Building and Environment 96 (2016) 198–205

Contents lists available at ScienceDirect

Building and Environment

journal homepage: www.elsevier.com/locate/buildenv

Source apportionment of CO_2, PM_{10} and VOCs levels and health risk assessment in naturally ventilated primary schools in Porto, Portugal

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ARTICLE INFO

Article history:
Received 28 September 2015
Received in revised form
24 November 2015
Accepted 26 November 2015
Available online 2 December 2015

Keywords:
Particulate matter
School building
PCA
Source apportionment
Exposure dose
Health risk

ABSTRACT

Children are by far more susceptible to the negative effects of air pollutants than adults. Building-level characteristics are structural factors largely beyond the control of those who live in them. Yet, there are gaps in understanding of the relationship of school building characteristics and/or occupant behaviour and indoor air parameters with implications for health and well-being.

The aims of the study were to investigate the potential sources of CO_2, PM_{10} and volatile organic compound (VOCs) in naturally ventilated primary schools and to assess the potential health hazards of PM_{10} on schoolchildren.

CO_2 and PM_{10} levels were determined in seventy three classrooms located in Porto city over a period of 8 h using low-drift NDIR sensors and light-scattering laser photometers, respectively. The VOCs samples were collected over 5-days in Tenax TA tubes and then analysed by gas chromatography coupled mass spectrometry.

Principal component analysis revealed the influence of activities or building features as major sources of indoor CO_2, PM_{10} and VOCs associated to the reduced airing of the classrooms which underlines the influence of indoor sources, occupant behaviour and maintenance/cleaning activities in schools and the high density of occupants.

The hazard quotient calculated based on the formula suggested by the United States Environmental Protection Agency is higher than the acceptable level of 1; being for children almost twelve times higher than the safe level. This indicates that the inhalation exposure to PM_{10} by children and adults occupying the school environment is not negligible.

1. Introduction

Children are by far more susceptible to the negative effects of air pollutants than adults, as they inhale more air per unit of body weight at a given level of exertion and are also less able to deal with toxic chemicals [1]. Studies have shown that there is a correlation between exposure to air pollutants in the school environment and the performance of students [2–4]. In addition, indirect indicators, such as school absenteeism, provide evidence of the impact of air pollutants on school children's health [5,6]. As children spend most of their time in an indoor environment, it is clear why indoor air quality (IAQ) has been recognised as one of the key influences on their health. The classrooms are one of the main places where children spend their time when they are not at home [7,8]. Air quality inside classrooms has been associated to several diseases common in childhood, such as asthma, rhinitis and rhino-conjunctivitis [9–11]. The latest statistics from the World Health Organization (WHO) shows that 36% of the respiratory diseases and 22% of chronic diseases are caused by the indoor environment [12].

Each indoor school environment has unique characteristics determined by the local outdoor air, specific building related-characteristics, such as the condition, maintenance, and cleaning of the school building, and occupant behaviour [8,13]. The pollutants in the air within a classroom are predominantly the same of

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http://dx.doi.org/10.1016/j.buildenv.2015.11.031

the outdoor air when the latter comes in though natural ventilation and infiltration. Many studies demonstrating that the level of indoor air parameters, such as carbon dioxide (CO_2), particulate matter (PM) and volatile organic compounds (VOCs) in classrooms is influenced by the increasing levels outside the school building, such as traffic and industrial emissions [14–17]. This represents an addition to the pollutants which originate from inside the classroom environment, such as those from furniture, paint, chalk, cleaning agents and re-suspension of soil dust [14,18,19]. Other factors, such as students' activities, level of occupancy, ventilation systems and temperature were also found to affect classroom's air quality [7,20].

Receptor models such as chemical mass balance [21], positive matrix factorisation [22,23] and principal component analysis (PCA) [24] have been applied in order to identify and quantify potential sources, including those found in the indoor environment. The main advantage of PCA is the ability to identify different sources without any prior knowledge regarding them. There is a lack of detailed analysis between the relationship of checklist reports on different types of building characteristics and indoor air parameters in Portugal where most of IAQ problems in schools were identified [20,25–29].

In terms of health effects, PM as well as VOCs pollutants could affect the respiratory system, which is the principal route of entry for air pollutants that are further exacerbated in the children or elderly [11,30]. Even in low concentrations, the presence of PM and VOCs indoors can lead to significant impacts on the respiratory health of children or susceptible individuals, such as those with low lung function, asthma and bronchitis [12]. In order to examine the potential risks resulting from children exposure to indoor air pollutants, health risk assessment has been applied worldwide [31]. Many studies have shown that PM contaminated with heavy metals and other pollutants such as polycyclic aromatic hydrocarbons (PAHs) can directly enter the body through inhalation, dermal contact and oral ingestion exposure pathways [32–34]. Besides the increasing number of studies reporting pollutant concentrations in different micro-environments, the integrated dose of the people was not successfully estimated. Moreover, as far as we know the assessment of the daily exposure to PM_{10} and the estimation of the daily dose was never done, even for children which are the most studied population's group.

This study aims to investigate the potential sources of CO_2, PM_{10} and VOCs, in naturally ventilated schools focused on school building characteristics and occupant behaviours, applying PCA to scrutinize the datasets of the samples collected. In addition, in the current study it was applied the risk assessment methodology to evaluate the intake and toxicological risk of PM_{10} in children and adults.

2. Materials and methods

2.1. Sampling sites and sampling protocol

Seventy three classrooms from 20 primary public schools located in the city of Porto (Fig. 1S), the second largest city in Portugal, located in the north of the country (41.16 °N, 8.62 W), were included in the study. The sample includes 40% of the primary schools in Porto. In each school, four classrooms of third and/or fourth grades (children aged 8–10 years) were selected among the classrooms with similar conditions. Preference was given to classrooms with high densities of occupation (m2/occupant) and full-week occupation time by the same class, and, if possible, on different floors.

The study included walkthrough surveys of schools grounds, buildings, and individual classrooms, as well as indoor and outdoor air monitoring [VOCs, aldehydes, $PM_{2.5}$, PM_{10}, CO_2, carbon monoxide (CO), bacteria, fungi, temperature, and relative humidity]. A checklist was applied in order to characterize the buildings and indoor spaces. This checklist included information about: ventilation systems, types of indoor materials, ventilation and cleaning practices, type of building construction, thermal isolation of the building and characterization of the building envelope.

At present, the primary schools as well as most public buildings in Portugal were covered by smoke-free law [35]. Table 1S presented in supplementary material summarizes detailed buildings' and classrooms' characteristics.

The walkthrough survey and the IAQ sampling in each school occurred within the same visiting period during the winter seasons, from November to March, during the years 2011–2013. Further information is described in detail elsewhere [36].

2.2. Principal component analysis

To investigate the relationship between building/classroom characteristics, occupant behaviour and IAQ parameters, PCA with varimax rotation was applied, as a first approach, to understand how the indoor air parameters were aggregated. The Scree Plot criterion was used to determine the number of components retained. If the factor loading was 0.40 or higher (in absolute value), an item was considered in the indicator. Considering the asymmetry in the distributions of the input variables, it was applied a logarithmic transformation to each of the IAQ parameters. After choosing indicators that represented each factor, multiple linear regression was used to assess the factor associated with each variable [37–39]. The stepwise forward method was used to assess which factors were associated with each input variable (data not shown). In a second approach, a multilevel linear regression with two levels-classroom and school (random effect)-was used to determine which factors explained each input variable and to evaluate the aggregation within schools. The aggregation was estimated using Intraclass Correlation Coefficient (ICC). Four models of multilevel analysis were considered: the first concerned the characteristics of the classroom; the second was represented by characteristics of the school; the third model considered characteristics both of the classroom and school for each indicator; and the fourth and final model was represented by the totality of the classroom and school features, that had a significant effect on the levels of each of the IAQ parameter in order to summarize the effect of all variables resulting from each of IAQ parameters analysed individually. Statistical analysis was performed using the software R, and multilevel analysis was implemented using the function lme (linear mixed effects) in the nlme library. Statistical significance was defined as $p < 0.05$.

2.3. Health risk assessment

The concentration of PM_{10} is used as input data in order to assess the impact on children's health exposure to PM_{10} through the inhalation pathway (the primary route) of PM_{10}.

PM_{10} dose rates were calculated using Eq. (1), which was validated in previously published studies [40–42]:

$$D = \left(\frac{BR_{WA}}{BW}\right) \times C_{WA} \times OF \times N \tag{1}$$

where, D represents the age-specific dose rate (μg/kg day); BR_{WA} is the age-specific weighted average breathing rate (L/min); BW is the age-specific body weight (kg); C_{WA} is the weighted average PM_{10} concentrations (μg/L); OF is the occupancy factor (considered 1, as children and adults kept their schedules and associated locations tightly); N is the total time per day spent by age-specific individual (min/day).

The main daily activity patterns (including residence time and type of performed activities) of the children and adults were registered and analysed. The BR_{WA} is characterized by the intensity of the activity practiced at the time of exposure. Children and adults spend their time having a nap, sleeping or seated normally (e.g. writing, reading, watching TV, and drawing), so, the "sedentary/passive" activity level was selected. The age-specific inhalation factors (male and female combined) were retrieved from the US EPA exposure factors handbook [43] since there is no available information concerning the Portuguese population. Thus, BR_{WA} was considered as 4.8 L/min for sedentary activities for 8 to 10-year-old children. The BR_{WA} values for adults were considered 4.2 L/min for ages between 21 and 30; 4.3 L/min between 31 and 40, 4.8 L/min between 41 and 50, and 5.0 L/min between 51 and 60 years old for sedentary activities. C_{WA} was estimated using the PM_{10} average concentrations and the OF was always considered as 1, since both children and adults kept their schedules and their respective locations tightly.

Schoolchildren (8–10 years old) and adults (21–60 years old) had similar daily schedules and/or activity patterns, spending about 6 h indoors. Body weight of schoolchildren was measured in kilogrammes (Kg), to the nearest tenth, using a digital scale and 33.4 kg was used. Body weight of 70.8 kg was used for adults according to US Environmental Protection Agency [44].

Non-carcinogenic health risks were assessed by calculation of hazard quotient (HQ, μg/kg day). The D was divided by the specific reference doses (RfD). With a lack of consensus regarding the specific RfC of PM_{10}, the estimated reference dose (RfD) was derived from the RfC of diesel particles (5 $\mu g/m^3$) – proportioning 0.60 μg/kg day in the winter conditions [45].

$$\mathrm{HQ} = \mathrm{D}/\mathrm{RfD} \tag{2}$$

If the HQ is less than one, it is believed that there is no increased risk of non-cancer health effects at a site. However, if HQ exceeded one, there is a possibility that some non-cancer effects may occur. Generally, larger HQs are cause for greater levels of concern.

3. Results

3.1. CO_2, PM_{10} and VOCs concentrations

The descriptive statistics of indoor and outdoor parameters are presented in Table 2S in supplementary material. Carbon dioxide levels ranged widely and, among the 73 classrooms surveyed, 86% of the classrooms ($n = 63$) had median CO_2 concentrations exceeding 1000 ppm [46]. The CO_2 levels changes in the classroom throughout the day and, depending on the occupancy and ventilation, following a path that is theoretically predictable for both the CO_2 accumulation in the room during the time of teaching and for the CO_2 reduction during the breaks. In the present study, CO_2 levels exceeded 1000 ppm during 70% of the occupation time measured. Maximum CO_2 levels should be interpreted cautiously as they may reflect events such as occupants clustering around and/or breathing proximate to the sensor during occupancy. As expected, indoor CO_2 levels were significantly higher than outdoor levels ($p < 0.05$) with an I/O ratio higher than 3 (Table 3S, supplementary material). Higher values were measured in classrooms with higher occupancy density for the longest teaching periods between breaks.

The indoor median concentration of PM_{10} in all of the classrooms exceeded the 50 $\mu g/m^3$ guideline values suggested by WHO (2010) for a sampling period of 24 h. There was a statistically significant difference between PM_{10} levels measured outdoors and inside the classrooms (75 *vs.* 127 $\mu g/m^3$, $p = 0.001$). Indoor concentrations exceeded outdoor levels, indicating an I/O ratio higher than the unity, which suggests contribution from possible indoor sources (Table 3S).

Benzene, naphthalene and styrene were detected in less than 25% of classrooms; formaldehyde and acetaldehyde were detected in all samples (Table 1S). Most of the individual VOCs had median levels lower than 5 $\mu g/m^3$. The most abundant VOC in schools was D-limonene (23 $\mu g/m^3$), followed by toluene (6.4 $\mu g/m^3$). As expected, indoor concentrations usually exceeded outdoor levels, although the differences were statistically significant only for D-limonene ($p = 0.001$) (Table 3S). Median outdoor concentrations of benzene and toluene were 2.2 $\mu g/m^3$ and 4.1 $\mu g/m^3$ respectively, reflecting the urban areas sampled. The high indoor/outdoor ratios (I/O > 6) for D-limonene, formaldehyde and acetaldehyde, and the moderate I/O ratio (~2) for total VOCs and toluene suggest that indoor sources are the main origin for these VOCs. In contrast, the I/O ratio for benzene (0.84) indicates that outdoor sources were the primary contributor for this species.

3.2. Source apportionment

The Scree Plot suggested the existence of three components (Table 1). The first component explains 19% of variance and was characterized by these variables: total VOCs, toluene, m/p-xylene and o-xylene; the second factor has 16% of variance explained and CO_2, relative humidity, $PM_{2.5}$, PM_{10}, trichloroethylene and bacteria characterized this component. Finally, the third factor explains 10% of variance and was characterized by six IAQ parameters: CO_2, CO, temperature, benzene, styrene and D-limonene. Three of these indicators (CO_2, PM_{10} and total VOCs) were selected for multilevel analysis.

The results of multilevel analysis are presented by the estimated linear regression coefficients of the classroom and school features and the respective 95% CI as well as intra-class correlation coefficient. Ceiling height, window area and the number of windows usually open in the cooling season were the characteristics that

Table 1
Rotated component matrix with *varimax* rotation obtained of the principal components analysis method.

	Component		
	1	2	3
CO_2, ppm	0.277	0.549	**0.487**
CO, mg/m^3	0.000	−0.116	0.486
Temperature, °C	−0.008	−0.353	−0.707
Relative humidity, %	−0.023	0.567	0.482
$PM_{2.5}$, $\mu g/m^3$	−0.020	−0.794	0.052
PM_{10}, $\mu g/m^3$	0.062	**−0.766**	0.042
Total VOC, $\mu g/m^3$	**0.927**	0.072	0.075
Benzene, $\mu g/m^3$	−0.081	0.135	0.574
Toluene, $\mu g/m^3$	0.975	0.012	−0.044
T4CE, $\mu g/m^3$	−0.050	−0.447	0.099
m/p-xylene, $\mu g/m^3$	0.964	−0.030	−0.080
Styrene, $\mu g/m^3$	−0.178	−0.161	0.509
o-xylene, $\mu g/m^3$	0.975	−0.002	−0.074
α-pinene, $\mu g/m^3$	0.226	0.249	0.087
D-limonene, $\mu g/m^3$	0.029	0.181	0.671
Naphthalene, $\mu g/m^3$	0.169	0.352	0.155
Formaldehyde, $\mu g/m^3$	−0.038	0.365	−0.280
Acetaldehyde, $\mu g/m^3$	0.000	0.180	0.068
Ventilation rate, L/s per person	−0.150	0.098	−0.328
Fungi, CFU/m^3	−0.132	−0.220	−0.145
Bacteria, CFU/m^3	−0.212	0.437	−0.096
Variance explained (%)	**19**	**16**	**10**

Note: The item in bold was the parameter selected to represent each component; the shaded cells represent if the factor loading was 0.40 or higher (in absolute value), an item was considered in the indicator; T4CE: Tetrachloroethylene.

showed an effect on classroom CO_2 levels, explaining 16% of the differences among schools buildings (Table 2). None of the school characteristics represented a significant effect for this parameter.

Regarding PM_{10} levels, the characteristics that showed a significant effect in the classroom, explaining 28% of the differences among schools were: the number of windows usually open in the heating season; visible damp spots on walls, ceilings or floors; main ceiling surface material; visible mould growth in the room; and, the presence of a closet or shelves with gouaches, inks, etc., for graphic arts. Gasoline dispensing facilities nearby, car park sources of outdoor air pollution and the existence of a laboratory were the characteristics that most contributed to the school levels of PM_{10}, explaining 33% of differences between schools (Table 3).

When the total VOC parameter was analysed, it was found that the characteristics of the school building had no significant effect on it. The number of windows usually open in the cooling season, the main floor surface material, the number of windows usually open before classes, and the presence of a closet or shelves with gouaches, inks, etc., for graphic arts were the variables that explained 21% of the differences among the schools evaluated (Table 4).

To summarize the effect of all variables resulting from each of the parameters analysed individually, a multilevel regression analysis was performed using the same model for each of the parameters studied. There was an increase in the value of the ICC, for the parameter CO_2 to 20% and parameter PM_{10}, reaching the ICC of 40%. On the other hand, a decrease was perceived, assuming a value of 16% of this coefficient, when the total VOC parameter was analysed (Table 5).

3.3. Health risk assessment

Only the inhalation pathway was focused on this study as it is an important pathway for exposure to PM_{10} indoors. The inhalation dose rates of PM_{10} were estimated for 8–10-year-old children and for adults (21–60 years old).

For children the dose rate was 7.19 μg/kg day considering activities as sedentary; while for adults the dose rate was 2.97 μg/kg day for ages between 21 and 30; 3.04 μg/kg day for ages between 31 and 40, 3.39 μg/kg day for ages between 41 and 50 years and 3.53 μg/kg day for adults between 51 and 60 years old.

The toxicological risk of PM_{10} was 12.0 for children. Regarding the adults the HQ values were 4.9 for ages between 21 and 30; 5.1 for ages between 31 and 40 years, 5.7 for ages between 41 and 50 and 5.9 for ages between 51 and 60. These results showed, that in the winter time, children and adults were clearly exposed to levels of PM_{10} concentration that have the ability to cause adverse health effects; being the HQ higher for children one of the most susceptible groups of the population.

4. Discussion

In this study, it was documented the concentrations of a large set of environmental parameters and the potential relationship between building/classrooms characteristics and occupant behaviour and the concentrations of CO_2, PM_{10} and VOCs in indoor air by larger sample size (73 classrooms from 20 public schools) than ever. Little information on the latter is available in the published literature.

4.1. Levels and source apportionment of CO_2, PM_{10} and VOCs

Based on CO_2 levels, inadequate ventilation appears to be a common IAQ problem encountered in the studied classrooms, reinforcing earlier studies [4,18,20,47,48]. Based on a 1000 ppm CO_2 limit [46] and using school-day averages, 86% of the classrooms were inadequately ventilated. Average levels of CO_2 exceed the value recommended by the Portuguese legislation (1250 ppm) in 47 classrooms (66.2%) [49]. In addition, classrooms were monitored under “closed” conditions, keeping windows and doors closed as best possible during the occupied hours. During the occupation period it was observed that CO_2 concentration produced by the occupants build up until reaching equilibrium with levels greater than 1000 ppm and decreased to levels below 1000 ppm during breaks (data not shown). The present study showed higher CO_2 concentrations in classrooms with higher ceiling height and an inverse association with windows area. Although almost all classrooms have the same ceiling height (range = 2.9–3.6 m), the classroom area/volume and the density of occupation varied between classrooms. Ceiling height may be associated with a higher volume of the space and consequently a higher dispersion and lower CO_2 concentrations, however it was observed (data not shown) that classrooms with higher ceiling height have a higher number of students and thus could be associated with increased CO_2 levels. Moreover, the difficulty associated to heating a high space volume might also explain and determine the occupant behaviour, reflected in the reduced number of times that the windows were opened (introduction of “fresh” air), thus suggesting a potential stagnation of the indoor air. Consequently, taking into account that the school staff reported that opening windows was not so frequent due to noise problems and/or weather conditions, the results of the present study underlined the relevance of classroom management or occupant behaviour influencing indoor CO_2 concentrations. Kvisgaard et al. [50] and Iwashita et al. [51] reported that occupant behaviour may account for 63–87% of the total ventilation rate.

Several studies have been assessing the health risk caused by hazardous exposure levels to PM_{10} and, more recently, $PM_{2.5}$ in schools indoors and outdoors. An increasing number of data has shown that increased levels may result in increased prevalence of acute and chronic health effects, including asthma, among children [5,15]. In general, the indoor PM_{10} concentrations obtained in the current study were consistent with data reported in other studies [52,53], but higher than those reported by Stranger et al. [54]. Particular attention should be paid to PM_{10} as it is well known that PM_{10} enhances adverse health effects and it is unclear whether a

Table 2
Estimated linear regression coefficients of the classroom features and respective 95% confidence intervals for the parameter CO_2, assuming a multilevel model with “school” as a random effect.

ln(CO_2)	Classroom estimates (95% CI)
Ceiling height (m)	0.724 (0.26; 1.19)
Windows area (m^2)	−0.022 (−0.04; −0.01)
No. of windows usually open in the cooling season	0.048 (−0.03; 0.12)
Variance of school (%)	**1.5**
ICC (%)	**16**

ICC: Intra-class correlation coefficient.

Table 3
Estimated linear regression coefficients of the classroom/school features and respective 95% confidence intervals for the parameter PM_{10}, assuming a multilevel model with “school” as a random effect.

ln(PM_{10})	Classroom estimates (95% CI)	School estimates (95% CI)	Total estimates (95% CI)
No. of windows usually open in the heating season	0.176 (0.05; 0.31)	–	0.106 (−0.03; 0.24)
Visible damp spots on walls, ceiling or floor	0.357 (0.12; 0.60)	–	0.281 (0.06; 0.51)
Main ceiling surface material	−0.345 (−0.67; −0.03)	–	−0.306 (−0.59; −0.02)
Visible mould growth in the room	−0.247 (−0.47; −0.03)	–	−0.145 (−0.37; 0.07)
Existence of a closet or shelves with gouaches, inks etc. for graphic arts	−0.147 (−0.40; 0.10)	–	−0.054 (−0.28; 0.17)
Gasoline dispensing facilities nearby	–	−0.485 (−0.85; −0.12)	−0.385 (−0.71; −0.06)
Proximity of a car park	–	−0.319 (−0.57; −0.07)	−0.173 (−0.40; 0.06)
Existence of a laboratory room	–	0.366 (0.01; 0.72)	0.264 (−0.05; 0.57)
Variance of school (%)	**2.6**	**3.2**	**1.6**
ICC (%)	**28**	**33**	**17**

ICC: Intra-class correlation coefficient.

Table 4
Estimated linear regression coefficients of the classroom features and respective 95% confidence intervals for the parameter total VOC, assuming a multilevel model with “school” as a random effect.

ln(Total VOC)	Classroom estimates (95% CI)
No. of windows usually open in the cooling season	−0.186 (−0.35; −0.03)
Main floor surface material	0.601 (0.13; 1.08)
Windows usually open before classes	−0.698 (−1.31; −0.08)
Existence of a closet or shelves with gouaches, inks etc. for graphic arts	0.438 (−0.06; 0.93)
Variance of school (%)	**8.3**
ICC (%)	**21**

ICC: Intra-class correlation coefficient.

Table 5
Estimated linear regression coefficients of the classroom/school features and respective 95% confidence intervals for the three parameters of indoor air quality, assuming a multilevel model with “school” as a random effect.

	ln(CO_2) estimates (95% CI)	ln(PM_{10}) estimates (95% CI)	ln(total VOC) estimates (95% CI)
Ceiling height (m)	0.657 (0.15; 1.16)	−0.290 (−0.84; 0.26)	0.057 (−0.94; 1.05)
Windows area (m^2)	−0.024 (−0.05; 0.01)	0.003 (−0.03; 0.03)	−0.006 (−0.06; 0.04)
No. of windows usually open in the cooling season	0.067 (−0.02; 0.16)	0.045 (−0.05; 0.14)	−0.113 (−0.29; 0.06)
No. of windows usually open in the heating season	−0.123 (−0.28; 0.03)	0.127 (−0.02; 0.28)	−0.227 (−0.53; 0.08)
Visible damp spots on walls, ceiling or floor	−0.012 (−0.26; 0.24)	0.371 (0.10; 0.64)	0.340 (−0.15; 0.83)
Main ceiling surface material	−0.025 (−0.38; 0.33)	−0.312 (−0.70; 0.08)	−0.009 (−0.72; 0.70)
Visible mould growth in room	0.066 (−0.18; 0.31)	−0.245 (−0.50; 0.01)	0.046 (−0.43; 0.52)
Existence of a closet or shelves with gouache, inks etc. for graphic arts	0.082 (−0.22; 0.39)	−0.106 (−0.46; 0.25)	0.555 (−0.05; 1.16)
Windows usually open before classes	−0.034 (−0.36; 0.29)	0.160 (−0.20; 0.52)	−0.661 (−1.30; −0.02)
Main floor surface material	0.035 (−0.28; 0.35)	−0.001 (−0.37; 0.37)	0.557 (−0.05; 1.17)
Variance of school (%)	**1.88**	**4.24**	**6.02**
ICC	**19.6**	**40.0**	**15.5**

ICC: Intra-class correlation coefficient.

threshold concentration exists for PM below which no effects on health are likely. Moreover, schoolchildren are considered more susceptible to air pollutants, and they spend most of their time in schools.

The indoor PM_{10} concentrations might either originate from particle generation by occupants themselves, resulting from their school activities or re-suspension of deposited particles [55]. Several studies concluded that the introduction of new particulate matter including soil material brought in with shoes, blackboard dust, skin flakes, and cloth and furniture fragments seems to be the main reason for high indoor PM_{10} concentrations [56–58]. Fromme et al. [59] reported that high PM_{10} levels in schools were correlated with less frequent cleaning and inefficient removal of deposited particles that thus became re-suspended. According to the same authors, occupancy strongly influences the indoor concentration level of PM_{10} through re-suspension. Besides, delayed deposition/settlement may be due to turbulence induced by occupant's movement and the reduced ventilation could also affect the dispersion of PM_{10}, causing their accumulation indoors. The PM_{10} indoor concentration profiles showed peaks within the time slots when the studied classrooms were occupied [19]. However, the data from current study should be observed with caution taking into account that indoor and outdoor particulate matter could not be sampled in parallel, affecting the accuracy of the estimated I/O ratios. In the current study, specific characteristics from both the classrooms and the school such as the number of open windows in the heating season, the visible damp spots on the wall or ceiling, and the presence of a laboratory room were associated with significantly higher PM_{10} concentrations. On the contrary, the values are significantly lower if the main ceiling surface material is painted (instead of wooden) and if gasoline dispensing facilities and car parks exist on the proximities of the school. These relationships pointed out for conditions promoting the penetration of particulate matter from outdoors and re-suspension of coarse particles indoors resulting from occupant activities, as well as for the presence of other potential indoor sources of PM_{10} such as the degradation/peeling of coating materials in the walls, ceiling (e.g. paint) resultant from dampness problems. The inverse association regarding the proximity of gasoline dispensing facilities or car park may reflect that in these schools there is a tendency to avoid opening the windows which prevents the entrance to the indoor environment of particulate matter from outside.

Given the potential impact of exposure to VOC on children health, it is important to increase the understanding of the factors that influence their indoor concentrations. According to Mendel [60] and Zhang et al. [61] indoor total VOC levels might be due to the furnishing, floor covering, insulating materials, adhesives, paints and glues as well as other solvents and cleaning products. In addition to these indoor sources, the insufficient ventilation is likely to favour the increase of total VOC levels [62]. Total VOCs levels measured in this study were higher than in previous studies [62–64], but lower than those measured by Yang et al. [65]. Comparisons of total VOC levels across studies can be problematic due to differences in definition, sampling times, measurement and analysis [62], thus examination of specific VOC species is often more informative. The current study showed an increase in total VOC levels when the floor surface material was PVC/vinyl or linoleum. Decreases in total VOC levels were associated with the increase of ventilation measured by the number of windows usually open in the cooling season and if the windows were usually open before classes. These results, the room-to-room variability, and the outdoor levels (Supplementary material) suggest classroom (indoor) sources rather than building-wide or outdoor sources underlining the importance of occupant behaviours in the control and assurance of good IAQ.

4.2. Health risk assessment

Calculating health risk assessment of air pollutants poses particular difficulties. In exposure rates risk, the particles exposed to the population must be identified and the danger and behaviour of these substances in the human body must be assessed. Moreover, calculating the dose reaching the target organ remains a challenge, while a theoretical exposure dose can be calculated from the air pollution concentration and exposure time; significant limitations still remain in the approach. For instance, although the applied model allows estimating the inhalation rates of PM_{10}, the areas of particle deposition are not considered in this model (alveoli, bronchia, trachea, etc.). However, this study is one of the first to assess the risk of PM_{10} inhalation in Portuguese schools located in an urban area and presents a methodology that could be highly applicable to estimate and compare levels of risk across emission source and populations.

Based on the exposure assessment, it was determined that the exposure dose is higher in children when compared to adults (7.2 μg/kg day and between 2.97 and 3.53 μg/kg day, respectively). The HQs exhibited the highest values for children, being twelve times higher than the safe level ($HQ = 1$), with a value of 12.0 for children and between 4.9 and 5.9 for adults. This suggests that the presence of PM_{10} indoors raises concerns with regard to potential adverse health effects. As previously mentioned, in the current study, PM_{10} would be related with building conditions promoting the penetration of particulate matter from outdoors and re-suspension of coarse particles indoors resulting from occupant activities, as well as for the presence of other potential indoor sources of PM_{10} such as the degradation/peeling of coating materials in the walls, ceiling (e.g. paint) resultant from dampness problems.

The results also signify that a degree of exposure to metals, PAHs and other components in PM_{10} might contribute to the risk of developing cardiovascular and respiratory diseases, as well as lung cancer, particularly among schoolchildren who are exposed to PM_{10} over the long-term [33,34].

This study is unique in that it jointly assessed a comprehensive array of school classroom characteristics and a large set of objective measurements of IAQ parameters. Additionally, the sample size of 73 classrooms from 20 school building enabled us to conduct a complete analysis, even if the results are not necessarily generalizable to Portuguese schools as a whole.

Limitations that should be highlighted are the lack of consensus in the literature about PM RfC and RfD and inhalation rate measurements obtained by studies conducted in other countries. In relation to the RfC, the reference concentration of diesel particles was used. Still, additional studies that incorporate objective measurements of PM composition and observations of multiple IAQ-related school environmental conditions would be useful to validate these findings.

In public health, the exposure quantification and risk estimates provide resources for planning and formulation of protection health policies where comparisons across emission sources and location can be evaluated and compared to find more accurate global assessments.

5. Conclusions

The present study suggest the influence of activities or building features as major sources of indoor CO_2, PM_{10} and VOCs levels associated to the reduced airing of the classrooms which underlines the influence of indoor sources, occupant behaviour and maintenance/cleaning activities in schools and the high density of occupants.

Whilst PM_{10} levels might be explained by the mixed source from indoor activities (such as students particulate matter re-suspension) and potential transport from outside into classroom environment; for VOCs, identified sources included floor surface material (PVC/vinyl, linoleum). As expected CO_2 levels is related to the natural ventilation of the classrooms which is a common problem in Portuguese schools due to most common closed windows and doors.

Health risks associated with the inhalation of indoor PM_{10} emissions were estimated for children and adults. The results of HQ were found to be greater than the acceptable level of 1, as proposed by USEPA. This indicates that children in the school classrooms face a potentially high health risk, and that the presence of high levels of PM_{10} is cause for concern. The results from this study provide a valuable evaluation of the health risk associated with exposure to PM. Consequently, certain mitigation procedures should be conducted to reduce the distribution of indoor pollutants in the school classrooms. It is recommended the implementation of more breaks and recesses between classes, decreasing the occupancy per room to 2.0 m^2/occupants according ASHRAE [46], increasing the exchange of indoor air with the outdoor, and improving the cleanliness of facilities which might benefit the IAQ. Further monitoring with a larger number of samples and sampling points needs to be undertaken, by way of a comprehensive investigation associated with source apportionment approaches and the investigation of health risks, particularly in the school environment.

Acknowledgements

This work was supported by Fundação para Ciência e a Tecnologia (FCT) through ARIA project (FCT PTDC/DTP-SAP/1522/2012, FCOMP-01-0124-FEDER-028709) and the scholarships SFRH/BD/108605/2015 & SFRH/BD/112269/2015.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.buildenv.2015.11.031.

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