



Indoor/Outdoor Relationships between PM₁₀ and Associated Organic Compounds in a Primary School

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ABSTRACT

Airborne particulate matter (PM₁₀) samples were collected daily, indoors and outdoors, in a primary school at Aveiro, Portugal, from February 28 to May 27, 2011. The carbonaceous content (organic and elemental carbon) was determined by a thermo-optical technique. The organic speciation of PM₁₀ was performed by gas chromatography-mass spectrometry. Mean PM₁₀ levels of 107 and 36 µg/m³ were obtained in the schoolroom and outdoors, respectively. On average, organic carbon accounted for 30.0% of the mass of PM₁₀ indoors, whereas a lower mass fraction of 21.3% was found outdoors. The lack of correlation between indoor and outdoor organic carbon and the much higher indoor levels suggest significant contributions by indoor sources. The most abundant organic compound classes were acids, sugars, polyols and *n*-alkanes. Infiltration of outdoor particles leads to contamination of the schoolroom with vehicle emissions, as well as emissions related to the burning of biofuel in nearby restaurants and bakeries. However, the much higher indoor concentrations than in the outdoor air for the majority of compounds suggest that the origin of much of the particulate matter is from within the school building (due to school activities and materials, skin debris, microorganisms, and so on), and this also includes the formation of secondary organic aerosols. Based on the concentrations of polycyclic aromatic hydrocarbons, a negligible cancer risk was estimated in relation to the air within the school.

Keywords: PM₁₀; Organic carbon; Elemental carbon; Classroom; Organic tracers.

INTRODUCTION

The impact of air pollution on school children's health is currently one of the key focus of international organisations. Children spend up to ten hours per day at school and the health impact of air pollutants is much higher for pupils than for adults in similar environments (Guo *et al.*, 2010). Children breathe higher volumes of air relative to their body weights. Their physiological vulnerability to air pollution derives from the narrower airways and the fact that their lungs are still developing (Foos *et al.*, 2008; Ginsberg *et al.*, 2008). Moreover, many children breathe through their mouths, bypassing the filtering mechanism of the nose. Thus, children are more likely to suffer the consequences of indoor pollution (Selgrade *et al.*, 2008). Particulate matter is one of the most important pollutants in indoor air. In spite of the various studies performed worldwide to assess

the pupils' exposure to indoor particles, only a few aimed at characterising their chemical composition and most of them were mainly focused on the elemental content (e.g., Kuruvilla *et al.*, 2007; Molnár *et al.*, 2007; Stranger *et al.*, 2008; Almeida *et al.*, 2011; Tran *et al.*, 2012). Comparatively almost nothing is known about the organic matter in particulate matter, which can encompass irritant, carcinogenic and/or mutagenic compounds. As far as we know, the organic speciation of airborne particles in schoolrooms has only been done by Pegas *et al.* (2012a). The analysis of organic compounds in indoor environments besides schools (residences, offices or commercial spaces) was the target of a few studies (Naumova *et al.*, 2002; Ohura *et al.*, 2004; Johannesson *et al.*, 2009; Jung *et al.*, 2010; Khedidji *et al.*, 2013; Sangiorgi *et al.*, 2013), but these have focused on the quantification of polycyclic aromatic hydrocarbons (PAHs), due to their carcinogenic potential. The large number of compounds likely present, the very time-consuming and expensive analyses, the complex formation mechanisms and the uncertain role of organic particulates in climate (Calvo *et al.*, 2013), have centred the attention almost exclusively on the outdoor air. Since organic compounds are abundant components of atmospheric aerosols (Alves, 2008), our

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understanding of the numerous sources, formation processes, health implications and other environmental issues surrounding airborne particles remains seriously hindered without more information on this fraction. This is even more striking for indoor environments.

In this study, a sampling campaign of indoor and outdoor airborne particulate matter was undertaken in a school classroom to: (i) investigate mass concentrations, (ii) obtain the speciated organic composition, (iii) assess the potentially harmful effects and (iv) evaluate the influence of outdoor air pollution on the indoor air, as well the possible contribution of indoor sources.

METHODOLOGIES

Sampling

An intensive sampling campaign of indoor and outdoor airborne particulate matter (PM₁₀) was carried out in a classroom of a city centre primary school of Aveiro, Portugal, from February 28 to May 27, 2011. The classroom had a wood flooring, water-based paint covering the walls, blackboard with chalk, white board with markers, and five wooden windows. It was naturally ventilated. The area of the room was 52.5 m². The number of students in the class was 25. The school is surrounded by commercial and residential buildings and adjacent to a parking lot. It is located at the interception of two busy roads controlled by traffic lights.

On working days, during the occupancy periods, simultaneous indoor and outdoor sampling of particulate matter with equivalent aerodynamic diameter less than 10 µm (PM₁₀) was done. To avoid noise inside the classroom, a Gent sampler operating at about 12.4 L/min was used, but the pump and flow control devices were placed in the outside hallway. Following the teacher' demand, aiming at allowing the free movement within the classroom, the sampling head was positioned at a distance of about 1 m from the lateral windowless wall and at an approximate height of 1.5 m above floor. This location avoided direct contamination of the filters with chalk dust, especially when blackboards on the front wall were used. An Echo TCR Tecora sampler (38.3 L/min), following the EN 12341 norm, was used outdoors. The instrument was located at a height of about 2 m. The regression slopes for the results obtained from parallel measurements involving both samplers over one week, before the sampling campaign, differed by less than 5%. The correlation coefficient was 0.97. The PM₁₀ samples were collected onto pre-baked (6 h at 500°C) 47 mm diameter quartz filters.

Analytical Techniques

After gravimetric determination of the PM₁₀ mass under controlled temperature and relative humidity conditions, filter punches were analysed by a thermo-optical transmission system in order to obtain the carbonaceous content (organic carbon, OC, and elemental carbon, EC). These determinations were done after passive exposure of filters to HCl vapours to eliminate interferences from carbonates. The analytical protocol was described in detail elsewhere (Alves *et al.*,

2011).

The five consecutive filters of each week were combined to perform the organic extractions. Thus, "average" weekly concentrations were obtained. The combined filters were extracted together by refluxing 300 mL of dichloromethane (Fisher Scientific) for 24 h to obtain non-polar and semi-polar, and, to a lesser extent, polar compounds. Taking into account that the extraction efficiency of dichloromethane for polar compounds is around 75% (Gonçalves *et al.*, 2011), after filtration, the filters were extracted 3 times with methanol (Fisher Scientific) in an ultrasonic bath (75 mL for 10 min, each extraction) to guarantee a 100% recovery of polar compounds. All the 4 extracts were then combined, vacuum concentrated and dried under a gentle nitrogen stream. The total organic extracts were subsequently separated into five different organic fractions by flash chromatography with silica gel (230–400 mesh, 60 Å Merck Grade 9385) and various solvents of increasing polarity: 1st - aliphatics (hexane); 2nd - aromatics (hexane: toluene); 3rd - carbonyls and methyl esters (hexane: dichloromethane); 4th - hydroxyl compounds (hexane: ethyl acetate); 5th - acids and sugars (formic acid: methanol). Following each elution, the different fractions were vacuum concentrated and evaporated under an ultra-pure nitrogen stream. Before injection, the fractionated extracts that included more polar compounds were derivatised to trimethylsilyl ethers. Finally, the extracts were analysed by gas chromatography-mass spectrometry (GC-MS) with a TRB-5MS 60 m × 0.25 mm × 0.25 µm column (Alves *et al.*, 2011). The GC-MS system was accurately calibrated using about 150 high purity individual compounds (Sigma-Aldrich, TSI and Chiron) at four/five different concentration levels. All samples and authentic standards were injected with two internal standards: tetracosane-d50 (Sigma-Aldrich) and 1-chlorohexadecane (Merck). Additionally, the EPA 8270 semi-volatile internal standard mix (Supelco), containing six deuterated compounds (1,4-dichlorobenzene-d4, naphthalene-d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12, perylene-d12), has been used for polycyclic aromatic hydrocarbon (PAH) analysis. Each organic extract was injected using whether the scan mode or the selected ion monitoring (SIM) mode. Compound identification was made by comparison with authentic standards, matching against Wiley and NIST spectral libraries and analysis of fragmentation patterns. For those compounds with no authentic standards available, relative response factors were obtained as an average from the overall homologous series or from compounds of similar chemical structure and retention time.

RESULTS AND DISCUSSION

The daily indoor PM₁₀ concentrations during occupancy periods ranged from 37 to 229 µg/m³, averaging 107 µg/m³ (Fig. 1). Much lower levels, from 9.3 to 94 µg/m³ (average = 36 µg/m³), were registered outdoors. 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. It has been pointed out that the high indoor levels are likely associated with the physical activities of pupils, which

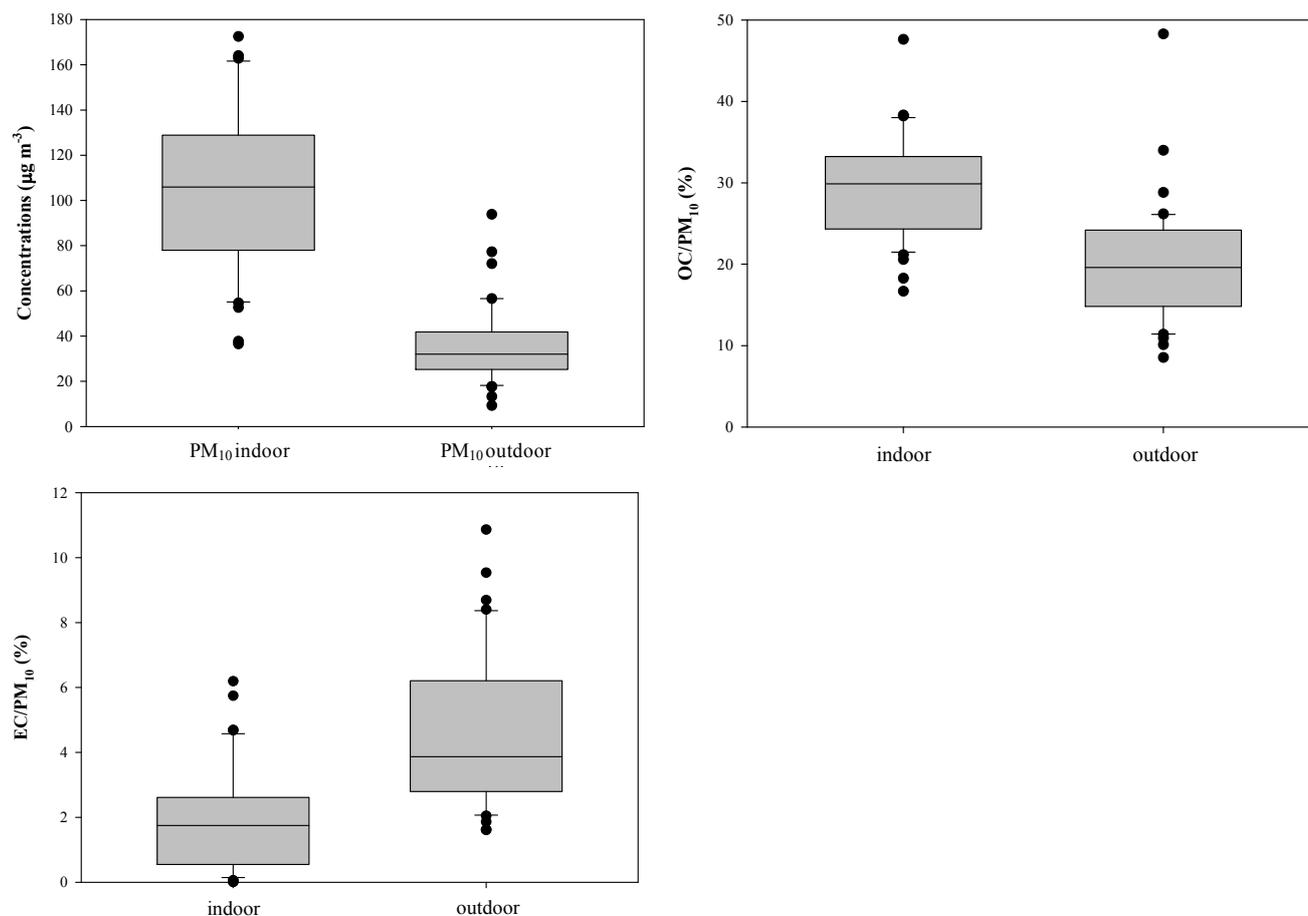


Fig. 1. Box plots of particulate matter concentrations and OC or EC-to-PM₁₀ percentage ratios obtained throughout the campaign.

contribute to a constant process of resuspension of sedimented material from the daily tasks, from skin desquamation or clothing, and from outdoor sources (Lee and Chang, 2000; Ekmekcioglu and Keskin, 2007; Fromme *et al.*, 2007; Diapouli *et al.*, 2008). Continuous measurements of PM₁₀ by light scattering monitors in 9 elementary schoolrooms, carried out in January 2012, in which the classroom of the present study was also included, indicated that maximum concentrations coincided with the cleaning activities or were registered when students were entering or leaving the classrooms (Alves *et al.*, 2013). Among the cleaning practices, sweeping was considered one of the main contributors to the PM₁₀ peaks. The room of the present study was swept daily.

On average, the OC accounted for 30.0% of the mass of PM₁₀ indoors, whereas a lower mass fraction (OC/PM₁₀ = 21.3%) was found outdoors. EC represented 1.9% of the PM₁₀ measured in the classroom, while the influence of traffic at roadside is reflected in higher EC mass fractions (EC/PM₁₀ = 4.6%). The *r*-square of the correlations between PM₁₀ and OC indicates that this carbonaceous component can explain 55 and 60% of the variation of the indoor and outdoor particle concentrations, respectively (Fig. 2). The outdoor EC explains 32% of the variance of PM₁₀ levels, whilst a lack of correlation between these two variables was observed indoors. OC and EC were somewhat correlated

outdoors ($r^2 = 0.41$), while no relationship was found indoors. A higher correlation indicates that these carbonaceous fractions have common sources or are affected by the same processes. The correlation between indoor and outdoor EC ($r^2 = 0.39$) suggests that outdoor EC can explain 39% of the variation for indoor EC concentrations. This fact, associated with a slightly higher proportion of indoor concentrations (1.2 times) than those registered outdoors, is indicative that most of the EC detected in the classroom originates outdoors. However, part of the emissions of EC in the classroom may be related to the sharpening of graphitic pencils. The lack of correlation between indoor and outdoor OC is probably due to significant contributions by indoor sources. Clearly, OC is enriched in indoor as compared to outdoor air.

Indoor enrichments (IEs) were defined as the fractional difference between the concentrations in the classroom and outside:

$$IE_i(\%) = \frac{x_i^I - x_i^O}{x_i^I} \times 100 \quad (1)$$

where *i* is a given chemical component (variable), *x* is the concentration of the component *i*, and I and O stand for indoor and outdoor, respectively.

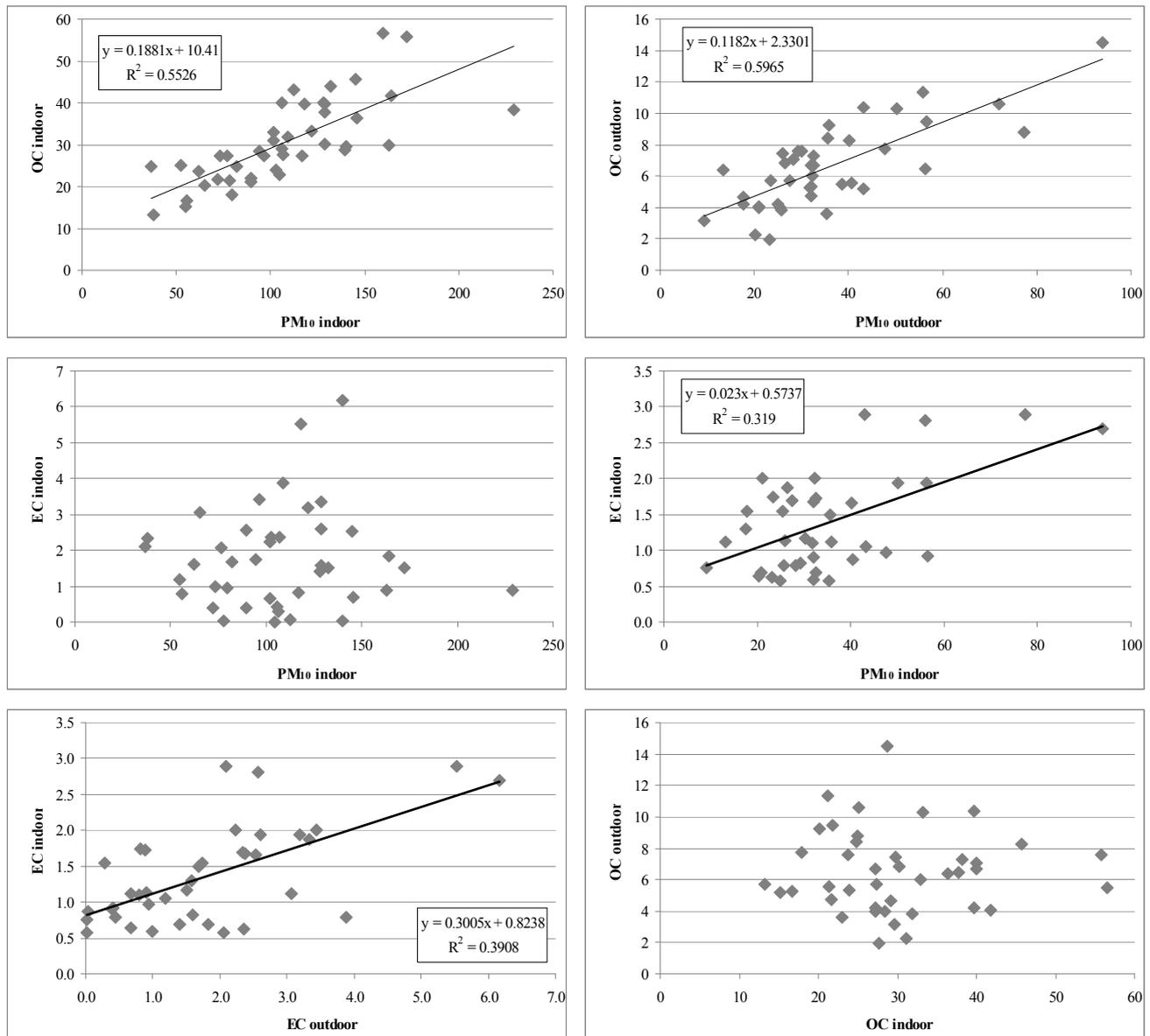


Fig. 2. Correlations between concentrations ($\mu\text{g}/\text{m}^3$) of PM₁₀ and its carbonaceous fractions.

It can be clearly seen that concentrations of most organic compounds were many times higher than their homologous outdoor levels (Fig. 3). This may be explained by active indoor sources, activities/movements of the occupants and reduced ventilation rates as a result of closed doors and windows, so causing accumulation. Among the indoor sources of organic compounds, submicrometer fragments of paper, skin debris, clothing fibres, cleaning products and waxes may be considered (Schneider, 2008; Weschler, 2012; You *et al.*, 2013). Air exchange rates below 1 h^{-1} , such as those usually registered in Portuguese classrooms, including the school focused in this study (Alves *et al.*, 2013), have been reported to contribute to the enrichment of particulate pollution in indoor environments (Abt *et al.*, 2000; Rojas-Bracho *et al.*, 2000). A previous work conducted in the classroom focused in the present study, in which gaseous pollutants and particulate inorganic constituents were

monitored, found out low ventilation rates, even with the window open (Pegas *et al.*, 2012b). The estimated ventilation rates ranged from 11 (window closed) to 23 L/s (window open). The maximum ventilation value, which corresponded to about 0.9 L/s per person, represented only 18% of the minimum value of 5 L/s per person recommended by the ANSI/ASHRAE Standard 62-2001.

It has been demonstrated that the indoor/outdoor (I/O) particle relationship is significantly influenced by the air exchange rate. Chen and Zhao (2011) provided a revision for both experimental and modelling studies on this subject. Some of the studies reviewed by these authors show that the I/O ratio with windows open is higher than the ratio with windows closed, while the opposite is concluded in other works. Also the I/O ratios under natural ventilation were compared with ratios under mechanical ventilation and no uniform conclusion could be drawn. These disparities

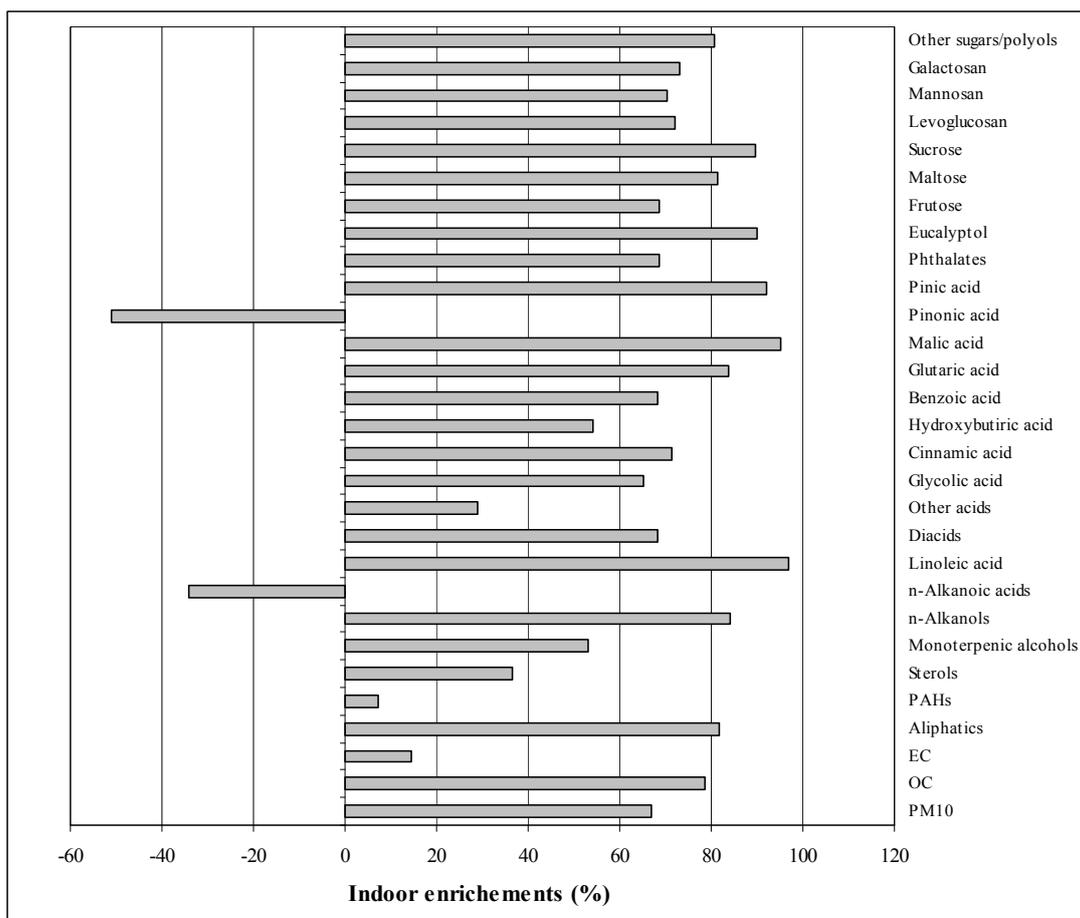


Fig. 3. Fractional differences between average concentrations of diverse compounds or group of compounds registered indoors and outdoors.

may stem from many factors, including the use of different measurement methodologies. In addition, the air exchange rates depend on many influencing aspects, such as penetration factor, deposition rate, indoor particle source emission rate and outdoor levels. According to Chen and Zhao (2011), if there is no indoor particle source, the I/O ratio will increase with the increase of air exchange ratio. On the other hand, if the indoor particle source emission rate is high and the outdoor levels are low, the I/O ratio will decrease with the increase of the air exchange rate.

The OC enrichment in the classroom is also confirmed by high OC/EC ratios (> 4.6). To justify the high OC/EC values found in an Australian school, Crilley *et al.* (2009) suggested, in addition to the influence of primary sources, the presence of secondary organic aerosols (SOA). The formation of SOA in indoor environments was demonstrated and confirmed in many test chamber experiments (e.g., Destailats *et al.*, 2006; Aoki and Tanabe, 2007). Sorption of semi volatile organic compounds could be as well an important indoor source of organic matter. Sorption processes seem to be more important in the indoor air compared to the outdoor air (Naumova *et al.*, 2003). Thus, the combination of active indoor sources, sorptive processes and SOA formation leads to an enrichment of the indoor particles in OC and to high OC/EC ratios inside the classroom.

The dominant organic compound classes were acids, sugars, polyols and *n*-alkanes (Fig. 4). The aliphatic fraction of particulate matter comprised *n*-alkanes from C_{16} or C_{17} to C_{36} . The maximum concentrations were observed for the homologues in the range C_{24} – C_{30} (Table 1). Whereas the *n*-alkanes with a carbon number around C_{20} typically derive from unburnt fuel, the homologues around C_{25} are mainly originated from lubricating oil. *n*-Alkanes with these anthropogenic origins do not reveal any clear odd or even carbon number predominance. Another significant source of atmospheric *n*-alkanes is represented by the higher plant waxes, which are emitted due to abrasion from leaves or as products of incomplete biomass combustion (Alves *et al.*, 2010; and references therein). The main homologues in these biogenic emissions are C_{27} , C_{29} or C_{31} . The carbon preference indices of *n*-alkanes around 1, both indoors and outdoors, obtained in this study suggest that infiltration of outdoor particulates leads to contamination of classrooms with vehicle emissions.

PAHs were detected at indoor and outdoor concentrations of 893 ± 650 and 830 ± 421 pg/m^3 , respectively, never exceeding the benzo[a]pyrene (BaP) equivalent carcinogenic threshold of $1 \text{ ng}/\text{m}^3$ set by the World Health Organisation. The BaP dose equivalent (BaPE) for each PAH is calculated by multiplication of the measured concentrations by the

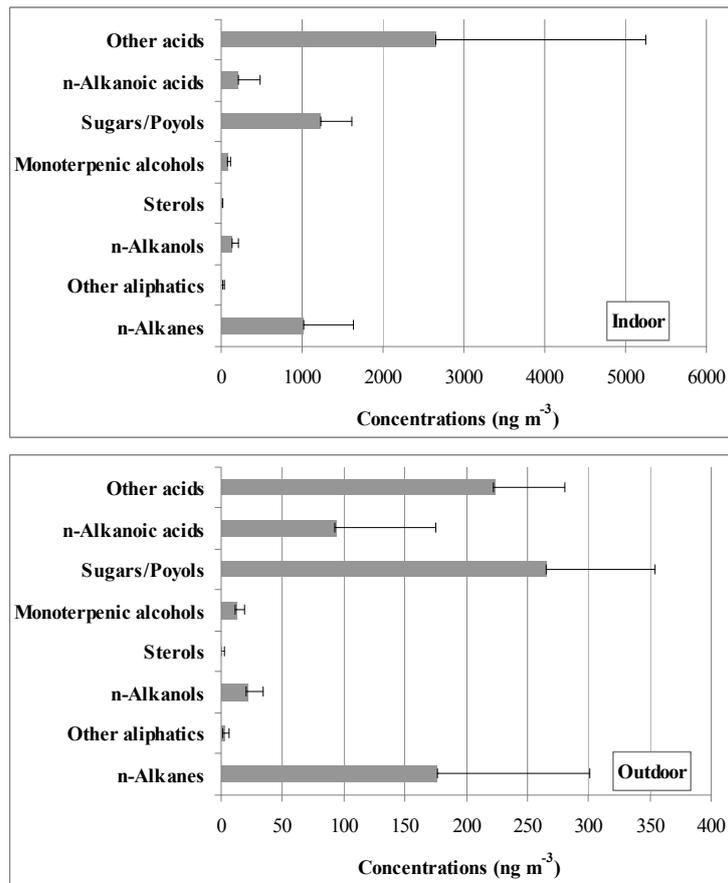


Fig. 4. Concentrations (ng/m³) of the main organic compound classes in PM₁₀.

Table 1. Homologous compound series and diagnostic parameters with constituents of the PM₁₀ samples.

	Indoor	Outdoor
<i>n</i> -alkanes	C ₁₇ –C ₃₆ Max € [C ₂₄ –C ₃₀]; C _{max} = C ₂₇ , C ₂₈ CPI = 0.98 ± 0.04	C ₁₆ –C ₃₆ Max € [C ₂₄ –C ₃₀]; C _{max} = C ₂₄ , C ₂₇ CPI = 1.01 ± 0.05
<i>n</i> -alkanols	C ₁₀ , C ₁₅ , C ₁₈ , C ₂₂ , C ₂₃ , C ₂₅ , C ₂₇ , C ₂₈ C _{max} = C ₁₈	C ₁₀ , C ₁₅ , C ₁₈ , C ₂₂ , C ₂₃ , C ₂₅ , C ₂₇ , C ₂₈ C _{max} = C ₁₈ , C ₂₈
<i>n</i> -alkanoic acids	C ₈ –C ₂₄ C _{max} = C ₁₈ CPI = 7.50 ± 3.81	C ₈ –C ₂₄ C _{max} = C ₁₆ , C ₁₈ CPI = 11.3 ± 7.55
BaPE (pg/m ³)	76.9 ± 92.2	52.1 ± 53.4
Flu/(Flu + Pyr)	0.71 ± 0.20	0.64 ± 0.26
L/M	5.33 ± 2.74	5.15 ± 0.94
L/(M + G)	4.33 ± 2.35	3.83 ± 0.89

CPI (carbon preference index) – summation of odd carbon number homologues divided by the summation of even carbon, for *n*-alkanes, and the inverse for *n*-alkanoic acids; C_{max} – homologues with maximum concentrations; BaPE – benzo[a]pyrene equivalent concentration; Flu – Fluoranthene; Pyr – Pyrene; L – Levoglucosan; M – Mannosan; G – Galactosan.

respective toxic equivalent factor (TEF). The TEFs were taken from Bari *et al.* (2011). The BaP equivalent dose is then calculated as a sum to express the carcinogenicity of the mixture:

$$\text{BaPE} = \sum \text{PAH}_i \times \text{TEF}_i \quad (2)$$

The TEFs were also used to calculate the proportion of total

carcinogenic potential represented by each individual PAH:

$$(\% \text{Carc. Potential})_i = \frac{(\text{RC} \times \text{TEF})_i}{\sum_{i=1}^N (\text{RC} \times \text{TEF})_i} \times 100 \quad (3)$$

where RC is the ratio of the individual PAHs to the

carcinogenic marker BaP. The compound that contributes most to the total carcinogenic potential of the PAH mixture was always BaP, with average values of 72% indoors and 59% outdoors (Fig. 5). The second highest contributor to the total carcinogenicity was benzo[j]fluoranthene with average shares of 9% and 19% in the classroom and outdoor air, respectively. Benzo[b]fluoranthene contributed to 15% of the total carcinogenic risk outdoors, but its representativeness in the schoolroom was much lower (less than 2%). The carcinogenic risk was estimated as follows:

$$\text{Carcinogenic risk} = \text{PAH}_i \times \text{TEF}_i \times \text{UR} \quad (4)$$

where UR represents the cancer unit risk, i.e., the excess cancer risk associated with an inhalation of $1 \mu\text{g}/\text{m}^3$ of a compound. It is obtained by multiplication of the cancer potency factor for BaP [$3.9 (\text{mg}/\text{kg}/\text{d})^{-1}$] by the reference child inspiration rate per day (12.4 m^3) and dividing by the reference child body weight (21 kg) multiplied by a conversion factor from mg to ng of 10^6 (Bari et al., 2011; Elert et al., 2011). The average carcinogenic risk to occupants of the classroom was found to be 1.8×10^{-7} . A slightly lower value was obtained outdoors (Fig. 5). In general, USEPA considers excess cancer risks that are below about one chance in a million (1×10^{-6}) to be so small as to be negligible.

The PAHs with highest concentrations in the outdoor air were fluoranthene ($1127 \pm 77 \text{ pg}/\text{m}^3$), chrysene ($1263 \pm 108 \text{ pg}/\text{m}^3$) and pyrene ($1052 \pm 140 \text{ pg}/\text{m}^3$). The corresponding

indoor levels were 1474 ± 173 , 622 ± 123 and $836 \pm 187 \text{ pg}/\text{m}^3$. Thus, the concentration of fluoranthene outdoors was lower compared to indoors, but chrysene and pyrene showed the opposite results. It should be noted that fluoranthene consists of naphthalene and a benzene unit connected by a five-membered ring. It is a non-alternant PAH, because it has rings other than those with six carbon atoms, and is a structural isomer of the alternant PAH pyrene. It is not as thermodynamically stable as pyrene, which (like chrysene) consists of four fused benzene rings. It has been shown that, depending on the physical and chemical characteristics, different compounds, although belonging to the same class, may present different affinities for surfaces and different sorptive tendencies (Zhang et al., 2002; Singer et al., 2007; Matt et al., 2011). Thus, sorption and re-emission of fluoranthene from indoor surfaces and materials may be more intense compared to those of pyrene and chrysene. However, this hypothesis requires further investigation or confirmation. Fluoranthene/(fluoranthene + pyrene) ratios > 0.5 , both indoors and outdoors, denote a dominant influence of coal/wood combustion (Katsoyiannis et al., 2011). The polyaromatic compound with maximum concentrations in the classroom was phenanthrene, which is emitted by incomplete combustion of petroleum products or in the tobacco smoke. The indoor concentrations ($1205 \pm 253 \text{ pg}/\text{m}^3$) exceeded those measured outdoors ($436 \pm 86 \text{ pg}/\text{m}^3$). Its detection in the indoor air may be due to infiltration of outdoor air and sorption on materials and surfaces.

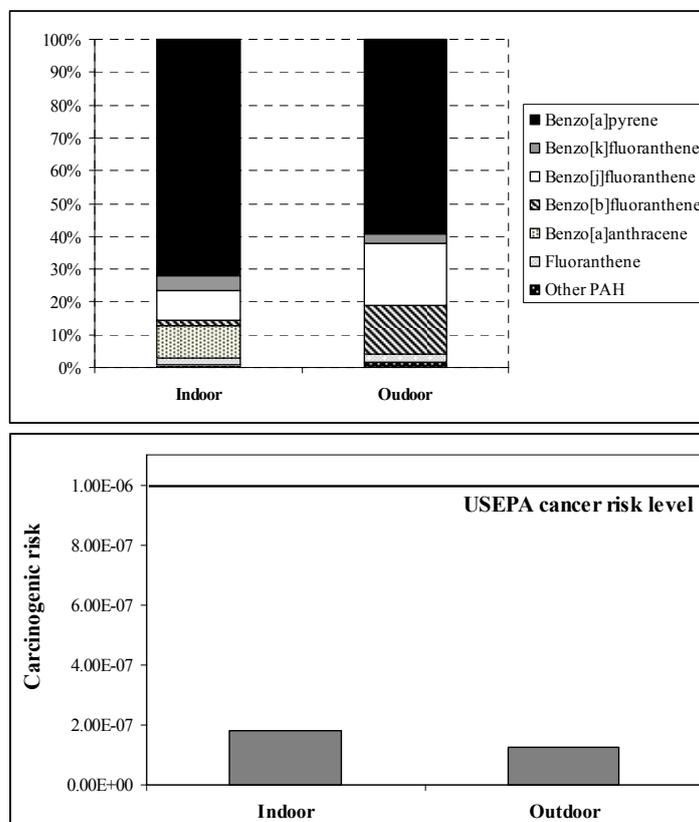


Fig. 5. Contribution of individual carcinogenic activities to the total carcinogenic potential of the PAH mixture (upper panel); comparison of carcinogenic risks with the United States Environmental Protection Agency safety value (lower panel).

Discontinuous series of *n*-alkanols from C₁₀ to C₂₈ were detected in the PM₁₀ samples, maximising at C₁₈ indoors and at C₁₈ or C₂₈ outdoors. This latter compound is derived from vegetation waxes. Octadecanol has been isolated from plants, insects, and human sebaceous lipids. It is also a constituent of many household or personal care products, such as pharmaceuticals, cosmetic creams, emulsions, textile oils and finishes, antifoam agents, etc. (Anonymous, 1985).

A series of *n*-alkanoic acids from C₈ to C₂₄, maximising at C₁₆ or C₁₈, were identified. While docosanoic and tetracosanoic acids have been associated with plant waxes, the other homologues are ubiquitous, involving either combustion processes or wax-like extracts of fungi (moulds), bacteria, spores, pollen, and algae (Alves et al., 2012). A possible source of organic acids in indoor environments includes direct emission from people (e.g., skin oils) (Arhami et al., 2010). The strong even over odd carbon number predominance, i.e., high CPI values, suggests the prevalence of biogenic sources. The ratio of C_{18:2} to C_{18:0} can be used as an indicator for aerosol aging. Average values of 0.06 and 0.11 were observed in the schoolroom and outside the building, respectively. The much lower ratio obtained inside the school corresponds to a more processed aerosol as unsaturated fatty acids can be photo-chemically degraded, while saturated fatty acids are more stable in the atmosphere (Alves et al., 2012). The lack of correlation ($r^2 = 0.07$) between indoor and outdoor concentrations and a negative indoor enrichment are indicative of different sources or processes affecting this type of acids in both environments.

A sequence of *n*-alkanedioic acids (C₃–C₁₀) were also identified as PM₁₀ constituents (Table 2). Diacids may have different origins including primary emissions from biomass burning and fossil fuel combustion, as well as photochemical oxidation of organic precursors of both anthropogenic and biogenic origin (Winterhalter et al., 2009; and references therein; Fu et al., 2013). With regard to the influence of biomass burning, no significant correlations were observed between the measured diacids (individually or as a sum) and levoglucosan, a specific tracer for biomass burning (Fabbri et al., 2009). This indicates that biomass burning is not a major source of diacids in our samples. The only exception was obtained for succinic acid in the outdoor air ($r^2 = 0.70$). Among diacids, the compound with highest concentrations was malic acid. It has been proposed as a primary pollutant in the exhaust of motor vehicles and a secondary product from photochemical degradation of aromatic hydrocarbons, such as toluene emitted by automobiles (Ho et al., 2011). However, the fact that this acid is found in many sour or tart-tasting foods can eventually justify its detection at such high levels in indoor particles. The most common use of malic acid is in candy and potato chips. Azelaic acid (C₉) was another diacid found at significant concentrations, especially indoors. It is formed upon oxidation of unsaturated fatty acids with a double bond at the ninth carbon (Winterhalter et al., 2009). In addition to anthropogenic sources (e.g., cooking), these unsaturated fatty acids may have an origin associated with the outer layer of the human skin. A typical adult sloughs 200,000–600,000 squames per minute, equivalent

Table 2. Concentrations (ng/m³) of di-, keto- and aromatic acids.

	Indoor	Outdoor
Glycolic	169 ± 98.8	58.9 ± 23.3
Cinnamic	20.9 ± 5.65	5.97 ± 2.19
Levulinic	13.2 ± 6.73	4.89 ± 1.91
Hydroxybutyric	7.29 ± 3.88	3.33 ± 2.48
Malonic	6.53 ± 3.03	2.88 ± 1.48
Methylmalonic	3.06 ± 1.49	0.612 ± 0.360
Benzoic	11.3 ± 5.66	3.57 ± 1.23
Succinic	13.1 ± 3.78	5.49 ± 4.23
Glutaric	6.51 ± 0.246	1.05 ± 0.428
Malic	1919 ± 734	91.0 ± 52.2
Adipic	6.03 ± 2.59	1.54 ± 0.489
Pinonic	3.02 ± 2.04	4.56 ± 0.218
Pinic	151 ± 92.2	11.7 ± 4.05
Pimelic	8.97 ± 1.63	1.43 ± 0.358
Suberic	4.13 ± 1.67	1.51 ± 0.747
Phthalic	0.714 ± 0.498	0.237 ± 0.137
Azelaic	20.7 ± 6.96	7.47 ± 3.73
Sebacic	1.14 ± 0.752	0.278 ± 0.081

to 30–90 mg per hour (Weschler, 2012). These compounds are readily oxidised, contributing to SOA products in occupied rooms. Other diacids identified in PM₁₀ samples were adipic (C₆) and phthalic (Pht) acids. Adipic acid has been proposed as mainly originating from anthropogenic emissions, namely through oxidation of cyclohexene (Mkoma and Kawamura, 2013). Phthalic acid (an aromatic diacid) is formed by the oxidation of PAHs, such as naphthalene, anthracene, and BaP. Another formation pathway could be the hydrolysis of phthalate esters, which are widely used as plasticizers (Winterhalter et al., 2009). The C₆/C₉ and Pht/C₉ ratios have been used previously to estimate the source strength of biogenic versus anthropogenic emissions, since adipic and phthalic acids have been proposed as mainly originating from anthropogenic sources and azelaic acid as mainly from biogenic inputs (Winterhalter et al., 2009). The higher are the ratios, the stronger are the influences of anthropogenic emissions (Fu et al., 2013). In this study, average C₆/C₉ ratios of 0.29 (indoor) and 0.22 (outdoor) were obtained, suggesting the dominance of biogenic contributions. The same average Pht/C₉ ratio (0.036) was determined for both the indoor and outdoor samples, likewise denoting the input of biogenic emissions as an important contributor to diacids. Much higher C₆/C₉ and Pht/C₉ ratios (up to 8) have been reported for polluted urban environments (Winterhalter et al., 2009; Zhang et al., 2010a).

Benzoic acid was found in both indoor and outdoor samples. It is a secondary product from photochemical degradation of aromatic hydrocarbons emitted by automobiles and has also been measured as primary pollutant in the exhaust of motor vehicles (Ho et al., 2011). Glycolic acid, an isomer of glyoxal monohydrate, formed from acetic acid and OH radicals (Tan et al., 2012), was also detected at higher concentrations indoors than outdoors. Since acetic acid is an intermediate product from isoprene oxidation, glycolic acid may be pointed out as a SOA product of this

typical biogenic volatile organic compound. Nevertheless, Björkqvist *et al.* (1997) proved that, in indoor environments, isoprene originates predominantly from expired air, in line with experiments carried out in the sixties. Exhalation results in significant concentrations of isoprene and its products in indoor environments, such as classrooms, whereas outdoor concentrations are negligible due to rapid dilution.

Two ketoacids from the oxidation of pinenes (Alves, 2008) were detected: pinonic and pinic acid. However, while pinic acid presented an indoor enrichment in relation to outdoor, the opposite was observed for pinonic acid. Among other reasons, competitive reaction, formation or degradation pathways, a wide range of concentrations of oxidants, and different volatilities and temperature dependences may explain this behaviour. It has been observed, for instance, that pinic acid exhibits pronounced temperature dependence, whereas the temperature dependence of pinonic acid is very weak (Zhang *et al.*, 2010b). On the other hand, besides the known outdoor biogenic sources, consumer products, such as cleaners, can contain hundreds of compounds, and reactive terpenes in many formulations. In indoor environments these terpenes or other volatile constituents can oxidise in the gas phase or on surfaces to form oxygenated organic species. Even though the chemistry of terpenes has been investigated for decades, insight into the dynamic nature of terpene oxidation continues to emerge and is needed by both the indoor and outdoor research communities (Wells, 2012).

The presence of biomass combustion tracers (e.g., levoglucosan, L, mannosan, M, and galactosan, G) in indoor particles suggest that infiltration of outdoor particulates

leads to contamination of classrooms with biomass burning smoke likely coming from nearby restaurants and bakeries with charcoal and firewood use. Taking into account that the sampling campaign took place, in large part, during spring, the contribution from domestic woodstoves and fireplaces should be minor. However, it is noteworthy that levoglucosan was detected in emissions from Chinese cooking (He *et al.*, 2004a, b; Hou *et al.*, 2008). Probably, the high temperatures reached during cooking processes would lead to thermal degradation of vegetable cellulose and, consequently, to the emission of levoglucosan. Spices such as *Cuminum cyminum* and *Capsicum* (powder of dry vegetables) used as condiments can also decompose to form levoglucosan (Hou *et al.*, 2008). Thus, in addition to woodstoves and/or open-hearth cooking in restaurants, food preparation in these commercial spaces may also generate anhydrosugar emissions. The L/M and L/(M + G) ratios are within the ranges reported for softwood burning (Fabbri *et al.*, 2009). The influence of biomass burning is also evidenced by the presence of diterpenoids (abietic acid and isopimaric acid), triterpenoids (lupeol and ursolic acid), phytosterols (stigmasterol and sitosterol) and phenolic compounds (syringic acid, vanillic acid, sinapic acid, coniferyl alcohol and sinapyl alcohol) (Fig. 6). All these compounds have been pointed out as unequivocal biomarkers from biomass burning. The biomarkers are source specific and may be used as confirming tracers for transport and fate studies of smoke emissions from biomass fuel sources (Simoneit, 2002).

Monosaccharides, such as erythrose (C₄), xylose (C₅), lyxose (C₅), arabinose (C₅), ribose (C₅), fructose (C₆), galactose (C₆) and mannose (C₆), and disaccharides, such

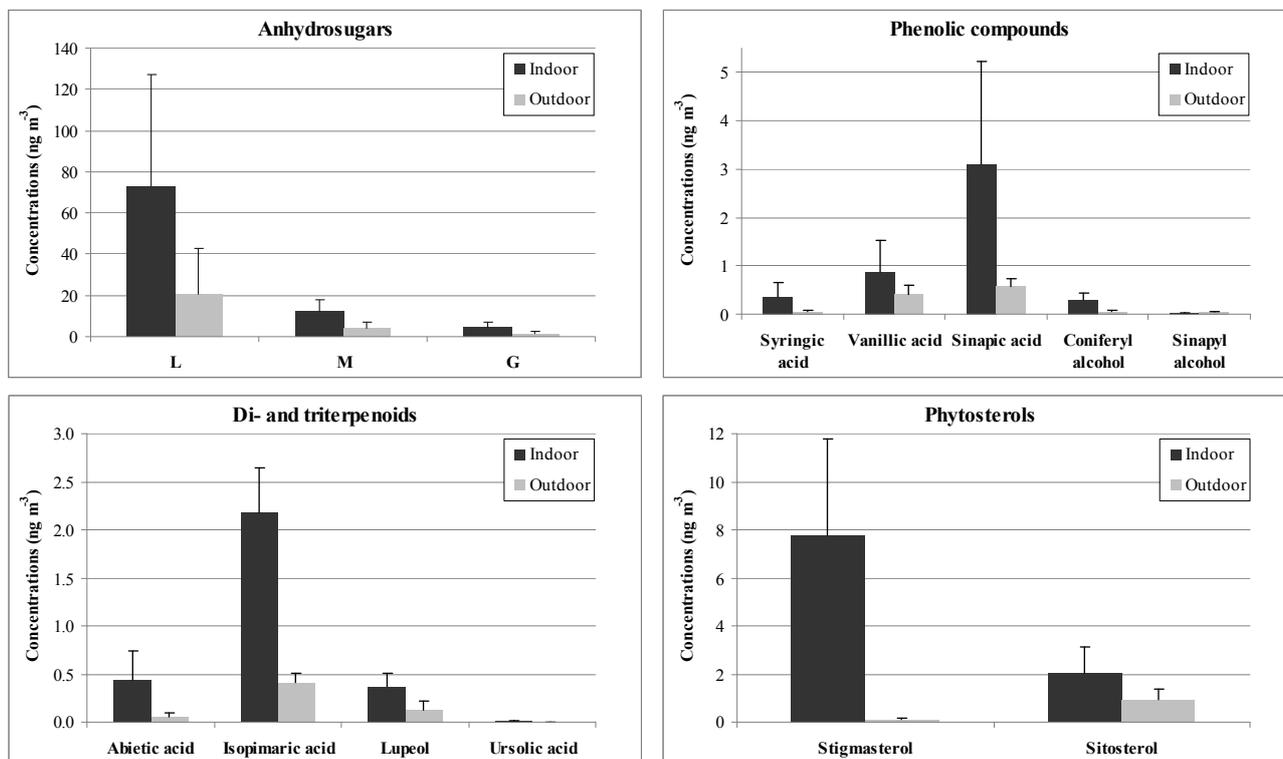


Fig. 6. Concentrations of biomass burning tracers.

Table 3. Concentrations (ng/m³) of sugars and polyols.

	Indoor	Outdoor
<i>Monosaccharides</i>		
erythrose	124 ± 85.0	38.9 ± 14.9
lyxose	36.6 ± 31.1	4.30 ± 2.85
xylose	14.1 ± 8.94	2.95 ± 1.86
arabinose	33.7 ± 15.2	10.4 ± 6.87
ribose	65.3 ± 36.5	11.9 ± 5.49
fructose	215 ± 176	43.3 ± 24.6
galactose	53.5 ± 27.7	10.3 ± 8.65
mannose	21.5 ± 10.1	5.07 ± 3.91
<i>Disaccharides</i>		
maltose	133 ± 77.6	34.7 ± 15.4
sucrose	16.4 ± 8.29	40.2 ± 23.6
<i>Polyols</i>		
glycerol	34.2 ± 15.3	10.0 ± 5.56
ribitol	3.04 ± 2.40	0.736 ± 0.617
xylitol	11.7 ± 8.47	3.17 ± 1.74
arabitol	17.5 ± 14.1	2.45 ± 1.99
mannitol	3.48 ± 2.72	0.820 ± 0.551
sorbitol	17.1 ± 8.04	3.19 ± 1.49

as maltose (C₁₂) and sucrose (C₁₂) were also present in PM₁₀ (Table 3). Saccharidic polyols included ribitol (C₅), xylitol (C₅), arabitol (C₅), mannitol (C₆) and sorbitol (C₆). Primary saccharides can enter the atmospheric compartment through numerous sources or processes, including burning of biomass, resuspension of soil particles, sea spray, and primary biological aerosols such as fungal spores, pollen, vegetative debris, bacteria and viruses (Calvo *et al.*, 2013). For example, it has been proposed that arabitol and mannitol are tracers for airborne fungal spores (Bauer *et al.*, 2008a). In this study, mannitol and arabitol were reasonably correlated (r^2 indoor = 0.87; r^2 outdoor = 0.55), indicating that the two sugar alcohols derive from the same source. The fungal contributions were roughly estimated using the conversion factor and ratio proposed by Bauer *et al.* (2008a): 1.7 pg mannitol/spore and 13 pg mannitol OC/pore. The number of fungal spores in PM₁₀ collected in the classroom was estimated to be 2621 ± 1414 spores/m³, whilst lower values (527 ± 316 spores/m³) were obtained for the playground. The relative contribution of fungal spores to OC was found to be always lower than 1%. Higher contributions of fungal spores to particulate matter were obtained in a tropical forest (Zhang *et al.*, 2010a) and at suburban and urban sites in Vienna (Bauer *et al.*, 2008b), using the same methodology. In contrast, Cheng *et al.* (2009) used ergosterol as a biomarker for fungal spores, providing substantially lower (0.2%) mean fungal contributions to OC in PM₁₀ in Hong Kong.

CONCLUSIONS

The results found within the elementary school are consistent with other studies where indoor PM₁₀ concentrations were greater than ambient levels measured simultaneously. It was observed that indoor PM₁₀ was significantly influenced by indoor OC sources, while indoor EC was predominantly of outdoor origin. Infiltration of

outdoor particulates and subsequent sorption on materials and surfaces may lead to contamination of a classroom with anthropogenic emissions (e.g., alkanes and PAHs from vehicle exhausts, and anhydrosugars from biomass burning). A multitude of pollutants with strong indoor sources (e.g., unsaturated fatty acids from human skin, terpenic compounds from cleaning products) joins those originating from infiltration, leading to much higher indoor concentrations compared to outdoors. In this study, indoor enrichments higher than 60% were estimated for most compounds or organic classes. The results also suggest that SOA formation and yields may be significant. Yet, the processes and species leading to SOA formation and growth are not fully understood, which precludes reliable quantitative predictions of their impacts and the correct development of control strategies to reduce indoor-generated pollution.

Taking into account the unique characteristics of schools (insufficient ventilation, relatively small room sizes, large number of children who are more susceptible to air pollutants, different and intense indoor activities, etc.), there is a need for better health exposure assessments. Although the carcinogenic risk might be low, as happened in this study, other health hazardous compounds besides PAHs whose toxicity or actuation mechanisms are not completely known, should also be investigated.

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