentrations were compared with Portuguese legislation and WHO guidelines for IAQ 106 107 and children's health.

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#### 109 2. Materials and methods

IAQ measurements were made in three different rural nursery schools (RUR1, RUR2 and
RUR3) located in *Bragança* district and without significant influence of traffic emissions

and in one urban nursery (URB) located in *Porto* and influenced by traffic emissions.

113 Table 1 shows a general description of each studied microenvironment of RUR1, RUR2,

114 RUR3 and URB, namely regarding the type of use, children's age, building floor, area,

number of occupants, period of occupation, ventilation routines and sampling time.

116 Measurements were performed in two classrooms in RUR1 and RUR3, one classroom in RUR2, and three classrooms in URB, as well as in the lunch rooms of all nursery schools. 117 118 Indoor air gaseous compounds, namely CO<sub>2</sub>, CO, NO<sub>2</sub>, O<sub>3</sub>, CH<sub>2</sub>O and TVOC, were continuously measured (at least 24 h in each ME) using an Haz-Scanner IEMS Indoor 119 Environmental Monitoring Station (SKC Inc., USA), equipped with high sensitive 120 121 sensors using the following methods: i) CO<sub>2</sub> – nondispersive infrared (NDIR) detection; 122 ii) CO, O<sub>3</sub>, NO<sub>2</sub> and CH<sub>2</sub>O – electrochemical detection; iii) VOC – photoionization detection (PID). All concentrations were converted from ppb and ppm (units from data 123 log), to ease the comparisons with legislation and guidelines using conversion factors 124 (Nota Técnica NT-SCE-02 2009; Tiwary and Colls, 2010). These conversion factors are 125 normalized to 293 K and 101.3 kPa, therefore concentrations were corrected for 126 127 temperature, using the values measured, and admitting an atmospheric pressure of 1 atm.

The equipment was submitted to a standard zero calibration (available in the equipment) and data were validated prior to each new measurement (in each new room). Indoor measurements were performed in each room studied, and, in some cases, both on weekdays and weekends, between April and June 2014. Measurements were recorded each minute and hourly means were calculated. In RUR1 measurements were made in full occupation and partial occupation for one of the classrooms and in the lunch room.
Full occupation concerned the usual period of nursery attendance and partial occupation
concerned to one week before the period of the Easter holidays, where the classroom's
occupation was reduced.

The indoor hourly mean values were compared with reference standards and guidelines 137 138 for general indoor environments, aiming to evaluate exceedances. Comparisons were 139 performed considering national and international reference values for general indoor environments, namely: i) Portuguese legislation (8 hour means) (Portaria n°353-A/2013) 140 for CO<sub>2</sub> (2250 mg m<sup>-3</sup>, plus 30% of margin of tolerance (MT) if no mechanical ventilation 141 system was working in the room), CO (10 000  $\mu$ g m<sup>-3</sup>), CH<sub>2</sub>O (100  $\mu$ g m<sup>-3</sup>), and TVOC 142 (600 µg m<sup>-3</sup>, plus 100% of MT if no mechanical ventilation system was working in the 143 room); ii) WHO guidelines (WHO, 2010) for CO (35000 µg m<sup>-3</sup> for hourly mean), NO<sub>2</sub> 144 (200  $\mu$ g m<sup>-3</sup> for hourly mean) and CH<sub>2</sub>O (100  $\mu$ g m<sup>-3</sup> for 30 minutes mean). For the 145 146 Portuguese legislation, 8-hour running means for all pollutants were calculated and the daily maximum was compared to the standard. 147

Hourly O<sub>3</sub> and NO<sub>2</sub> outdoor concentrations were obtained to calculate I/O ratios for rural 148 nursery schools in the subsequent days after indoor measurements and with the same 149 150 equipment used indoors; for the urban nursery outdoor concentrations were monitored at the nearest air quality station from the Air Quality Monitoring Network of the Porto 151 Metropolitan Area, managed by the Regional Commission of Coordination and 152 153 Development of Northern Portugal (Comissão de Coordenação e Desenvolvimento Regional do Norte) under the responsibility of the Ministry of Environment. This station 154 155 is classified as urban traffic and is representative of the urban area studied.

Mean, minimum, maximum, median and standard deviation (SD) values were calculated 156 157 for the hourly mean data of indoor air pollutants' concentrations in occupation and nonoccupation periods and weekend periods. Data was tested for normality using both the 158 159 Shapiro-Wilk and Anderson-Darling tests. Whenever measurements were performed for more than one day differences between mean hourly concentrations measured in different 160 161 sampling days were tested using t-test for normal distributions and Mann–Whitney's Utest for the other distributions. Differences between weekdays and weekends as well as 162 between rural and urban context were also studied using *t*-test for normal distributions 163 and Mann–Whitney U test for the other distributions. For all analyses a significance level 164 165 of 0.05 was considered. Descriptive statistics for the parameters were calculated using MS Excel<sup>®</sup> (Microsoft Corporation, USA), and other statistical analyses were computed 166 167 using R software, version 3.1.2 (R Foundation for Statistical Computing, 2014).

### 168 **3. Results and Discussion**

Tables 2, 3 and 4 summarize the main statistical parameters (minimum, maximum, mean, 169 170 median and standard deviation) of the hourly mean data for each room considering the entire sampling period for weekdays occupation periods, weekdays non-occupation 171 periods and weekends, respectively. Mean daily profiles were performed to represent 172 mean IAQ scenarios for weekdays and weekends. When comparing two or more 173 consecutive sampling days (weekdays and weekends), statistical differences regarding 174  $CO_2$  (p > 0.05 in all cases) were not found; however, for CO, NO<sub>2</sub>, O<sub>3</sub>, CH<sub>2</sub>O and TVOC 175 176 statistical differences (p < 0.05) were found in 50, 75, 50, 25 and 50% of the cases, respectively. Nevertheless, similarly to what was performed by Branco et al. (2015), a 177 178 daily mean scenario was assumed for all pollutants allowing the following analyses.

# 179 **3.1 Average daily profiles**

#### 180 **3.1.1 Ventilation indicator - CO**<sub>2</sub>

Average daily profiles of CO<sub>2</sub> for all the studied nursery schools are represented in Figure
1: a) RUR1, b) RUR2, c) RUR3 and d) URB.

183 Two peaks of CO<sub>2</sub> concentrations were observed in the classrooms: i) during morning – 184 rising in early morning and decreasing before lunch time; and ii) during afternoon - rising 185 after lunch time and decreasing until the end of the afternoon. In lunch rooms, three peaks 186 were observed corresponding to the breakfast, lunch and snack times in RUR1 and RUR2, 187 and two peaks corresponding to the lunch and snack times in the remaining nursery 188 schools. Consequently, all these peaks were found during occupation periods.

A large difference was found between daily profiles on weekdays and weekends. For the latter, concentrations were usually found bellow 1000 mg m<sup>-3</sup> in all studied nursery schools. During meals lower concentrations were observed in the classrooms, although never lower than in non-occupation periods (night, dawn and weekends).

193 Exceptions for the general profiles above described were found for: i) classroom B of RUR3, characterized by a small increase during the occupation period, which might have 194 195 been due to the usage of this room as a support room, namely for material storage and for only one baby sleeping period, thus having low occupation for a short period; ii) 196 classroom B of URB, characterized by a continuous increase of CO<sub>2</sub> concentrations 197 198 throughout the day from early morning until late afternoon. This was probably due to the lack of ventilation (closed windows) during the morning and sleeping period (12h to 15h), 199 leading to the highest concentrations (7448 mg m<sup>-3</sup>). Before the sleeping period the 200 201 windows were opened and CO<sub>2</sub> concentrations started to decrease until the early evening when they stabilized close to the minimum value ( $708 \text{ mg m}^{-3}$ ). 202

Windows and doors closed during classrooms' occupation periods (to avoid noise and 203 reducing/increasing indoor temperatures) also caused the highest CO<sub>2</sub> concentrations 204 found by Yang et al. (2009) (5813 mg  $m^{-3}$ ) and by Yoon et al. (2011) (3088 mg  $m^{-3}$ ) for 205 urban nursery schools, although lower than those reported for classrooms B and C of 206 URB. The same was verified by Branco et al. (2015) for urban nursery schools. 207 Gładyszewska-Fiedoruk (2011) reported similar CO<sub>2</sub> concentrations in a nursery school 208 209 at north-eastern Poland, and also highlighted the importance of good natural ventilation. 210 Classroom C of URB that had natural ventilation, recorded on average, for occupation periods, higher concentrations (mean  $\pm$  SD: 2087 $\pm$ 971 mg m<sup>-3</sup>) than the rooms of rural 211 212 nursery schools also naturally ventilated, which might have been due to the occupational densities. This is in fact another determining factor for the indoor air concentrations of 213 CO<sub>2</sub> in classrooms. Therefore, it was possible to observe that classroom C of URB had 214 215 higher occupation density than the above referred rural classrooms. In general, the 216 occupational densities were found higher in the urban nursery school than in the rural 217 ones (Table 1).

218 Global concentrations found in rural nursery schools were on average lower than those in the urban nursery school during occupation periods (mean  $\pm$  SD: 1408  $\pm$  388 mg m<sup>-3</sup> and 219  $2273 \pm 943$  mg m<sup>-3</sup>, respectively), non-occupation periods (mean  $\pm$  SD: 795  $\pm$  97 mg m<sup>-3</sup> 220 and  $976 \pm 83$  mg m<sup>-3</sup>, respectively) and weekends (mean  $\pm$  SD:  $676 \pm 14$  mg m<sup>-3</sup> and 799 221  $\pm$  60 mg m<sup>-3</sup>, respectively). Yoon et al. (2011) reached the same conclusion in their study, 222 reporting higher concentrations of CO<sub>2</sub> in urban pre-schools (1525 mg m<sup>-3</sup>) than in rural 223 ones (995 mg m<sup>-3</sup>). Theodosiou and Ordoumpozanis (2008) that studied the thermal 224 environment and IAO in kindergartens and primary schools in Kozani (Greece) and 225 226 Carreiro-Martins (2014) that evaluated the association between reported wheezing and 227 measured indoor CO<sub>2</sub> and other environmental comfort parameters in day care centres of

Porto and Lisbon, reported higher mean concentrations (2700 mg m<sup>-3</sup> and 2592 mg m<sup>-3</sup>, 228 229 respectively) than those recorded in all nursery schools in this study. Cano et al. (2012) reported for urban pre-schools of Porto a higher mean concentration (3145 mg m<sup>-3</sup>) than 230 that registered in URB (mean  $\pm$  SD: 2273  $\pm$  943 mg m<sup>-3</sup> for occupation periods). Zuraimi 231 and Tham (2008) reported for nursery schools in Singapore concentrations similar to 232 those found in classrooms A and B of URB. Yoon et al. (2011) reported for rural nursery 233 234 schools similar concentrations to those found in classrooms A of RUR1 (full occupation) and RUR2 and in classroom B of RUR3. 235

### 236 3.1.2 Traffic related pollutants - CO, NO<sub>2</sub> and O<sub>3</sub>

Figures 2, 3 and 4 show CO, NO<sub>2</sub> and O<sub>3</sub> hourly mean concentrations in: a) RUR1, b) 237 RUR2, c) RUR3 and d) URB. For O<sub>3</sub>, daily distributions of hourly mean concentrations 238 239 were only represented for a) RUR1 - Classroom A in full occupation (week), Classroom A in partial occupation, Classroom B (week and weekend) and lunch room in full 240 occupation and partial occupation; b) RUR2 - Classroom A (week) and lunch room; c) 241 242 RUR3 – Classroom A (week) and d) URB. The remaining profiles were not represented because O<sub>3</sub> concentrations were below or very close to the minimum detection limit of 243 244 the equipment (1 ppb).

Regarding CO concentrations, it was possible to distinguish a similar daily profile in all the studied buildings, especially on weekdays, when concentrations increased early in the morning until the end of the afternoon, matching with anthropogenic activities mainly related with work/school-to-home-to-work/school routes, thus showing the probable outdoor influence. During night and early morning concentrations tended to decrease.

For NO<sub>2</sub> concentrations, statistically different average daily profiles were found for all nursery schools (p < 0.05). Oscillations might have been related with ventilation (door

and /or windows opening), reducing NO<sub>2</sub> concentrations, and with accumulation (in 252 253 RUR2 for example), increasing NO<sub>2</sub> concentrations. In RUR3, slight increases were only verified in occupation periods; however, concentrations were on average significantly 254 higher (p < 0.05) than in RUR1 and RUR2. In URB, NO<sub>2</sub> daily profiles corresponded to 255 the daily traffic patterns, where there were increases in the morning and late afternoon. 256 The major cause for the increase of indoor NO<sub>2</sub> concentrations was the exhaust gases' 257 emission from outdoors, which was very clear in classroom A of RUR3, with windows 258 facing the parking area, recording 291.66 µg m<sup>-3</sup> at 18h (time of parents' arrival). 259 Nevertheless, in some cases the accumulation of this pollutant and lack of rooms' 260 261 ventilation were the factors that contributed the most to the concentrations recorded (as in classroom B in RUR1). 262

263 CO and NO<sub>2</sub> concentrations obtained in URB for occupation, non-occupation and weekend periods were on average higher (CO -  $3608.7 \pm 1175.1 \,\mu g \, m^{-3}$ ,  $3053.0 \pm 1024.0$ 264  $\mu$ g m<sup>-3</sup> and 3218.4 ± 669.5  $\mu$ g m<sup>-3</sup>, respectively; NO<sub>2</sub> – 63.87 ± 43.21  $\mu$ g m<sup>-3</sup>, 62.99 ± 265 45.40  $\mu$ g m<sup>-3</sup> and 104.80  $\pm$  47.61  $\mu$ g m<sup>-3</sup>, respectively) than those registered in rural 266 nursery schools (CO –  $3109.3 \pm 830.5 \ \mu g \ m^{-3}$ ,  $2817.5 \pm 863.9 \ \mu g \ m^{-3}$  and  $2439.8 \pm 195.6$ 267  $\mu$ g m<sup>-3</sup>, respectively; NO<sub>2</sub> – 47.13 ± 43.04  $\mu$ g m<sup>-3</sup>, 44.96 ± 41.59  $\mu$ g m<sup>-3</sup> and 61.51 ± 39.95 268  $\mu$ g m<sup>-3</sup>), which can be explained by the influence of road traffic from outdoor air, as no 269 270 indoor sources were present. Yang et al. (2009) and Wichmann et al. (2010) also pointed to road traffic as responsible for high concentrations of CO and NO<sub>2</sub> in South Korean 271 272 urban pre-schools and in Sweden homes, pre-schools and schools, respectively. In the 273 lunch rooms, CO and NO<sub>2</sub> concentrations were on average lower than those recorded in the classrooms, except for RUR1, probably due to emissions from gas stoves in the lunch 274 275 room of this school.

Regarding weekend periods, CO and NO<sub>2</sub> daily profiles seemed almost constants 276 277 throughout the day, however, CO concentrations in URB showed a similar profile to that recorded for weekdays. Despite this, in all nursery schools concentrations on weekends 278 279 were significantly lower (p < 0.05 in all cases) than those recorded on weekdays, once the nursery schools were closed being the influence from outdoors restricted. For NO<sub>2</sub>, 280 281 all nursery schools showed profiles with the same order of magnitude as those recorded 282 on weekdays. CO concentrations obtained in the four nursery schools studied, both for 283 weekdays and weekends, were substantially higher than those reported by Yoon et al. (2011) (1512.0 µg m<sup>-3</sup>). Cano et al. (2012) reported much lower concentrations in Porto 284 pre-schools (478  $\mu$ g m<sup>-3</sup>) than those recorded in the nursery schools here studied; 285 however, for Lisbon pre-schools the reported concentrations (3888 µg m<sup>-3</sup>) were similar 286 to some of those here recorded (classrooms A of RUR2 and RUR3 and in classrooms A 287 288 and C of URB). Wichmann et al. (2010) reported for kindergartens in Stockholm, Sweden, similar NO<sub>2</sub> concentrations  $(12.4 \,\mu g \, m^{-3})$  to some of those here stated (classroom 289 290 A in full occupation and in the lunch rooms in full occupation of RUR1, and RUR2 and 291 RUR3).

Regarding O<sub>3</sub>, concentrations were higher during the afternoons in all nursery schools. 292 The maxima concentrations were recorded between 16h and 19h related with cleaning 293 294 activities and windows opening. The peak observed in RUR3 was similar to that recorded for NO<sub>2</sub>, and the outdoor air appeared to be the main cause for the O<sub>3</sub> indoor air 295 296 concentrations. In RUR2 and URB, the maximum concentrations were recorded in lunch 297 rooms during clean-up activities, which were frequently associated with windows opening and consequent influence of outdoor air. Rural nursery schools recorded 298 significantly higher (p < 0.05) concentrations than URB. In the inexistence of indoor 299 300 sources, which happened in all nursery schools of this study, outdoor air is expected to have been the main determinant of indoor  $O_3$  concentrations, as already identified in several studies (Sousa et al., 2009; Bayer-Oglesby et al., 2004; Duenas et al., 2004; Syri et al., 2001).  $O_3$  outdoor concentrations are clearly higher in rural areas than urban ones, thus reproducing the same behaviour indoors. Generally,  $O_3$  concentrations were lower on weekends than on weekdays, being zero in most cases, which reinforces the influence of outdoor air.

307 Zuraimi and Tham (2008) reported higher  $O_3$  concentrations (59 µg m<sup>-3</sup>) than those in the 308 nursery schools of this study, probably due to cleaning routines and outdoor air 309 contributions.

In general, the influence of outdoor air could be associated with the observed indoor concentrations of CO, NO<sub>2</sub> and O<sub>3</sub>, which could be supported by the inexistence of indoor sources (in majority of cases) as well as by the I/O ratios results (presented in Section 3.4).

### 314 **3.1.3 TVOC and CH<sub>2</sub>O**

Figure 5 shows TVOC hourly mean concentrations determined for all the studied class and lunch rooms of the four nursery schools: a) RUR1, b) RUR2, c) RUR3, and d) URB).

RUR1 and RUR2 showed a nearly constant profile throughout the day; in RUR3 maxima 317 318 concentrations were found during dawn and in URB occurred mainly during occupation periods. In lunch rooms, the highest concentrations were recorded immediately after 319 lunch and snack times. Although it was not possible to find a typical profile for TVOC 320 321 concentrations in the studied nursery schools, all recorded peaks seemed to be related with: i) the cleaning activities (products emitting VOC), which were performed mostly in 322 the late afternoon in the classroom and after lunch in the lunchrooms; ii) with the 323 accumulation phenomenon caused by the lack of ventilation after these activities; for 324

partial occupation period in RUR1, a deeper cleaning was performed, thus higher concentrations of TVOC were recorded in the lunch room as well as in classroom A; and iii) in URB, the frequent peaks during occupation periods seemed to be related with insufficient ventilation, boosting accumulation during sleeping time (12h to 15h). After sleeping time concentrations decreased, as the room was ventilated.

330 The concentrations recorded on weekends, when above the detection limit, seemed to be 331 constant and almost the same as those recorded for non-occupation periods (night and dawn) during the week, except for RUR3. In this nursery school there were probably 332 specific indoor sources of those pollutants, namely building materials such as wood 333 334 clusters, plywood and furniture materials, because besides increasing during weekends 335 (classroom A), a progressive increasing from the end of the day until the following morning on weekdays was observed in classrooms A and B. Classroom A of RUR2 and 336 337 Classroom C of URB reported concentrations for occupation periods (mean ± SD: 117.66  $\pm$  17.59 µg m<sup>-3</sup> and mean  $\pm$  SD: 124.58  $\pm$  89.62 µg m<sup>-3</sup>, respectively), similar to those 338 reported by Yang et al. (2009) (123.00 µg m<sup>-3</sup>). Classroom B of RUR1 reported 339 concentrations for occupation periods (mean  $\pm$  SD: 155.22  $\pm$  115.11 µg m<sup>-3</sup>) similar to 340 those reported by Cano et al. (2012) (181.00 µg m<sup>-3</sup>). Roda et al. (2011) that investigated 341 IAQ in Paris child day care centers and St-Jean et al. (2012) that studied IAQ in Montréal 342 343 reported lower TVOC concentrations than those reported for nursery schools in this study (12.75 µg m<sup>-3</sup> and 22.90 µg m<sup>-3</sup>, respectively). In these two studies, the authors concluded 344 345 that indoor TVOC concentrations were caused by emissions from building materials and 346 furniture, worsened by insufficient ventilation rates.

TVOC mean concentrations recorded for occupation periods in rural nursery schools were lower (mean  $\pm$  SD: 145.18  $\pm$  128.15 µg m<sup>-3</sup>) than those reported by Yoon et al. (2011) (351.00 µg m<sup>-3</sup>) and similar to those reported by Yang et al. (2009) (162.69 µg m<sup>-3</sup>). However, both these studies concluded that those concentrations were caused by emissions from building materials and furnishing. Cano et al. (2012) reported in Lisbon much higher concentrations (3339.00  $\mu$ g m<sup>-3</sup>) than those found in this study. In general, for occupation periods the urban nursery recorded higher mean TVOC concentrations (mean ± SD: 271.66 ± 216.42  $\mu$ g m<sup>-3</sup>) than rural ones (mean ± SD: 145.18 ± 128.15  $\mu$ g m<sup>-3</sup>), and Yoon et al. (2011) found the same for the South Korean nursery schools.

Figure 6 shows CH<sub>2</sub>O hourly mean concentrations for: a) classroom A (weekend), classroom B (weekdays) and the lunch room in full occupation of RUR1; b) the lunch room of RUR2; c) the lunch room of RUR3; and d) URB. CH<sub>2</sub>O concentrations for the remaining studied rooms are not represented because they were below the minimum detection limit of the equipment (0.05 ppm).

361 For CH<sub>2</sub>O concentrations a daily pattern was found in URB. In this nursery school it was possible to identify three concentration peaks in classrooms A and B and in the lunch 362 363 room, recorded on weekdays. In classroom A the peak was between 11h and 13h, 364 corresponding to the lunch and cleaning time as well as to the preparation for the sleeping period (children were less than 2 years old and spent the entire school day inside the 365 366 classroom). CH<sub>2</sub>O concentrations' increase might have been related with the products used for cleaning. Furthermore, certain activities such as dragging wood furniture 367 368 (scraping the floor) to prepare the classroom for sleeping time could be connected with 369 the emission of this pollutant. The other two peaks corresponded to the cleaning periods 370 in the lunch room (before lunch) and cleaning before sleeping time and children's hygiene in classroom B. Besides the recorded peaks, concentrations increased at the end of the 371 372 day, in agreement with the period of general cleaning of the entire building. After that, the building was closed and the concentrations of CH2O increased due to the 373 374 accumulation at the end of the night, and gradually decreased throughout the morning. In

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RUR1 (full occupation), RUR2 and RUR3 two concentrations peaks were recorded in the 375 lunch rooms at lunch time probably related with furniture dragging (furniture finishing -376 377 varnished wood). On average, URB registered higher CH2O concentrations for occupation and non-occupation periods (44.19  $\pm$  42.99 µg m<sup>-3</sup> and 60.63  $\pm$  60.83 µg m<sup>-3</sup>, 378 respectively) than those registered in rural nursery schools ( $18.42 \pm 34.03 \,\mu g \,m^{-3}$  and 1.54379  $\pm 2.77 \ \mu g \ m^{-3}$ , respectively) and lower for weekend periods (1.33  $\pm 1.15 \ \mu g \ m^{-3}$  and 3.68 380  $\pm$  6.37 µg m<sup>-3</sup>, respectively). CH<sub>2</sub>O mean concentrations recorded for occupation periods 381 382 in URB were much higher than those reported by the European Commission in AIRMEX project (European Indoor Air Monitoring and Exposure Assessment) (2003-2008) for 383 schools and kindergartens (the maximum mean value recorded was  $31.9 \,\mu\text{g} \,\text{m}^{-3}$  in Leipzig 384 during July). For Athens, Budapest and Helsinki schools and kindergartens, (20.2 µg m<sup>-</sup> 385 <sup>3</sup>, 18.23  $\mu$ g m<sup>-3</sup> and 21.23  $\mu$ g m<sup>-3</sup>, respectively) (Kotzias et al., 2009) as well as in the 386 387 kindergartens analysed in the SINPHONIE project (Schools Indoor Pollution and Health: Observatory Network in Europe)  $(15 \pm 10 \,\mu g \, m^{-3})$  (Jantunen et al., 2008), concentrations 388 389 were similar to those recorded in occupation periods of rural nursery schools. 390 Furthermore, CH<sub>2</sub>O mean concentrations in schools of Nijmegen, Catania, Thessaloniki and Nicosia (6.1  $\mu$ g m<sup>-3</sup>, 13.0  $\mu$ g m<sup>-3</sup>, 13.8  $\mu$ g m<sup>-3</sup>, 12.0  $\mu$ g m<sup>-3</sup>, respectively), also reported 391 in AIRMEX project, were lower than those reported in this study (Kotzias et al., 2009). 392

Concluding, TVOC higher concentrations in RUR1, RUR2 and URB were mainly caused by cleaning activities (products used). In RUR3 internal sources, namely building materials and furniture finishing, were the probable causes for the concentrations recorded. The lack of ventilation increased even more significantly the concentrations recorded. Regarding CH<sub>2</sub>O, dragging of furniture in RUR1, RUR2 and RUR3 lunch rooms during meal time appeared to have been the main responsible for the CH<sub>2</sub>O concentrations recorded. In URB, concentrations seemed to have been related withcleaning activities and poor ventilation.

A careful choice of materials which do not emit VOC must be prioritized to improve IAQ
and to protect children's health. Furthermore, improved ventilation rates will allow the
reduction of indoor concentrations of these pollutants.

### 404 **3.3 Comparison with standards and guidelines**

Table 5 shows the number of non-compliances and exceedances (%) to the standards andguidelines referred to in the Material and methods section.

407 WHO guidelines for CO, NO<sub>2</sub> and O<sub>3</sub> concerning 1h, 8h and 24h means were never exceeded. However, WHO guideline for 30 minutes CH<sub>2</sub>O mean was always exceeded 408 409 during occupation periods in the lunch rooms of RUR1, RUR2 and RUR3, probably due to furniture dragging (tables and chairs) during meal times. In URB, exceedances were 410 411 around 28% in all the studied rooms during occupation periods, being lower than in rural 412 nursery schools. Classrooms A and B also recorded CH<sub>2</sub>O exceedances for the Portuguese 413 legislation (100% in both classrooms). These values were probably due to emissions from 414 cleaning products, furniture and flooring which were varnished wood. Missia et al. (2010) reported CH<sub>2</sub>O concentrations (5.8-62.6  $\mu$ g m<sup>-3</sup>) below the WHO guideline. CH<sub>2</sub>O 415 concentrations reported in AIRMEX and SINPHONIE projects could not be compared 416 417 with the WHO guideline (30 minutes), because measurements were one week long. Exposure to CH<sub>2</sub>O concentrations may cause inflammation of the airways and adverse 418 pulmonary effects (Venn et al., 2003, Jones, 1999). To minimize the concentrations 419 recorded, higher ventilation in the rooms with the highest concentrations should be 420 implemented, and materials free from CH<sub>2</sub>O emissions should be preferred whenever 421 422 possible.

According to Portuguese legislation for CO<sub>2</sub>, exceedances were recorded in classrooms 423 424 A and B of URB (50% and 100%, respectively). Classrooms A of RUR3 and C of URB had natural ventilation so the margin of tolerance was applied, thus no exceedances were 425 426 observed. A determining factor for the indoor air concentrations of CO<sub>2</sub> that is not taken into account by the Portuguese legislation is the occupation density of the classrooms. In 427 428 fact, Portuguese legislation regarding the number of children per classroom for infants under 3 years old (Portaria nº 262/2011) and for pre-schoolers (Despacho nº 5048-429 430 B/2013) which only considers educational and economic criteria, legislate for infants, groups aged 1 to 2 year old (until the acquisition of march) and groups aged 2 to 3 years 431 432 old, a maximum of, respectively, 10, 14 and 18 children per group. For pre-schoolers a minimum of 20 and a maximum of 25 children per classroom is legislated. Beyond that, 433 there is a guideline recommended by ASHRAE for classrooms (for children between 5 434 435 and 8 years old) that considers the density of occupation (25 occupants per 100  $m^2$ ) (ASHRAE, 2007). Although only classroom C of URB has exceeded the value of the 436 437 Portuguese legislation regarding the number of children per classroom for pre-schoolers (Despacho nº 5048-B/2013), all classrooms of this study exceeded ASHRAE 438 recommended guideline. Branco et al. (2015) also found exceedances to ASHRAE 439 440 recommended guideline for all urban nursery schools analysed. This circumstance as well as ventilation habits referred in Section 3.1, led to the increase of CO<sub>2</sub> concentrations in 441 classrooms to values above the Portuguese legislated standards. Theodosiou and 442 Ordoumpozanis (2008) and Zuraimi and Tham (2008) also pointed out the higher 443 444 occupation densities as an important factor for the increase of indoor CO<sub>2</sub> concentrations. St-Jean et al. (2012) also referred a high occupational density when comparing with 445 446 ASHRAE recommendation and indicated high occupation density as an important factor for the increase of CO<sub>2</sub> concentrations. 447

Headache, nausea, breathlessness and loss of concentration are possible health symptoms
for children attending these nursery schools (USEPA 2009; Griffiths and Eftekhari,
2008).

For TVOC, exceedances were recorded in classroom A of RUR3 (33%) and in classrooms 451 A and B of URB (50% and 100%, respectively). According to the analysis previously 452 453 performed, these exceedances may have been caused by specific point activities 454 performed in the classrooms (A and B of URB), associated with the use of paints and glues, as well as by the probable existence of internal sources of these pollutants such as 455 furniture materials, finishing (paints and varnishes), decoration and construction products 456 (classroom A of RUR3). Missia et al. (2010) and Zhang and Niu (2003) also associated 457 458 finishing products, coatings and building materials (carpets, acoustic and thermal 459 insulation) as sources of TVOC. These high concentrations may result in irritation of the upper airways and/or the lower respiratory tract and adverse lung effects (Rumchev et al., 460 461 2004; Nurmatov et al., 2013).

462 On weekends, no exceedances were observed. In general, most exceedances and non-463 compliances were registered in the urban nursery school.

#### 464 **3.4 Indoor/Outdoor ratios**

The concentrations measured indoors were compared with those outdoors using the I/O ratio for NO<sub>2</sub> and O<sub>3</sub>. Median, minimum (min) and maximum (max) I/O ratios were obtained for each studied room in the four nursery schools and are presented in Table 6.

Median I/O ratios of NO<sub>2</sub> were lower than 1 in classroom A in full occupation and for
RUR1 lunch room in full occupation and partial occupation, meaning that lower
concentrations were observed indoors. In classrooms A during partial occupation and B,
both for weekdays and weekends, indoor air concentrations of NO<sub>2</sub> were higher than

outdoors, which was probably due to accumulated indoor concentrations that did not
decrease as fast as the outdoor concentrations, leading to ratios higher than 1. Whichmann
et al. (2010) that studied indoor-outdoor relationships at homes, pre-schools and schools
in Stockholm (Sweden) also reported mean I/O ratio for pre-schools higher than 1.

I/O ratios found during the weekend were higher than those on weekdays, once again
demonstrating the contribution of accumulation observed in non-occupation periods
resulting from the lack of proper ventilation.

Maxima values of NO<sub>2</sub> I/O ratios in URB were higher than in rural nursery schools. This
was expected since URB was strongly affected by traffic emissions and it is known that
NO<sub>2</sub> is one of the main components of exhaust gases. Furthermore, the accumulation
phenomenon (lack of proper ventilation) increased even more the already high indoor
concentrations (influenced by outdoor air) in URB.

484 Concerning  $O_3$ , I/O median ratios calculated for all microenvironments were lower than 485 1. This behaviour was expected, because in the absence of indoor sources, indoor  $O_3$ 486 concentrations are mainly due to outdoor air. On average, I/O ratios were higher in rural 487 nursery schools than in the urban one.

488

# 489 **4.** Conclusions

Indoor concentrations of CO<sub>2</sub>, CO, CH<sub>2</sub>O, NO<sub>2</sub>, O<sub>3</sub> and TVOC were monitored in rural
nursery schools and in one urban nursery school allowing a better understanding of the
effect that those two different contexts have on IAQ.

Regarding CO<sub>2</sub>, the urban nursery recorded, on average, higher concentrations than rural
 nursery schools, reaching maxima peaks in occupation periods of around 7500 mg m<sup>-3</sup>.

The Portuguese legislation reference value was exceeded only in the classrooms from the 495 496 urban nursery, which was due to the higher occupation densities than in rural nursery schools. CO and NO<sub>2</sub> concentrations obtained in URB for occupation periods were on 497 498 average higher than those registered in rural nursery schools, which can be explained by the influence of road traffic from outdoor air. The inexistence of indoor sources as well 499 500 as I/O ratio results indicated that the influence of outdoor air was the main determinant 501 NO<sub>2</sub> and O<sub>3</sub> indoor concentrations. Regarding O<sub>3</sub>, rural nursery schools registered higher 502 indoor concentrations than the urban nursery school. Several studies have concluded that the outdoor O<sub>3</sub> concentrations are clearly greater in rural areas than in urban ones (Sousa 503 504 et al., 2008). Thus, as no indoor sources were present (referred above) the higher rural indoor concentration were due to the outdoor contribution. 505

High concentrations of CH<sub>2</sub>O and TVOC were occasionally observed associated mainly
to cleaning activities and in some cases indicating the presence of internal sources of these
pollutants, such as furniture finishing (emission of CH<sub>2</sub>O).

509 From this study it is possible to conclude that there is a need to implement measures to reduce critical situations regarding IAQ and consequent children's risk of exposure, 510 511 mainly in the urban context. Measures such as changing materials and consumer products 512 that emit VOC can be applied to promote children's and childcare workers overall life quality. More efficient ventilation (by mechanical or natural systems) could also be 513 514 applied to reach the same goal. Besides that, it could also be necessary to review the 515 Portuguese legislation on the number of children per classroom, having into account the occupation density and children's health issues. 516

517 The study allowed to communicate the results to the staff of the nursery schools involved518 as well as to provide mitigation measures when necessary. The authors believe that these

519	findings may be useful to improve IAQ in other nursery schools and to support future
520	research. More nurseries need to be studied to help supporting these findings.

521

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# 680 Figure captions

- Figure 1. Daily profile of CO<sub>2</sub> concentrations registered indoors for a) RUR1, b) RUR2,
  c) RUR3, and d) URB.
- Figure 2. Daily profile of CO concentrations registered indoors for a) RUR1, b) RUR2,c) RUR3, and d) URB.
- Figure 3. Daily profile of NO<sub>2</sub> concentrations registered indoors for a) RUR1, b) RUR2,
  c) RUR3, and d) URB.
- Figure 4. Daily profile of O<sub>3</sub> concentrations registered indoors in a) classroom A in FO
- 688 (week), classroom A in PO, classroom B (week and weekend) and lunch room in FO and
- 689 PO of RUR1, b) classroom A (week) and lunch room of RUR2, c) classroom A (week),
- 690 and d) URB.
- Figure 5. Daily profile of TVOC concentrations registered indoors for a) RUR1, b) RUR2,c) RUR3, and d) URB.
- Figure 6. Daily profile of CH<sub>2</sub>O concentrations registered indoors in a) classroom A
- 694 (weekend), classroom B (weekdays) and lunch room in FO of RUR1, b) lunch room of
- 695 RUR2, c) lunch room of RUR3; and d) URB.

Nursery	Room	Type of use	Children's age (years)	Floor	Area (m <sup>2</sup> )	Occupation (Children + staff)	Period of occupation	Ventilation	Sampling time (weekdays + weekend days)
	А	Classroom	4-5	Ground floor	63	$FO^{a}: 25+2$ $PO^{b}: 6+2$	09h – 12h 14h – 15h30	DNV <sup>c</sup> (Door to inner corridor frequently closed; Windows frequently open; AVAC system off)	2 + 2
RUR1	В	Classroom	5	Ground floor	48	20+2	09h - 12h 14h - 15h30	DNV (Door to inner corridor frequently closed; Windows frequently open; AVAC system off)	3 + 2
	LR	Lunch room	3-5	Ground floor (back)	56	FO : ~200 PO : ~21	12h – 14h	DNV (Open to kitchen and to inner corridor; Windows open during the occupation; AVAC system off	1 + 0
RUR2	А	Classroom	3-6	Ground floor (back)	32.5	14+2	09h - 11h30 12h15 - 16h	DNV (Door to inner corridor always open; Windows frequently closed; A/C <sup>d</sup> and heating off)	4 + 2
	LR	Lunch room	3-6	Ground floor	26	14+2	11h30 - 12h15	DNV (Door to inner corridor always open; Windows open during the occupation)	3 + 0
	А	Classroom	<1-2	Ground floor	23.5	23+2	08h - 11h30 13h30 - 18h 12h30 - 15h30 (sleeping time)	DNV (Door to inner corridor frequently closed; Windows frequently open; AVAC system off)	4 + 2
RUR3	В	Classroom	2-3	Ground floor	37.5	1 (Functioned as support room)	8h - 11h30 12h30 - 18h	DNV (Door to inner corridor always closed; Windows always closed; AVAC system off)	3 + 0
	LR	Lunch room	<1-3	Ground floor (back)	104	24	11h30 - 12h30	DNV (Door to inner corridor always open; Windows always closed; AVAC system off)	3 + 0

Table 1 – Summary of the main characteristics of each studied microenvironment and sampling periods.

	А	Classroom	<2	1st floor (back)	38	23+2	07h30 - 19h30 12h - 13h (sleeping time)	DFV <sup>e</sup> (Door to inner corridor always closed; A/C and dehumidifier frequently used)	4 + 2
URB1	В	Classroom	2-3	1st floor (back)	21	23+2	08h30 - 10h50 11h45 - 18h30 12h - 15h (sleeping time)	DFV (Door to inner corridor always closed. Windows sometimes open; A/C and dehumidifier frequently used)	4 + 0
	С	Classroom	4	2nd floor (front)	59	29+2	09h - 11h30 14h - 18h	DNV (Door to inner corridor always closed; Windows sometimes open)	3 + 2
	LR	Lunch room	2-5	Ground floor (back)	38	21 to 74	11h30 - 13h30	DNV (Opening to the kitchen and to the inner corridor; No direct opening to the outside)	3+0

<sup>a</sup> FO – full occupation; <sup>b</sup> PO – partial occupation; <sup>c</sup> DNF – Dominate natural ventilation; <sup>d</sup> A/C – Air Conditioner; <sup>e</sup> DNF – Dominate forced ventilation.

Note: adapted from Nunes et al. (2005)

	Nursery			RU	J <b>R1</b>		RU	J <b>R2</b>		RUR3			URI	3	
	Room	AFO <sup>a</sup>	Apo <sup>b</sup>	Bc	LR <sub>F0</sub> <sup>d</sup>	LRPO e	А	LR <sup>f</sup>	A <sup>g</sup>	В	LR	Α	В	С	LR
	Min	700	701	728	1092	885	953	1195	691	695	1622	823	715	773	1002
2	Max	2288	1490	1418	2431	1178	2398	1484	3490	1375	2513	3171	7448	4571	1331
$CO_2 (mg m^{-3})$	Mean	1391	951	981	1808	1054	1565	1340	1883	1035	2068	2010	3791	2087	1202
	Median SD <sup>h</sup>	1397 535	828 248	932 209	1901 550	1068 107	1436 467	1340 144	1680 873	1021 215	2068 445	1940 635	4237 1938	2126 971	1237 122
	Min	3293.0	3496.6	1761.5	3172.2	2976.8	2772.5	1919.3	2667.2	2595.5	1535.7	2512.6	1646.7	1534.0	1591
	Max	5293.0 5972.0	4861.0	3206.3	3410.2	3515.8	3809.4	2259.8	3912.0	3109.9	1654.7	6686.5	6216.7	5386.6	1838
$CO(u = m^{-3})$	Mean	4417.7	4273.7	2583.9	3322.5	3309.6	3298.7	2239.8	3329.9	2872.0	1595.2	5031.7	3940.7	3692.4	1770
CO (µg m <sup>-3</sup> )		3907.0		2385.9		3309.6 3317.6									
	Median		4319.6		3385.2		3285.6	2089.5	3344.2	2917.3	1595.2	5065.4	3609.8	4176.6	1824
	SD	1030.0	441.8	431.9	106.8	175.5	261.5	170.3	325.6	141.1	59.5	1064.3	1167.6	1165.1	103
	Min	0.00	0.00	0.00	21.92	0.00	0.00	3.12	0.00	0.00	12.65	0.00	0.00	0.00	0.00
	Max	0.00	0.00	61.79	233.76	0.00	0.00	72.95	0.00	0.00	42.94	653.44	697.74	0.00	12.21
CH <sub>2</sub> O (µg m <sup>-3</sup> )	Mean	0.00	0.00	5.62	112.80	0.00	0.00	38.03	0.00	0.00	27.79	99.32	72.94	0.00	4.50
	Median	0.00	0.00	0.00	82.74	0.00	0.00	38.03	0.00	0.00	27.79	30.52	19.89	0.00	2.90
	SD	0.00	0.00	17.76	89.06	0.00	0.00	34.91	0.00	0.00	15.14	162.55	126.91	0.00	5.04
	Min	0.00	5.99	56.14	0.00	9.20	38.26	0.00	72.79	82.84	2.22	98.58	0.64	15.15	6.32
	Max	34.46	137.52	143.86	2.82	29.29	94.54	24.86	291.66	113.82	9.51	146.27	143.40	82.09	20.92
NO <sub>2</sub> (µg m <sup>-3</sup> )	Mean	16.67	41.18	96.48	0.94	15.71	55.54	12.43	125.17	101.32	5.87	125.57	79.40	40.07	10.45
	Median	17.07	24.26	94.11	0.00	13.42	50.67	12.43	117.77	101.65	5.87	127.74	89.26	35.52	7.27
	SD	13.03	39.33	21.42	1.33	6.42	16.76	12.43	44.06	7.32	3.65	13.84	38.23	21.32	6.07
	Min	2	0	6	0	1	0	0	0	0	0	0	0	0	2
	Max	32	38	29	0	10	17	3	27	0	0	6	1	2	6
O <sub>3</sub> (µg m <sup>-3</sup> )	Mean	11	8	18	0	4	3	1	2	0	0	0	0	0	4
	Median	8	4	16	0	3	1	1	0	0	0	0	0	0	4
	SD	10	11	7	0	3	5	1	5	0	0	1	0	0	2

Table 2 – Statistical parameters of the hourly mean data for each room studied at all nursery schools in weekdays for occupation periods.

	Min	142.01	182.75	79.23	0.00	201.00	77.55	0.00	0.00	0.00	0.00	87.10	139.39	0.00	0.00
	Max	174.17	325.63	488.33	0.00	450.12	140.20	0.00	1927.18	702.71	0.00	1245.68	1321.37	337.04	0.00
TVOC (µg m <sup>-3</sup> )	Mean	159.40	229.27	155.22	0.00	329.70	117.66	0.00	379.14	81.38	0.00	434.90	527.14	124.58	0.00
	Median	160.44	219.66	100.47	0.00	357.65	118.04	0.00	0.00	0.00	0.00	295.23	527.21	118.05	0.00
	SD	9.49	39.62	115.11	0.00	98.53	17.59	0.00	540.50	215.88	0.00	314.98	257.12	89.62	0.00

<sup>a</sup> A<sub>FO</sub> – Classroom A in full occupation; <sup>b</sup> A<sub>PO</sub> – Classroom A in partial occupation; <sup>c</sup> B – Classroom B; <sup>d</sup> LR<sub>FO</sub> – Lunch Room in full occupation; <sup>e</sup> LR<sub>PO</sub> – Lunch Room in partial occupation; <sup>f</sup> LR - Lunch Room; <sup>g</sup> A – Classroom A; <sup>h</sup> SD – Standard Deviation

	Nursery			RU	J <b>R1</b>		RU	J <b>R2</b>		RUR3			U	RB	
	Room	AFO <sup>a</sup>	Apo <sup>b</sup>	Bc	LR <sub>FO</sub> <sup>d</sup>	LRPO e	Α	LR <sup>f</sup>	A <sup>g</sup>	В	LR	Α	В	С	LR
	Min	684	653	633	700	700	647	645	622	695	697	778	708	727	766
2	Max	1077	1051	869	1068	987	1718	2437	873	1080	2582	1835	1202	3127	1718
$CO_2 (mg m^{-3})$	Mean	723	715	700	806	787	753	921	728	796	1023	1059	866	1052	925
	Median SD <sup>h</sup>	694 94	696 71	689 49	785 98	785 67	695 176	703 401	727 54	780 95	784 471	1031 259	789 162	819 591	848 214
	<u> </u>	3313.8	3700.5	1846.9	2058.1	2364.1	2461.2	891.6	2514.5	2537.7	1034.0	2545.4	1635.5	1576.8	704.22
		5624.7	4627.8	2871.1	3474.3	2304.1 3474.3		3511.4	2314.3 3397.1	2967.4			4101.2	4612.3	2301.91
<b>CO</b> ( -3)	Max						3366.2				1945.8	5808.0			
CO (µg m <sup>-3</sup> )	Mean	4294.5	4068.6	2418.8	2846.9	2879.0	2983.6	1660.8	2928.9	2749.3	1344.1	4347.4	3100.1	3281.5	1483.11
	Median	4212.1	4040.5	2422.6	2854.9	2854.9	3032.9	1700.4	2966.9	2749.6	1336.0	4463.4	3194.8	3530.7	1358.22
	SD	603.0	221.4	289.7	282.4	281.5	185.2	498.6	206.4	112.4	207.9	918.0	788.8	941.3	452.88
	Min	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Max	0.00	0.00	2.08	54.47	0.00	0.00	178.13	0.00	0.00	34.88	158.14	703.33	219.29	332.57
CH <sub>2</sub> O (µg m <sup>-3</sup> )	Mean	0.00	0.00	0.06	3.54	0.00	0.00	8.94	0.00	0.00	2.87	23.59	165.78	31.71	21.42
	Median	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	61.04	0.00	0.00
	SD	0.00	0.00	0.36	11.70	0.00	0.00	28.70	0.00	0.00	7.63	44.07	199.73	57.11	54.55
	Min	0.00	1.58	44.10	0.00	0.32	11.38	0.00	91.06	92.64	0.00	102.73	5.13	17.77	1.26
	Max	34.25	66.58	126.06	52.21	52.21	92.01	39.05	224.80	115.40	26.65	173.62	107.88	65.90	37.12
NO <sub>2</sub> (µg m <sup>-3</sup> )	Mean	4.36	26.78	85.56	22.50	24.49	48.42	5.24	123.27	103.69	5.31	136.48	60.40	40.25	14.82
	Median	0.00	21.40	87.22	23.96	27.92	48.54	0.00	120.15	103.39	2.55	133.02	68.20	41.09	12.98
	SD	8.62	19.35	20.20	15.49	14.31	19.22	9.06	21.17	5.89	6.38	17.60	34.27	13.83	9.96
	Min	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	Max	23	16	19	20	20	23	22	5	0	0	6	3	6	14
O <sub>3</sub> (µg m <sup>-3</sup> )	Mean	3	1	2	1	2	2	3	0	0	0	1	0	0	2
	Median	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	SD	6	3	5	4	5	4	5	1	0	0	2	1	1	4

Table 3 – Statistical parameters of the hourly mean data for each room studied at all nursery schools in weekdays for non-occupation periods.

	Min	138.20	183.51	74.23	0.00	162.86	91.61	0.00	0.00	0.00	0.00	86.72	64.68	0.00	0.00
	Max	174.75	274.50	112.69	431.92	431.92	180.69	0.00	1827.79	535.84	0.00	460.00	1329.23	567.03	187.34
TVOC (µg m <sup>-3</sup> )	Mean	156.62	240.18	101.71	288.37	330.12	116.10	0.00	530.06	103.25	0.00	224.79	542.80	159.32	8.60
	Median	160.44	245.34	101.58	349.63	359.23	114.60	0.00	46.03	0.00	0.00	210.81	622.90	144.45	0.00
	SD	8.96	24.39	9.06	136.36	84.26	14.21	0.00	683.31	167.83	0.00	90.52	349.42	147.17	38.48

<sup>a</sup> A<sub>FO</sub> – Classroom A in full occupation; <sup>b</sup> A<sub>PO</sub> – Classroom A in partial occupation; <sup>c</sup> B – Classroom B; <sup>d</sup> LR<sub>FO</sub> – Lunch Room in full occupation; <sup>e</sup> LR<sub>PO</sub> – Lunch Room in partial occupation; <sup>f</sup> LR - Lunch Room; <sup>g</sup> A – Classroom A; <sup>h</sup> SD – Standard Deviation

	Nursery	RU	R1	RUR2	RUR3	U	RB
	Room	Α	В	Α	Α	Α	В
	Min	638	616	629	621	756	699
	Max	700	685	781	780	962	790
CO <sub>2</sub> (µg m <sup>-3</sup> )	Mean	684	653	680	688	859	739
	Median SD <sup>a</sup>	694 18	656 26	684 29	695 43	863 61	721 38
	 Min	2059.1	1765.4	29 2174.7	2429.8	2713.0	
	Max	2688.0	2600.4	2865.3	2936.5	4984.2	3530.0
CO (µg m <sup>-3</sup> )	Mean	2340.7	2179.1	2547.3	2692.2	3887.8	2548.9
	Median	2333.6	2185.7	2529.2	2700.5	3940.2	2582.7
	SD	160.5	236.1	199.6	132.5	586.5	468.5
	Min	0.00	0.00	0.00	0.00	0.00	0.00
	Max	24.92	0.00	0.00	0.00	84.39	8.51
CH <sub>2</sub> O (µg m <sup>-3</sup> )	Mean	14.71	0.00	0.00	0.00	2.48	0.18
	Median	12.46	0.00	0.00	0.00	0.00	0.00
	SD	6.00	0.00	0.00	0.00	12.92	1.22
	Min	0.00	58.57	24.11	76.20	126.14	45.40
	Max	16.50	124.79	64.08	141.01	171.21	77.94
NO <sub>2</sub> (µg m <sup>-3</sup> )	Mean	4.50	89.16	45.04	107.32	152.40	57.19
	Median	3.17	81.96	46.00	111.61	155.71	55.52
	SD	4.71	19.55	8.45	15.90	13.66	7.38
	Min	0	0	0	0	0	0
	Max	0	10	0	1	10	2
O <sub>3</sub> (µg m <sup>-3</sup> )	Mean	0	2	0	0	2	0
	Median	0	0	0	0	0	0
	SD	0	12	0	0	2	0

Table 4 – Statistical parameters of the hourly mean data for each room studied at all nursery schools in weekends.

	Min	0.00	56.92	92.31	0.00	118.44	0.00
	Max	0.00	116.53	115.76	579.55	384.97	86.30
TVOC (µg m <sup>-3</sup> )	Mean	0.00	96.41	105.12	64.90	169.78	6.80
	Median	0.00	100.58	107.84	0.00	159.90	0.00
	SD	0.00	16.25	8.75	148.63	44.77	18.55

<sup>a</sup> SD - Standard Deviation

				Weekda	ys			During occupation
Nursery	Room	WHO			Portuguese Le	egislation		WHO
		CH <sub>2</sub> O [30 min] <sup>a</sup>	CO <sub>2</sub> <sup>b</sup>	CO2MT <sup>c</sup>	CH <sub>2</sub> O <sup>d</sup>	TVOC <sup>e</sup>	TVOC MT <sup>f</sup>	CH <sub>2</sub> O [30 min]
	A <sub>FO</sub> <sup>g</sup>	bdl <sup>h</sup>	0%	na <sup>i</sup>	bdl	0%	na	0%
	A <sub>PO</sub> <sup>j</sup>	bdl	0%	na	bdl	0%	na	bdl
RUR1	В	0%	0%	na	0%	0%	na	bdl
	LR <sub>FO</sub> <sup>k</sup>	93%	na	na	na	na	na	100%
	LR <sub>PO</sub> <sup>1</sup>	bdl	0%	na	bdl	0%	na	bdl
RUR2	Α	bdl	0%	na	bdl	0%	na	bdl
KUK2	LR <sup>m</sup>	4%	0%	na	0%	0%	na	100%
	Α	bdl	33%	0%	bdl	67%	33%	bdl
RUR3	В	bdl	0%	na	bdl	0%	na	bdl
	LR	19%	0%	na	0%	0%	na	100%
	Α	17%	50%	na	100%	50%	na	28%
URB	В	37%	100%	na	100%	100%	na	28%
UKB	С	7%	50%	0%	0%	0%	na	0%
	LR	4%	0%	na	0%	0%	na	29%

Table 5 - Non-compliances and exceedances (%) of values to ASHRAE and WHO guidelines. as well as to Portuguese legislation. on weekdays and only during occupation periods.

<sup>a</sup> % of 30 minute mean concentrations above the reference value of 100  $\mu$ g m<sup>-3</sup>; <sup>b</sup> % of 8-hour running mean concentrations above the reference value of 2250 mg m<sup>-3</sup>; <sup>c</sup> % of 8-hour running mean concentrations above the reference value of 2925 mg m<sup>-3</sup> (2250 mg m<sup>-3</sup> + 30% of margin of tolerance (MT)); <sup>d</sup> % of 8-hour running mean concentrations above the reference value of 100  $\mu$ g m<sup>-3</sup>; <sup>e</sup> % of 8-hour running mean concentrations above the reference value of 100  $\mu$ g m<sup>-3</sup>; <sup>e</sup> % of 8-hour running mean concentrations above the reference value of 100  $\mu$ g m<sup>-3</sup>; <sup>e</sup> % of 8-hour running mean concentrations above the reference value of 100  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (600  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (600  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (600  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>; <sup>f</sup> % of 8-hour running mean concentrations above the reference value of 1200  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>) (100  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>) (100  $\mu$ g m<sup>-3</sup>)

N		N	02	<b>O</b> <sub>3</sub>				
Nursery	Room	Weekdays	Weekend	Weekdays	Weekend			
	A <sub>FO</sub> <sup>a</sup>	0.00 (min-max: 0.00-1.41)	0.06 (min-max: 0.00-0.92)	0.02 (min-max: 0.00-0.80)	-			
	A <sub>PO</sub> <sup>b</sup>	0.33 (min-max: 0.13-9.51)	-	0.00 (min-max: 0.00-0.52)	-			
RUR1	В	1.08 (min-max: 0.62-18.22)	1.06 (min-max: 0.81-13.06)	0.02 (min-max: 0.00-0.63)	0.06 (min-max 0.00-0.16)			
	LR <sub>FO</sub> <sup>c</sup>	0.17 (min-max: 0.00-0.41)	-	0.17 (min-max: 0.00-0.29)	-			
	LR PO <sup>d</sup>	0.26 (min-max: 0.16-4.29)	-	0.00 (min-max: 0.00-0.40)	-			
DUDA	А	0.66 (min-max: 0.33-7.52)	0.53 (min-max: 0.33-7.22)	0.00 (min-max: 0.00-0.40)	-			
RUR2	LR	0.04 (min-max: 0.00-2.90)	-	0.02 (min-max: 0.00-0.53)				
	А	1.35 (min-max: 1.13-21.17)	1.19 (min-max: 0.91-17.29)	0.00 (min-max: 0.00-0.44)	0.00 (min-max 0.00-0.01)			
RUR3	В	1.21 (min-max: 0.90-14.37)	-	-	-			
	LR <sup>e</sup>	0.06 (min-max: 0.00-2.34)	-	-	-			
	А	5.14 (min-max: 1.11-38.67)	10.40 (min-max: 2.44-23.61)	0.00 (min-max: 0.00-0.06)	0.00 (min-max 0.00-0.10)			
UDD	В	7.66 (min-max: 0.06-22.84)	-	0.00 (min-max: 0.00-0.05)	-			
URB	С	3.27 (min-max: 1.00-9.08)	6.09 (min-max: 2.92-11.86)	0.00 (min-max: 0.00-0.09)	0.00 (min-max 0.00-0.03)			
	LR	0.62 (min-max: 0.05-2.66)	-	0.00 (min-max: 0.00-0.19)	-			

Table 6 – I/O ratios for  $NO_2$  and  $O_3$ : median values observed in each studied site for weekdays and weekends and respective minimum (min) and maximum (max) ratio.

<sup>a</sup> A<sub>FO</sub> – Classroom A in full occupation; <sup>b</sup> A<sub>PO</sub> – Classroom A in partial occupation; <sup>c</sup> LR<sub>FO</sub> – Lunch Room in full occupation; <sup>d</sup> LR<sub>PO</sub> – Lunch Room in partial occupation; <sup>e</sup> LR - Lunch Room

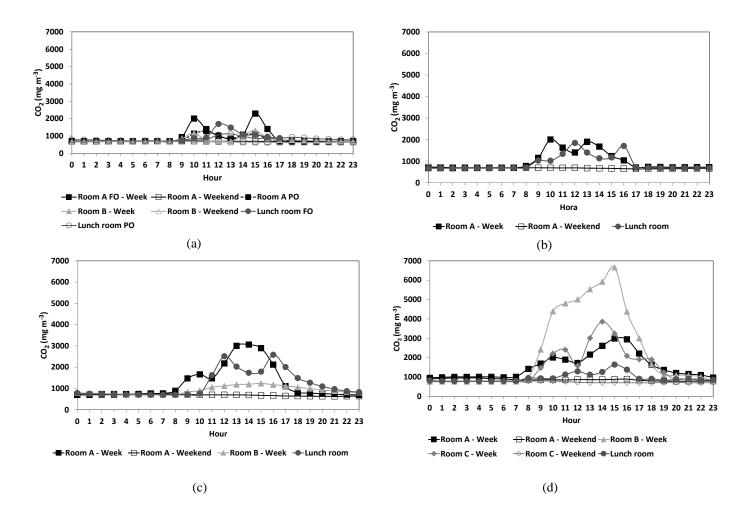


Figure 1.

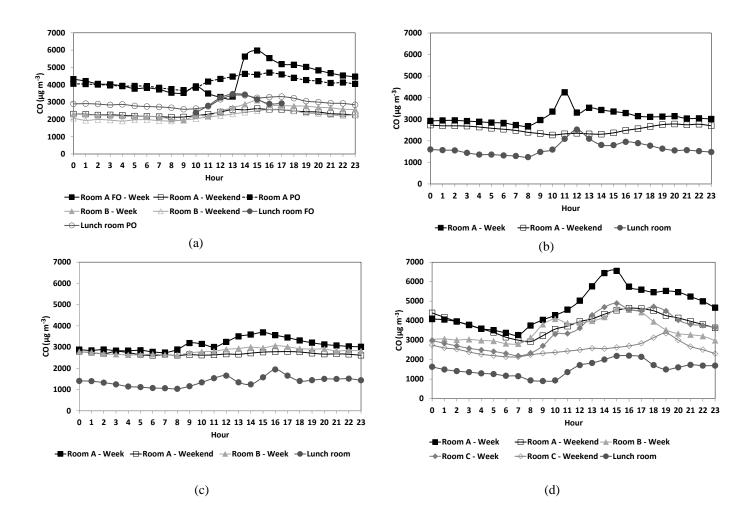


Figure 2.

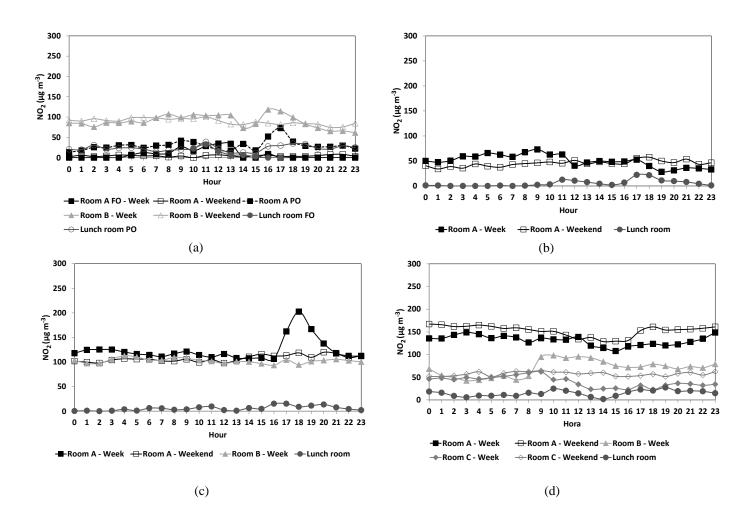


Figure 3.

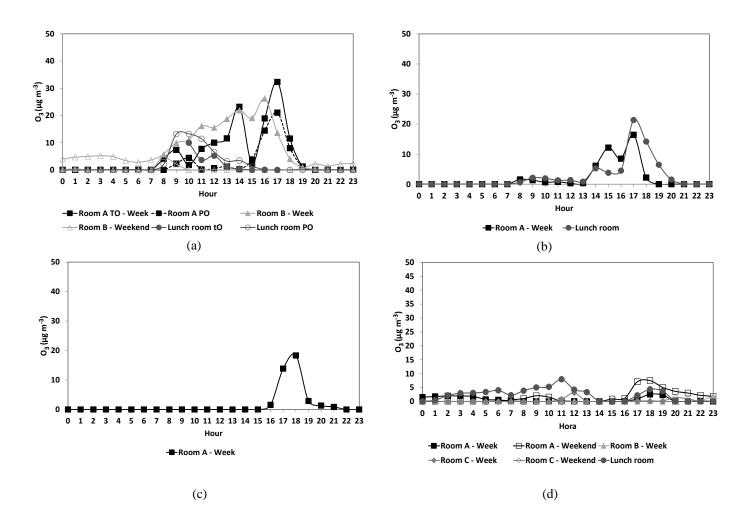
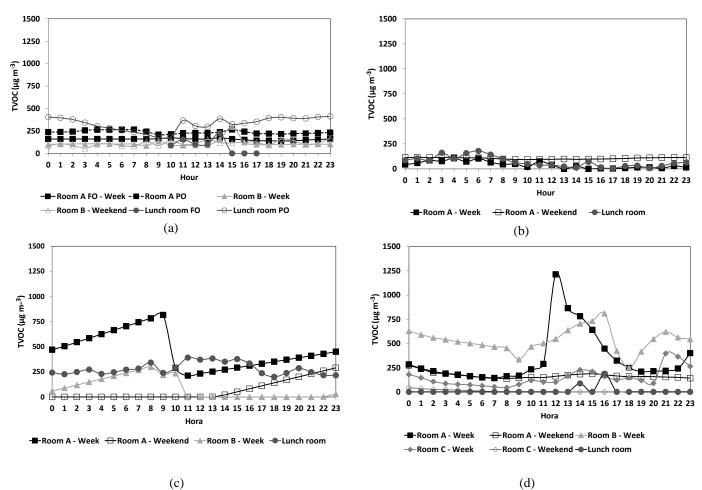


Figure 4.



(c)

Figure 5.

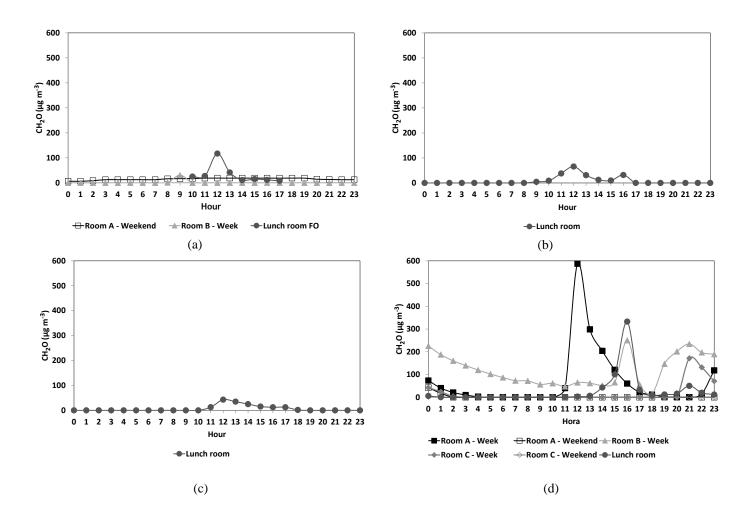


Figure 6.