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Comfort Parameters and Particulate Matter (PM_{10} and $PM_{2.5}$) in School Classrooms and Outdoor Air

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ABSTRACT

In January 2012, one kindergarten and eight elementary school classrooms were monitored. The campaign included simultaneous measurements, indoors and outdoors, of comfort parameters, CO, CO_2 and particles. Automatic monitors using a light scattering technique were employed to measure PM_{10} continuously. During occupied periods, low volume samplers were used to daily collect $PM_{2.5}$ samples, which were subsequently analysed for carbonates, organic carbon (OC), elemental carbon (EC) and water soluble inorganic ions. With regard to comfort, the schools did not meet the recommended levels in many rooms. Indoor-outdoor CO_2 ratios between 3 and 12, and indoor levels much higher than 1000 ppm during the occupied periods, indicate the highly inadequate ventilation in these locations. The results clearly demonstrate that there is a high level of exposure to particulate matter in these schools. The continuous measurements of PM_{10} suggest that the physical activity of pupils, which is assumed to be more marked in younger children, contributes to a constant process of resuspension of sedimented particles. In addition, peak PM_{10} concentrations coincident with cleaning activities suggest the need to change certain practices to improve cleanliness. Around 40% of the $PM_{2.5}$ mass is composed of carbonaceous matter, with 4–5 times higher OC mass fractions than EC. It was observed that both OC and EC were significantly influenced by indoor sources. Water-soluble inorganic ions represented around 10–20% of the $PM_{2.5}$ mass measured in classrooms. Excluding calcium, in general the ionic species were present at indoor-outdoor ratios of less than 1, suggesting the major origin in the outdoor air.

Keywords: Indoor air quality; Air exchange rates; PM_{10}; $PM_{2.5}$; OC/EC; Water soluble ions.

INTRODUCTION

Studies carried out by the USA Environmental Protection Agency (EPA) of human exposure to air pollutants indicate that indoor air levels of many pollutants may be 2–5 times, and occasionally more than 100 times, higher than outdoor levels. These levels of indoor air pollutants are of particular concern because it is estimated that most people, including children, spend as much as 90% of their time indoors (Bronsema *et al.*, 2004). Over the past several decades, the exposure to indoor air pollutants is believed to have increased due to a variety of factors, including the construction of more tightly sealed buildings, reduced ventilation rates to save energy, the use of synthetic building materials and furnishings, and the use of chemically formulated personal care products, household cleaners, etc.

Unlike other buildings, managing schools involves not only maintenance related concerns, but also child safety issues. Many aspects are unique to schools (EPA, 2005): (i) classrooms have about four times as many occupants as office buildings for the same floor area; (ii) in general, maintenance suffers from financial cuts; (iii) the presence of a wide variety of emission sources, including art and science supplies, and gyms; (iv) highly variable heating and ventilation systems; (v) sometimes portable classrooms or buildings that were not originally designed to meet the educational requirements are used.

Failure to prevent or respond promptly to indoor air quality (IAQ) problems can increase short- and long-term health outcomes (Norbäck *et al.*, 2000; Simoni *et al.*, 2006; Tillet, 2010; Simoni *et al.*, 2010), impact both teacher and student's attendance, comfort and performance (Gilliland *et al.*, 2001; Shaughnessy *et al.*, 2006; Mohai *et al.*, 2011), and accelerate the deterioration of school buildings and equipments (Lohbeck, 2008). Among the indoor air pollutants, there is an increasing interest in the measurement of particulate matter concentrations. Aerosol exposure via the inhalation route is of prime importance, representing a major potential source of hazard for human health of a greater or lesser severity, depending on the airborne concentrations, duration of exposure and chemical composition. Therefore, it is of

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utmost significance to measure particle concentrations in the size fractions known to affect the human health (Abdel-Salam, 2006). There are some recent studies reporting indoor and outdoor aerosol behaviour and its chemical composition (e.g., Chao and Wong, 2002; Na *et al.*, 2004; Gemenetzis *et al.*, 2006; Martuzevicius *et al.*, 2008; Olson *et al.*, 2008; Smolík *et al.*, 2008 Lai *et al.*, 2010; Saraga *et al.*, 2010; Zhu *et al.*, 2010; Huang *et al.*, 2012), but only a few refer to elementary schools (Fromme *et al.*, 2008; Almeida *et al.*, 2012; Oeder *et al.*, 2012; Pegas *et al.*, 2012; Smolík *et al.*, 2012). In the present study, the simultaneous characterisation of inhalable (< 10 μm, PM_{10}) and thoracic (< 2.5 μm, $PM_{2.5}$) suspended particles, along with measurements of comfort parameters, were carried out. This aims to deepen the knowledge of indoor air quality in schools and childcare settings and, thus, to contribute to the elaboration of a prolific set of proposals leading to new policies, guidelines and practices.

METHODOLOGY

Selected Schools

The selected schools are located in the municipality of Aveiro, central Portugal. This coastal municipality has a population of about 78,500 inhabitants within its administrative limits and a total area of 200 km². A total of nine classrooms were monitored, eight integrated in elementary schools and one in a kindergarten (Table 1). All the educational settings started their activities in the sixties or seventies, although, in the meantime, were somewhat refurbished and routine maintenance works were carried out. All the school buildings are naturally ventilated through openable windows and doors, and have white painted concrete walls, wooden window frames, wood or cork flooring and portable electric heaters. The number of pupils per class ranged from 20 to 25, either for kindergarten or primary levels.

Sampling Campaign

The sampling campaign took place between 9 and 29 January, 2012. Continuous measurements of temperature, relative humidity (RH), CO_2, CO and total volatile organic compounds (TVOCs) were performed with an Indoor Air IQ-610 Quality Probe (Gray Wolf® monitor) in the classrooms of every school. The same measurements, excepting TVOCs, were continuously carried out outside on the playgrounds using an IAQ-CALC monitor (model 7545) from TSI. These equipments were supplied with a factory calibration certificate, but were further checked prior to their use with appropriate calibration kits.

Three automatic monitors using a light scattering technique were employed to measure PM_{10} continuously in the three classrooms of each school: (i) a TSI Model 8533 DustTrak DRX, (ii) an AIR-AIDE model AA-3500, and (iii) a Topas monitor from Turnkey Instruments. This latter monitor is designed to continuously record environmental total suspended particles (TSP), PM_{10}, $PM_{2.5}$ and PM_1. A "demonstration of equivalence" exercise was carried out before the monitoring campaign. These three instruments were intercompared with gravimetric PM_{10} measurements, following the EN 12341 (the reference procedure). The automatic monitors were initially calibrated in the factory using a "standard" test dust. The side-by-side intercomparison allowed the verification or correction of the calibration factors, which were then used to scale the readings of each monitor to give "true" results.

From Monday to Friday, during the occupancy periods, simultaneous sampling of $PM_{2.5}$ was performed in 3 classrooms and one outdoor site per school. The daily $PM_{2.5}$ samples were collected onto pre-baked (6 h at 500°C) 47 mm diameter quartz filters using Echo TCR Tecora samplers, following the EN 14907 norm. The four low-volume samplers were intercompared before the IAQ evaluation programme. The regression slopes for the results obtained from parallel measurements over one week differed by less than 5%. All the correlation coefficients were higher than 0.95.

Following the teachers' request in order to allow the free movement within the classrooms, the sampling equipments were installed in a central location at the bottom of the rooms. This location avoids direct contamination of the filters with chalk dust, principally when blackboards on the front wall are being used. The sampling heads or inlets were positioned at a height of about 1.2 m above floor and at a distance of around 1 m from the bottom wall.

Analytical Methodologies

Before weighing, the filters were conditioned in a desiccator at least for 24 h in a temperature- and humidity-controlled room. Before and after sampling, gravimetric determination was performed with a microbalance Mettler Toledo AG245 (readability 0.1 mg/0.01 mg). Filter weights were obtained from the average of 10 measurements, with weight variations less than 5%. After gravimetric determination of the $PM_{2.5}$ mass, filter punches were analysed by a home-made thermo-optical transmission system in order to obtain the carbonaceous content. After exposure of filter punches to HCl vapours to remove carbonate carbon, controlled heating in anoxic and oxic conditions was performed to separate, respectively, OC into two fractions of increasing volatility and EC. The first fraction corresponds to the volatilisation at T < 200°C of lower molecular weight organics (OC_1). The second fraction is related to the decomposition and oxidation of higher molecular weight species at temperatures ranging from 150 to 600°C (OC_2). The last fraction of OC is identified by transmittance and corresponds to pyrolysed organic carbon (PC) produced in the previous heating steps. Separation between OC and EC was achieved by initially heating the filter punches under an inert atmosphere to evaporate first the OC fraction. The remaining fraction is sequentially evaporated/burnt under a gas flow containing O_2. This last carbon fraction contains initial EC plus OC that has pyrolysed during heating under an inert atmosphere. The interference between PC and EC can be controlled by continuous evaluation of the blackening of filter using a laser beam and a photodetector measuring the filter light transmittance. Carbonates present in $PM_{2.5}$ samples were analysed through the release of CO_2, and measurement by the same non-dispersive infrared analyser coupled to the thermo-

Table 1. School classrooms selected for the indoor air quality monitoring campaign in Aveiro, Portugal.

School	Coordinates	Year of construction	School characteristics	Code (School.Room)	Classroom characteristics
Glória Elementary	N 40°38'16" W 08°39'10"	1966	Traffic City centre	1.1	4th grade; 1st floor; floor area 44.3 m^2; ceiling height 3.5 m; blackboard with chalk and electronic interactive board; occupied 7.5 h per day
				1.2	4th grade; 2nd floor; floor area 52.2 m^2; ceiling height 3.23–4.9 m; blackboard with chalk and electronic interactive board; occupied 7.5 h per day
				1.3	3rd grade; 1st floor; floor area 54.5 m^2; ceiling height 3.0 m; blackboard with chalk; occupied 7.5 h per day
Esgueira Kindergarten	N 40º38'47" W 08º37'52"	1974	Urban Residential	2.1	Children 3–6 years old; 1st floor; floor area 50.8 m^2; ceiling height 2.7 m; occupied 6.5 h per day
Esgueira Elementary	N 40º38'47" W 08º37'52"	1974	Urban Residential	2.2	Two 3rd grade classes (one in the morning; another in the afternoon); 2nd floor; floor area 49.5 m^3; ceiling height 2.7–6.7 m; whiteboard with markers; occupied 10.5 h per day
				2.3	3rd grade; 2nd floor; floor area 49.5 m^3; ceiling height 3.5 m; whiteboard with markers and electronic interactive board; occupied 5–10 h per day, depending on the activities
Eixo Elementary	N 40º37'39" W 08º34'07"	1979	Rural	3.1	2nd and 3rd grades simultaneously; 1st floor; floor area 50.7 m^3; ceiling height 4.9 m; blackboard with chalk and whiteboard with markers; occupied 7.25 h per day
				3.2	4th grade; 1st floor; floor area 51.5 m^3; ceiling height 2.8 m; blackboard with chalk, whiteboard with markers and electronic interactive board; occupied 7.25 h per day
				3.3	3rd grade; 2nd floor; floor area 51.5 m^3; ceiling height 2.8 m; blackboard with chalk and whiteboard with markers; occupied 7.25 h per day

optical system, when a punch of each filter was acidified with orthophosphoric acid (20%) in a free CO_2 gas stream.

It should be mentioned that results of an international round robin test on the analysis of carbonaceous particles positioned the University of Aveiro within the group with "best estimates". In addition, the comparison between the methodology of the University of Aveiro and the "European Supersites for Atmospheric Aerosol Research" (EUSAAR-2) protocol (Cavalli *et al.*, 2009) for the quantification of the different carbon fractions in atmospheric particulate matter samples gave similar results for OC and EC, without significant differences at a 95% confidence level (Nunes *et al.*, 2010). In the present study, two replicate carbon analyses were carried out. The analyses were repeated when the coefficient of variation was higher than 5%.

For the determination of water soluble inorganic ions, small parts of the filters were extracted by sonication during 15 min with 5 mL of ultra pure Milli-Q water. The liquid extracts were filtered through a 13 mm PVDF syringe filter (pore size 0.2 μm) and analysed by ion chromatography. An AS4 (4 mm) column with an AG4 guard column were used coupled to a Dionex AMMS II suppressor for Cl^-, NO_3^- and SO_4^{2-} determination. Cations (Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) were analysed using a CS12 (4 mm) column with a CG12 guard column. A Dionex CMMS III suppressor was used in these analyses. The anionic system comprised a Model 401 Dilutor (injector pump), a Gilson sampling injector Model 231, a Shimadzu oven CTO-6A, a Shimadzu conductivity detector CDD-6A, a Dionex interface UCI-100 and a Jasco pump PU-980. The cationic system integrates a Gilson injector 234 and a Dionex DX-100 chromatograph. Sets of multi-cation and anion standards were prepared from a stock solution with 1000 mg/L for each ion. The ion concentrations in the samples were obtained through individual calibration curves ($r^2 \geq 0.99$). For the final results, blank values were taken into account. Samples were analysed by two different analysts on two different days. The inter-day precision, expressed as coefficient of variation, was ≤ 3%.

Ventilation Rates

The ventilation Eq. (1) has been used to calculate the fresh air ventilation rate (Griffiths and Eftekhari, 2008). For a well-mixed space the change in CO_2 concentration with time is given by:

$$C_\tau = C_{ext} + \frac{q_{CO_2} \times 10^6}{Q} - \left(C_{ext} - C_0 + \frac{q_{CO_2} \times 10^6}{Q} \right) e^{\left(-Q_t/V\right)} \quad (1)$$

where C_t is the indoor concentration of CO_2 at time t (ppm), C_{ext} is the outdoor concentration of CO_2 (ppm), C_0 is the concentration of CO_2 in the indoor air at time 0 (ppm), Q is the volume flow rate of air entering the space (m^3/s), q_{CO_2} is the volumetric indoor emission rate of CO_2 (m^3/s), V is the volume of the classroom (m^3) and t is the interval since $t = 0$ (s). When the classroom is unoccupied there is no CO_2 emission from the occupants, and $q_{CO_2} = 0$. Thus, Eq. (1) can be rearranged to give the following expression, which allows the ventilation rate (Q) to be calculated from measured concentration values time t apart:

$$Q = -\frac{V}{t} \times \ln\left(\frac{C_t - C_{ext}}{C_0 - C_{ext}} \right) \quad (2)$$

RESULTS AND DISCUSSION

Comfort Parameters and Ventilation Rates

The human sensation of warmth depends on the thermal balance of the body as a whole. Thermal comfort occurs when body temperatures are held within narrow ranges, and the physiological effort of regulation is minimised. A number of variables interact to determine whether people are comfortable with the temperature and relative humidity of the indoor air. Factors such as clothing, activity level, age, and physiology of people in schools vary widely, so the thermal comfort requirements differ for each individual. The American Society of Heating, Refrigerating, and Air-Conditioning Engineers (ASHRAE) Standard 55–1992 describes the temperature and humidity ranges that are comfortable for 80% of people engaged in largely sedentary activities, assuming "normal indoor clothing". At winter conditions, the recommended temperature values for a relative humidity of 30% range from 20.3 to 24.2°C. An interval between 19.7 and 23.3°C is suggested when the relative humidity value is 60%. For many rooms, the schools did not meet ASHRAE recommended levels for winter conditions (Table 2). The average relative humidity values varied depending on the school and room, some of them exceeding the recommended upper limit of 60%. Previous studies suggest that thermal abnormal conditions can cause discomfort or distraction among students, and have consequent effects on performance (Mendell and Heath, 2005). Elevated relative humidity can be responsible for IAQ problems resulting from mould and other microbiological amplification, and subsequent allergic incidents.

At concentrations occurring in most indoor environments, CO_2 build-up can be considered as a surrogate for other occupant-generated pollutants, particularly bioeffluents, and for ventilation rate per occupant, but not as a direct causal factor in human health responses (Apte and Erdmann, 2002). The outdoor air in most locations contains down to about 380 parts per million carbon dioxide. While levels below 5,000 ppm are considered to pose no serious health threat, experience indicates that individuals in schools with elevated CO_2 concentrations tend to report drowsiness, lethargy and a general sense that the air is stale (Mahyuddin and Awbi, 2012). Researchers have been looking for links between elevated CO_2 concentrations and reduced attendance, productivity or achievement (Shendell *et al.*, 2004; Wargocki and Wyon, 2006).

CO_2 concentrations during school hours at a given day and classroom varied from 1300 ppm up to 7000 ppm (Fig. 1), largely exceeding the limit of 1000 ppm stipulated by the Portuguese legislation (RCESE, 2006). Regardless of classroom, the air exchange rates were not higher than 0.2 h^{-1}, showing an effective building tightness with closed

Table 2. Comfort parameters, total volatile organic compounds and carbon oxides measured in indoor and outdoor environments.

School	Measurement site	Temperature (°C)	Relative Humidity (%)	TVOC (μg/m^3)	CO (μg/m^3)	CO_2 (ppm)
Glória (traffic, city centre)	Classroom 1.1	19.9 ± 1.9	47.8 ± 3.2	167 ± 41	391 ± 211	1556 ± 360
	Classroom 1.2	16.9 ± 0.9	65.7 ± 3.3	866 ± 472	714 ± 214	3574 ± 1026
	Classroom 1.3	17.0 ± 1.5	50.9 ± 2.5	338 ± 127	514 ± 224	1123 ± 345
	Outdoor	10.6 ± 4.5	65.0 ± 14.4	-	320 ± 505	448 ± 22
Esgueira (urban, residential)	kindergarten	18.8 ± 4.3	61.9 ± 8.1	723 ± 189	524 ± 170	2144 ± 525
	Classroom 2.2	17.6 ± 1.4	71.6 ± 2.0	1543 ± 399	905 ± 283	4957 ± 890
	Classroom 2.3	17.7 ± 1.5	57.8 ± 4.6	483 ± 198	479 ± 164	2032 ± 780
	Outdoor	8.5 ± 2.9	86.0 ± 10.2	-	405 ± 437	415 ± 17
Eixo (rural)	Classroom 3.1	17.1 ± 1.1	51.9 ± 5.8	163 ± 97	333 ± 135	1175 ± 363
	Classroom 3.2	17.2 ± 0.7	65.8 ± 4.6	1002 ± 1402	519 ± 150	1792 ± 651
	Classroom 3.3	20.5 ± 2.7	56.0 ± 6.1	624 ± 209	609 ± 122	2332 ± 494
	Outdoor	11.1 ± 4.6	70.7 ± 17.0	-	192 ± 185	415 ± 17

windows that is responsible for the high levels of CO_2 and the extremely low outdoor air infiltration of not more than 38 L/min per person (Fig. 2). In Europe, depending on the country, the required outdoor airflow rates range from 300 to 500 L/min per person (Jones, 2011), while the recommended value in the US was set at 426 L/min per person (ASHRAE, 2004).

Particulate Matter Levels and Chemical Composition

The daily average $PM_{2.5}$ concentrations obtained during the occupancy periods ranged from 44 ± 3.2 μg/m^3 to 117 ± 16 μg/m^3. The highest values were obtained in the school with the highest occupancy rates. In addition, this school, located in a residential area, is the only one surrounded by an unpaved playground. Soil particles are tracked in on shoes or clothing from the outdoors, contributing to indoor-outdoor ratios (I/O) in the range 1.3–5 (average = 2.7). Lower average I/O ratios of 1.3 and 1.7 were, respectively, obtained in the city centre and rural schools, where comparable $PM_{2.5}$ concentrations have been measured, while an I/O value of 2.3 was determined for the establishment located in the residential area. I/O ratios greater than one indicate the presence of indoor sources, activities/movements of the occupants and reduced ventilation rates as a result of closed doors and windows, so causing accumulation. No significant differences were observed in outdoor $PM_{2.5}$ concentrations among schools with averages in the range 37–42 μg/m^3.

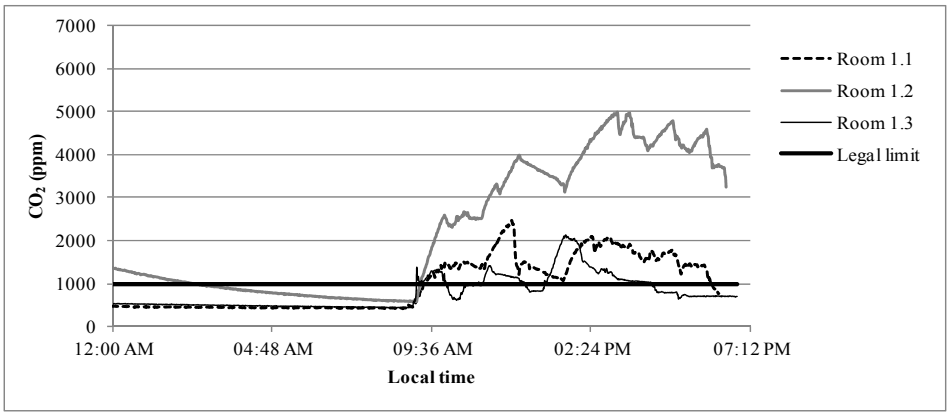
In classrooms (# 2.2 and 2.3) of the school in the periphery of the city, the PM_{10} concentrations (Table 3 and Fig. 3) frequently exceeded the limit value of 150 μg/m^3 established by the Portuguese legislation for indoor air (RSECE, 2006). In the classroom with highest occupancy rates (# 2.2), the average PM_{10} levels for daytime and night-time periods were, respectively 362 ± 84 and 111 ± 37 μg/m^3. $PM_{2.5}$ represented, on average, 12.7 and 28.0% of the PM_{10} concentrations during the teaching hours and vacant periods, respectively (Table 4). This suggests that high PM_{10} levels found in classrooms are probably due to resuspension of soil material or other coarse particles. The very high TSP concentrations, up to 3227 μg/m^3, registered in classroom # 2.2, confirm the presence of resuspended coarse particles. In this classroom, PM_{10} accounted for 12.7 ± 1.9% (period of school activities) and 28.0 ± 4.6% (night-time) of the TSP concentrations. Slightly higher averages for daytime and lower at night-time were obtained in the other classrooms, where a Topas monitor has been used. Human activity is seemingly the most important factor to account for the indoor levels of coarse particles. Without this source, at night the indoor particulate material is composed predominantly of $PM_{2.5}$. Significant differences between night and day were also observed for the finest particle size fraction. The settling of coarser particles after school hours contributed to increased submicron particles PM_1-to-$PM_{2.5}$ ratios during the night.

A wide range of $PM_{2.5}/PM_{10}$ and $PM_1/PM_{2.5}$ ratios has been reported worldwide for different schools, depending on season, meteorology, occupancy rates, physical characteristics of buildings, activities inside the classrooms and ventilation.

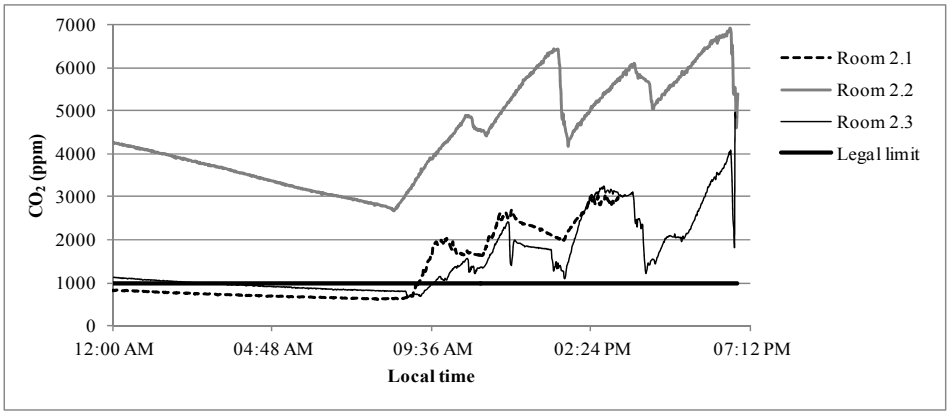
In all schools of the present study, the maximum concentrations of PM_{10} during teaching hours were registered when students were entering or leaving the classrooms, when the blackboard was being used or when craft activities involving paper or paperboard cutting were underway (Fig. 3). Intense physical activities of occupants in classrooms cause either resuspension of previously deposided particles or delayed deposition/settling of coarse particles indoors (hindered settling), enhancing the indoor concentrations (Goyal and Khare, 2009). However, the highest PM_{10} levels coincided with the cleaning activities at the end of the afternoon. In fact, the patterns of diurnal variation depend, in addition to other features, on the cleaning practices. Concentrations tend to be higher in rooms where sweeping is carried out than in those that are vacuumed. Mopping contributes to the lowest levels during the cleaning periods and favours steeper decays during nigh-time.

The particle concentrations observed in the Portuguese schools are consistent with results reported for other countries. It has been pointed out that the high levels are likely associated with the physical activities of pupils, which contribute to a constant process of resuspension of sedimented material from the daily tasks, from skin desquamation or clothing, and from outdoor sources (Brunekreef *et al.*, 1997;

Glória (traffic, city centre)



Esgueira (urban, residential)



Eixo (rural)

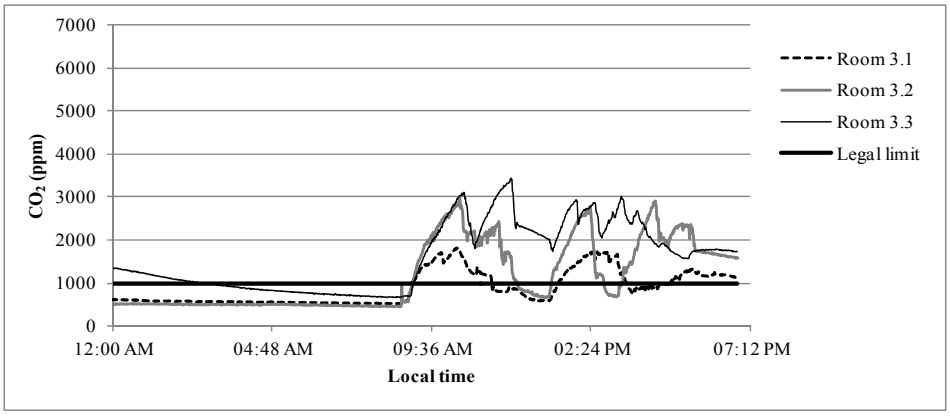


Fig. 1. CO_2 levels recorded during night-time and one typical working day in the 9 classrooms.

Lee and Chang, 2000; Janssen *et al.*, 2003; Peacock *et al.*, 2003; Pénard-Morand *et al.*, 2005; Ekmekcioglu and Keskin, 2007; Fromme *et al.*, 2007; Van Roosbroeck *et al.*, 2007; Diapouli *et al.*, 2008; Richmon-Bryant *et al.*, 2009). Brunekreef *et al.* (1997) studied 13 schools located 35–645 m from a motorway. The PM_{10} levels ranged from 6.73 to 20.8 μg/m^3 in schools farthest from the road, and from 9.20 to 32.8 μg/m^3 in schools nearest from the road. Lee and Chang (2000) found PM_{10} concentrations in 5 schools in Hong Kong above the 24-h average value (180 μg/m^3) stipulated by the local legislation. Braniš *et al.* (2005) studied the effects of outdoor air and human activities on PM_{10}, $PM_{2.5}$ and PM_1 concentrations in schools in Prague. The results confirmed that human activities are an important factor for high indoor particulate levels and that outdoor concentrations influence the indoor environment. Five elementary schools in Istanbul, Turkey, showed PM_{10} levels varying from 27.9 to 289 μg/m^3 according to the traffic intensity (Ekmekcioglu and Keskin, 2007). In 64 schools in Munich, Fromme *et al.* (2007) observed indoor

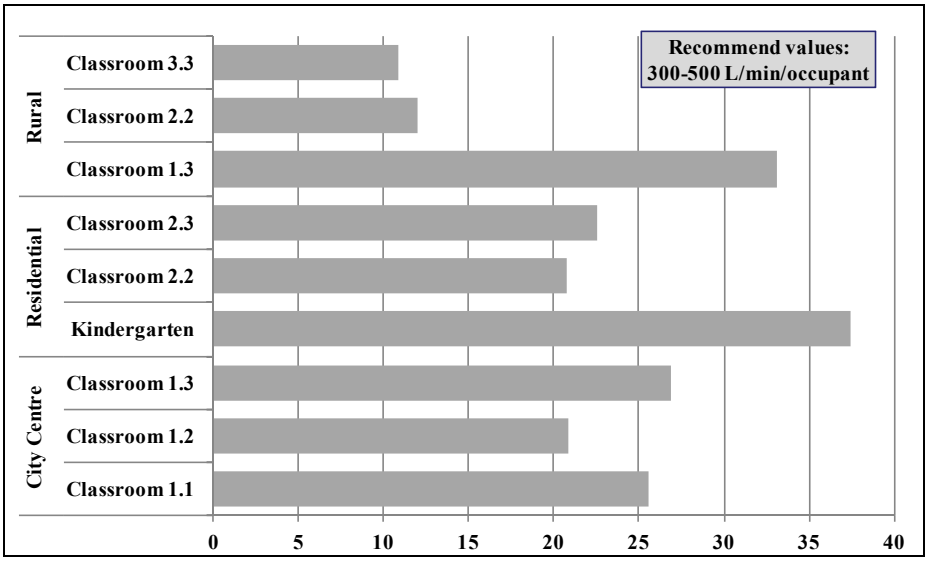


Fig. 2. Ventilation rates (L/min per capita).

Table 3. PM_{10} concentrations (μg/m^3) during occupied and vacant periods.

School	Measurement site	PM_{10} Occupancy	PM_{10} Unoccupancy
Glória (traffic, city centre)	Classroom 1.1	81.0 ± 11.7	70.5 ± 4.00
	Classroom 1.2	104 ± 63.5	45.2 ± 4.73
	Classroom 1.3	70.1 ± 25.2	26.8 ± 21.3
Esgueira (urban, residential)	kindergarten	97.0 ± 15.9	58.2 ± 12.1
	Classroom 2.2	362 ± 83.7	111 ± 36.9
	Classroom 2.3	177 ± 75.0	172 ± 72.8
Eixo (rural)	Classroom 3.1	68.0 ± 19.6	39.7 ± 9.49
	Classroom 3.2	73.7 ± 13.8	53.0 ± 32.6
	Classroom 3.3	62.2 ± 8.93	14.6 ± 4.80

particle concentrations lower in the summer and twice higher during the winter. The high occupants' number associated with the small size of classrooms and poor ventilation, were pointed out as contributors to increased concentrations of PM in the winter. Seven primary schools in Athens, Greece, have been reported to present higher indoor PM_{10} (229 μg/m^3) than outdoor levels (166 μg/m^3) (Diapouli *et al.*, 2008). In a study carried out in a school building near an urban traffic roadway in Delhi city, Goyal and Khare observed maximum PM_{10} concentrations of 14,474 ± 637.5 μg/m^3 at 11:00 to 11:30 a.m., in winter, when recess hour started and finished and children left and entered to the classrooms.

The carbonaceous fraction (TC = OC + EC) accounted for 43 ± 10% of the $PM_{2.5}$ mass indoors. Similar values (39 ± 12%) were found outdoors (Fig. 4). Higher carbonaceous contents (52–80%, averaging 62%) have been reported for $PM_{2.5}$ sampled in an American elementary school in Montana (Ward *et al.*, 2007). Variable TC mass fractions, from about 23 to 60% of $PM_{2.5}$, were observed in seven schoolrooms in Riverside, California (Sawant *et al.*, 2004). A wide interval of values were obtained for the percent amount of TC in PM_{10} (median = 27%) in two schoolrooms in Munich (Fromme *et al.*, 2008). In the present study, while EC represented, on average, 10% of the $PM_{2.5}$ mass collected at the playground of the city centre school, around half of this percentage was obtained for the other two institutions. Lower OC/EC ratios (average = 3.31) for the outdoor air of the city centre school also denote a more pronounced influence of traffic emissions than those observed for the periphery (OC/EC = 4.18) and rural (OC/EC = 4.03) schools. Much higher OC/EC ratios (average values ranging from 4.2 to 9.7) were obtained in the classrooms of all schools (Fig. 5), suggesting that the organic carbonaceous fraction is produced more by indoor sources rather than outdoor pollutant penetration. These indoor OC/EC ratios are within the range (5.3–10.8) measured by Na and Cocker (2005) in four schoolrooms in Riverside, California. Crilley *et al.* (2012) reported OC/EC ratios from 2.72 to 13.6 in $PM_{2.5}$ sampled in 17 schools in different suburbs in the city of Brisbane, Australia. Higher ratios (OC/EC from 5.3 to 33.6) were observed in $PM_{2.5}$ samples collected in elementary and middle schools in Libby, Montana (Ward *et al.*, 2007). To justify the high OC/EC values found in an Australian school, Morawska *et al.* (2009) suggested, in addition to the influence of primary sources, the presence o secondary organic aerosols (SOA). Several research groups have conducted investigations into the formation of SOA in indoor environments through reactions of O_3 with VOCs (Morawska *et al.*, 2009; Waring *et al.*, 2011; and references therein). It can be argued that, unless there is office equipment, the O_3 concentrations in

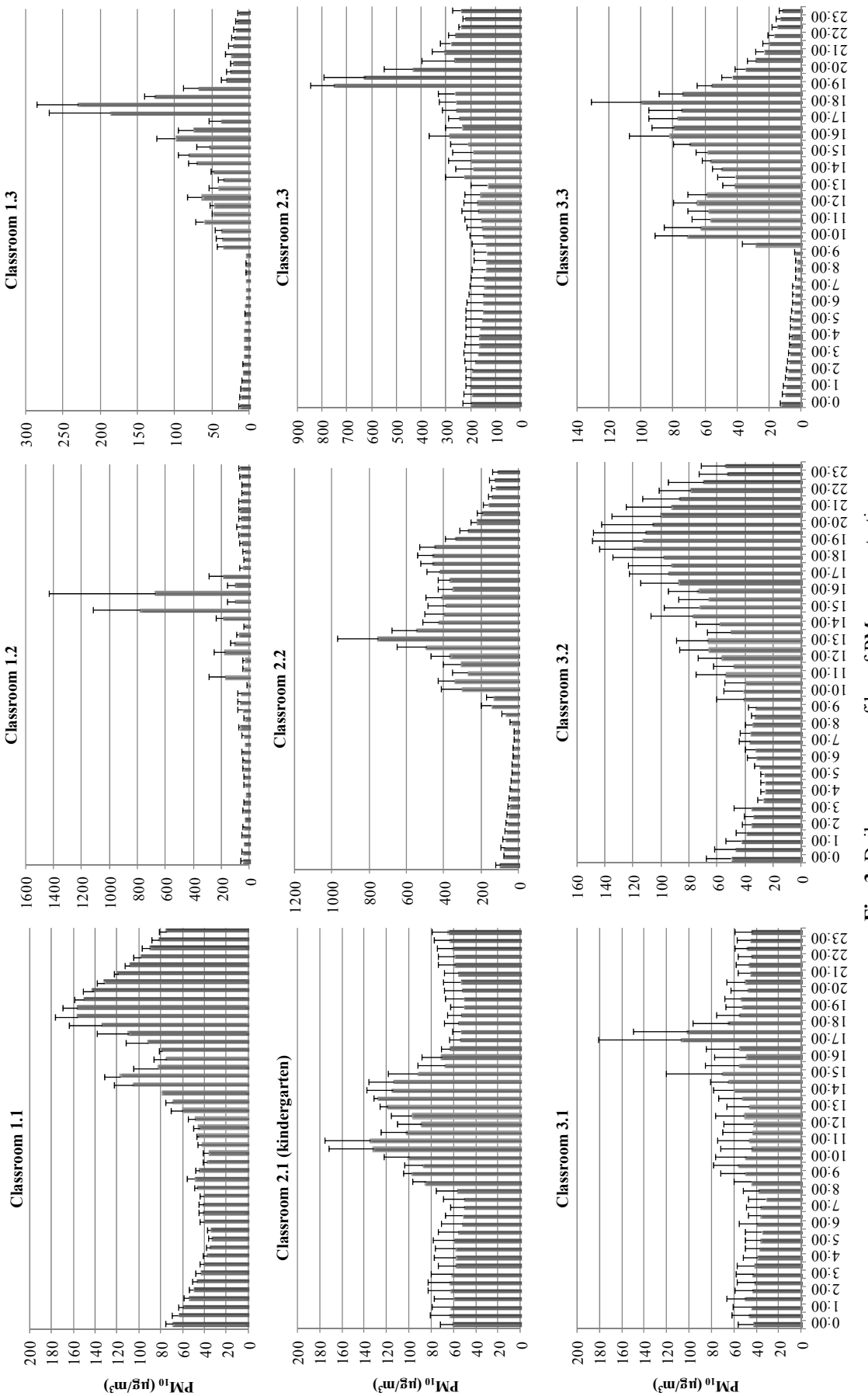


Fig. 3. Daily average profiles of PM_{10} concentrations.

Table 4. Mass concentration ratios (%) between different particle size fractions in this study (classrooms where a Topas monitor was used) and mean values reported in the literature for teaching hours.

	This study			Other studies			
	City centre (Room 1.3)	Residential (Room 2.2)	Rural (Room 3.3)	Goyal and Khare (2009) (a)	Fromme *et al.* (2007) (b)	Halek *et al.* (2009) (c)	Braniš *et al.* (2005) (d)
PM_{10}/TSP	45.6 ± 3.6 (day)	55.4 ± 2.4 (day)	50.3 ± 1.7 (day)	-	-	-	-
	80.9 ± 1.3 (night)	72.3 ± 0.95 (night)	75.2 ± 1.5 (night)				
$PM_{2.5}$/PM_{10}	18.2 ± 3.6 (day)	12.7 ± 1.9 (day)	23.57 ± 3.1 (day)	17.3 (non-winter)	21.6 (winter)	15.3 (winter)	51.8 (day)
	54.8 ± 9.9 (night)	28.0 ± 4.6 (night)	51.5 ± 3.7 (night)	30.5 (winter)	19.6 (summer)		91.4 (night)
PM_1/$PM_{2.5}$	25.8 ± 3.3 (day)	16.3 ± 2.4 (day)	24.3 ± 3.5 (day)	64.5 (non-winter)			62.6 (day)
	41.2 ± 7.2 (night)	20.5 ± 3.4 (night)	39.2 ± 2.3 (night)	81.5 (winter)	-	45.2 (winter)	79.6 (night)

(a) Classroom of a school near an urban roadway in Delhi City; (b) 64 schools (primary and secondary) in Munich; (c) Five elementary schools in Tehran; (d) University lecturing room (Oct.–Nov.)

classrooms are very low and, thus, the SOA formation potential is also very low. However, Waring (2009) and Langer *et al.* (2008) showed that *d*-limonene, used in air fresheners, detergents and other consumer products, even at ozone-very limited conditions, yields large SOA formation events. It is also known that ozone reacts with indoor-emitted terpenoids to form SOA, process that contributes to the rapid decay of this oxidant inside buildings and to the detection of I/O ratios substantially lower than 1 (Waring *et al.*, 2011). On the other hand, taking into account that one of the main primary contributors to indoor particles is probably resuspended dust, some of which associated with soil, high OC/EC ratios are not surprising. In fact, it has been reported that particles from dust resuspension present high OC/EC ratios (e.g., Amato *et al.*, 2009; Chen *et al.*, 2012).

The poor indoor-outdoor correlations of OC (r^2 between 0.22 and 0.30) indicate the presence of multiple carbon sources, and different contributions to indoor and outdoor carbonaceous particles. For the school located in the residential area, a modest correlation (r^2 = 0.30) between the EC concentrations measured inside and outside was obtained, whereas lower correlations were found for the other educational settings. This indicates that, probably, the use of graphite pencils and their sharpening may represent a much more important indoor source of EC than that associated with penetration of outdoor traffic emissions. Concerning the relationships between indoor and outdoor measurements, each intercept roughly reflects EC concentrations that originate exclusively from indoor emission sources because intercepts are the concentration values when outdoor EC contributions are zero (Cao *et al.*, 2012). The percentage of the EC intercept in the average EC concentration indicates the contribution of indoor sources to measured indoor EC levels. For the school located in the residential area, where a better indoor-outdoor relationship was obtained, the intercept/average value was 0.25, indicating that about 25% of indoor EC results from the input of indoor sources. The same procedure applied to OC led to similar results across schools. This crude assessment pointed out that around 15% of the indoor OC has an origin in indoor sources, which we believe to be an underestimation, taking into account the very high I/O ratios.

The slopes of the relationships between anions (Cl^-, NO_3^- and SO_4^{2-}) and cations (Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) expressed by the sum of the equivalent concentration are lower than 1 (average = 0.8). In addition to the contribution of carbonates in some samples, the anion deficits are thought to have been affected by the absence of data on F^-, PO_4^{3-}, NO_2^-, and organic acid salts (Huang *et al.*, 2012). The water soluble inorganic ions represented, as a sum, 16.5 ± 1.5%, 11.0 ± 4.8% and 19.2 ± 5.0% of the $PM_{2.5}$ mass measured in classrooms of the city centre, residential and rural schools, respectively. Sulphate and nitrate were always the dominant ionic species (Fig. 6). These two secondary ions were highly correlated (r^2 = 0.82), suggesting that their precursors were released from similar emission sources. Furthermore, there was no correlation between Ca^{2+} and SO_4^{2-} and between Ca^{2+} and NO_3^-, suggesting there was sufficient ammonia (NH_3) to completely neutralise the

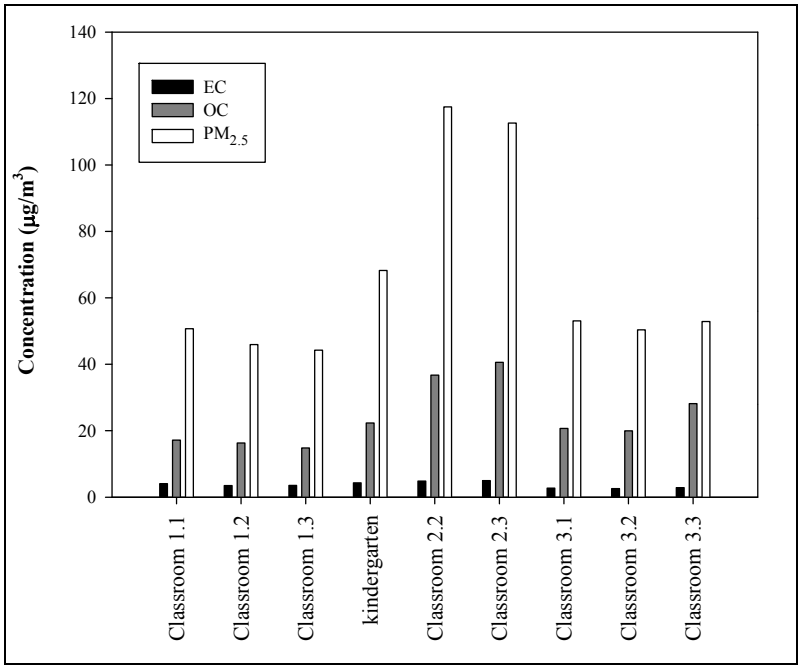


Fig. 4. Average levels of $PM_{2.5}$, OC and EC during occupancy periods.

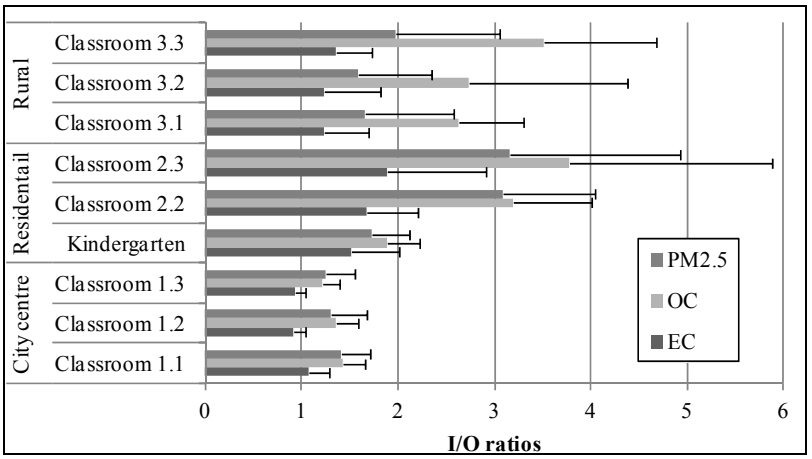


Fig. 5. Indoor-outdoor ratios for $PM_{2.5}$, OC and EC.

sulphuric acid formed from oxidation of SO_2. The molar ratios of outdoor NH_4^+ to SO_4^{2-} (3.40 ± 1.47) indicate their complete neutralisation to $(NH_4)_2SO_4$. However, the NH_4^+ to SO_4^{2-} molar ratios obtained for indoor particles (1.74 ± 0.69) point out that the neutralisation process produced a mixture of $(NH_4)_2SO_4$ and $(NH_4)_3H(SO_4)_2$ (Alves *et al.*, 2007). In addition, the global good correlation between NH_4^+ and NO_3^- (r^2 = 0.79) also suggests aerosol in the form of NH_4NO_3. It has been argued that ammonium nitrate aerosol will partially vaporise as it enters a building, which contributes to I/O ratios lower than 1. The decay of concentrations inside buildings has been attributed to transformation of indoor ammonium nitrate to ammonia and nitric acid gases, which are further lost by sorption and deposition on indoor surfaces (Lunden *et al.*, 2003).

The moderate global correlations between Mg^{2+} and Ca^{2+} (r^2 = 0.50), Mg^{2+} and Cl^- (r^2 = 0.53), and Na^+ and Mg^{2+} (r^2 = 0.45), indicate that these species may be associated with crustal dust. Excepting calcium in all schools, the I/O ratios for the other ions were generally lower than 1, indicating that the sources of these species were mainly outdoor air penetration rather than indoor sources. The very weak indoor-outdoor relationships ($r^2 < 0.20$) indicated that the indoor and outdoor water-soluble ion components had different sources. The outdoor calcium could be related to the resuspension of road dust caused by moving vehicles and exhaust gases. In addition to the calcium delivered by outdoor air penetration, the detection of this species in classrooms may be related to the use of chalk in blackboards. This indoor source could also be responsible for the detection of carbonate and potassium in classrooms. Possible outdoor sources for these two species are, respectively, soil dust and biomass burning. Carbonates were only detected in some samples, especially those obtained in classrooms with

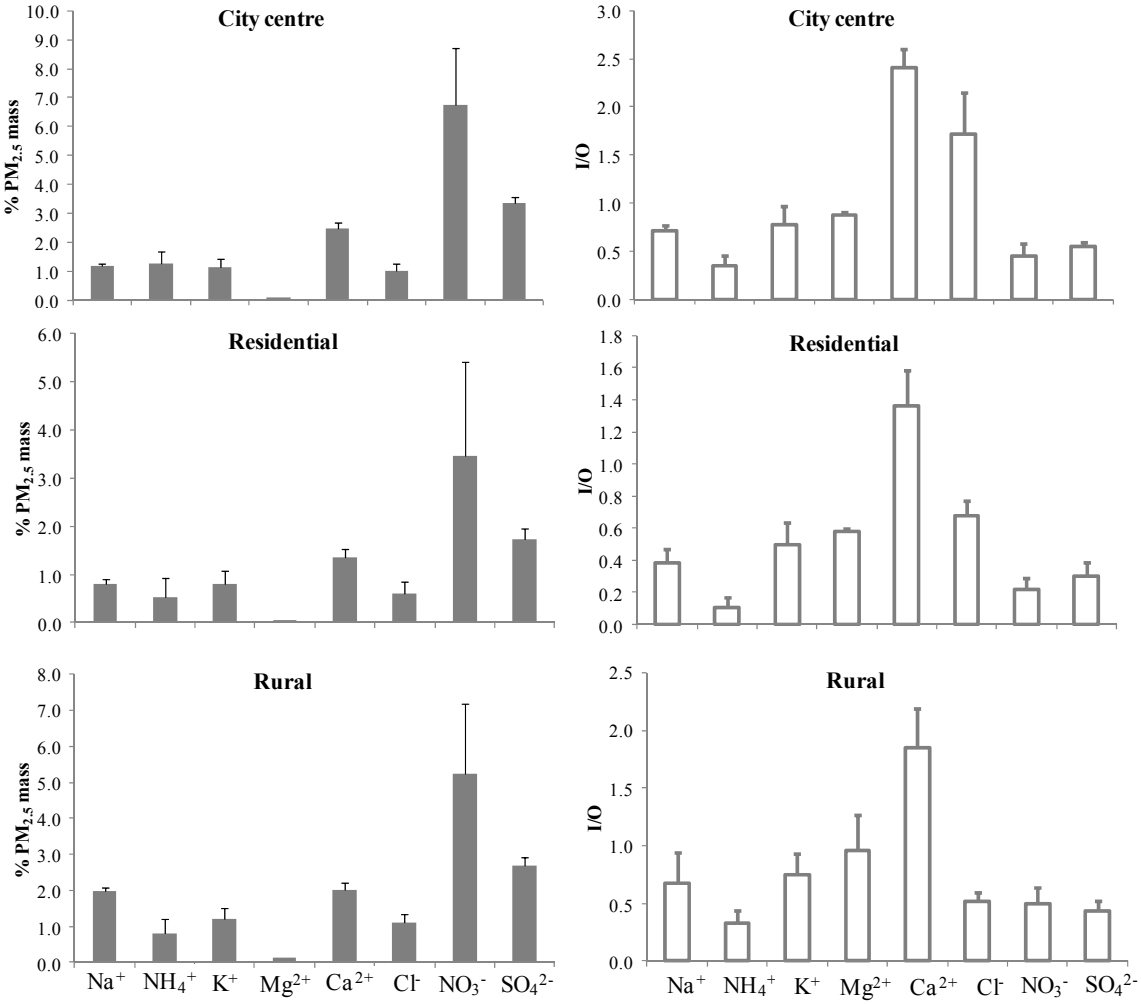


Fig. 6. Mass fractions (percentage) of water soluble ions in indoor $PM_{2.5}$ and indoor-outdoor ratios.

use of chalk or in outdoor samples from the school with unpaved playground. In these samples, carbonates represented up to 9% of the $PM_{2.5}$ mass.

Correlations between Indoor $PM_{2.5}$ and Comfort or Chemical Parameters

The $PM_{2.5}$ concentrations were correlated either with their chemical constituents or with comfort parameters (Table 5). No significant correlations between meteorological parameters and $PM_{2.5}$ levels were observed. Although not statistically significant, the negative correlations between $PM_{2.5}$ and RH indicate that aerosol levels decrease with increasing water vapour in the air. Inconsistent results have been reported for other schools. While Fromme *et al.* (2007) also observed negative correlations for schools in the city of Munich and a neighbouring district, Braniš *et al.* (2005) presented positive associations between the two parameters. It has been argued that RH promotes the formation of ammonium nitrate, but an increase in precipitation causes a decrease in all $PM_{2.5}$ components through scavenging (Tai *et al.*, 2010). Some days of rain throughout the sampling campaign in the Aveiro region may have contributed to particle scavenging in the outdoor air and, consequently, to the decrease of $PM_{2.5}$ levels indoors.

Statistically significant positive correlations between CO_2 and $PM_{2.5}$ were found in all schools, confirming that the human presence and related activities represent an important source of particles indoors. This also indicates, once again, that inadequate ventilation plays a major role in the establishment of poor IAQ.

The *r*-square of the correlations between $PM_{2.5}$ and OC indicates that this carbonaceous component can explain 44, 91 and 45% of the variation of the indoor particle concentrations in the city centre, residential and rural schools, respectively. The statistically significant positive correlations between these two variables highlight the relevance of organic matter sources or formation processes to the particulate matter in classrooms.

Significant positive associations were found between $PM_{2.5}$ and both K^+ and Ca^{2+}, which may be related to the use of chalk. In addition, these two water soluble ions may be associated with crustal dust. Calcium may also derive from the gradual decay of construction materials in the interior of buildings. A very significant correlation between $PM_{2.5}$ and Mg^{2+} was observed in the rural school. Besides being a crustal element, magnesium is part of the chemical

Table 5. Correlations (r) and p-values (in brackets) between $PM_{2.5}$ concentrations and chemical constituents or comfort parameters for the 3 schools.

	City centre	Residential	Rural
T	0.052 (0.85)	–0.154 (0.58)	0.028 (0.92)
RH	–0.221 (0.43)	–0.264 (0.34)	–0.320 (0.25)
CO_2	0.515 0.049)*	0.684 (0.0049)**	0.547 (0.035)*
OC	0.667 (0.0066)**	0.955 (0.0001)***	0.670 (0.0063)**
EC	–0.065 (0.81)	0.270 (0.33)	0.560 (0.030)
Na^+	0.231 (0.41)	–0.470 (0.077)	0.352 (0.20)
NH_4^+	–0.362 (0.18)	0.085 (0.76)	–0.010 (0.97)
K^+	0.643 (0.0097)**	0.557 (0.031)*	0.360 (0.19)
Mg^{2+}	0.526 (0.044)	–0.359 (0.19)	0.817 (0.0002)***
Ca^{2+}	0.417 (0.12)	0.544 (0.036)*	0.736 (0.0018)**
Cl^-	0.441 (0.099)	–0.172 (0.54)	0.179 (0.52)
NO_3^-	–0.255 (0.36)	0.425 (0.11)	–0.229 (0.42)
SO_4^{2-}	0.125 (0.66)	0.073 (0.80)	–0.044 (0.88)

* $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$

formulation of many consumer products and construction materials (e.g., inks, toners, sealants, alloys, some types of bricks, etc.). No significant correlations between other chemical constituents and $PM_{2.5}$ levels were obtained.

CONCLUSIONS

In most indoor spaces, the average temperatures and relative humidity values were outside the comfortable ranges stipulated by international organisations. Very high CO_2 levels and extremely low outdoor air infiltration indicate excessive building tightness, very deficient ventilations and excessive room occupancy. Exposure to particulate matter in schools is high. I/O ratios greater than one for particles indicate the presence of indoor sources other than the contribution from outdoor. Indoor concentrations are strongly influenced by activities and movement of occupants, which may allow resuspension of previously deposited particles or their delayed deposition or settling. Implementation of more breaks and recesses between classes, decreasing the occupancy per room, increasing the exchange of indoor air with the outdoor, reducing human activities and improving cleanliness of facilities might alleviate the high CO_2 and particle levels. Despite there being a large literature on class-size effects on academic achievement, in times of austere budgets, legislative actions to increase the number of pupils per class have been undertaken. However, taking into account the IAQ problems observed in most schools, policymakers need to bear in mind the possible negative effects of any increase in class size. The need to judiciously weigh costs and benefits is imperative. To develop effective risk assessment and management strategies, toxicological evaluations of particles is highly recommended in future researches. Moreover, strategies to minimise the exposure must be implemented and assessed for effectiveness in intervention studies aiming at reducing health risks of children. It should be taken into account that the benefits resulting from the adoption of strategies to improve IAQ outweigh the investment. In fact, there is growing evidence that interventions induce significant health benefits, which are largely greater than their costs.

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