

1 **Children's exposure to indoor air in urban nurseries - Part II: gaseous pollutants'**
2 **assessment**

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13

14 **Abstract**

15 This study, Part II of the larger study “Children’s exposure to indoor air in urban nurseries”,
16 aimed to: i) evaluate nursery schools’ indoor concentrations of several air pollutants in class
17 and lunch rooms; and ii) analyse them according to guidelines and references. Indoor
18 continuous measurements were performed, and outdoor concentrations were obtained to
19 determine indoor/outdoor ratios. The influence of outdoor air seemed to be determinant on
20 carbon monoxide (CO), nitrogen dioxide (NO₂) and ozone (O₃) indoor concentrations. The peak
21 concentrations of formaldehyde and volatile organic compounds (VOC) registered (highest
22 concentrations of 204 and 2320 µg m⁻³ respectively), indicated the presence of specific indoor
23 sources of these pollutants, namely materials emitting formaldehyde and products emitting
24 VOC associated to cleaning and children’s specific activities (like paints and glues). For
25 formaldehyde, baseline constant concentrations along the day were also found in some of the
26 studied rooms, which enhances the importance of detailing the study of children’s short and
27 long-term exposure to this indoor air pollutant. While CO, NO₂ and O₃ never exceeded the
28 national and international reference values for IAQ and health protection, exceedances were
29 found for formaldehyde and VOC. For this reason, a health risk assessment approach could be
30 interesting for future research to assess children’s health risks of exposure to formaldehyde and
31 to VOC concentrations in nursery schools. Changing cleaning schedules and materials emitting
32 formaldehyde, and more efficient ventilation while using products emitting VOC, with the
33 correct amount and distribution of fresh air, would decrease children’s exposure.

34 **Keywords**

35 Indoor air, nursery school, children, exposure, health risk assessment

36

38 **1. Introduction**

39 Exposure to air pollutants in indoor environments may lead to health effects, from discomfort
40 symptoms to the prevalence of respiratory or even cardiovascular diseases and/or carcinogenic
41 effects, mainly lung cancer and childhood leukaemia (Franklin, 2007; Jones, 1999; Lin et al.,
42 2013). The World Health Organization (WHO) selected particulate matter (PM) and some
43 gaseous compounds as crucial to verify Indoor Air Quality (IAQ), namely radon, carbon
44 monoxide (CO), nitrogen dioxide (NO₂), polycyclic aromatic hydrocarbons, formaldehyde and
45 other volatile organic compounds (VOC) as benzene, naphthalene, trichloroethylene, and
46 tetrachloroethylene (WHO, 2010). The increasing concern about those pollutants led WHO and
47 national governmental organizations, like the United States Environmental Protection Agency
48 (USEPA) and Health Canada, to define guidelines and standards to protect people's health by
49 ensuring a better IAQ.

50 There were found some studies on children's exposure to indoor air in nursery schools, but
51 some of them were merely focusing on ventilation, CO₂ and/or comfort parameters, PM or even
52 biological compounds (Branco et al., 2014; Branco et al., 2015; Carreiro-Martins et al., 2014;
53 Fonseca et al., 2014; Gładyszewska-Fiedoruk, 2011; Madureira et al., 2015; Nunes et al., 2015;
54 Theodosiou and Ordoumpozanis, 2008). Nevertheless, Zuraimi and Tham (2008) investigated
55 indoor concentrations of several air pollutants, evaluating their sources in child care centres in
56 the tropical region of Singapore. Despite the large number of child care centres and air
57 pollutants assessed, samplings were only conducted in the middle of the week and during
58 occupation periods, which did not allow understanding potential differences between
59 occupation and non-occupation periods. Yoon et al. (2011) measured indoor air concentrations
60 of several chemical compounds (including TVOC and formaldehyde) besides PM in Korean
61 pre-schools. However, NO₂ (also considered crucial to IAQ by WHO) was not considered in
62 that study. Roda et al. (2011) investigated IAQ of Paris child care centres to compare it with
63 dwellings by measuring biological and chemical pollutants, besides comfort parameters.
64 However, chemical pollutants were measured passively during an entire week (except the
65 weekend), which did not allow to understand pollutants variations along the day. St-Jean et al.
66 (2012) also studied IAQ in day care centres of Montréal (Canada) to determine its associations
67 with building characteristics. Despite considering a few different chemical compounds as well
68 as a VOC selection, passive sampling was also used for formaldehyde and VOC sampling,

69 which did not allow understanding pollutants variations along the day, and no outdoor
70 measurements were used to understand the outdoor influence on nursery schools' indoor air.
71 Also in the AIRMEX study (Geiss et al., 2011), in which 23 different VOC were measured in
72 public buildings including schools and kindergartens in eleven European cities, passive
73 sampling was used with the duration of a full 7-days week, not allowing to understand variations
74 along the day and between occupation and non-occupation periods.

75 Accordingly, following the study already reported focusing on the PM assessment (Branco et
76 al., 2014) in the scope of INAIRCHILD project (Sousa et al., 2012), and aiming to reduce the
77 lacks above referred, this study aims to assess children's exposure to indoor air pollution in
78 urban nursery schools. To meet this goal, the study was divided in two parts: i) Part I – CO₂
79 and comfort assessment; and ii) Part II (the present study) which aimed to: i) evaluate indoor
80 concentrations of several gaseous air pollutants in different microenvironments of urban
81 nursery schools in Porto city; and ii) analyse those concentrations according to guidelines and
82 references for IAQ and children's health.

83

84 **2. Materials and methods**

85 2.1. Sites description, sampling and analysis

86 This study was carried out in the city of Porto (Portugal) on four different nursery schools
87 located at urban sites influenced by traffic emissions (N_URB1, N_URB2, N_URB3 and
88 N_URB4), from March to June 2013 in N_URB1, N_URB2 and N_URB3, and in November
89 2013 in N_URB4. Its main characteristics (including occupation, ventilation and cleaning
90 habits and other specific activities), indoor microenvironments considered, and sampling
91 periods were fully described in Part I of the present study (Branco et al., 2015).

92 Indoor gaseous air compounds, namely CO, formaldehyde, NO₂, O₃, and total volatile organic
93 compounds (TVOC), were continuously measured using an Haz-Scanner IEMS Indoor
94 Environmental Monitoring Station (SKC Inc., USA), equipped with high sensitive sensors.
95 Sampling methods and main characteristics of each sensor are summarized in Table 1. Sampling
96 procedures, periods and duration were fully described in Part I (Branco et al., 2015).

97

98 **Table 1 – Sampling methods and main characteristics of each sensor.**

Sensor	Detection methods	Sensor minimum resolution	Sensor accuracy	Measurement range
CO	Electrochemical detection	< 1746 $\mu\text{g m}^{-3}$	< +/- 10% of reading or 2% of full scale – whichever is greater	0-58200 $\mu\text{g m}^{-3}$
Formaldehyde	Electrochemical detection	62.5 $\mu\text{g m}^{-3}$	< +/- 10% of reading or 2% of full scale – whichever is greater	0-5000 $\mu\text{g m}^{-3}$
NO ₂	Electrochemical detection	41 $\mu\text{g m}^{-3}$	< +/- 10% of reading or 2% of full scale – whichever is greater	0-41000 $\mu\text{g m}^{-3}$
O ₃	Electrochemical detection	2.14 $\mu\text{g m}^{-3}$	< +/- 10% of reading or 2% of full scale – whichever is greater	0-1070 $\mu\text{g m}^{-3}$
TVOC	Photoionization detection (PID)	230 $\mu\text{g m}^{-3}$	< +/- 10% of reading or 2% of full scale – whichever is greater	0-115385 $\mu\text{g m}^{-3}$

99

100 The mean values were compared with reference standards and guidelines aiming to evaluate
 101 exceedances and/or non-compliances. Comparisons were performed considering national and
 102 international reference values for general indoor environments, namely: i) Portuguese 2006
 103 legislation (hourly means) (*Decreto-Lei n° 79/2006*) for CO (12 500 $\mu\text{g m}^{-3}$), O₃ (200 $\mu\text{g m}^{-3}$),
 104 formaldehyde (100 $\mu\text{g m}^{-3}$), and TVOC (600 $\mu\text{g m}^{-3}$); ii) Portuguese 2013 legislation (Portaria
 105 n° 353-A/2013) for CO (10 000 $\mu\text{g m}^{-3}$), formaldehyde (100 $\mu\text{g m}^{-3}$), and TVOC (600 $\mu\text{g m}^{-3}$,
 106 plus 100% of margin of tolerance (MT) if no mechanical ventilation system was working in the
 107 room); iii) WHO guidelines (WHO, 2010) for CO (35000 $\mu\text{g m}^{-3}$ for hourly mean), NO₂ (200
 108 $\mu\text{g m}^{-3}$ for hourly mean) and formaldehyde (100 $\mu\text{g m}^{-3}$ for 30 minutes mean); and iv) Health
 109 Canada guidelines (HealthCanada, 2013) for NO₂ (480 $\mu\text{g m}^{-3}$ for hourly mean) and
 110 formaldehyde (123 $\mu\text{g m}^{-3}$ for hourly mean). For the Portuguese 2013 legislation, 8-hour
 111 running means were calculated and the daily maximum was compared with the reference value.
 112 Although Portuguese 2006 legislation was officially replaced by the new Portuguese 2013
 113 legislation, comparisons were made with both due to the clear differences between them, which
 114 allowed concluding on the expected impacts from the application of the new one.

115 Simultaneously, hourly NO₂ and O₃ outdoor concentrations were obtained from the nearest air
 116 quality station, classified as urban traffic and representative of the area (CCDR-N, 2011),
 117 because only one equipment was available inhibiting simultaneous measurements outside the
 118 nursery schools. These measurements were conducted by the Air Quality Monitoring Network
 119 of Porto Metropolitan Area, managed by the Regional Commission of Coordination and
 120 Development of Northern Portugal (*Comissão de Coordenação e Desenvolvimento Regional
 121 do Norte*) under the responsibility of the Ministry of Environment. These concentrations
 122 allowed calculating the correspondent indoor/outdoor (I/O) ratios.

123

124 2.2 Statistical analysis

125 Data were tested for normality with both Shapiro-Wilk and Anderson-Darling tests. If normal,
126 the differences between hourly mean concentrations in different sampling days for each
127 microenvironment were analysed by a parametric unpaired *t*-test; in the other cases, the non-
128 parametric Kruskal-Wallis test was used for the microenvironments where there were more than
129 two complete sampling days, and the Wilcoxon Rank Sum Test (also called Mann-Whitney *U*
130 test) was used for those where there were only two complete sampling days.

131 The one-sample parametric *t*-test was used to analyse if the differences along the day were
132 significant for normal distributions; for other distributions, the non-parametric Wilcoxon
133 Signed Rank Test was used.

134 To analyse other differences, namely between weekdays and weekends, as well as between
135 different microenvironments and nursery schools, the parametric unpaired *t*-test or the non-
136 parametric Wilcoxon Rank Sum Test was used, respectively when distributions were normal or
137 not. In all cases, a significance level (α) of 0.05 was considered. Descriptive statistics was
138 calculated using MS Excel[®] (Microsoft Corporation, USA), and other statistical analyses were
139 determined using R software, version 3.1.2 (R Development Core Team, 2014).

140

141 3. Results and discussion

142 Table 2 summarizes the main statistical parameters (minimum, maximum, mean, median and
143 standard deviation) of the hourly mean for each room of the four nursery schools.

144 When comparing two or more consecutive sampling days of the studied microenvironments,
145 statistically significant differences were found ($p < 0.05$) in 83.3%, 50% and 75% of the cases
146 regarding CO, NO₂ and O₃, respectively. For formaldehyde and TVOC, it was not possible to
147 make these statistical comparisons because concentrations were usually specific in time.
148 Despite this, a daily mean scenario in each microenvironment was assumed for the following
149 analyses of all the studied pollutants.

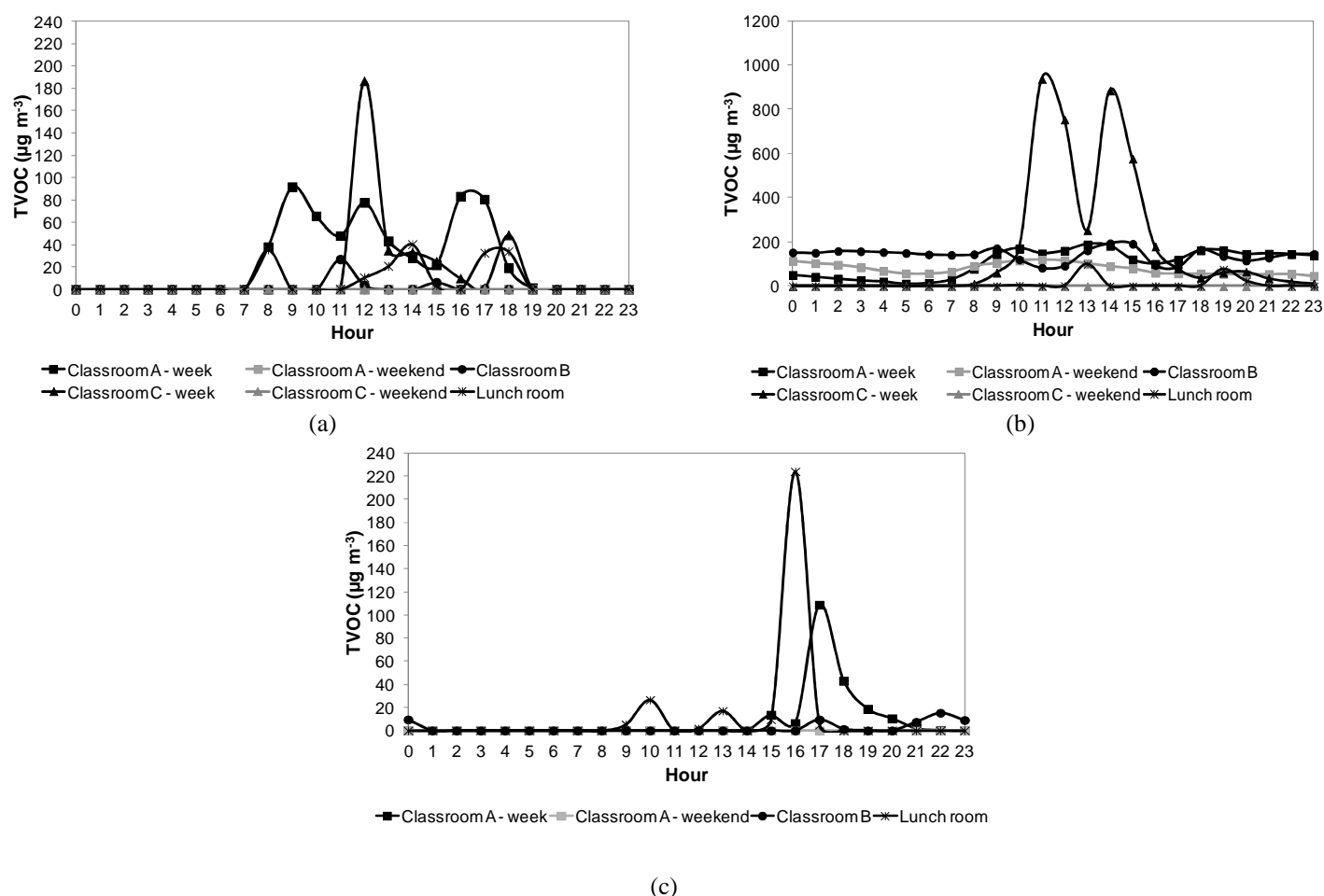
Table 2 – Statistical parameters of the hourly mean data for each room studied in the four nurseries.

Nursery	Room	N_URB1				N_URB2				N_URB3			N_URB4		
		A	B	C	LR	A	B	C	LR	A	B	LR	A	B	LR
CO ($\mu\text{g m}^{-3}$)	Min	913	1577	0	0	1498	1996	0	1949	1240	3077	734	0	0	0
	Max	4956	4347	2578	2879	3711	3902	2689	3211	2618	3916	2544	1972	89.9	1165
	Mean	2599	2765	463	1230	2359	2786	971	2552	1960	3477	1513	604	4.2	83
	Median	2476	2571	158	1152	2297	2723	893	2511	1984	3487	1438	669	0.00	0
	StDev	940	1043	608	531	521	520	577	333	329	224	541	444	15.7	221
Formaldehyde ($\mu\text{g m}^{-3}$)	Min	0	0	0	0	0	0	0	0	0	0	0	0	12	0
	Max	146	0	0	9	0	0	204	0	2	0	6	50	87	77
	Mean	2	-	-	0	-	-	8	-	0	-	0	35	35	2
	Median	0	-	-	0	-	-	0	-	0	-	0	38	35	0
	StDev	12	-	-	1	-	-	33	-	0	-	1	9	18	11
NO ₂ ($\mu\text{g m}^{-3}$)	Min	0	-	1	0	87	49	36	57	80	109	114	-	-	-
	Max	57	-	75	84	148	131	171	142	138	189	155	-	-	-
	Mean	6	-	40	22	121	73	62	93	113	136	138	-	-	-
	Median	0	-	41	18	124	72	58	90	115	133	140	-	-	-
	StDev	13	-	19	21	15	15	16	22	13	20	9	-	-	-
O ₃ ($\mu\text{g m}^{-3}$)	Min	0	15	2	4	1	8	1	9	9	10	17	7	5	12
	Max	20	32	53	49	23	39	28	61	48	25	57	27	13	32
	Mean	13	24	18	23	13	17	20	26	18	16	38	9	10	19
	Median	15	23	14	22	12	15	20	20	16	15	40	8	10	18
	StDev	5	5	10	9	3	7	4	14	7	4	7	3	2	5
TVOC ($\mu\text{g m}^{-3}$)	Min	0	0	0	0	0	52	0	0	0	0	0	0	0	0
	Max	354	54	373	132	202	276	2320	197	307	20	388	0	0	12
	Mean	17	3	8	8	92	141	104	8	5	2	12	-	-	0
	Median	0.00	0	0	0	90	115	0	0	0	0	0	-	-	0
	StDev	59	10	42	27	54	62	310	36	31	6	58	-	-	1

A – Classroom A; B – Classroom B; C – Classroom C; LR – Lunch Room

152 3.1 TVOC and formaldehyde

153 TVOC mean concentrations from the studied class and lunch rooms in N_URB1, N_URB2 and
 154 N_URB3 are represented in Figure 1 a), b) and c), respectively. N_URB4 is not represented in
 155 Figure 1 because concentrations were zero or very close to zero (maximum concentration
 156 observed equal to $4 \mu\text{g m}^{-3}$) (Table 2).

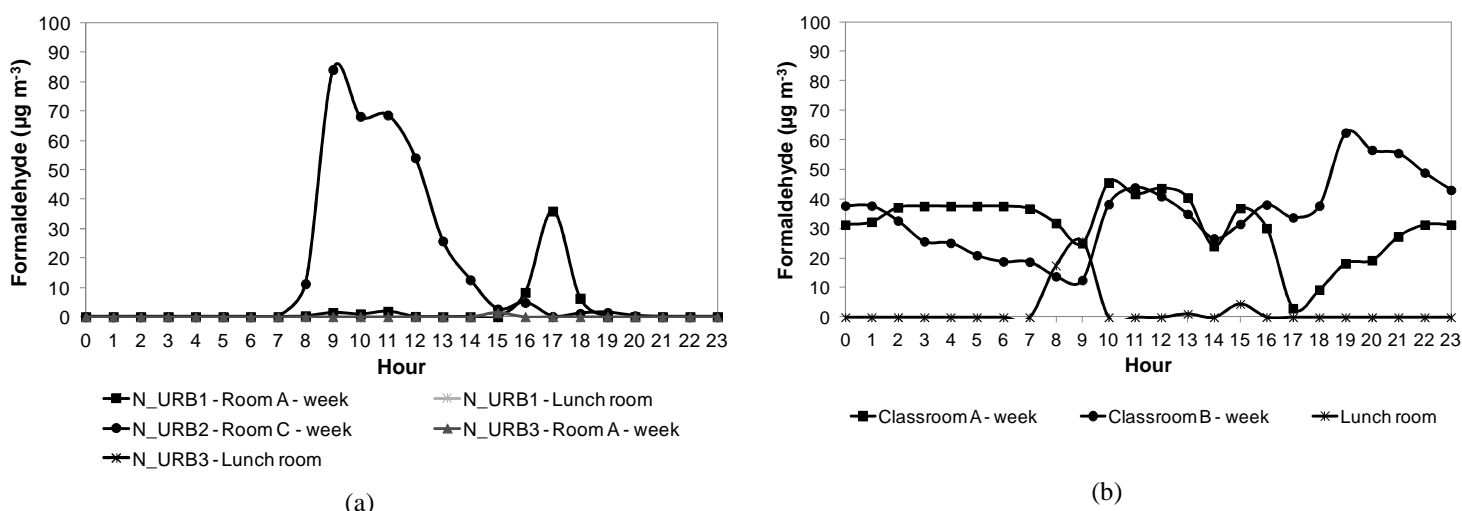


157 **Figure 1 - Daily profile of TVOC mean concentrations registered indoors of a) N_URB1,**
 158 **b) N_URB2, and c) N_URB3.**

159 Although different concentrations and daily profiles were observed, it is clear that the presence
 160 of TVOC occurred mainly during occupation periods, which seemed to be result of typical
 161 children activities associated with the use of paints and glues. The concentrations measured
 162 while the nursery schools were closed (night and weekend) were very close to zero, with
 163 exception of classrooms A (both on weekdays and weekend), B and C (on weekdays) of nursery
 164 school N_URB2 (Table 2) in which it seemed to exist a continuous source of VOC.
 165 Additionally, peak concentrations were observed in the beginning of the morning, during or

166 immediately after lunch time and in the afternoon. These TVOC concentrations in the indoor
 167 air immediately before and/or after the occupation periods in the classrooms were associated
 168 with the cleaning activities using products that emitted VOC.

169 Figure 2 shows the formaldehyde mean concentrations for a) classroom A (weekdays) and
 170 lunch room of N_URB1, classroom C (weekdays) of N_URB2 and classroom A (weekdays)
 171 and lunch room of N_URB3, and b) N_URB4. The formaldehyde concentrations for the
 172 remaining studied rooms are not represented because concentrations were close to zero (Table
 173 2) in all those cases, except for weekend on classroom A of N_URB4 which was due to
 174 instrument error. No daily profile was found for formaldehyde concentrations on the different
 175 studied rooms. The highest concentrations were observed in classroom C of N_URB2 during
 176 weekdays, where there was a peak in the morning (after the opening hour), which decreased
 177 through the morning until the period after lunch and a second peak (lower) was found about 5
 178 p.m.. These peaks matched the periods of entrance and exit from the classroom. In the other
 179 rooms represented in Figure 2 a) concentrations were close to zero. Regarding N_URB4, in the
 180 lunch room, concentrations were close to zero, except at the beginning of the morning, during
 181 and after lunch, also periods of entrance and exit. Indoor formaldehyde concentrations seemed
 182 to indicate the presence of specific indoor sources for this pollutant, namely the use of materials
 183 emitting formaldehyde (mainly furniture). The higher concentrations during occupation
 184 periods, characterized by some peaks, seemed to be mainly related to entrance and/or exit
 185 periods, associated with moving the furniture (tables and chairs).



186 **Figure 2 – Daily profile of formaldehyde mean concentrations registered indoors in a)**
 187 **classroom A (weekdays) and lunch room of N_URB1, classroom C (weekdays) of**
 188 **N_URB2, classroom A (weekdays) and lunch room of N_URB3; and b) N_URB4.**

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Table 3 – Exceedances (%) to WHO guidelines and Portuguese legislation (2006 and 2013) reference values of formaldehyde and TVOC measured on weekdays and only during occupation periods.

Nursery	Room	Weekdays					During occupation		
		Portuguese 2006 legislation		WHO	Portuguese 2013 legislation		Portuguese 2006 legislation		WHO
		Formaldehyde ^a	TVOC ^b	Formaldehyde ^c	Formaldehyde ^d	TVOC ^e	Formaldehyde ^a	TVOC ^b	Formaldehyde ^c
N_URB1	A	1	0	1	0	0	2	0	2
	B	0	0	15	0	0	0	0	22
	C	0	0	0	0	0	0	0	0
	LR	0	0	0	0	0	0	0	0
N_URB2	A	0	0	0	0	0	0	0	0
	B	0	0	0	0	0	0	0	0
	C	6	11	6	33	33	18	29	17
	LR	0	0	0	0	0	0	0	0
N_URB3	A	0	0	0	0	0	0	0	0
	B	0	0	0	0	0	0	0	0
	LR	0	0	0	0	0	0	0	0
N_URB4	A	0	0	0	0	0	0	0	0
	B	0	0	0	0	0	0	0	0
	LR	0	0	1	0	0	0	0	0

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a) % of the hourly mean concentrations above the reference value of 100 µg m⁻³; b) % of the hourly mean concentrations above the reference value of 600 µg m⁻³; c) % of the 30-min mean concentrations above the reference value of 100 µg m⁻³; d) % of 8-hour running mean concentrations above the reference value of 100 µg m⁻³; e) % of 8-hour running mean concentrations above the reference value of 600 µg m⁻³.

195 Table 3 shows the number of non-compliances and exceedances (%) to the standards and
196 guidelines referred in section 2.2. The values presented on the table are the percentage (%) of
197 the measured hourly means which were above the Portuguese 2006 reference values, the
198 percentage (%) of the 30-min means which were above the WHO reference value (only for
199 formaldehyde), and the percentage (%) of the daily maximum 8-hour running means which
200 were above the Portuguese 2013 reference values.

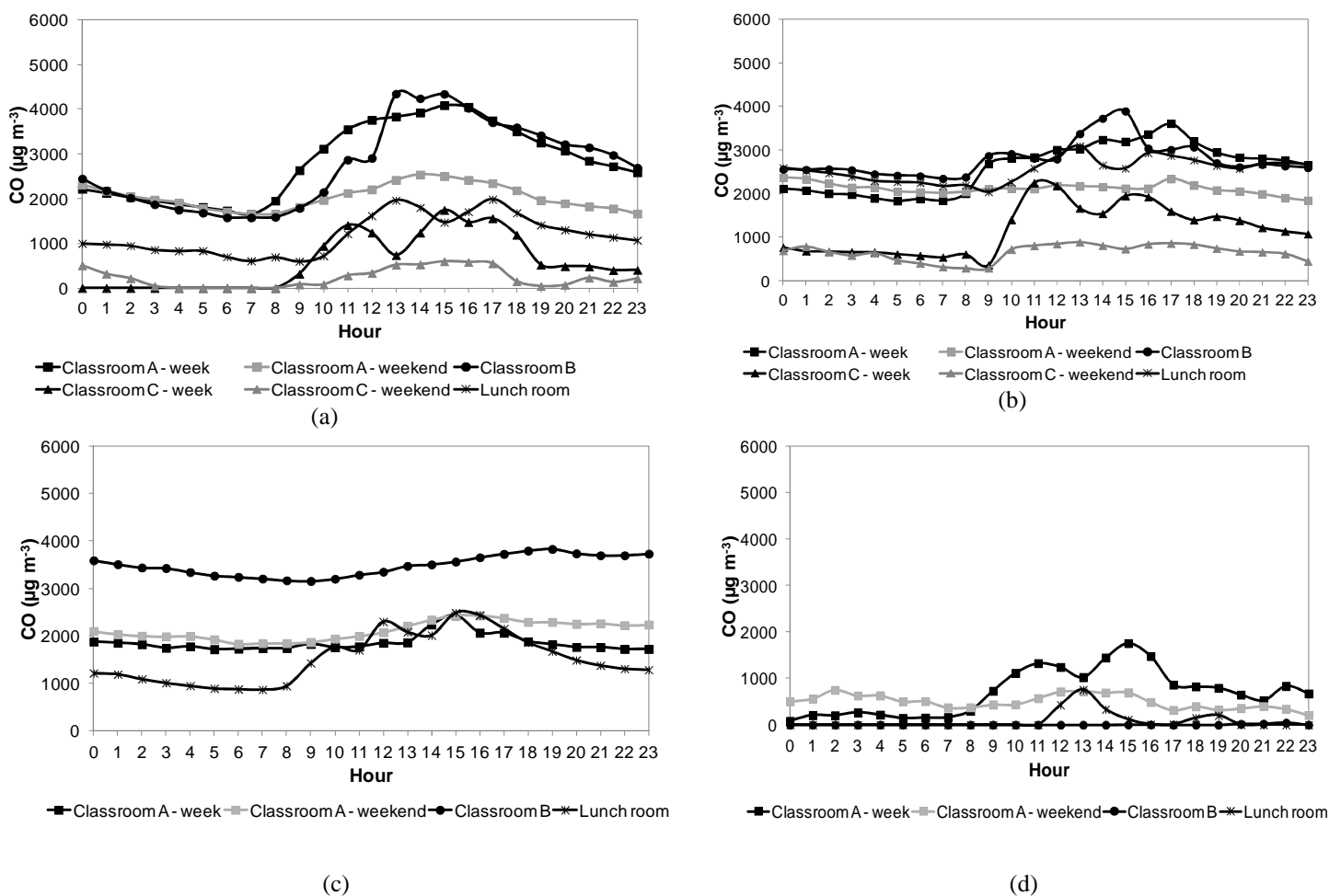
201 In few situations the recommended standard and guideline values for formaldehyde and TVOC
202 were exceeded. In the case of formaldehyde, the exceedances were mainly found during
203 occupation periods and mainly for the WHO reference value (WHO, 2010). A health risk
204 assessment approach could be important to assess the children's health risks of short-term
205 exposure to those high concentrations, and to confirm if they are expected to cause mild or
206 moderate eye irritation.

207 Formaldehyde concentrations in N_URB4 were similar to those registered by Yoon et al. (2011)
208 in Korean urban pre-schools ($45.27 \mu\text{g m}^{-3}$), but far from those registered in Korean
209 kindergartens ($162.69 \mu\text{g m}^{-3}$) (Yang et al., 2009). Both of those studies found much higher
210 TVOC concentrations ($591.2 \mu\text{g m}^{-3}$ and $642.11 \mu\text{g m}^{-3}$ respectively), and both also concluded
211 that those problems in indoor air were caused by emissions from building materials and
212 furnishing, worsened by insufficient ventilation as concluded in Part I (Branco et al., 2015).
213 Formaldehyde concentrations found in classroom C of N_URB2 and in N_URB4 were often
214 found higher than those reported by Roda et al. (2011), both in hot and cold season (10.7 and
215 $14.8 \mu\text{g m}^{-3}$, respectively), and higher than those reported by St-Jean et al. (2012) ($22.9 \mu\text{g m}^{-3}$).
216 The selection of classroom materials to use in nursery schools' indoor environments should
217 be performed with extreme caution by choosing formaldehyde-free materials to safeguard
218 children's health. Moreover, better ventilation (amount of fresh air and its distribution) could
219 help to reduce indoor formaldehyde and TVOC concentrations. It is important to notice that the
220 analysis performed in the present study were made for TVOC, but further investigations in
221 specific VOC are needed, as made in previous studies (Pegas et al., 2012; Roda et al., 2011; St-
222 Jean et al., 2012) which reported considerable indoor concentrations in nursery and primary
223 schools. That will allow comparing the results to better understand sources and pathways of
224 children's exposure to specific VOC inside nursery schools.

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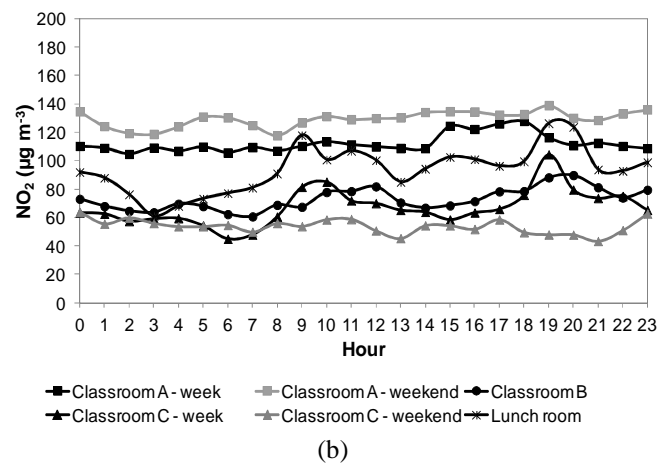
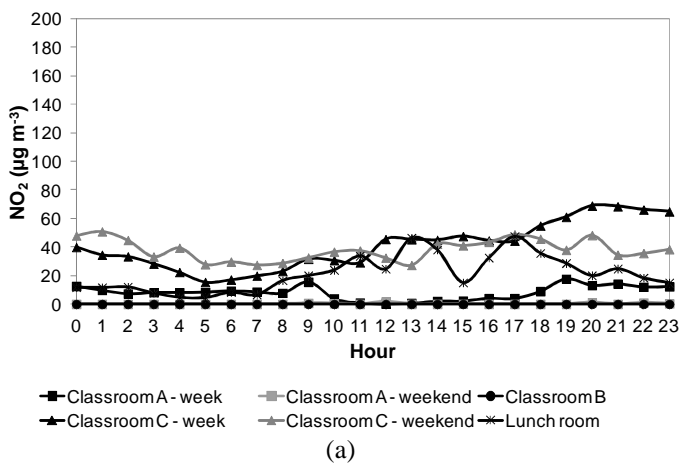
226 3.2 CO, NO₂ and O₃

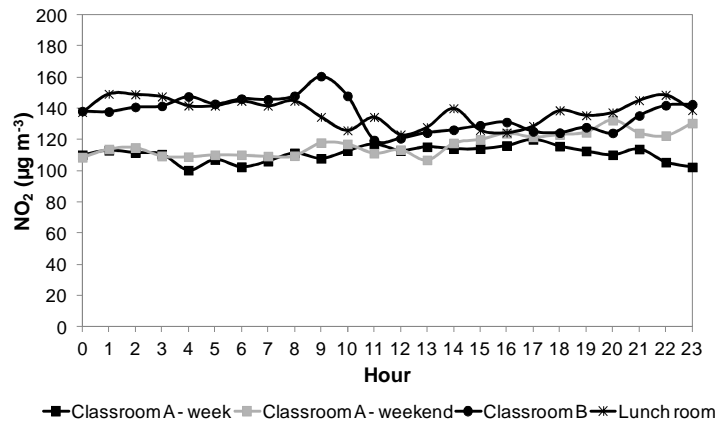
227 Figure 3 shows the CO mean concentrations in all the studied rooms of the four nursery schools
 228 ((a) N_URB1, (b) N_URB2, (c) N_URB3, and (d) N_URB4). It is possible to distinguish a
 229 similarity in the daily profile, especially during weekdays, in all the studied rooms – an increase
 230 in CO concentrations in the early morning and a decrease starting at the evening. During
 231 weekend, CO concentrations seemed to have an almost constant profile along the day. In
 232 general, CO concentrations were significantly lower ($p < 0.05$) in N_URB4 than in the other
 233 three nursery schools. The highest concentrations were found on weekdays in classrooms A and
 234 B of N_URB1 (respectively 4956 and 4347 $\mu\text{g m}^{-3}$) and the lowest were found in classroom B
 235 of N_URB4 (close to zero) (Table 3). In N_URB2, CO concentrations in classroom C were
 236 significantly lower ($p < 0.05$) than in the remaining rooms of that nursery school. In N_URB3,
 237 CO concentrations in classroom B were significantly higher ($p < 0.05$) than in the other rooms
 238 of that nursery school. As there were not found any indoor sources, outdoor CO concentrations
 239 were expected to be the main determinant of the indoor concentrations registered.



240 **Figure 3** – Daily profile of CO mean concentrations registered indoors of a) N_URB1, b)
 241 N_URB2, c) N_URB3, and d) N_URB4.

242 NO₂ mean concentrations registered in N_URB1, N_URB2 and N_URB3 are represented in
 243 Figure 4 a), b) and c), respectively. NO₂ mean concentrations in N_URB4 and in classroom B
 244 of N_URB1 are not represented due to instrument error. The lowest concentrations were found
 245 in N_URB1 and the highest in N_URB3 (Table 2). In fact, in classrooms A (both weekend and
 246 weekdays) and B of N_URB1 concentrations were always very close to zero. Although with
 247 significant differences amongst them ($p = 0.06$), classrooms of N_URB2 (weekdays), as well
 248 as the studied rooms of N_URB3, showed higher values and significantly different profiles (p
 249 < 0.05) than those observed in N_URB1. All of these three buildings were located in a busy
 250 traffic street (N_URB1 and N_URB2 were located in the same street), but N_URB2 and
 251 N_URB3 had a road junction with traffic lights next to the front façade of the building, which
 252 could indicate higher NO₂ emissions from the vehicles exhaust and consequently higher
 253 concentrations of this compound entering into the building. In classroom A of N_URB2, both
 254 in weekdays and weekend, there were found significantly higher values ($p < 0.05$) than in the
 255 rest of that building, probably due to the location of this classroom (in the ground floor and with
 256 windows in the front façade of the building). In the weekend at some classrooms, indoor NO₂
 257 concentrations were higher than in weekdays because as there was no ventilation during the
 258 weekend, the high concentrations observed in Friday did not decrease maintaining a high and
 259 almost constant daily profile during the whole weekend.

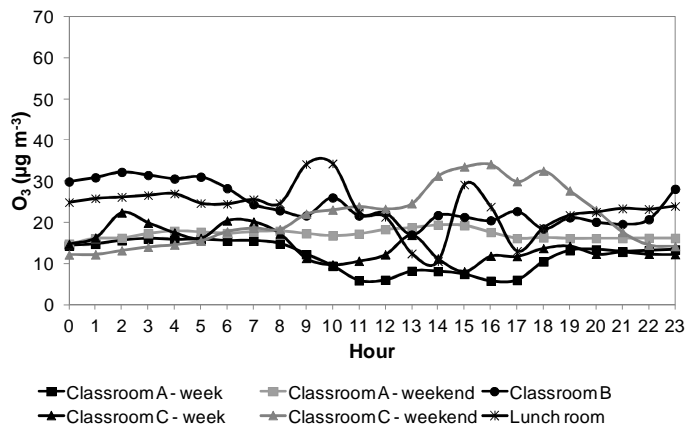




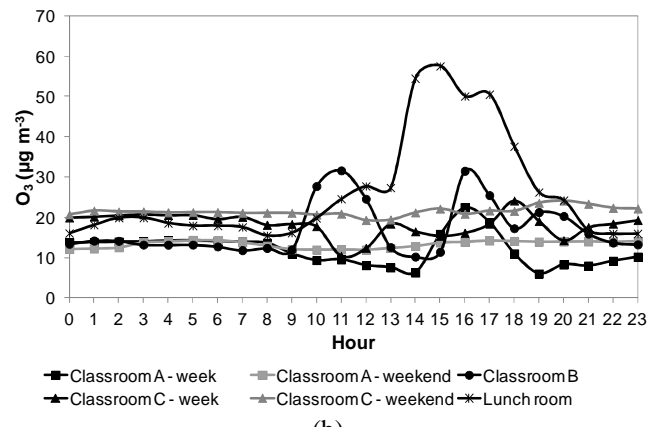
(c)

260 **Figure 4 – Daily profile of NO₂ mean concentrations registered indoors of a) N_URB1, b)**
 261 **N_URB2, and c) N_URB3.**

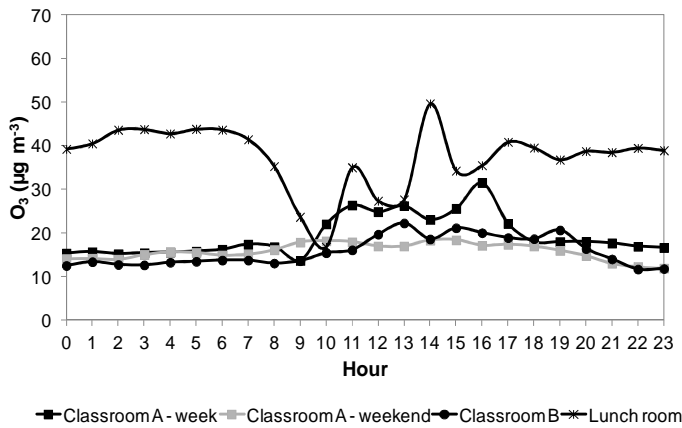
262 Figure 5 a), b) c) and d) shows the O₃ mean concentrations determined in the studied rooms of
 263 N_URB1, N_URB2, N_URB3 and N_URB4, respectively. It is possible to observe O₃
 264 concentrations with a similar order of magnitude among the different studied rooms in the four
 265 nursery schools, and with no relevant variations along the day in all the studied classrooms. The
 266 highest values were often found in the lunch rooms (Table 2) during or immediately after lunch
 267 time, which in the absence of indoor sources might be associated with higher ventilation to
 268 outdoors during daytime. The accumulation in those indoor microenvironments led to the O₃
 269 highest concentrations during the night and dawn found in the lunch rooms of N_URB3 and
 270 N_URB4. In N_URB4, no relevant variations in O₃ concentrations were found in the
 271 classrooms. As there are no indoor sources, O₃ concentrations indoors seemed to be associated
 272 with outdoor concentrations.



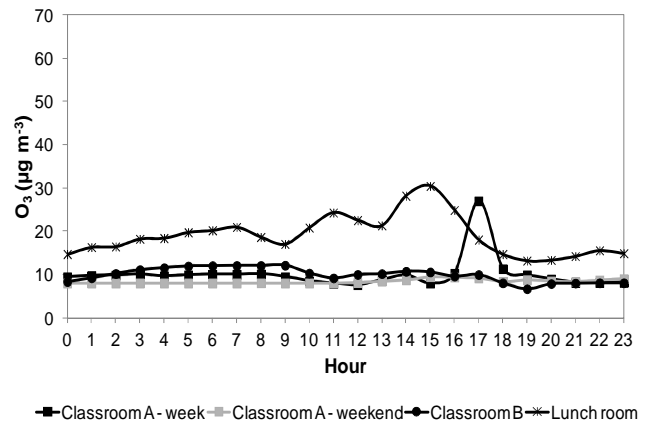
(a)



(b)



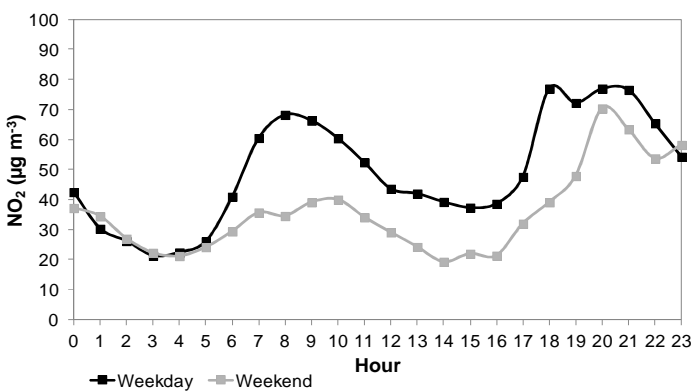
(c)



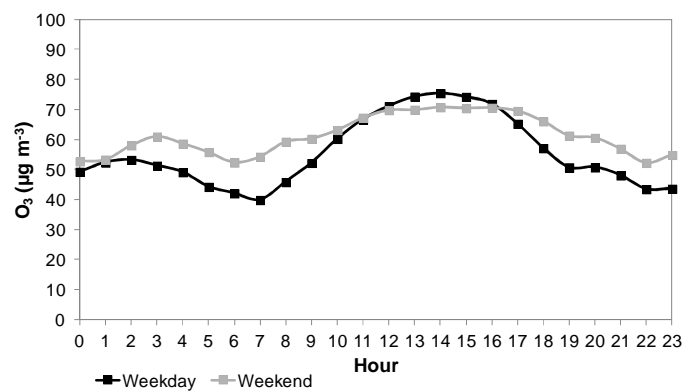
(d)

273 **Figure 5 – Daily profile of O₃ mean concentrations registered indoors of a) N_URB1, b)**
 274 **N_URB2, c) N_URB3, and d) N_URB4.**

275 The outdoor mean concentrations of NO₂ and O₃ allowed obtaining a mean daily profile,
 276 represented in Figure 6 a) and b) respectively. In both NO₂ and O₃ profiles a similar pattern was
 277 found between weekdays and weekend with NO₂ concentrations usually higher on weekdays
 278 and with O₃ concentrations usually higher on weekend. Daily variations in NO₂ concentrations
 279 boiled down to two significant peaks – one in the morning and another at the end of the
 280 afternoon, matching the two traffic rush periods, as expected for urban areas (Wichmann et al.,
 281 2010). From the O₃ outdoor profiles, it is possible to observe the highest concentrations along
 282 the afternoon, as expected (Sousa et al., 2009). These profiles were generally similar to those
 283 typically found indoors, thus outdoor air seemed to be the main contributor to those
 284 concentrations found indoors.



(a)



(b)

285 **Figure 6 – Daily profile of outdoors mean concentrations for a) NO₂ and b) O₃.**

286
287

Table 4 – I/O ratios for NO₂ and O₃: median values observed in each studied site for weekdays and weekends, and respective minima (min) and maxima (max) values.

Nursery	Room	NO ₂		O ₃	
		Weekday	Weekend	Weekday	Weekend
N_URB1	A	0.02 (min-max: 0.00-2.26)	0.00 (min-max: 0.00-0.23)	0.16 (min-max: 0.00-1.15)	0.19 (min-max: 0.16-0.22)
	B	0.00 (min-max: 0.00-0.00)	-	0.33 (min-max: 0.21-0.50)	-
	C	1.88 (min-max: 0.50-4.41)	1.50 (min-max: 0.42-4.88)	0.24 (min-max: 0.03-6.11)	0.22 (min-max: 0.14-0.62)
	LR	0.41 (min-max: 0.00-3.43)	-	0.31 (min-max: 0.05-1.78)	-
N_URB2	A	3.80 (min-max: 1.18-7.88)	6.19 (min-max: 1.63-13.73)	0.25 (min-max: 0.06-0.99)	0.20 (min-max: 0.16-0.50)
	B	2.94 (min-max: 0.93-7.67)	-	0.30 (min-max: 0.10-1.70)	-
	C	2.33 (min-max: 0.36-5.11)	2.42 (min-max: 0.71-4.68)	0.28 (min-max: 0.01-1.07)	0.27 (min-max: 0.18-0.65)
	LR	1.98 (min-max: 0.79-4.20)	-	0.42 (min-max: 0.11-2.49)	-
N_URB3	A	4.20 (min-max: 1.46-15.93)	4.08 (min-max: 1.25-9.77)	0.33 (min-max: 0.17-1.08)	0.30 (min-max: 0.17-3.97)
	B	2.70 (min-max: 1.07-5.65)	-	0.22 (min-max: 0.14-1.07)	-
	LR	6.79 (min-max: 2.99-17.49)	-	0.57 (min-max: 0.26-1.07)	-
N_URB4	A	-	-	0.15 (min-max: 0.13-0.64)	0.19 (min-max: 0.12-0.53)
	B	-	-	0.79 (min-max: 0.12-8.03)	-
	LR	-	-	2.53 (min-max: 0.30-19.88)	-

288

289 Indoor concentrations were compared with those obtained outdoors using the I/O ratio. Outdoor
290 concentrations were obtained from an air quality station instead of measured simultaneously
291 outside each nursery school. Although the air quality station was representative of the study
292 area (CCDR-N, 2011), this might be a study limitation and results should be interpreted with
293 care. Table 4 shows mean I/O ratios (and minima and maxima) for NO₂ and O₃ in each studied
294 room. In N_URB1, NO₂ I/O ratios were usually below 1, showing indoor concentrations lower
295 than outdoors, with the exception of classroom C, both in weekdays and weekend, although
296 there were ratios below 1 in these cases. In the case of N_URB2 I/O median ratios were often
297 above 1, and in N_URB3 all the I/O ratios were also above 1, which might be due to the steep
298 decrease of outdoor concentrations which were not followed by the same decrease indoors. As
299 indoor concentrations of NO₂ in N_URB4 were usually zero, I/O ratios were not represented.
300 O₃ I/O ratios in N_URB1, N_URB2 and N_URB3 were usually below 1 both during weekdays
301 and weekend. In N_URB4, the same was found in classroom A and B, but different results were
302 found in the lunch room (2.53), which might be also due to the steep decrease of outdoor
303 concentrations which were not followed by the same decrease indoors as referred for NO₂.

304 Table 3 shows the exceedances (%) to the standards and guidelines referred in the section 2.2.
305 When there were more than one standard or guideline for the same pollutant, like in the case of
306 CO and NO₂, Table 4 reports the most restrictive one (Portuguese 2006 legislation for CO and
307 WHO guideline for NO₂). Although influenced by outdoor concentrations, the indoor O₃, CO
308 and NO₂ concentrations did not exceed the standards and guidelines used for comparison
309 referred in section 2.2. Zuraimi and Tham (2008) found much higher O₃ concentrations (62.65
310 µg m⁻³), mainly determined by outdoor concentrations, shelf area and table cleaning, but CO
311 concentrations observed in classrooms A and B of N_URB1, classrooms A, B and lunch room
312 of N_URB2 and in N_URB3 were higher than those found in that study (1266.38 µg m⁻³ only
313 determined by outdoor air). On the opposite, lower CO concentrations were found by Yang et
314 al. (2009) (524.42 µg m⁻³) and by Yoon et al. (2011) (812.89 µg m⁻³). Roda et al. (2011)
315 registered indoor NO₂ concentrations comparable to those found in N_URB1 but much lower
316 than those detected in N_URB2 and N_URB3, ranging between 9.0 and 41.0 µg m⁻³, which
317 were determined by outdoor air influence in the absence of indoor sources, mainly due to the
318 proximity to roadways with heavy traffic and by the fact that most of nursery schools'
319 classrooms were located on the ground floor. There were not found exceedances to the
320 Portuguese 2006 and 2013 standards for CO, O₃ or NO₂, which indicates that the registered
321 concentrations of those pollutants are not expected to cause health effects on children attending

322 these nursery schools. As expected, it is possible to observe from the results that the new
323 Portuguese legislation is less restrictive. Exceedances to Portuguese 2006 standards were
324 always higher during occupation periods than on weekdays in general. Moreover, it is also
325 important to refer that the results here presented were similar to those obtained in Portuguese
326 primary schools by Pegas et al. (2012) for NO₂ and VOC. School activity and indoor sources
327 were also identified as increasing loadings of air pollutants in those primary schools, being
328 inadequate ventilation, specific indoor sources (especially for VOC) and outdoor influence
329 (NO₂) the main determinants of IAQ.

330

331 **4. Conclusions**

332 This study allowed a better understanding of the behaviour of several indoor air pollutants in
333 the studied nursery schools, with and without occupation. The influence of outdoor air seemed
334 to be determinant on O₃, CO and NO₂ indoor concentrations, and the observed formaldehyde
335 and TVOC peak concentrations indicated the presence of specific indoor sources for these
336 pollutants, namely materials emitting formaldehyde (mainly furnishing) and products emitting
337 VOC associated to cleaning and children's specific activities (like paints and glues). For
338 formaldehyde, baseline constant concentrations along the day were also found in some of the
339 studied rooms, which enhances the importance of detailing the study of short and long-term
340 children's exposure to this indoor air pollutant.

341 While CO, NO₂ and O₃ never exceeded the national and international reference values for IAQ
342 and health protection, exceedances were found for formaldehyde and TVOC. For this reason, a
343 health risk assessment approach could be interesting for future research to assess the children's
344 health risks of short-term exposure to formaldehyde and to VOC concentrations.

345 Thus, improving IAQ is needed in the studied nursery schools. Besides the measures proposed
346 in Part I of this study, others like changing cleaning activities schedule (after the occupation
347 period), changing materials emitting formaldehyde and better ventilation while using products
348 emitting VOC (amount of fresh air and its distribution), could also be applied to reach the same
349 goal. It is important to take into consideration when applying these changes the conclusions
350 reported by Branco et al. (2014) concerning the concentrations of PM in the rooms. The study
351 of individual VOC should be done to better understand IAQ inside these nursery schools. These
352 recommendations can also be applied in a broader perspective, as the problems found in these

353 nursery schools are similar to others being reported in other nursery and even in primary
354 schools. Additionally, it could be also important to study other nursery schools to help
355 supporting these findings, not only considering urban traffic influence, but also including
356 suburban and rural contexts for comparison.

357

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366 **References**

- 367
- 368 Branco, P. T. B. S., Alvim-Ferraz, M. C. M., Martins, F. G., Sousa, S. I. V., 2014. Indoor air
369 quality in urban nurseries at Porto city: Particulate matter assessment. *Atmospheric*
370 *Environment*. 84, 133-143.
- 371 Branco, P. T. B. S., Alvim-Ferraz, M. C. M., Martins, F. G., Sousa, S. I. V., 2015. Children's
372 exposure to indoor air in urban nurseries-part I: CO₂ and comfort assessment. *Environmental*
373 *Research*. 140, 1-9.
- 374 Carreiro-Martins, P., Viegas, J., Papoila, A. L., Aelenei, D., Caires, I., Araujo-Martins, J., et
375 al., 2014. CO₂ concentration in day care centres is related to wheezing in attending children.
376 *Eur J Pediatr*. 173, 1041-9.
- 377 CCDR-N, 2011. Remodelação da Rede de Medida da Qualidade do Ar da Região Norte -
378 Documento técnico. Comissão de Coordenação e Desenvolvimento Regional do Norte.
379 <http://www2.ccdr-n.pt/fotos/editor2/ambiente/remodelacaorede.pdf>.
- 380 Fonseca, J., Slezakova, K., Morais, S., Pereira, M. C., 2014. Assessment of ultrafine particles
381 in Portuguese preschools: levels and exposure doses. *Indoor Air*. 24, 618-628.
- 382 Franklin, P. J., 2007. Indoor air quality and respiratory health of children. *Paediatric Respiratory*
383 *Reviews*. 8, 281-286.
- 384 Geiss, O., Giannopoulos, G., Tirendi, S., Barrero-Moreno, J., Larsen, B. R., Kotzias, D., 2011.
385 The AIRMEX study - VOC measurements in public buildings and schools/kindergartens in
386 eleven European cities: Statistical analysis of the data. *Atmospheric Environment*. 45, 3676-
387 3684.
- 388 Gładyszewska-Fiedoruk, K., 2011. Analysis of stack ventilation system effectiveness in an
389 average kindergarten in north-eastern Poland. *Energy and Buildings*. 43, 2488-2493.
- 390 HealthCanada, Residential Indoor Air Quality Guidelines. 2013.
- 391 Jones, A. P., 1999. Indoor air quality and health. *Atmospheric Environment*. 33, 4535-4564.
- 392 Lin, L.-Y., Chuang, H.-C., Liu, I. J., Chen, H.-W., Chuang, K.-J., 2013. Reducing indoor air
393 pollution by air conditioning is associated with improvements in cardiovascular health among
394 the general population. *Science of The Total Environment*. 463–464, 176-181.
- 395 Madureira, J., Paciência, I., Rufo, J. C., Pereira, C., Teixeira, J. P., de Oliveira Fernandes, E.,
396 2015. Assessment and determinants of airborne bacterial and fungal concentrations in different
397 indoor environments: Homes, child day-care centres, primary schools and elderly care centres.
398 *Atmospheric Environment*. 109, 139-146.

399 Ministério das Obras Públicas, Transportes e Comunicações. Diário da República - I Série, Nº
400 67, 2416-2468. , Decreto-Lei nº 79/2006.

401 Ministérios do Ambiente, Ordenamento do Território e Energia, da Saúde e da Solidariedade,
402 Emprego e Segurança Social - Diário da República - 1ª Série, Nº 253, 6644(2)-6644(9). ,
403 Portaria nº 353-A/2013.

404 Nunes, R. A. O., Branco, P. T. B. S., Alvim-Ferraz, M. C. M., Martins, F. G., Sousa, S. I. V.,
405 2015. Particulate matter in rural and urban nursery schools in Portugal. *Environmental*
406 *Pollution*. 202, 7-16.

407 Pegas, P. N., Nunes, T., Alves, C. A., Silva, J. R., Vieira, S. L. A., Caseiro, A., et al., 2012.
408 Indoor and outdoor characterisation of organic and inorganic compounds in city centre and
409 suburban elementary schools of Aveiro, Portugal. *Atmospheric Environment*. 55, 80-89.

410 R Development Core Team, 2014. R: A Language and Environment for Statistical Computing.
411 R Foundation for Statistical Computing, Vienna, Austria. URL: <http://www.Rproject.org/>.

412 Roda, C., Barral, S., Ravelomanantsoa, H., Dusséaux, M., Tribout, M., Le Moullec, Y., et al.,
413 2011. Assessment of indoor environment in Paris child day care centers. *Environmental*
414 *Research*. 111, 1010-1017.

415 Sousa, S.I.V., Alvim-Ferraz, M.C.M., Martins, F.G., Pereira, M.C., 2009. Ozone exposure and
416 its influence on the worsening of childhood asthma. *Allergy* 64, 1046-1055.

417 Sousa, S. I., Ferraz, C., Alvim-Ferraz, M. C., Vaz, L. G., Marques, A. J., Martins, F. G., 2012.
418 Indoor air pollution on nurseries and primary schools: impact on childhood asthma--study
419 protocol. *BMC Public Health*. 12, 435.

420 St-Jean, M., St-Amand, A., Gilbert, N. L., Soto, J. C., Guay, M., Davis, K., et al., 2012. Indoor
421 air quality in Montréal area day-care centres, Canada. *Environmental Research*. 118, 1-7.

422 Theodosiou, T. G., Ordoumpozanis, K. T., 2008. Energy, comfort and indoor air quality in
423 nursery and elementary school buildings in the cold climatic zone of Greece. *Energy and*
424 *Buildings*. 40, 2207-2214.

425 WHO, WHO guidelines for indoor air quality: selected pollutants. World Health Organisation,
426 Regional office in Europe, European Series, Copenhagen, Denmark, 2010.

427 Wichmann, J., Lind, T., Nilsson, M.A.M., Bellander, T., 2010. PM2.5, soot and NO2 indoor–
428 outdoor relationships at homes, pre-schools and schools in Stockholm, Sweden. *Atmospheric*
429 *Environment* 44, 4536-4544.

430 Yang, W., Sohn, J., Kim, J., Son, B., Park, J., 2009. Indoor air quality investigation according
431 to age of the school buildings in Korea. *Journal of Environmental Management*. 90, 348-354.

- 432 Yoon, C., Lee, K., Park, D., 2011. Indoor air quality differences between urban and rural
433 preschools in Korea. *Environmental Science and Pollution Research*. 18, 333-345.
- 434 Zuraimi, M. S., Tham, K. W., 2008. Indoor air quality and its determinants in tropical child
435 care centers. *Atmospheric Environment*. 42, 2225-2239.