Indoor air quality in preschools (3- to 5-year-old children) in the Northeast of Portugal during spring–summer season: pollutants and comfort parameters

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ABSTRACT

Indoor air quality at schools (elementary, primary) has been the subject of many studies; however, there are still relative few data regarding preschool (3- to 5-year-old children) environments. This investigation determined the concentrations of particulate matter (PM)2.5, total volatile organic compounds (TVOC), formaldehyde, carbon monoxide (CO), and ozone (O3) as well as the levels of carbon dioxide (CO2), temperature, and relative humidity (RH) in the indoor and outdoor air of two preschools situated in different geographical regions of Portugal. The indoor concentrations of TVOC, CO, O3, and CO2 were predominantly higher at the end of school day compared to early morning periods. The TVOC and CO2 concentrations were higher indoors than outdoors suggest- ing predominantly an indoor origin. Outdoor air infiltrations were the major contributing source of CO and O3 to indoor air in both preschools. The concentrations of all pollutants were within the limits defined by national regulations and international organizations, except for TVOC that exceeded 8–12-fold higher than the recommendation of 0.2 mg/m3 proposed by European Commission. The levels of CO2 were below the protective guideline of 2250 mg/m3 (Portuguese legislation); however, the observed ranges exceeded the Portuguese margin of tolerance (2925 mg/m3) at the end of school days, indicating the impact of occupancy rates particularly at one of the preschools. Regarding comfort parameters, temperature exerted a significant influence on O3 concentrations, while RH values were significantly correlated with TVOC levels in indoor air of preschools, particularly during the late afternoon periods.

Introduction

Outdoor air pollution is a worldwide concern, which was recently classified as carcinogenic to humans by the International Agency for Research on Cancer (IARC) (group 1; IARC, 2016) as there is sufficient evidence to indicate that air contaminants are attrib- uted to cause lung cancer. Exposure to outdoor air pollutants has also been associated with an increased risk of developing cancer of the urinary bladder (IARC, 2016). Several investigators have focused on indoor air quality (IAQ) since individuals spend large portion of time indoors. Factors that predomi- nantly affect IAQ include the existence of various indoor emission sources such as various indoor equipment, human activities, emissions released from buildings through its construction materials, infiltration of outdoor air, and ventilation deficien- cies (Fernández et al., 2013; WHO, 2010). Special attention needs to be given to compounds that may produce and/or potentiate the development of adverse health problems (WHO, 2010). It is note- worthy that the WHO (2010) included formalde- hyde (group 1—carcinogenic to humans; IARC, 2006) and carbon monoxide (CO) as relevant indoor air pollutants, while monitoring of particulate matter (PM), CO, and ozone (O_3) was recommended for outdoor air (WHO, 2006). As a consequence, PM_{10} and $PM_{2.5}$ (particulate matter with 10 and 2.5 µm of aerodynamic diameter, respectively), total volatile organic compounds (TVOC), formaldehyde, CO, and O_3 are among the compounds included in the list of pollutants that need to be monitored in indoor air in Portuguese buildings (Decreto Lei nº 18-2013; Portaria no. 353-A/2013). Once inhaled, PM_{10} is deposited in the upper respiratory system, whereas $PM_{2.5}$ and smaller particles penetrate deeply into the lungs and deposit in the smaller conducting airways and gas exchange regions of lungs (Costa et al., 2014; Kim et al., 2013). The indoor levels of pollutants are influenced by occupation rates and local atmo- spheric conditions. Indoor temperature (T) and rela- tive humidity (RH) may also contribute to accumulation of indoor pollutants (Huang et al., 2016; Sakai et al., 2004; Wolkoff & Nielsen, 2010).

Young children are one of the most vulnerable population groups as their respiratory, immune, reproductive, central nervous, and digestive sys- tems are not completely developed (Bateson & Schwartz, 2008; Burtscher & Schüepp, 2012; Madureira et al., 2015; Makri et al., 2004; Salvi, 2007). In modern society, 3- to 5-year-old children spend up to 85% of their time at homes and/or premises in education settings, namely nurseries, kindergartens, day-care centers, and preschools. These environments constitute children first social integrations, after homes being the places where they spend the majority of their times (approxi- mately 7–8 hr per day). Most of the available studies have focused on children’s exposure to indoor air pollutants at primary and elementary schools (Alves et al., 2014; Annesi-Maesano et al., 2012, 2013; Bakó-Biró et al., 2012; Demirel et al.,2014; Ferreira and Cardoso, 2013, 2014; Guo et al.,2010; Macedo et al., 2013; Madureira et al., 2012,2014, 2015; Pegas et al., 2010, 2011a, 2011b, 2012; Cavaleiro Rufo et al., 2015). IAQ in preschools may vary from primary and higher learning schools due to different conducted activities and behavioral patterns, building, and environmental characteristics including construction materials, cleaning, and ventilation habits (Yang et al., 2009; Vassura et al., 2015; Wichmann et al., 2010). Thus, it would seem that children exposure in preschools may vary from older students’ envir- onments. During the last years, some studies con- cerning exposure of young children, that is, less than 5 years, to indoor air pollutants at educa- tional microenvironments

were reported for: 1) nurseries (Branco et al., 2014, 2015a, 2015b; Mainka et al., 2015; Nunes et al., 2015, 2016), 2) day-care centers and kindergartens (Araújo- Martins et al., 2014; Carreiro-Martins et al., 2014; Cyprowski et al., 2013; Gładyszewska-Fiedoruk, 2013; Yang et al., 2009), and 3) preschools (Fonseca et al., 2014; Latif et al., 2014; Mainka &Zajusz-Zubek, 2015; Oliveira et al., 2015a, 2015b, 2015c, 2016a, 2016b, 2016c; Slezakova et al., 2015; Rawi et al., 2015; Vassura et al., 2015; Wichmann et al., 2010; Yoon et al., 2011). Some of these investigations mainly focused on ventilation and/ or carbon dioxide (CO_2) levels as a global IAQ indicator (Branco et al., 2015a; Carreiro-Martins et al., 2014; Gładyszewska-Fiedoruk, 2013; Latif et al., 2014; Mainka & Zajusz-Zubek, 2015). In Portugal, limited information is available regard- ing preschool children (3–5 years old) exposure to some of the most relevant indoor contaminants at preschools (Fonseca et al., 2014; Oliveira et al., 2015a, 2015b, 2015c, 2016a, 2016b, 2016c;
Slezakova et al., 2015). Further, none of the avail- able studies apparently examined different periods of the day and did not explore the influence of comfort parameters on the levels of indoor pollutants.

The aim of the present study was to: i) monitor the levels of $PM_{2.5}$, TVOC, formaldehyde, CO, and O_3 simultaneously in indoor and outdoor air in two urban Portuguese preschools; ii) examine the impact of comfort parameters, namely CO_2, T, and RH on indoor levels of pollutants; iii) determine the concentrations of studied $PM_{2.5}$ and gaseous pollutants according to the existing guidelines and references for IAQ.

Materials and methods

Characterization of sampling sites

Preschools are educational establishments dedi- cated to learning and social development of 3- to 5-year-old children before their attendance at pri- mary schools. In 2013, a total of 6 429 public and private preschools operated in Portugal with 266,666 registered children (3–5 years old) (Pordata, 2016). Particulate matter$_{2.5}$, four gaseous pollutants (TVOC, formaldehyde, CO, and O_3) and three comfort parameters (CO_2, T, and RH) were simultaneously monitored in indoor and outdoor air of two Portuguese preschools located in different geographical regions (Figure 1). One of the selected preschools (P1) was located in Oporto Metropolitan Area (north of Portugal), the second most densely populated city of the country. In 2013, 44,467 children attended in a total of 151 public and private preschools located in that metropolitan area (Pordata, 2016).

Figure 1. Geographical location of preschools P1 (Oporto) and P2 (Chaves).

Preschool P1 was situated in an urban zone of Oporto city in a close proximity (less than 1000 m) to an exit of one of the major highways sur- rounding the city. The mean traffic density in the streets surrounding the preschool was 16 cars/ min, which increased to 27 and 25 cars/min dur- ing peak hr of 8 a.m. and 6 p.m., respectively. Preschool P2 was situated in Chaves, the second largest city of the district of Vila Real (north of Portugal) where a total of 787 children were regis- tered in 2013 at 35 public and private preschools (Pordata, 2016). P2 was located in the main traffic street that provides direct access to Chaves city center. Immediately in the back area of that pre- school, there was a main city shopping center with a petrol station, which generated a moderate traf- fic density throughout the day (5 cars/min). The traffic density also rose at peak rush hr (7 cars/ min around 8 a.m. and 10 cars/min at 6 p.m.). Both urban-traffic preschools were constructed before 1950 and present a two-floor building structure with a 2–3 m brick-wall base. P1 pre- sented a fence with 4 m above the brick wall. The main structural characteristics of both preschools and meteorological conditions data (solar radia- tion, wind speed, and precipitation) are provided in Table 1S (Supplementary Material).

A total of 173 and 44 preschool children (3–5 years old) were enrolled at P1 and P2, respectively.

An example of children activity and time patterns of 3-year-old children from both preschools is presented in Table 2S. During the sampling cam- paigns, a total of 18–40 and 16–22 children (< 1 student/m^2) were present in the rooms of P1 and P2, respectively.

The selected indoor microenvironments were ventilated through the opening of windows during two particular periods of the school day: i) 5–10 min before children arrivals at school (around 8 a.m.) and approximately during 5–20 min after the end of school day when the room was empty (after 6 p.m.). None of the preschool rooms had mechanical ventilation system. During classes including when some physical activities were per- formed indoors the doors were closed except dur- ing recesses. At night, all windows and doors were kept closed. Both preschool rooms were equipped with closets and/or shelves with stock of art-craft supplies (gouaches, inks, glues). In addition, chil- dren' hand-made

paintings were hung in rooms and/or glued to the walls at P2 classroom. P1 and P2 rooms were cleaned daily at the end of the school day.

Sample collection

Gaseous pollutants and comfort parameters were monitored during 10 consecutive weeks between April and June 2013 at both preschools. Sampling campaigns were performed in an educational playroom (P1) and in one classroom (P2). Both selected rooms were used during the school day for classes and several activities including handi- works with different materials, musical classes, reading, playing, and physical exercises.

$PM_{2.5}$ masses were gravimetrically determined by collection on polytetrafluoroethylene mem- brane filters (Ø47 mm, SKC Ltd., Dorset, UK) with a setup of low-flow (38.3 L/min) pumps (models Bravo H2; TCR TECORA, Paris, France) that were connected to PM EN LVS sampling head (in accordance with norm EN14907). $PM_{2.5}$ sam- pling was conducted continuously for a period of 24 hr over 5 week days (n=50). The concentrations of TVOC, CO, CO_2, and O_3 were measured twice per day at each preschool: i) in early morning between 8 and 9 a.m. which corresponded to the period of children arrivals and ii) in late afternoon between 4 and 5 p.m. before the children left pre- schools. Monitoring was performed for 15–20 min periods (with a logging interval of 30 sec by a multi-gas sensor probe (model TG 502; GrayWolf Sensing Solutions, Shelton, USA) during each weekday, i.e., from Monday morning to Friday afternoon). The concentrations of formaldehyde were registered with a Formaldemeter™ (model htV-M; PPM Technology, Caernarfon, UK). Equipment was calibrated by manufacturers before the beginning of sampling campaigns, and the analytical response was checked daily according to manufacturer specifications. All parameters were monitored at the breathing level of preschool children (approximately at 0.4–1 m above the floor) and preferably in the center of the rooms in order to minimize the influence of outdoor sources. The levels of $PM_{2.5}$ (n=50) gaseous pollu- tants and CO_2 were concomitantly monitored in the outdoor areas of each preschool. Measurements were conducted in places where the sampling interferences from trees, walls, and/ or fences would be minimal. Indoor and outdoor T and RH were monitored by Testo mini data- logger (model 174H; Testo AG, Lenzkirch, Germany). Precaution was taken to ensure the safety of children. Indoor potential source activ- ities, number of children, door, and window posi- tions, as well as ventilation status were registered daily in selected indoor rooms.

Data analysis

Statistical treatment was performed using SPSS (IBM SPSS Statistics 20) and statistical software (v. 7, StatSoft Inc., USA). Statistical significance was defined as $p \leq 0.05$.

Results

Pollutants and comfort parameters

The mean and range concentrations of $PM_{2.5}$ in indoor air of preschools P1 and P2 are illustrated in Figure 2a.

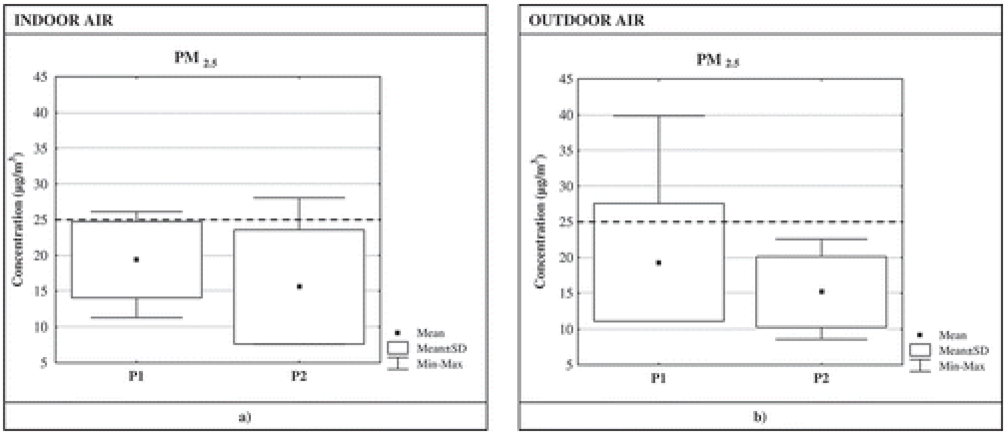

Figure 2. PM2.5 concentrations in indoor (a) and outdoor (b) air of preschools P1 and P2. The horizontal dashed lines represent guideline values of 25 µg/m3 (Portuguese Regulation, 2013; WHO, 2006 WHO. 2006. World Health Organization. Air Quality Guidelines Global Update 2005. Copenhagen, Denmark: World Health Organization Regional Office for Europe.

There were no significant differences (11.2–26.1 µg/m^3 at P1 and 7.58–28.1 µg/m^3 at P2) detected. The levels of indoor TVOC, CO, and O_3 during early morning and late afternoon periods at preschools P1 and P2 are presented in Figure 3. The concentrations of gaseous pollutants inside rooms ranged widely throughout the day. At both preschools, the levels of TVOC, CO, and O_3 were significantly higher at the end of the school day than at early morning period, except for TVOC and CO at P2 (Figure 3). Inter-comparison between preschool indoor air revealed that con- centrations of TVOC (P1: 1.84 ± 1.24 mg/m^3 and 2.5 ± 0.81 mg/m^3; P2: 2.5 ± 0.52 mg/m^3 and 1.67 ±

0.67 mg/m^3, respectively, for early morning and late afternoon periods) and formaldehyde (0.03 ± 0.02; 0.01–0.06 mg/m^3 at P1 *versus* 0.04 ± 0.02; 0.01–0.09 mg/m^3 at P2) were higher at P2 (Figure 3; Table 1). The concentrations of formal- dehyde in indoor air of both preschools reached maximal values of 0.06 and 0.09 mg/m^3, respec- tively, at P1 and P2 (Table 1). At preschool yards, formaldehyde concentrations were always below the limit of detection (LOD) of the equipment (12.5 µg/m^3). In addition, P1 displayed markedly higher levels of CO (2.5-fold) and O_3 (2.4-fold) than P2. Regarding CO_2, indoor concentrations (796–3606 mg/m^3 at P1; 686–2686 mg/m^3 at P2) were significantly greater between both preschools only for late afternoon periods (Figure 4). Finally, regarding other comfort parameters, both indoor means of T and RH rose numerically (11 for T and 1.2% for RH) from the morning to the afternoon periods at P1. At P2, the indoor T increased 23%, while RH fell by 43% (Table 2).

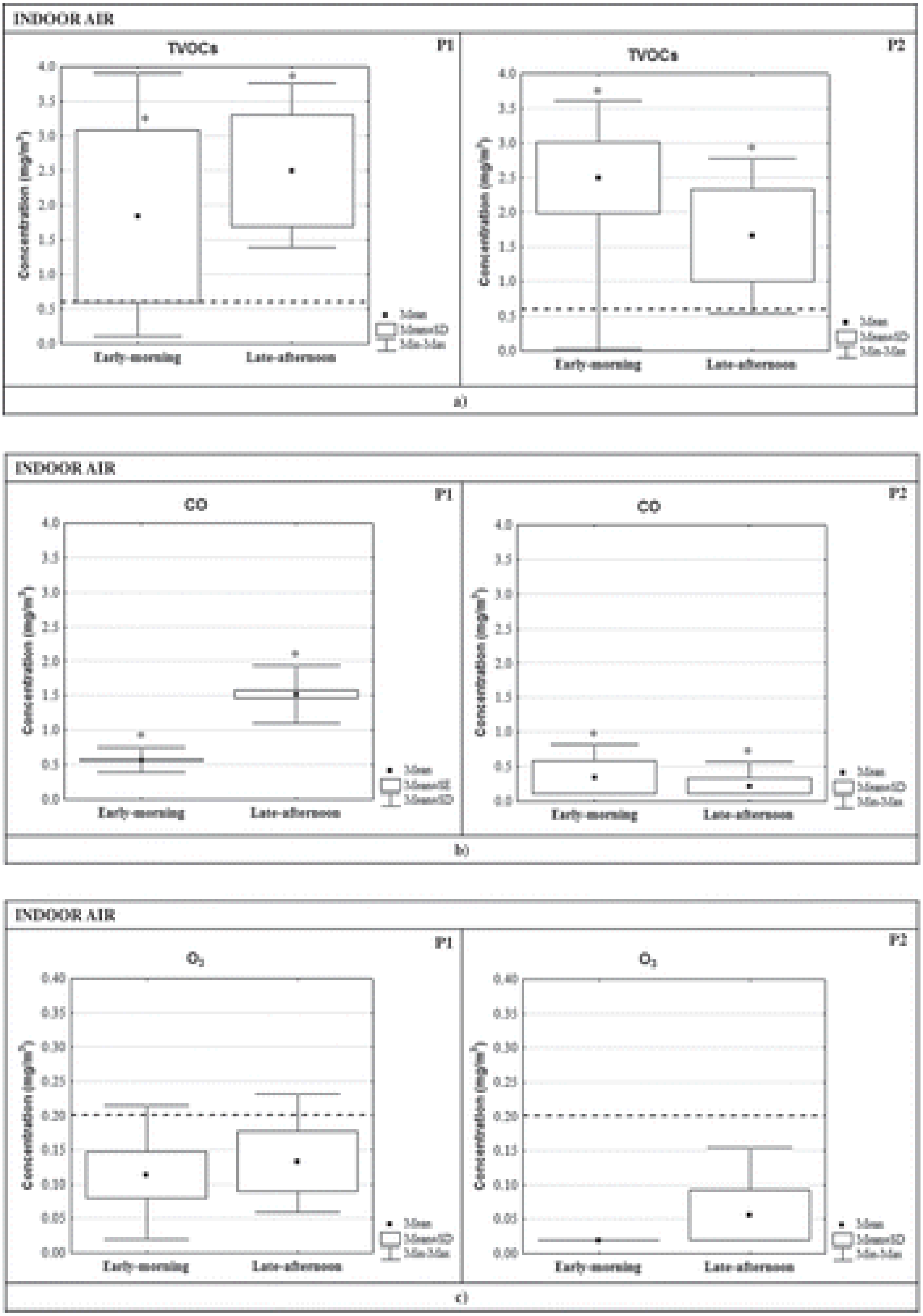

Figure 3. Levels of indoor gaseous pollutants: a) total volatile organic compounds (TVOC); b) carbon monoxide (CO); c) ozone (O3) in indoor air of the studied preschool P1 and P2 (mg/m3). The horizontal dashed lines represent guideline values of 0.6 mg/m3 for TVOC and maximum reference concentration of 0.2 mg/m3 for O3 in indoor air of Portuguese buildings (Nota Técnica NT-SCE-02, 2006 Nota Técnica NT-SCE-02. 2006; Portaria no. 353-A/2013). *Significant differences ($p \leq 0.05$) for nonparametric Mann–Whitney U-test between early morning and late afternoon pollutant concentrations for each preschool.

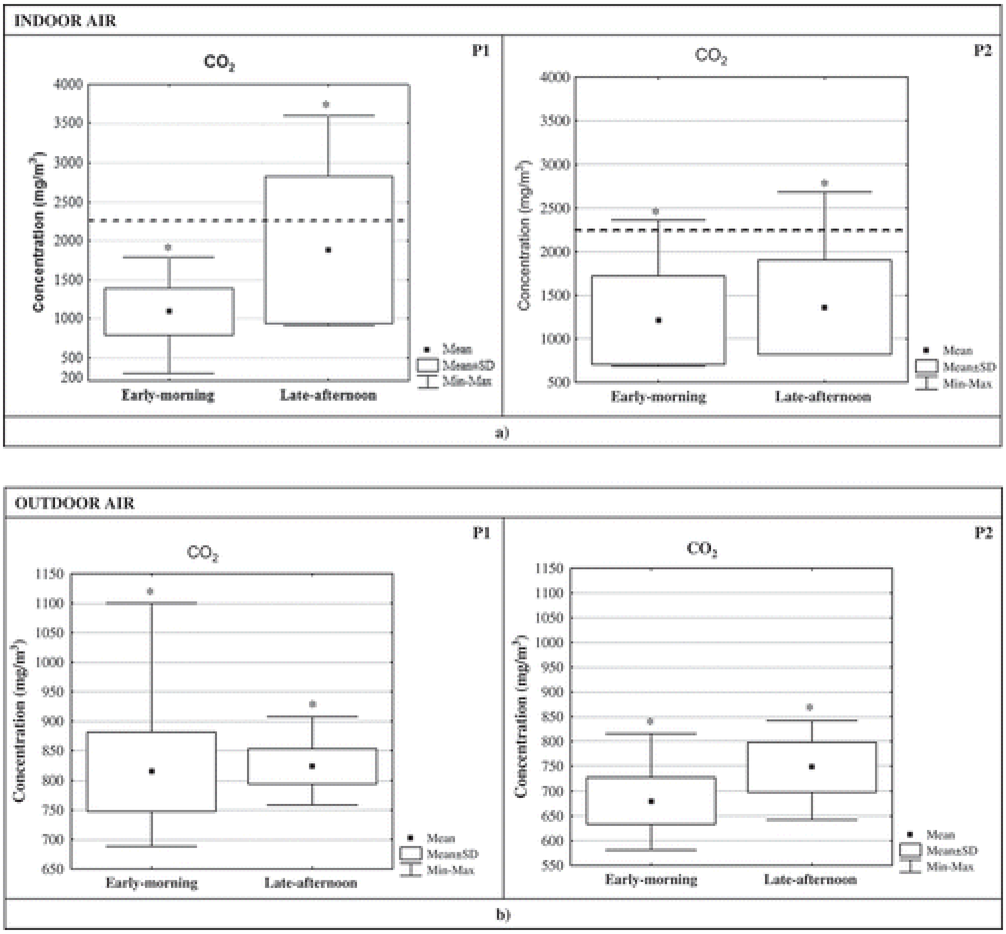

Figure 4. Concentrations of carbon dioxide (CO2) at preschools P1 and P2 in: a) indoors; b) outdoor school areas. The horizontal dashed line represents Portuguese indoor guideline values of 2250 mg/m3 (Portaria no. 353-A/2013)*Significant differences at p≤0.05 for nonparametric Mann–Whitney U-test between early morning and late afternoon levels for each preschool.

The concentrations of $PM_{2.5}$ in outdoor air of P1 and P2 are given in Figure 2b. No significant differences were observed between both schools. The concentrations of gaseous pollutants in ambi- ent air of both preschools are shown in Figure 5. The ambient levels of TVOC were markedly higher in the early morning hr than at the end of the day at both preschools. Further, early morning TVOC concentrations were significantly higher at P2 than at P1. CO and O_3 levels were significantly higher in late afternoon period than in early morn- ing hr at P1, while at P2, only the mean concen- trations of O_3 were significantly higher at the end of the school day. Similar to the findings for indoor air, early morning and late afternoon ambi- ent concentrations of CO_2 (814 and 824 mg/m^3 at P1 *versus* 680 and 748 mg/m^3 at P2), CO (0.62 and 1.81 mg/m^3 at P1 *versus* 0.41 and 0.62 mg/m^3 at P2; 1.5–3-fold higher), and O_3 (0.15 and 0.26 mg/ m^3 at P1 *versus* 0.05 and 0.19 mg/m^3 at P2; 1.4–3- fold higher) were significantly greater at P1 than at P2. Throughout the day, mean values of outdoor T rose 29 and 41%, while

RH decreased 29 and 37%, respectively, at P1 and P2 (Table 2).

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Table 2. Indoor and outdoor levels (mean and standard deviation; range) of temperature (T) and relative humidity (RH) at the preschools P1 and P2.

	Indoor air		Outdoor air	
	T (ºC)	RH (%)	T (ºC)	RH (%)
P1				
	25.4 ± 2.1	42.7 ± 13.7	25.5 ± 4.3	42.1 ± 18.8
	(21.8–32.0)	(19.6–71.2)	(17.3–36.1)	(17.0–89.6)
Early morning	24.8 ± 1.6	42.6 ± 14.6	23.2 ± 2.8	45.5 ± 21.1
	(21.8–28.2)	(19.6–71.2)	(17.3–30.8)	(17.0–89.6)
Late afternoon	27.5 ± 2.3	43.1 ± 9.6	30.1 ± 3.0	35.2 ± 10.1
	(24.6–32.0)	(30.9–57.2)	(26.5–36.1)	(22.0–51.6)
P2				
	20.6 ± 3.1	54.0 ± 11.8	19.9 ± 5.7	51.4 ± 15.7
	(14.4–29.4)	(24.6–74.5)	(12.4–29.4)	(24.7–77.3)
Early morning	18.7 ± 1.9	62.5 ± 4.9	16.4 ± 2.9	59.6 ± 10.6
	(14.4–21.5)	(48.8–74.5)	(12.8–23.0)	(42.5–77.3)
Late afternoon	23.0 ± 2.7	43.7 ± 9.1	23.2 ± 5.7	43.5 ± 15.7
	(19.9–29.4)	(24.6–56.2)	(12.4–29.4)	(24.7–76.5)

Indoor-to-outdoor ratios

The levels of TVOC and CO_2 were significantly higher in indoor air compared with outdoors (TVOC: 1.84 *versus* 1.4 mg/m^3 for early morning hr and 2.5 *versus* 1.3 mg/m^3 during late after- noon periods at P1, 2.5 *versus* 1.91 mg/m^3 at morning hr and 1.67 *versus* 1.21 mg/m^3 at the end of the school day at P2; CO_2: 1090 *versus* 814 mg/m^3 during the morning and 1881 *versus* 824 mg/m^3 at the end of afternoons at P1, 1213 *versus* 680 mg/m^3 during early morning periods and 1362 *versus* 748 mg/m^3 during the end of school day at P2). Mean indoor-to-outdoor (I/O) ratio of concentrations of both pollutants was higher than unity at both preschools (TVOC: I/ O = 1.3–1.9 at P1, 1.3–1.4 at P2; CO_2: I/O = 1.3– 2.3 at P1, 1.7–1.8 at P2). In addition, I/O ratios of $PM_{2.5}$ concentrations > 1 were attained with values ranging between from 1.06 to 1.69 at P1 and from 1.27 to 1.87 at P2. A different profile was noted for levels of CO which were signifi- cantly higher in preschool outdoor compared with indoor air of the selected rooms (respec- tively, for early morning and late afternoon per- iods: 0.62 *versus* 0.57 mg/m^3 and 1.81 *versus* 1.52 mg/m^3 at P1; 0.41 *versus* 0.36 mg/m^3 and 0.62 *versus* 0.23 mg/m^3 at P2). Similar observations were also found for concentrations of O_3 (0.15 *versus* 0.11 mg/m^3 during morning hr and 0.26 *versus* 0.13 mg/m^3 for afternoons at P1; 0.05 *versus* 0.02 mg/m^3 for early morning periods and 0.19 *versus* 0.06 mg/m^3 at the end of the school day at P2). For these pollutants (CO and O_3), I/O ratios were below unity at both pre- schools (CO: I/O = 0.84–0.92 and 0.37–0.89; O_3: I/O = 0.50–0.73 and 0.31–0.4, respectively, at P1 and P2).

Table 1. Concentrations of formaldehyde (mg/m^3) in indoor air of preschools P1 and P2.

	Mean	Min	Percentile 25	Percentile 75	Maximum	*p*
P1	0.03	0.01	0.02	0.04	0.06	0.002
P2	0.04	0.01	0.02	0.05	0.09	

*Significant differences at $p \leq 0.05$ for nonparametric Mann–Whitney U-test between both preschools.

Correlations between comfort parameters and pollutants

Since both preschools were naturally ventilated, Spearman correlation coefficients (r) were determined to estimate the influence of T, RH, and CO_2 on levels of gaseous pollutants in indoor and outdoor air of both preschools. The degree of correlation may be used as an indicator of pollutant concentration dependency on the comfort parameters. A significant negative cor- relation was observed between T and RH in outdoor air of both preschools. Similar findings were noted in indoor air of rooms with correla- tions significant during the late afternoon per- iod. In outdoor air of both preschools, CO_2 levels were significantly associated with T values with stronger correlations observed during early morning periods when the impact of traffic emissions was expected to be lower. Similar findings were found in indoor air of P2.

$PM_{2.5}$ levels in indoor air were inversely related to respective concentration outdoors at both pre- schools indicating once again the influence of indoor sources. RH and CO_2 values also showed a negative correlation with $PM_{2.5}$ concentrations in indoor and outdoor air at both preschools.

Regarding the levels of gaseous pollutants in indoor air of preschools, T values were markedly associated with concentrations of O_3, with greater correlations during the late afternoon periods. RH values were significantly correlated with TVOC in indoor air of both preschools. A significant asso- ciation was also noted between RH and CO_2 levels during the afternoon period at P1 and P2. In addition, CO_2 levels were markedly associated with concentrations of TVOC and CO monitored during the afternoon periods in indoor air of both preschools.

For preschool ambient air, T values were sig- nificantly associated with CO and O_3 levels in both early morning and late afternoon periods. The correlations appeared to be greater during the afternoon periods at both preschools.

Discussion

Air pollutants

In what specifically applies to school environ- ments, the air, school activities, cleaning activities, cooking and emissions from printers, and photo- copy machines are the predominant PM indoor sources (Blondeau et al., 2005; Destaillats et al., 2008; Yi et al., 2016). Regarding preschools in this study, it was found that indoor levels were numerically higher than outdoors for both pre- schools.

The determination of I/O ratios enable estimation of an indoor/outdoor impact of a pol- lutant on indoor areas and the potential identification of their origin, with I/O ratios >1 indicating the existence of indoor emission sources. $PM_{2.5}$ ratios suggested the presence of indoor sources predominantly due to re-suspen- sion of indoor dusts such as soil particles, cloth fibers, and building materials deterioration asso- ciated with schoolchildren activities. Yoon et al. (2011) and Amato et al. (2014) also noted that $PM_{2.5}$ I/O ratios were higher than unity in Korean and Spanish preschools. $PM_{2.5}$ mean levels in indoor and outdoor air of both preschools were lower than the guideline quantities of 25 $\mu g/m^3$ (Figure 2) (Decreto Lei nº 118-2013, Portaria no. 353-A/2013; WHO, 2006). At both preschools, sampling campaigns were performed over 24-hr; however, the inclusion of the periods without stu- dents and their activities such as during nights when classrooms were empty and all windows and doors were closed may contribute to lower mean levels of $PM_{2.5}$. The results obtained in this study were also lower than concentrations reported by Mainka and Zajusz-Zubek, (2015), Oliveira et al. (2015b, 2015c), and Rawi et al. (2015), which may be attributed to varying sam- pling strategies (8- or 24-hr sampling periods), different seasonal and/or meteorological condi- tions and to various environmental characteristics. The findings of this study are in agreement with those of Madureira et al. (2012), who also detected negative correlations between RH and CO_2 levels with PM_{10}, $PM_{2.5}$, and PM_1 in indoor air of Portuguese primary schools.

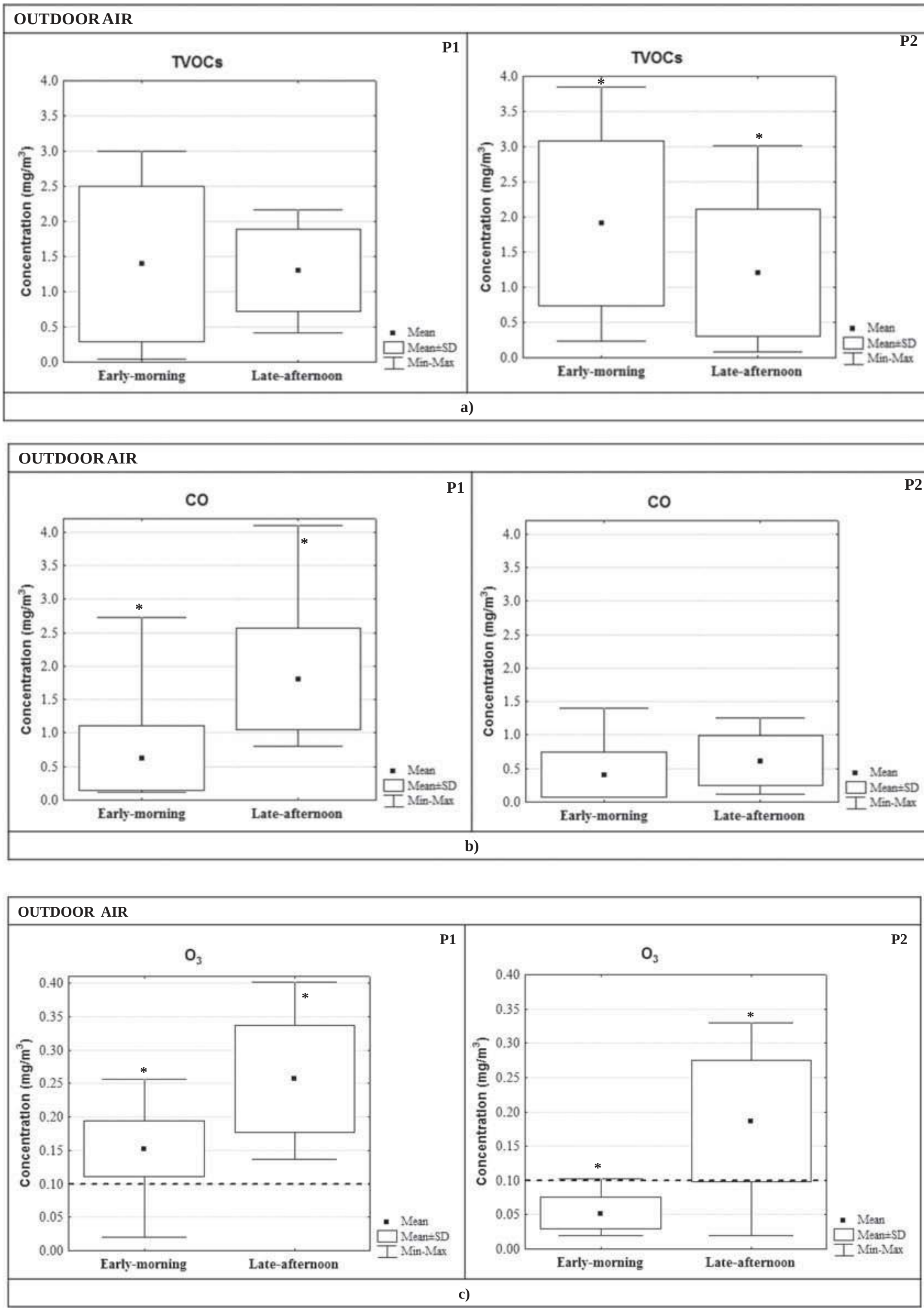

Figure 5. Levels of outdoor gaseous pollutants: a) total volatile organic compounds (TVOC); b) carbon monoxide (CO); c) ozone (O_3) in the ambient air of preschool P1 and P2 (mg/m^3). The horizontal dashed line represents WHO guideline of 0.1 mg/m^3 for O_3 (WHO, 2006). *Significant differences at $p \leq 0.05$ for nonparametric Mann–Whitney U-test between early morning and late afternoon pollutant concentrations for each preschool.

At both preschools TVOC concentrations were significantly higher indoors than outdoors. TVOC I/O ratios suggested the prevalence of indoor sources, which is in agreement with observations of other investigators (Araújo-Martins et al., 2014; Mainka et al., 2015; Nunes et al., 2016; Rawi et al., 2015; Vassura et al., 2015; Yoon et al., 2011). Among the many gaseous pollutants examined by Vassura and coworkers (2015), VOC and princi- pally aldehydes were the compounds that pre- sented the highest indoor source emissions. Furniture and wooden products containing VOC- based resins as well as insulating materials, textiles, and products including paints, wallpapers, glues, adhesives, varnishes, and lacquers constitute the major sources of VOC in indoor environments (Franklin, 2007; McGwin Jr et al., 2010; Sarigiannis et al., 2011; Paciência et al., 2016). Cleaning products are a mixture of water and surfactants that contain VOC in their composi- tion. In addition, some electronic equipment such as computers, printers, and copier machines constitutes potential indoor sources of VOC (Destaillats et al., 2008). Outdoor TVOC levels observed at P1 were in similar range during early morning and late afternoon periods, while the respective levels indoors were significantly higher at the end of the school day, possibly due to accumulation of TVOC in indoor air as a conse- quence of deficient ventilation throughout the day. Regarding P2, higher concentrations of TVOC were found during the morning period than at the end of the day indicating a sufficient ventila- tion of the classroom throughout the day. At both preschools, early morning concentrations were higher than the respective outdoor levels, which might be due to accumulation of emissions during the night period as well as from cleaning (at the end of the classes). P2 classroom presented higher levels of TVOC than P1 educational playroom possibly due to room decoration and materials (children' handmade drawings and the presence of art craft supply that were suspended from the ceiling and/or glued to the walls). These findings are in agreement with the results reported by Rawi et al. (2015) and Godwin & Batterman (2007). Indoor morning and afternoon concentrations of TVOC were higher than levels reported in the indoor air of Malay (0.08 ppm at Balakong area and 0.11 ppm at Bangi area (Rawi et al., 2015)), Italian (0.008–0.04 ppm (Vassura et al., 2015)), and Korean (0.59 mg/m^3, 0.07–1.93 mg/m^3 (Yoon et al., 2011)) preschools. Regarding Portuguese data, P1 and P2 TVOC indoor levels were also higher than concentrations reported in 14 primary schools situated in Lisbon during the spring season (37–317 µg/m^3 (Pegas et al., 2011b), 300 ± 330 µg/m^3 (Pegas et al., 2011a; Paciência et al. 2016). The mean concentrations of TVOC in indoor air of both preschools were 3–4 and 8–12- fold higher than the available recommendations for IAQ at Portuguese public buildings (0.6 mg/ m^3 over 8-hr sampling period; Decreto Lei nº 118-2013, Portaria no. 353-A/2013) and European Collaborative Action report (0.2 mg/ m^3; ECA, 1997), respectively. It is important to mention that sampling of the gas pollutants was performed during short periods (15–20 min in the early morning and late afternoon hr) as recom- mended by the national guidelines of that time (minimum of 5 min sampling periods; Nota Técnica NT-SCE-02, 2006). At the end of 2013, the national guidelines were changed to 8-hr con- tinuous sampling when assessing indoor air pollu- tion. Thus, the concentrations reported in this study need to be considered with caution as these values might not represent 8-hr exposure noted in the current national guidelines (Decreto Lei nº 118-2013; Portaria no. 353-A/2013). However, the observed levels may represent risks for the respec- tive population of children at P1 and P2 since exposure to VOC

may irritate eyes, nose, and throat and produce headaches, nausea, damage to the liver, kidneys, as well as some neurological symptoms (Annesi-Maesano et al., 2013; Demirel et al., 2014; Le Cann et al., 2011; Paciência et al., 2016; Sarigiannis et al., 2011; WHO, 2010); increased asthma severity, allergy, and airway inflammation in children were also reported pre- viously (Annesi-Maesano et al., 2013; Franklin, 2007; Lee et al., 2014; McGwin Jr et al., 2010; Zhao et al., 2008; Paciência et al., 2016). Further, highly reactive unsaturated carbon-carbon VOC may react with O_3, which results in production of short lived and highly irritating pollutants that may also produce long-term adverse health effects. Future studies need to include the assessment of some other specific (individual) VOC, such as benzene, toluene, styrene, trichloroethylene, and tetrachloroethylene that are known to produce adverse health effects (Portaria no. 353-A/2013; Valcke & Haddad, 2015).

Formaldehyde is one of the most studied VOC (Weschler, 2009). The predominant sources of for- maldehyde in indoor air in Portuguese preschools are insulating materials, plywood furniture con- taining formaldehyde-based resins as well as water-based paints, and cleaning agents among other building materials that contain urea formal- dehyde resins (Franklin, 2007; Sarigiannis et al., 2011). Indoor levels of formaldehyde at Portuguese preschools were within the range reported by Yoon et al. (2011) in Korean pre- schools but higher than those observed for Italian preschools (Vassura et al., 2015). As formaldehyde exposure may induce both short- and long-term adverse health effects, a guideline value of 0.1 mg/ m^3 was recommended in order to prevent sensory irritation for short-term exposure (30 min) in the general population; 0.21 mg/m^3 was recommended for chronic effects, including cancer (Decreto Lei nº 118-2013, Portaria no. 353-A/2013; WHO, 2010). In the rooms of both preschools, formalde- hyde levels were well below those limits.

Indoor levels of CO are mostly attributed to emissions of combustion sources that may occur in indoor environments and from infiltration of ambient air. The mean early morning and late afternoon indoor levels of CO at both preschools were similar to indoor concentrations reported for urban Korean preschools (Yoon et al., 2011) and lower than levels found at Balakong and Bangi area at Malay preschools (Rawi et al., 2015). Regarding European countries, Vassura et al. (2015) examined the IAQ in Italian preschools located at a suburban area of the province of Bologna and detected lower levels of CO than in this study. It should be noted that indoor levels of CO depend upon the interaction of different inter- related factors, namely type, nature, and number of CO sources as well as building characteristics, infiltration, and/or ventilation rates and levels of CO in outdoor areas (EHC, 1999) which might account for the observed differences. Overall, both indoor and outdoor CO concentrations were lower than the established value of 10 mg/ m^3 (8-hr mean) set in national legislation (Decreto Lei nº 118-2013, Portaria no. 353-A/2013; European Union, 2008; WHO, 2010). I/O ratios demonstrated that outdoor air penetration was the predominant indoor source at P1 and P2, in agree- ment with findings reported by Branco et al (2015b), Nunes et al. (2016), and Yoon et al (2011). Differences between indoor and outdoor CO levels were higher at P1 than at P2 (Figures 3 and 5), possibly due to greater traffic density. Traffic emissions were reported as the major source of ambient CO in Oporto, Portugal (Slezakova et al., 2011).

According to Portuguese legislation, a maxi- mum reference concentration of 0.2 mg/m^3 is

recommended for O_3 indoor air in Portuguese buildings (Nota Técnica NT-SEE-02, 2006). During the sampling campaigns, O_3 indoor con- centrations were significantly higher at the end of the school day than during the morning periods, with total mean always below the maximum limit of 0.2 mg/m^3.

However, at P1, a maximal concen- tration of 0.23 mg/m^3 was reached at end of the school day (Figure 3). The predominant indoor sources for O_3 include electronic equipment such as old printers and photocopy machines as well as air humidifiers (Destaillats et al., 2008), but such equipment was not used in the rooms of both preschools. In addition, P1 levels were higher than those observed at P2. The I/O ratios sug- gested infiltration of outdoor emissions as the major source of O_3 indoors, which agrees with data reported in previous studies (Branco et al., 2015b; Demircigil et al., 2014; Demirel et al., 2014). In 2014, the WHO daily maximum 8-hr mean concentration was lowered from 0.12 to 0.1 mg/m^3 based on conclusive associations between daily mortality and O_3 concentrations (EEA, 2015). The total mean outdoor concentrations exceeded the guideline of 0.1 mg/m^3 (WHO, 2006), reaching maximal levels of 0.4 mg/m^3 and 0.33 mg/m^3 during the end of school day at P1 and P2, respectively. The EEA pointed out that during 2013 the daily maximum of 0.1 mg/m^3 was exceeded more than 25-fold in 18 of the 28 European Union countries (EEA, 2015) with the highest concentrations observed in some Mediterranean countries, including Portugal.

The sampling campaigns performed in both preschools monitored indoor and outdoor levels of relevant pollutants during limited periods of the school day (15–20 min in the early morning and 15–20 min at late afternoon). Since the national and international organizations recommend mon- itoring during a consecutive period of 8-hr, the levels reported in this study may not represent the overall 8-hr mean preschool children exposure. However, it is expected that early morning and late afternoon levels might include the overall ranges of schoolchildren exposure at preschools.

Comfort parameters

Thermal comfort may exert a significant impact on the general well-being and daily performance of building occupants (Mendes et al., 2013). Concerning IAQ, the Portuguese legislation recommends indoor T ranging from 20ºC for the cooling season (winter) to 25ºC during the hot season (summer) and RH of 50% during the overall year (Decreto Lei nº 118-2013). This investigation was performed during spring and beginning of summer, and the obtained means of T and RH at both P1 and P2 schools (Table 2) were generally within the recommen- dations for indoor air. The considered pre- schools were both naturally ventilated, and thus, fluctuations of T and RH were a conse- quence of local climate conditions. The negative correlations observed between T and RH were expected since the increase in T is frequently associated with a reduction in RH values. Humidity is one of the critical factors that affect emissions of VOC from building materials (Huang et al., 2016). In this study, it was found that RH values were significantly corre- lated with levels of TVOC in indoor air in both preschools. It is known that higher T, RH, and larger wood-based surfaces promote emission of VOC from different

construction materials (Sakai et al., 2004; Wolkoff & Nielsen, 2010). Several investigators examined wood-based pro- ducts containing urea–formaldehyde glue and reported that VOC emissions are proportional to RH at a specific T (Myers, 1985; Van Netten et al., 1989), while T dependence is more com- plex (Zhang et al., 2007).

The number of occupants and their activities inside a room also influences IAQ. The American Society of Heating, Refrigerating, and Air- Conditioning Engineers (ASHRAE) recommend a maximum of 25 occupants (5–8 years old) for an acceptable IAQ in a room with 100 m^2 (ASHRAE, 2013). National regulation estimates a mean occu- pation of 20–25 children per each classroom (Despacho nº 5048-1B/2013). Occupation rates at P1 exceeded those recommendations on some school days, particularly during the afternoon peri- ods at P1 (18–40 children). Occupants are respon- sible for the ventilation status of indoor environments through control of the ventilation system and/or the opening of windows. Exhaled air is usually the largest source of CO_2 in class- rooms, and thus, it is frequently used as a screening tool to evaluate if adequate volumes of fresh outdoor air are being introduced indoors. Therefore, CO_2 might be used as an efficient indicator of occupancy and ventilation status in indoor microenvironments. The early morning and late afternoon mean indoor concentrations of CO_2 were below the protective guideline of 2250 mg/m^3 (Portaria no. 353-A/ 2013). However, during the afternoon periods, levels of CO_2 exceeded that recommendation (Figure 4a). At P1, the maximal levels of CO_2 exceeded the 30% margin of tolerance (2925 mg/m^3) that might be applied when no mechanical ventilation is being utilized at the selected indoor rooms (Portaria no. 353-A/2013). It is important to note that during afternoon periods, 3- to 5-year-old preschool chil- dren (principally the youngest ones) slept for 2 hr after lunch (Table 2S). During that time, windows and doors were kept closed, which probably con- tributed to the lower air exchange rate and thus higher CO_2 indoors, especially at P1, where the number of children was greater. Similar observa- tions were reported previously in urban nurseries (Branco et al., 2015a; Yang et al., 2009; Yoon et al., 2011). Inadequate ventilation may be generated by insufficient air volume, high levels of recirculation, incorrect placement of the ventilation points, and deficient distribution that exits certain areas without ventilation (Fernández et al., 2013). Ambient levels of CO_2 ranged between from 688 to 1100 mg/m^3 at P1 and from 581 to 842 mg/m^3 at P2 (Figure 4b). The outdoor concentrations of CO_2 at the Oporto preschool (P1) were higher than levels reported during the summer season (180–443 mg/m^3 (Madureira et al., 2012) and 280–520 mg/m^3

(Mendes et al., 2013)), but similar to concentrations detected by Mendes et al. (2013) during a winter season (250–2050 mg/m^3). It is known that different atmospheric conditions and varying local sources markedly influence levels of CO_2 and ambient air gaseous pollutants. As expected, indoor CO_2 concentrations were always higher than outdoors at both preschools (Figure 4), mainly due to room occupancy and some apparent ventilation deficien- cies. WHO (2000) and ASHRAE (2013) recommended a minimum permissible CO_2 concentration of 1800 mg/m^3. At both preschools, differences between indoor and outdoor CO_2 means were, respectively, 276 and 1057 mg/m^3 at P1 for early morning and late afternoon periods, and 533 and 614 mg/m^3 at P2, always below the 1800 mg/m^3 permissible limit even at the end of the school day. Shendell and colleagues (2004) examined the asso- ciation of student absence with measurements of indoor minus outdoor CO_2 concentrations and found that 45% of the 434 American studied class- rooms had short-term CO_2 levels above 1800 mg/ m^3. Shendell et al (2004) indicated that an 1800 mg/ m^3 increase in CO_2 indoors compared to outdoor concentrations might promote a relative rise of 10–20% of student absenteeism. Recently, Bakó- Biró et al. (2012) and Ferreira and Cardoso (2014) presented evidence that low ventilation rates in classrooms, that is, CO_2 concentrations > 1800 mg/m^3, significantly reduced children's attention and vigilance, affecting negatively memory and con- centration. In conclusion, this study estimated IAQ in Portuguese preschools. Overall, the attained results suggested that insufficient ventilation throughout the school day and the impact of out- door air penetration indoors was the predominant factors that affected IAQ at the examined preschools.

Funding

This work was supported by European Union (FEDER funds through COMPETE) and National Funds (*Fundação para a Ciência e Tecnologia, FCT*) through projects UID/QUI/ 50006/2013, POCI-01-0145-FEDER-007265, and POCI-01- 0145-FEDER-006939, by the FCT/MEC with national funds and co-funded by FEDER in the scope of the P2020 Partnership Agreement. Additional financial support was provided by FCT through the fellowships SFRH/BD/80113/ 2011 and SFRH/BPD/105100/2014.

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