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# Indoor and outdoor air quality: a university cafeteria as a case study

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**Abstract:** A short but exhaustive air sampling campaign was conducted in a university cafeteria, an occupational environmental not yet studied. Carbonyls and volatile organic compounds were collected by passive diffusion samplers. Temperature, relative humidity, CO<sub>2</sub>, CO and particulate matter were continuously monitored indoors and outdoors. Simultaneous PM<sub>10</sub> sampling with high and low volume instruments, equipped with quartz and Teflon filters, respectively, was performed during working hours and at night. The quartz filters were analysed for their carbonaceous content by a thermo-optical technique and organic constituents by GC-MS. Water-soluble ions and elements were analysed in the Teflon filters by ion chromatography and PIXE, respectively. Low air change rates  $(0.31\text{-}1.5~\text{h}^{\text{-}1})$  and infiltration factors of 0.14, for both  $PM_{2.5}$  and  $PM_{10}$ , indicate poor ventilation conditions. Concentrations of both gaseous pollutants and particulate matter were much higher in the cafeteria than outdoors, showing strong variations throughout the day depending on occupancy and activities. The average concentration of indoor-generated PM<sub>10</sub> was estimated to be 32 µg m<sup>-3</sup>. Organic compounds in PM<sub>10</sub> included alkanes, PAHs, saccharides, phenolics, alcohols, acids, alkyl esters, triterpenoids, sterols, among others. The complex particle composition reveals the multiplicity of sources, formation reactions and removal processes, not yet fully known, and suggests the contribution from dust resuspension, abrasion and off-gassing of building materials, cooking emissions, tobacco smoke, and several consumer products. Many compounds are in the list of ingredients of personal care products, pesticides, plasticisers, flame retardants and psychoactive drugs.

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**Keywords:** Cafeteria, Indoor air quality, PM<sub>10</sub>, Metals, Organic compounds, Sources

The inhalation cancer risks of metals and PAHs were found to be negligible.

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#### 1. Introduction

In recent decades, Indoor Air Quality (IAQ) has been the focus of attention by the scientific community, governments and international organisations (Abdul-Wahab et al., 2015). It is a known fact that we spend more than 90% of our time indoors and that the levels observed are often higher than those recorded outdoors. Thus, the health risks from exposure to indoor air pollution may be larger than those associated with outdoor pollution. Poor IAQ can be especially detrimental to vulnerable groups such as children, young adults, the elderly, or those suffering from chronic respiratory and/or cardiovascular diseases (Cincinelli and Martellini, 2017). Furthermore, IAQ in the workplace is important because employees are occupationally exposed on a regular basis to a variety of pollutants with potential impact on comfort, work-related health problems, sickness absence, and productivity (Al horr et al., 2016; Carrer and Wolkoff 2018; Śmiełowska et al., 2017). IAQ in elderly care centres (e.g. Almeida et al., 2016; Bentayeb et al., 2015; Buczyńska et al., 2014; Mendes et al., 2015; Mui et al., 2008; Simoni et al., 2003), office buildings (e.g. Mandin et al., 2017; Śmiełowska et al., 2017; and references therein) and schools (e.g. Alves et al., 2013; Barmparesos et al., 2018; Bennet et al., 2019; Blondeau et al., 2005; Chatzidiakou et al., 2015; Fuoco et al., 2015; Kalimeri et al., 2016; Madureira et al., 2016; Yang et al., 2015) has been extensively investigated. Some studies were also conducted to evaluate the IAQ in restaurants with different cooking styles (Dai et al., 2018; Lee et al., 2001; Lee et al., 2011; Elsharkawy and Javed, 2018; Mugica et al., 2001; Sofuoglu et al., 2015; Taner et al., 2013; Wilson et al., 2011).

Restaurants not only have many of the same threats generally observed in office buildings, but they also encompass several other pollutants given off by cookware, open flames, cooking appliances, refrigeration units, strong chemical degreasers and aggressive cleaning solvents. Most of the published studies report the monitoring of comfort parameters, ventilation and/or some regulated pollutants. Gaseous compounds (CO, CO<sub>2</sub>, NOx and some volatile organic compounds), particles less than 2.5 and 10 µm in diameter (PM<sub>2.5</sub> and PM<sub>10</sub>, respectively), as well as some organic compounds in the particulate phase, normally from the group of polycyclic aromatic compounds (PAHs) with proven carcinogenic properties, are the most commonly monitored pollutants (Abdullahi et al., 2013; Marć et al., 2018; and references therein).

University cafeterias, a specific type of eating establishment, are popular meeting places for students and staff, providing space for leisure, relaxation and a room to have a quick lunch or snack. For this category of restaurant, as far as we know, no exhaustive evaluation has been made. Using optical counters, Zhao et al. (2010) measured PM concentrations in three different cafeterias located at Tsinghua University, Beijing, China. However, no information is given on the chemical composition of the particles and on the concentration of other pollutants. Hsu and Huang (2013) conducted a study in the student cafeteria at Chia-Nan University in southern Taiwan. Measurements of relative humidity, temperature, airflow velocity,  $CO_2$  and total suspended particles were made, although the

focus of the work was not centered on IAQ but rather on the efficiency of two disinfection treatments in removal of microorganisms.

This paper describes and discusses a short case study on a comprehensive air quality monitoring programme carried out in a university cafeteria. Besides the measurement of traditional parameters, such as regulated gaseous pollutants, the work had as main objective the detailed characterisation of PM<sub>10</sub>, including organic and inorganic components, in order to understand which sources and processes contribute to the measured levels and to estimate the carcinogenic and non-carcinogenic risks associated with inhalation of elements and PAHs by employees and customers. This preliminary study can serve as a basis for improving future monitoring campaigns, especially as regards the definition of source-marking chemical species, and for defining mitigation strategies to reduce exposure to pollutants in similar facilities.

# 2. Methodology

# 2.1. Sampling

A one-week extensive sampling campaign was carried out from 27<sup>th</sup> November to 1<sup>st</sup> December 2017 at the León (Spain) university cafeteria. The commercial establishment is continuously open for 12 hours (7:00-19:00, UTC) from Monday to Friday. The cafeteria is one large open room (410 m²) with fourteen tables, each capable of seating four to eight people, three standing round tables and stools along the 21 m long bar (Fig. 1). A separate kitchen provides the space to prepare the daily special menus and several dishes of fast food, such as burgers, sandwiches and French fries. The kitchen is equipped with a diesel fired range cooker, electric grills and toasters. The extractor hood of the kitchen has 4.8 × 2.4 m. There is no information regarding the brand, model or power. The cafeteria has two entrance doors, one opening to an adjacent alley and the other opening to an outdoor patio. A heat pump works as heater during the colder months and as air conditioner during the warmer months. Attendance indoors was highly variable, depending on the time of the day. Officially, the maximum occupancy allowed is 140 persons. People occupancy was logged manually throughout the campaign. In general, in the early morning, there was a simultaneous presence of 10 to 20 people, reaching a peak at lunch time (60 to 100 people). During the afternoon, the frequency of attendance varied from 10 to 40 clients, with a maximum around 16:00.

Temperature, relative humidity,  $CO_2$  and CO were continuously monitored with air quality probes inside (TSI, model 7545) and outside (Gray Wolf<sup>®</sup>, WolfSense IQ-610) the cafeteria. Real-time laser photometric instruments (TSI, DustTrak DRX 8533) were used to record particulate matter (PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub>) concentration profiles over time in the indoor and outdoor environments, simultaneously. All these continuous monitoring instruments were operated with 1-min resolution and were factory-calibrated before use. Simultaneous sampling with two PM<sub>10</sub> high volume air MCV

(model CAV-A/mb) instruments for gravimetric quantification was carried out on pre-weighed 150 mm quartz fibre filters (Pallflex®). The equipment was operated at a flow of 30 m³ h⁻¹. Samples were collected during the opening hours, simultaneously indoors and outdoors. Night time samples were also taken in order to compare the PM levels during occupancy and non-occupancy periods. Furthermore, PM₁0 samples were collected on Teflon membrane filters (47 mm diameter, Pall Corporation) with a TCR TECORA (model 2.004.01) operated at a flow of 2.3 m³ h⁻¹. The gravimetric quantification was performed with an analytical balance (Mettler Toledo, XPE105 DeltaRange®). Due to the possibility of vandalism and unfavourable weather conditions, with freezing temperatures, the outdoor equipment was shut down and sheltered at night. Thus, outdoor measurements correspond to the occupancy period in the cafeteria. All the data were registered in UTC.

Passive sampling of formaldehyde and acetaldehyde was done using Radiello<sup>TM</sup> diffusion tubes filled with 2,4-dinitrophenylhydrazine (2,4-DNPH) coated FLORISIL<sup>®</sup> over one week of continuous exposure, simultaneously indoors and outdoors. Aldehydes react with 2,4-DNPH to give the corresponding 2,4-dinitrophenylhydrazones. Volatile organic compounds (VOCs) were also collected with passive samplers from Radiello<sup>TM</sup>. To verify if there were large variations in the carbonyl and VOC levels, passive sampling was repeated two more times in the weeks following the campaign described above.

## 2.2. Chemical analyses

After gravimetric determinations, thermal-optical analysis of  $PM_{10}$  quartz filters was performed to obtain the carbonaceous content (organic and elemental carbon, OC and EC). This method is based on the  $CO_2$  quantification, by a non-dispersive infrared (NDIR) analyser, which is released from the volatilisation and oxidation of different carbon fractions under controlled heating. A laser beam and a photodetector that measure the filter light transmittance allow separating the EC formed by OC pyrolysis from the one that was initially in the filter (Alves et al., 2011).

Two circular punches of 47 mm from the quartz filters were sequentially extracted with dichloromethane and methanol. The resulting extract was separated into five different organic fractions by flash chromatography with silica gel and various solvents of increasing polarity. To meet the detection limits required by the speciated organic compounds, the night sampling filters were combined and extracted together. Thus, an average composition was obtained for non-occupancy periods at night. After each elution, the different fractions were concentrated in a TurboVap system from Biotage and evaporated to dryness by a nitrogen stream. Before chromatographic analysis, the fractionated extracts containing polar compounds were derivatised to trimethylsilyl ethers. All the extracts were analysed by gas chromatography-mass spectrometry (GC-MS). A detailed description of

the whole procedure for the speciation of organic compounds was previously reported by Alves et al. (2011).

Half of each Teflon filter was extracted with ultrapure Milli-Q water for ion chromatography analysis. A detailed description of the method can be found elsewhere (Vicente et al., 2018). All the elements with Z > 10 were determined in the other half of the filters by proton-induced X-ray emission (PIXE) (Lucarelli et al., 2014).

The carbonyl-DNPH derivatives were extracted from the Radiello tubes with 2 mL of acetonitrile for 30 min, filtered through a micropore filter membranes with porosity of 0.45  $\mu$ m, and analysed by high performance liquid chromatography. HPLC measurements were carried out with a Jasco PU-980 pump, a Rheodyne 7125 six-port valve with a 20  $\mu$ L loop and a Jasco MD-1510 diode array detector. An isocratic mixture of acetonitrile and water (60:40), at a flow rate of 1.5 mL min<sup>-1</sup>, was used to separate the 2,4-DNPH on a Supelcosil<sup>TM</sup> LC-18 column (5  $\mu$ m particle size; L  $\times$  I.D.: 250  $\times$  4.6 mm). The carbonyl concentrations were quantified with external calibrations curves constructed from standard solutions of carbonyl-DNPH derivatives in acetonitrile. The UV wavelength for detection was 360 nm. VOCs in activated charcoal cartridges were chemically desorbed with 2 mL of carbon disulphide (CS<sub>2</sub>) and 100  $\mu$ L of internal standard (2-fluorotoluene) for 30 min and analysed by gas chromatography coupled to flame ionisation detector (GC-FID), following the conditions described in Pegas et al. (2010).

## 2.3. Air change rates

In the present study, the air change rates per hour (ACR,  $h^{-1}$ ) were estimated by the  $CO_2$  concentration decay method (Alves et al., 2013). For a well-mixed space, the change in  $CO_2$  concentration with time is given by:

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$$C_{\tau} = C_{ext} + \frac{q_{CO_2} \times 10^6}{Q} - \left( C_{ext} - C_0 + \frac{q_{CO_2} \times 10^6}{Q} \right) e^{\left( -Q_{t/v} \right)}$$
 (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 (m³ s⁻¹),  $q_{CO_2}$  is the volumetric indoor emission rate of  $CO_2$  (m³ s⁻¹), V is the volume of the room (m³) and t is the interval since t = 0 (s). When the room 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 estimated from measured concentration values time t apart:

$$183 Q = -\frac{V}{t} \times \ln \left( \frac{C_t - C_{ext}}{C_0 - C_{ext}} \right)$$

184 (2)

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186 The air change rate is defined as:

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$$188 \quad ACR \qquad = \qquad Q/V$$

189 (3)

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## 3. Results and discussion

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#### 3.1. Comfort parameters and gaseous pollutants

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The indoor thermal environment is not only related to energy use in buildings, but is also linked to human satisfaction and wellbeing, health outcomes, and productivity. While temperature is the most influential factor in generating a phenomenological sense of thermal comfort, relative humidity (RH) contributes to a combined effect on the occupants' perception. Relative humidity levels below 25% are associated with increased discomfort and drying of the mucous membranes and skin, which can lead to chapping and irritation. Low relative humidity also increases static electricity, which can hinder the operation of computers and other equipment. High humidity levels can result in condensation within the building structure, providing the conditions for dust mite, mould and bacteria populations to grow. These, in turn, can cause respiratory problems and/or allergic reactions. Increasing RH is also associated with enhanced off-gassing of VOCs from building materials (Huang et al., 2016). The American Society of Heating, Refrigerating, and Air-Conditioning Engineers (ASHRAE Standard 55–2004) proposed indoor temperatures from 20 to 23 °C in the autumn/winter season, while the recommended RH values are in the 30-60% range for optimum comfort. The interdaily variations (mean±SD) of temperature during the occupancy periods were weak, ranging from 19.2±1.7 to 20.0±1.0 °C, while RH fluctuated between 31.2±2.8 and 36.1±5.3% (Fig. 2). Outside, the daytime temperatures and RH oscillated between -5.8 and 6.5 °C and from 46.2 to 90.9%, respectively.

 $CO_2$  at the concentrations usually observed in buildings is not a direct health risk, but its levels can be used as an indicator of occupant odours (bioeffluents).  $CO_2$ , primarily generated through human metabolism, provides a good indication of air change rates. Its build up in indoor environments is attributed to inefficient ventilation. ASHRAE (Standard 62.1-2016) reports that while not necessarily hazardous, the amount of  $CO_2$  in an indoor environment should remain under 1,000 ppm. The mean daytime carbon dioxide concentration indoors was  $891\pm226$  ppm, decaying to values around 400 ppm at night. A twofold difference in indoor concentration was observed between occupancy and non-

occupancy periods. Concentrations recorded throughout the weekend were similar to those observed in nocturnal periods. The inter-day fluctuations depended on the occupancy rates, reaching a maximum concentration of 3430 ppm in the early evening of Thursday (Fig. 2), the day the cafeteria closed later due to a party attended by about 150 people.

ACR ranged between 0.31 and 1.5 h<sup>-1</sup>, indicating poor ventilation conditions. For all spaces in general, a minimum ACR of 4 h<sup>-1</sup> is recommended, while specific values of 8-12 h<sup>-1</sup> and 12-15 h<sup>-1</sup> are suggested for restaurants and cafeterias, respectively (The Engineering ToolBox, 2005). Natural ventilation can be challenging during the wintertime in cooler climates. While open windows and doors can bring fresh air inside, this action obviously allows heat to escape. The installation of heating, ventilation, and air conditioning (HVAC) technologies may be the only solution to assure ventilation requirements for acceptable IAQ and thermal comfort.

Carbon monoxide levels were very low indoors. The maximum concentration ever recorded was 1.1 ppm. All the doors and windows were kept closed, due to the cold weather outside, preventing CO infiltration into the cafeteria from the nearby low traffic street and doorstep smokers. Low concentrations, always < 3 ppm, were also observed outdoors. ASHRAE (Standard 62.1-2016) lists a maximum allowable short-term limit of 9 ppm for CO in indoor air, while the WHO (2010) recommends CO concentrations based on exposure time ranging from 90 ppm (15-minute exposure) to 10 ppm (8-hour exposure). The American Conference of Governmental Industrial Hygienists (ACGIH) has assigned CO a threshold limit value (TLV) of 25 ppm as a time weighted average (TWA) for a normal 8-hour workday and a 40-hour workweek. The ACGIH limit is based on the risk of elevated carboxyhemoglobin levels. The National Institute for Occupational Safety and Health (NIOSH) has established a recommended exposure limit of 35 ppm as an 8-hour TWA and 200 ppm as a ceiling, based on the risk of cardiovascular effects.

Formaldehyde and acetaldehyde, two ubiquitous compounds emitted by combustion processes, meat cooking operations and consumer products, presented indoor mean concentrations of 8.8 and 17.8  $\mu$ g m<sup>-3</sup>, respectively, while the corresponding outdoor values were 1.3  $\mu$ g m<sup>-3</sup> and of the same order of the blanks. The inter-week variations were always <10%. Formaldehyde was below the air quality guideline of 100  $\mu$ g m<sup>-3</sup> (as a 30-min average value) recommended by the World Health Organisation (WHO) in 2010. However, short-time exceedance of this guideline level might have occurred in the indoor environment. Acetaldehyde was included by the WHO in Group 2, which encompasses pollutants of potential interest, but further investigation would be needed before it is clear whether there is enough evidence to warrant their inclusion in the guidelines. However, based on studies of short- and long-term exposure, countries such as Canada have set a maximum daily limit of 280  $\mu$ g m<sup>-3</sup>.

The indoor levels of VOCs (Table 1) were higher than those observed outdoors. Among these, ambient concentrations of benzene are regulated by law due to its toxicity (European Commission, 2009). In contrast, indoor levels of VOCs are not subject to any legislation. Of all the VOCs found in

buildings, BTEX (benzene, toluene, ethyl benzene, o-, m-, and p-xylene) are among the most common. Indoors, these compounds can originate from multiple sources, including building materials (paints, varnishes), consumer products (cleaning agents, fingernail polish, lacquers, adhesives), and human activities (smoking and combustion). In outdoor air, traffic is pointed out as a major source. The outdoor concentrations of benzene were far below the annual ambient air quality standard of 5 µg m<sup>-3</sup> set by the European Commission. Previous studies have reported the indoor BTEX levels at homes (Dai et al., 2017; Esplugues et al., 2010; Fan et al., 2018; Liu et al., 2013), libraries (Cincinelli et al., 2016), schools (Demirel et al., 2014; Pegas et al., 2011; Raysoni et al., 2017), printing shops and photocopy centres (El-Hashemy and Ali, 2018), and a confined parking area (Castro et al., 2015). Highly variable concentrations, up to hundreds of µg m<sup>-3</sup>, have been obtained, depending on the source strengths. Benzene levels of 18.4, 10.0, 9.3 and 3.7 µg m<sup>-3</sup> were measured in the dining areas of a Korean barbecue style restaurant, a Chinese hot pot restaurant, a Chinese dim sum restaurant and a Western canteen, respectively (Lee et al., 2001). The corresponding values reported for toluene were 156.1, 93.8, 81.6 and 17.6 μg m<sup>-3</sup>. In the present study, *n*-butanol and toluene were the VOCs with the highest indoor air concentrations. Butanol (or butyl alcohol) is present in many foods and beverages and is also used in a wide range of consumer products, such as cleaning products or cosmetics. Toluene is commonly added to gasoline and can enter the indoor spaces from outside. Toluene is also often used as solvent and can be released from a variety of products found in homes, including paints and other finishes, adhesives, and some personal care products. It is also found in tobacco smoke. Although detected at lower concentrations compared to these two VOCs, n-hexane showed very high I/O ratios (up to 13), suggesting the presence of active sources in the cafeteria. This aliphatic compound is primarily used as a solvent and as a component of certain glues and adhesives, cleaning agents, biocidal products, ink and toners, textile dyes, perfumes, fragrances and cosmetics (Federal Institute for Occupational Safety and Health, 2017). It is also present in volatile fractions of various plant species, such as apples and orange juice (Toxno, 2018). Moreover, Ahearn et al. (1996) reported that a variety of fungi found in buildings are capable of releasing gases that include *n*-hexane.

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# 3.2. Particulate matter

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#### 285 3.2.1. Mass concentrations

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From the gravimetric measurements, average  $PM_{10}$  concentrations of  $35.6 \pm 5.6$ ,  $12.4 \pm 3.4$  and  $23.3 \pm 10.0$  and  $\mu g$  m<sup>-3</sup> were recorded indoors during the occupancy and non-occupancy hours, and in the outdoor air, respectively, not exceeding the 24-h guideline of 50  $\mu g$  m<sup>-3</sup> set by the WHO. However, the real-time data collected by the photometric monitors show strong variations throughout the day. The highest values were reached at morning snack and lunch time, when the influx of students and staff was typically higher. Throughout the week, the maximum  $PM_{2.5}$  and  $PM_{10}$  values

were in the ranges 44-92 and 62-136 μg m<sup>-3</sup>, but peaks as high as 2560 and 2710 μg m<sup>-3</sup>, respectively, were registered in late afternoon and early evening of the day of a student party, during which even some firecrackers were launched Fig. 3). Throughout the week, a peak was observed between 19:00 and 20:00, time at which cleaning activities of the premises took place. Fine particles represented an overwhelming proportion of PM<sub>10</sub> indoors: PM<sub>2.5</sub>/PM<sub>10</sub>=0.82-0.91 (avg=0.88) and PM<sub>1</sub>/PM<sub>10</sub>=0.81-0.91 (avg=0.87). The PM<sub>2.5</sub>/PM<sub>10</sub> and PM<sub>1</sub>/PM<sub>10</sub> ratios were even higher outdoors, denoting the dominance of fine aerosols: 0.92-0.97 (avg=0.95) and 0.91-0.97 (avg=0.94). Regardless of particle size, indoor concentrations were 1.5-2 times greater than those found in outside air. During the night, in the cafeteria, the concentrations drop approximately 3 times.

The infiltration factor, which represents the equilibrium fraction of the outdoor PM that penetrates indoors and remains suspended (Othman et al., 2019), was calculated as follows:

$$C_{in} = F_{inf} \times C_{out} + C_{ig} \tag{4}$$

where  $C_{in}$  and  $C_{out}$  are the indoor and outdoor  $PM_{2.5}$  or  $PM_{10}$  concentrations,  $F_{inf}$  is the infiltration factor, and  $C_{ig}$  represent the concentrations of indoor-generated particles. The slopes of the correlations between indoor and outdoor concentrations for the occupancy periods, i.e. the infiltration factors, were 0.14 for both  $PM_{2.5}$  and  $PM_{10}$ . As inferred from the  $CO_2$  measurements, the low particle infiltration factors denote air tightness. On average, concentrations of indoor-generated particles were  $27 \mu g PM_{2.5} m^{-3}$  and  $32 \mu g PM_{10} m^{-3}$ . To evaluate the influence of outdoor particulate pollutants on indoor environment, the concept of contribution rate ( $\rho$ ) was applied, as suggested by Lv et al. (2017):

 $\rho = (F_{inf} \times C_{out})/C_{in} \times 100 \%$  (5)

The contribution rates of outdoor sources were estimated to be, on average, 17.7 and 16.1 % for  $PM_{2.5}$  and  $PM_{10}$ , respectively, whereas a major input from indoor sources was observed (82.3 and 83.9 %).

# 3.2.2. Chemical composition

To obtain a  $PM_{10}$  mass balance (Fig. 4), the measured element concentrations were converted into the respective mass concentrations of the most common oxides (SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO, MgO, MnO, Fe<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, K<sub>2</sub>O, etc.). Because the GC-MS analyses indicated the presence of highly oxygenated compounds, to derive the organic matter content in  $PM_{10}$ , a total organic mass to organic carbon ratio (OM/OC) of 1.8 was adopted (Polidori et al., 2008). Total carbon (TC = OC + EC) accounted for 36.0  $\pm$  5.8, 42.8  $\pm$  7.9 and 27.6  $\pm$  12.6 %wt. of the  $PM_{10}$  mass indoors during the occupancy and non-occupancy hours, and in the outdoor air, respectively (Fig. 5). Besides sampling and analysis artefacts affecting the attainment of chemical mass balance, the unaccounted mass can partly be explained by

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the presence of unanalysed constituents. Furthermore, part of the unaccounted PM mass is usually assigned to particle-bound water (Tsyro, 2005). On average, I/O ratios of 1.5 and 0.7 were obtained for OC and EC, respectively, suggesting the presence of organic matter emitting sources in the cafeteria, while elemental carbon is predominantly generated from combustion processes in the outdoor environment. Mean OC/EC ratios of 7.5, 3.5 and 3.6 were observed for the occupancy and night-time periods in the cafeteria and for the outdoor air, respectively. A much higher ratio (21.0) was registered for the weekend daytime period. Lower EC infiltration rates over the weekend due to the fact that the building is closed, absence of indoor EC sources (e.g. toasting bread) and off-gassing of VOCs from surfaces leading to secondary organic aerosol formation, may justify the higher proportion of OC compared to EC. It should be noted that OC and EC concentrations decreased from 10.7 and 1.1  $\mu$ g m<sup>-3</sup> in the periods of occupation to 6.1 and 0.29  $\mu$ g m<sup>-3</sup> in the weekend, respectively.

On average, water-soluble ions accounted for PM<sub>10</sub> mass fractions of 6.0, 15.8, 13.4, and 1.3 % during the occupancy of the cafeteria, at night-time indoors, in the daytime period on the weekend inside the building, and at regular working hours outdoors, respectively. Expect for Ca<sup>2+</sup> and K<sup>+</sup>, the I/O ratios for all water-soluble ions were less than 1. The higher I/O ratios for  $Ca^{2+}$  and  $K^{+}$  may be related to resuspension of fine dust from human movement. Concentration values in daytime were higher than at night-time (Fig. 6). Together with chloride, the secondary ions sulphate, nitrate, and ammonium were the dominant species. In addition to sea salt and biomass burning, chlorinated water and cleaning products containing bleachers can release significant amounts of chloride. Ammonium may also originate from cleaning products. For unknown reasons, in the outdoor sample of Tuesday, concentrations increased approximately 4-fold compared to other days, contributing to a high standard deviation. A good correlation between concentrations of total anions and total cations in PM<sub>10</sub> was observed, but an anion deficit was registered. Non-measured organic ions, such as formate and acetate, could contribute to the ionic balance. However, the anion deficit is most likely due to the presence of non-determined carbonates (CO<sub>3</sub><sup>2-</sup>/HCO<sub>3</sub><sup>-</sup>) from resuspended local soil. Outdoors, the molar ratio of NH<sub>4</sub><sup>+</sup> to SO<sub>4</sub><sup>2-</sup> was found to be greater than 2. This higher molar ratio suggests that in addition to (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, NH<sub>4</sub>NO<sub>3</sub> was formed because of excess quantity of NH<sub>4</sub><sup>+</sup>. Ammonium nitrate is produced in the presence of high NH<sub>4</sub><sup>+</sup> and HNO<sub>3</sub> concentrations, low temperatures and high relative humidity. A mean NH<sub>4</sub>+/SO<sub>4</sub><sup>2</sup>- molar ratio of 1.6 was obtained indoors, indicating aerosols in the form of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>HSO<sub>4</sub>. Due to the higher temperatures indoors, NH<sub>4</sub>NO<sub>3</sub> may decompose to form NH<sub>3</sub> and HNO<sub>3</sub>, which are then partially lost by deposition and sorption processes to indoor surfaces, especially those with higher roughness, while another part reacts with NaCl and Ca-containing particles. NaNO<sub>3</sub> and Ca(NO<sub>3</sub>)<sub>2</sub> have low volatility compared to NH<sub>4</sub>NO<sub>3</sub>

Both indoors and outdoors, the elements with highest concentrations were Na, Mg, Al, Si, S, Cl, K, Ca and Fe (Table 2). Elements detected at I/O much higher than 1, suggesting indoor sources, were Mg, P, K, Ca, Cr, Ni and Zn. Some of these elements (e.g. Ca, Mg, K) are present in mineral matter and can be associated with soil dust that is brought in from outdoors on shoes. It has been shown that

resuspended dust contributes to the airborne particulate matter in the "personal cloud" (i.e. personal breathing zone). Resuspension rates of settled dust depend on factors such as amounts accumulated, floor characteristics, walking style, shoe types, humidity and cleaning activities (Rasmussen et al., 2018). Some elements may result from abrasion of metallic components from furniture and other building materials. Cr, for example, is an important alloying element in stainless steel and is also present as chromated copper arsenate (CCA), a preservative for furniture and wood building materials against insect, bacterial, and fungal decline (Rivas et al., 2015; and references therein). P is likely associated with chlorinated phosphorus-based flame retardants and plasticisers, which are ubiquitous in the indoor environment (Araki et al., 2014). Many metals (e.g. Ni and Zn) are also in the list of ingredients of numerous personal care products (Borowska and Brzóska, 2015; Omenka and Adeyi, 2016).

Enrichment factors of each element with respect to crustal material (Wedepohl, 1995) were calculated using Fe as reference element:

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$$EF = (X/Fe)_{air}/(X/Fe)_{crust}$$
 (3)

where EF is the enrichment factor of element X, (X/Fe)<sub>air</sub> is the concentration ratio of X to Fe in the  $PM_{10}$  samples, and  $(X/Fe)_{crust}$  is the average concentration ratio of X to Fe in the continental crust. Whether for samples of the cafeteria or for outdoor PM<sub>10</sub> filters, EFs lower than 5 were obtained for Na, Mg, Al, Si, K, Ca, Ti, V, Mn, Rb, Sr, Y and Zr, indicating a dominant soil dust origin. Although the sampling campaign was carried out in winter, when the residential biomass combustion is a common practice, the EFs for potassium do not reveal the dominance of wood or coal burning. However, it should be borne in mind that residential combustion equipment is relatively inefficient, reaching temperatures generally below 700-800 °C. It has been extensively reported that, before condensing to form particles, potassium is released into the gas phase at temperatures above 727 °C and that only the full combustion of biomass fuels at elevated temperatures (>1327 °C) is expected to result in most of the potassium entering the gas phase (Mason et al., 2016). Compared to the dustderived elements, the EFs for S, Cl, Cu, Zn, As, Br, Mo and Pb were larger than 10, for both indoor and outdoor samples, illustrating the influence of anthropogenic sources. Se was a particularly enriched element with EF values > 1100. In addition to As and S, coal combustion is also a strong emitter of Se. More than 20% of the total energy consumption in the region of León come from coal combustion.

The emissions from coal burning increase during coldest months due to the use of domestic heating and cooking devices (Blanco-Alegre., 2019). In outdoor environments, many of these elements, in particular Cu, Zn, Pb and Mo, are mostly associated with traffic emissions (Pant and Harrison, 2013). In dwellings and in working and leisure places, in addition to infiltration from outdoors, these constituents can originate from the multiplicity of materials and consumer products

used indoors. Attention is drawn to the fact that the concentration of some elements has increased sharply on the day of the student party. Potassium reached a level of 2140 ng m<sup>-3</sup>, while in other occupancy periods concentrations ranged from 305 to 367 ng m<sup>-3</sup>. Cl, Zn and Cu increased from usual levels in the ranges 214-396, 21-24 and 2.7-5.7 ng m<sup>-3</sup> to values of 1870, 61 and 15 ng m<sup>-3</sup>, respectively, on the party day. These elements are abundant constituents of tobacco smoke (Misha et al., 1986), reason why there are suspicions that, although prohibited, this practice may have occurred during the event.

The particulate matter organic extracts encompassed several aliphatics, polycyclic aromatic hydrocarbons (PAHs), alcohols, acids, sterols, glycerol derivatives, phenolic compounds, saccharides, among others. Aliphatic compounds included the homologous series of *n*-alkanes from C<sub>11</sub> to C<sub>32</sub> and some alkenes. These are ubiquitous compounds in the environment with both natural and anthropogenic origin. Alkane homologues were detected at I/O ratios ranging from 1.0 (dodecane) to 54 (dotriacontane). The sum of their concentrations was as follows: 49.4 ng m<sup>-3</sup> (indoor, occupancy), 15.9 ng m<sup>-3</sup> (indoor, night-time), 16.5 ng m<sup>-3</sup> (outdoor, daytime), and 6.0 ng m<sup>-3</sup> (indoor, weekend, daytime). The maximum indoor concentration (13.4 ng m<sup>-3</sup>) was observed for dotriacontane. Alkenes (C<sub>14</sub>, C<sub>16</sub>, C<sub>18</sub>, C<sub>20</sub> and C<sub>23</sub>) were present at I/O ratios from 3 to 5 and very low concentrations, peaking in the cafeteria for hexadecene (0.47 ng m<sup>-3</sup>). Excepting hexadecene, alkenes were not detected in samples from non-occupancy periods.

Figure 7 depicts the mean concentrations of individual PAHs in the cafeteria and outdoors. Excluding naphthalene and acenaphthene, two volatile PAHs consisting of two benzene rings, the I/O ratios for all other polyaromatics were always < 1. Contrary to many other compounds, the average outdoor concentrations (7.8 ng m<sup>-3</sup>) was 3.3 times higher than those measured indoors (2.4 ng m<sup>-3</sup>). Levels in the cafeteria dropped to 0.49 ng m<sup>-3</sup> on the weekend, whereas the night-time value (4.3 ng m<sup>-3</sup>) was between the one measured during working hours in the cafeteria and that registered outdoors. Although nocturnal measurements in the ambient air were not carried out in the present study, outdoor concentrations usually higher at night may have contributed to the build-up of PAHs in the cafeteria through infiltration processes. Due to residential heating emissions, decreased atmospheric mixing height and nocturnal atmospheric stability, an appreciable increase in concentrations in the evening is likely to occur. On the other hand, at daytime, after emission by combustion sources (e.g. traffic), PAHs undergo chemical transformations. Better dispersion conditions and atmospheric reactivity during daytime, in general, lead to lower diurnal concentrations compared to nocturnal values. PAH oxidative transformations in ambient air are supposed to be started by gas-phase OH and NO<sub>3</sub> radicals, followed by reactions with NO<sub>2</sub> and O<sub>3</sub> leading to ring-retaining quinones, nitro-PAHs and ring-opened phthalic acids and their degradation products (Alam et al., 2015).

The homologous series of fatty alcohols from  $C_{10}$  to  $C_{30}$  was observed in the  $PM_{10}$  samples (Table 3). 1-Hexacosanol and 1-octadecanol were the most abundant homologues. Their indoor concentrations were more than 600 times higher than those measured outdoors. These n-alkanols are

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constituents of many household and personal care products. Furthermore, octadecanol was identified as one of the dominant compounds in fine organic aerosols from charbroilers and meat cooking operations (Rogge et al., 1991). The homologous series of alkanoic acids (C<sub>6</sub>.C<sub>26</sub>) and some alkenoic acids were also present. In general, the concentrations recorded during the occupancy period in the cafeteria far exceeded those observed outside. Palmitic, stearic and oleic were the dominant acids, in agreement with previous studies describing cooking emissions (Abdullahi et al., 2013; He et al., 2004; Rogge et al., 1991; Zhao et al., 2007).

Some dicarboxylic acids from C<sub>4</sub> to C<sub>16</sub>, at I/O ratios generally higher than 1, were present in  $PM_{10}$ , but at lower concentrations compared to those of fatty acids. He et al. (2004), who also detected diacids in emissions from cooking processes at lower levels in comparison with alkanoic and alkenoic acids, suggested that these compounds are formed by oxidation of dialdehydes during the autoxidation process of unsaturated lipids. Among diacids, azelaic (C<sub>9</sub>) was the most abundant in the cafeteria, reaching concentrations up to 67 ng m<sup>-3</sup>. The lab-engineered form of this acid is typically used in skincare products. It is also an abundant compound in emissions from electronic cigarettes, heat-notburn tobacco products, and conventional cigarettes (Ruprecht et al., 2017). Biomass combustion is also a source of diacids, in addition to emitting keto- (e.g. levulinic) and hydroxy-acids (e.g. glycolic, glyceric and lactic) (Alves et al., 2011). All these acids were not detected in the outdoor samples or were present in lower concentrations when compared to those observed inside the cafeteria. Liu et al. (2017) measured a wide variety of carboxylic acids in a university classroom, indoors and outdoors, including monoacids, diacids, hydroxy acids, carbonyl acids, and aromatic acids. Besides indoor sources such as off-gassing of building materials, evidence for acid formation from indoor chemical reactions with ozone was noticed. It was also suggested that human occupants may contribute to the abundance of carboxylic acids either through direct emissions or surface reactions. Although generally present at very low concentrations indoors, ozone may react with unsaturated organic compounds present in skin and other materials. As observed in the university classroom, the elevated indoor concentrations of lactic acid in the cafeteria during the occupancy period was likely human perspiration. A recent study conducted in an art museum revealed the unexpected build-up of lactic acid from sweat (Pagonis et al., 2019). It was found that this acid leaves the skin, travels through the air, and sticks to the walls and other surfaces at high rates. It should be also noted that lactic and other short chain acids (levulinic, glycolic, etc.) are frequently used in exfoliants, anti-wrinkle products and other cosmetics. Thus, both the emission and partitioning mechanisms between the gas and the particulate phases of these and many other compounds are complex and still far from being fully understood.

Some resin acids were detected as  $PM_{10}$ -bound organic components: dehydroabietic, isopimaric, abietic and podocarpic. Among these, dehydroabietic acid dominated. Its concentrations outside the cafeteria exceeded those observed indoors, pointing to the residential burning of coniferous wood (Vicente and Alves, 2018). Although it is an unlikely source in the cafeteria, it should be noted that

very high concentrations of free resin acids were detected in depilatory wax strips (Nilsson et al., 2008).

Levoglucosan and its stereoisomers, mannosan and galactosan, were one of the dominant groups of organic compounds. Levoglucosan has been recurrently used as a biomass burning tracer, since it is a thermal degradation product of cellulose (Vicente and Alves, 2018). However, it was also detected in particulate matter samples emitted from Sichuan, Dongbei, Hunan and Cantonese cooking styles, suggesting that this anhydrosugar can also be generated during the heating process of vegetables (He et al., 2004; Zhao et al., 2007). Mean I/O ratios lower than 1 reveal the strong impact of residential biomass burning on the ambient air of the surroundings due to the low temperatures.

Many other monosaccharides, disaccharides, polysaccharides and polyols, were detected in PM<sub>10</sub>. However, due to structural isomerism and identical fragmentation patterns in the mass spectra, their individual quantification was not possible. Thus, a mean response factor of the calibrated compounds with the same functional groups was applied. A much higher indoor concentration for these unidentified saccharides and a strong nocturnal decay was observed, suggesting that many of these compounds were produced by the day-to-day activities of the cafeteria.

Many phenolic compounds and derivatives were detected in both indoor and outdoor samples. These constituents are used in commercial or consumer products and in building materials. Methoxy-and dimethoxyphenols, such as vanillic acid, sinapyl alcohol and syringic acid, are also found in smoke particles from biomass burning (Vicente and Alves, 2018). Among aromatic compounds, benzyl alcohol is worth mentioning because of the high concentrations. It is used as a solvent, and a preservative. It is also employed in inks, textiles and sheet plastics, and as a fragrance in perfumes and in flavouring.

Glycerol and many oxygenated derivatives were present at much higher concentrations in the cafeteria compared to the outside. During the cooking process, triglycerides (i.e. fatty acids esterified to a glycerol backbone) of fat in uncooked meat are hydrolysed or thermally oxidised, forming free glycerol, free fatty acids and mono- and diglycerides (Abdullahi et al., 2013).

Cholesterol and phytosterols were abundant compounds in  $PM_{10}$ . Concentrations in the cafeteria during the occupancy period far exceeded those recorded outdoors. Cholesterol ( $C_{27}$ ) is biosynthesised by higher animals and found in body tissues, especially in oils and fats. Phytosterols, such as  $\beta$ -sitosterol and stigmasterol ( $C_{29}$ ), are present in plant lipid waxes and membranes. All these sterols were previously observed in  $PM_{2.5}$  emitted from Chinese cooking (He et al., 2004; Zhao et al, 2007). However, cholesterol is the only sterol that have been detected in meat cooking fumes (Rogge et al., 1991).

In the present study, many unusual compounds were detected in indoor aerosols, as far as we know, for the first time. Some of the substances are part of the list of ingredients of various cosmetics. Ethylene brassylate, for example, which was only present in indoor samples, is a macrocyclic compound mainly used as a fragrance ingredient for its musk-like odour. Parsol MCX (ethylhexyl

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methoxycinnamate) is used in the formulation of a wide variety of brands, including sunscreens and makeup products that contain ingredients to protect the skin from the sun. A mean I/O ratio of 122 suggests its emission from personal care products and concentration build-up in confined spaces, such as the cafeteria. Dibutyl adipate is the diester of butyl alcohol and adipic acid. In cosmetics and personal care products, it is used in nail polish and skin care products. Once again, much higher concentrations indoors than outdoors (I/O=23) indicate a possible origin in the products used by the occupants. Because of its floral fragrance, methyl dihydrojasmonate is an additive in a wide range of cleaning and personal care products. It was absent from outdoor samples, but the indoor concentrations did not show appreciable differences between occupancy and non-occupancy periods, suggesting the presence of a permanent source. Acetyl tributyl citrate is an organic compound that is employed as a solvent in paints, inks, and nail enamel. It may also be found in eye makeup and aerosol hair sprays. Moreover, it is used to make plastics more flexible (e.g. plasticisers in packaging films for food), added as a flavour ingredient in non-alcoholic beverages and applied in the manufacture of many pharmaceutical drugs. Acetyl tributyl citrate was only detected in indoor aerosols. Pyroglutamic acid, known either as pyrrolidone-5-carboxylic acid, 5-oxoproline, PCA or pidolic acid, is a ubiquitous natural amino acid derivative. It is used for dry skin and hair products, owing to its humectant characteristics. Pidolic acid is found in large quantities in skin and brain tissues. It is sold online as a nootropic dietary supplement to improve cognitive and executive functions, and memory. The presence of this acid was only observed indoors, and at much higher concentrations during the occupancy periods.

2-Propanol-1-chloro-phosphate, also known as Amgard TMCP or Hostaflam OP 820, is a chlorine containing alkyl phosphate ester. It is mainly employed as an additive flame retardant, especially in polyurethane foam, with some minor use in other plastics and as a back coating in textiles. It was only present in indoor samples collected during daytime, including weekend. This suggests that the emission of this compound can be promoted by sunlight, although its formation processes and detection in the particulate phase deserve further investigation.

Another compound that was only detected indoors was piperonyl butoxide (PBO). It is a manmade organic compound used as a component of pesticide formulations, particularly insecticides, and classified as a synergist. By itself, PBO does not harm insects. Instead, it acts with insect killers to intensify their effectiveness. Boric acid, which was present in both indoor and outdoor samples, is also a pesticide that can be used to control a wide variety of pests, including insects, spiders, mites, algae, moulds, fungi, and weeds.

Some psychoactive substances, including drugs of abuse (e.g. tetrahydrocannabinol, THC), were found in the particulate matter samples. THC is the principal psychoactive constituent of cannabis. THC was only present in two indoor samples collected during the occupancy periods, reaching 21.7 ng m<sup>-3</sup> on Thursday, the day of the student party. THC was previously detected as a constituent of outdoor airborne particles in 6 Spanish urban locations at concentrations ranging from 23 to 44 pg m<sup>-3</sup>

(Viana et al., 2012). Another psychoactive substance in airborne particles was caffeine. However, this compound was only detected in outdoor samples and during non-occupancy periods. Since caffeine is expected to partition between the gas and the particulate matter (Viana et al., 2012), its absence during working hours may be related to the higher temperatures in the cafeteria because of the use of heating. In the outdoor air across Italy, caffeine reached gross average concentrations of 15±8 ng m<sup>-3</sup> in winter and 1.2±0.6 ng m<sup>-3</sup> in summer, while in the metropolitan area of Rome the corresponding levels were 1.5±0.5 ng m<sup>-3</sup> and 0.3±0.1 ng m<sup>-3</sup> (Viana et al., 2012; and references therein).

Nicotinic acid, also denominated niacin or vitamin B3, is present in all living cells and found in whole and processed foods. It is used to make other chemicals and added to feeds and flours as a dietary supplement. It is also used as a medication to treat dyslipidaemic states. Niacin was not detected in outdoor air but reached concentrations up to 16.1 ng m<sup>-3</sup> in the cafeteria during the occupancy period. The mechanisms by which it is formed and remains airborne in the particulate phase are yet to be clarified.

Irgafos 168 (tris(2,4 di tert butylphenyl)phosphite), as one of the common antioxidants, is extensively used in polyolefin materials (e.g. polypropylene employed in food packaging), to protect polymers from aging and oxidation. However, irgafos 168 may degrade into several products, such as 2,4 di tert butylphenol (DP1), and tris(2,4 di-tert butylphenyl)phosphate (DP2), which is the oxidised form of irgafos 168 (Yan et al., 2018). In the present study, whilst DP1 was detected at 5-fold higher concentrations in the cafeteria than outside, DP2 was only present in two daytime indoor samples (both with 19 ng m<sup>-3</sup>) and in just one outdoor sample (39 ng m<sup>-3</sup>).

3.2.3. Health risk analysis

Noncarcinogenic and carcinogenic risks associated with inhalation exposure to trace elements in indoor  $PM_{10}$  by the employees were estimated following the methodology proposed by the United States Environmental Protection Agency (USEPA, 1989, 2009):

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$$THQ = (EF \times ED \times ET \times C)/(RfC \times AT)$$
 (6)

$$TR = (EF \times ED \times ET \times C \times IUR)/AT$$
 (7)

The target hazard quotient (THQ) and target carcinogenic risk (TR) are dimensionless, EF is the occupational exposure frequency (250 days per year, i.e. 5 days per 50 weeks), ED is the exposure duration (25 years), ET is the exposure time (8 h per day), C is the metal concentration in the air (mg m<sup>-3</sup>), and AT is the averaging time (365 days per year × 24 h per day × ED for noncarcinogenic effects and 612,000 h, i.e., 70 years × 365 days per year × 24 h per day, for carcinogenic effects). RfC is the USEPA reference concentration (mg m<sup>-3</sup>). Considering that for some elements, reference doses

for oral exposure (RfD, mg kg<sup>-1</sup> day<sup>-1</sup>) are available instead of RfC values (USEPA, 2017, 2019), these latter were derived using the following equation (USEPA, 2013):

$$891 RfC = (RfD \times BW) / IR (8)$$

where IR and BW are the inhalation rate and body weight of an adult (20 m³ day $^{-1}$  and 70 kg). Chronic inhalation unit risk (IUR) values for the carcinogenic elements found in the PM $_{10}$  samples were taken from USEPA (2017): arsenic  $4.3\times10^{-3}$  (µg m $^{-3}$ ) $^{-1}$ , lead  $1.2\times10^{-5}$  (µg m $^{-3}$ ) $^{-1}$ , hexavalent chromium  $8.4\times10^{-2}$  (µg m $^{-3}$ ) $^{-1}$  and Ni oxide  $2.6\times10^{-5}$  (µg m $^{-3}$ ) $^{-1}$ . The IUR of Cr(VI) is based on a Cr(III):Cr(VI) proportion of 1/6. Since in the present study total Cr was determined, one seventh of

the concentration obtained was used to estimate the risk.

The noncarcinogenic risks associated with inhalation exposure by employees to particulate trace elements in the indoor air during the occupancy period are presented in Figure 8. A THQ < 1 indicates no significant (acceptable) risk, a THQ > 1 suggests that the noncarcinogenic effect is likely to manifest, whereas a high chronic risk is evident when THQ > 10 (Slezakova et al., 2014). The estimated mean THQ for PM<sub>10</sub>-bound trace elements ranged from null values for many elements 0.92 for Cl. The total mean THQ of all eleven elements in PM<sub>10</sub> was 1.7. Cl and Mn were the most important contributors (55 and 38 %, respectively) to the total noncarcinogenic risk, followed by Si (6.0 %), and Al (1.3 %). If the exposure time and frequency assumed for the working hours were considered for non-occupancy periods and for outdoor air,  $\Sigma$ THQ values of 0.17-0.26 and around 2.0 would be obtained, respectively.

For carcinogenic constituents, USEPA considers that targeting a  $10^{-6}$  risk level for individual chemicals and pathways will usually lead to negligible risks. However, caution is suggested to guarantee that the cumulative cancer risk for all potential carcinogenic contaminants does not have a residual cancer risk above  $10^{-4}$  (Slezakova et al., 2014). In the present study,  $\Sigma TR$  was always  $< 2 \times 10^{-6}$ , suggesting that the risk associated with the exposure to  $PM_{10}$ -bound carcinogenic elements is negligible.

To estimate the carcinogenic risks associated with PAH inhalation, the benzo[a]pyrene equivalent carcinogenicity ( $BaP_{eq}$ ) was evaluated by multiplying concentration of each PAH with their toxicity equivalent factor (TEF) (Bari et al., 2010, and references therein). The exposure concentration (EC) is calculated as follows (CalEPA, 2005):

$$EC = (CA \times ET \times EF \times ED) / AT$$
(9)

where CA represent the benzo[a]pyrene equivalent concentrations (ng m<sup>-3</sup>). The inhalation unit risk (IUR) of respiratory cancer for  $BaP_{eq}$  is  $3.9 \times 10^{-6}$  (ng kg day)<sup>-1</sup>. This value is obtained multiplying the

inhalation cancer risk unit factor of BaP  $(1.1 \times 10^{-6} \text{ (ng kg day)}^{-1})$  by the inhalation rate  $(20 \text{ m}^3 \text{ per day})$  and dividing by the body weight (70 kg). The carcinogenic risk of the PAH mixture is then calculated using the following equation:

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$$Risk = IUR \times EC$$
 (10)

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The mean estimated excess inhalation cancer risk associated with total  $BaP_{eq}$  concentrations in the cafeteria during the occupancy periods was  $3 \times 10^{-8}$ , which is considered negligible. Outdoor and non-occupancy PAH levels also pose insignificant risks. However, it should be noted that other routes of exposure (dermal and ingestion) have not been considered and that only particle-bound PAH were used in the estimates. After formation and emission, these compounds partition between the gas phase and atmospheric aerosols.

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## 4. Conclusions

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A short, but comprehensive, air quality monitoring programme was carried out in a university cafeteria, a type of occupational environment for which no inspections have been made so far. The mean values obtained for comfort parameters and regulated pollutants were, generally, within the ranges stipulated by international organisations. However, real time measurements of CO2 and particulate matter revealed strong fluctuations, depending on activities and occupancy rates. Very low air change rates and particle infiltration factors from outdoors indicate inadequate ventilation. For most pollutants, the indoor levels during the working hours largely exceeded those measured at night and outdoors, pointing to the presence of multiple indoor sources. It was observed that more than 80 % of the particle mass concentrations were generated indoors. Several metals, either crustal or anthropogenic, were detected in the particulate matter. About 200 organic compounds were also identified, some of which, to the best of our knowledge, are described as indoor aerosol constituents for the first time. Many organic compounds detected in indoor air are components of various cosmetics and other personal care products, but also of pesticides, drugs of abuse, plastics, building materials (e.g. flame retardants), cooking emissions, sweat, among others. Some compounds can also be secondarily formed as by-products of surface reactions. However, the formation pathways of many compounds and the partitioning mechanisms between the gas and the particulate phases of some semivolatiles are complex and still far from being well elucidated, deserving extensive investigation in the future.

The total mean hazard quotient that represents the noncarcinogenic effects due to the inhalation of particle-bound metals during the occupancy period in the cafeteria was higher than the acceptable level (>1). The cumulative cancer risk for both carcinogenic metals and PAHs was lower than the

acceptable level (10<sup>-4</sup>). However, exposure to metals and PAHs occurs also via ingestion and dermal contact and if these routes are considered, then the estimated risks might be higher.

To protect the restaurant workers and other public, possible strategies to control and minimise the emissions of indoor pollutants at their point sources within the cafeteria, such as confining the cooking area and equipping the kitchen with a more efficient smoke exhaust system, should be targeted. Other measures could be the installation of appropriate ventilation and air cleaner systems in the dining room, the use of eco-labelled cleaning products, the adoption of integrated pest management (IPM) alternatives to pesticides, placing footwear sanitiser mats at the entrance, inspection of wall coverings and other building materials and, if necessary, replacement by low emitting materials, minimising the use of plastics and compliance with prohibitions (e.g. smoking). At the official level, the implementation of regular inspection protocols is also essential.

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# Acknowledgments

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Table 1
Weekly concentrations of volatile organic compounds (μg m<sup>-3</sup>) monitored by passive sampling

	n-hexane	1-butanol	benzene	toluene	ethyl benzene	m-xylene	p-xylene	2-ethoxyethyl acetate	o-xylene	1,2,4-trimethylbenzene
Indoor	0.4-1.1	3.7-13.8	0.9-1.4	3.4-10.4	0.7-1.0	1.8-2.4	0.7-1.4	<0.3	0.8-1.1	0.8-1.2
Outdoor	0.1	< 2.0	0.6-0.8	1.3-1.6	0.3-0.5	0.6-1.0	0.3-0.4	< 0.3	0.3-0.5	0.4-0.5
I/O	3.8-12.6	-	1.3-1.8	2.1-6.2	1.5-3.1	1.9-3.2	1.5-3.1	-	1.4-2.9	1.7-3.0

Table 2

Minimum, maximum and average concentrations of elements (ng m<sup>-3</sup>), enrichment factors and indoor/outdoor ratios for working hours

						~.																						
			Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Br	Rb	Sr	Y	Zr	Mo	Pb
or ancy	_	Min	197	69.0	173	347	20.5	100	214	305	452	11.1	-	0.418	3.93	93.4	0.356	2.73	10.3	-	-	1.24	0.426	1.26	-	0.900	-	0.603
	ancy	Max	356	776	1151	828	390	1070	1870	2140	850	33.9	0.682	5.47	8.72	271	3.91	14.9	60.9	1.41	3.89	4.31	1.25	4.94	0.304	2.03	2.05	5.98
Indoor	Occupancy Day	Avg	250	232	465	620	27.3	485	607	692	625	22.3	0.190	1.66	5.54	195	1.49	6.90	27.8	0.757	1.62	2.77	0.749	3.00	0.089	1.27	0.551	2.80
ŏ	ŏ	EF	1.6	2.4	0.88	0.33	7.0	86	137	3.0	3.5	1.1	0.67	6.8	1.7	-	12	73	80	59	3094	284	1.1	1.5	0.64	0.89	68	26
		Min	41.5	9.58	30.5	58.1	0.158	155	13.3	117	8.37	2.43	0.695	-	-	26.7	-	0.98	7.01	0.102	0.427	0.450	-	-	-	-	-	-
or	þt	Max	115	23.9	100	137	3.11	500	79.8	197	51.2	4.20	0.947	0.421	1.44	53.6	0.496	1.23	10.6	1.06	1.96	0.628	0.856	1.02	-	0.424	2.61	5.46
Indoor	Night	Avg	73.2	14.8	71.8	110	1.49	305	37.8	148	28.6	3.19	0.790	0.215	0.766	39.7	0.277	1.09	8.27	0.532	1.15	0.566	0.285	0.627	-	0.141	0.944	2.51
		EF	2.3	0.86	0.74	0.29	1.7	220	47.1	3.2	0.79	0.78	11	5.1	1.2	-	13	56	115	217	9397	260	1.5	1.4		0.34	541	95
		Min	403	79.3	142	243	5.32	152	470	60.6	123	6.25	0.239	-	2.17	136	-	2.36	6.16	-	0.011	1.77	0.334	1.18	-	0.507	-	0.189
oor	>	Max	766	130	619	1363	26.3	1711	988	365	641	31.5	2.76	2.55	10.6	660	1.00	14.8	32.9	2.76	9.69	9.24	2.57	5.31	1.62	3.35	6.35	6.04
Outdoor	Day	Avg	550	102	424	802	15.3	570	722	184	356	20.7	1.23	1.11	6.09	397	0.459	8.15	17.0	0.958	2.61	3.75	1.30	3.66	0.800	1.69	2.03	2.86
Ū		EF	2.3	0.75	0.44	0.20	1.8	32	106	0.39	0.91	0.52	1.7	1.7	0.90	-	1.5	42	24	23	1105	161	1.0	0.94	4.3	0.61	150	10
oor	-0c.	Concent.	224	24.7	41.0	72.6	5.45	182	114	35.5	21.0	2.43	-	-	0.621	25.7	0.450	0.867	1.13	0.585	0.924	0.819	-	-	-	-	1.72	-
Indoor Non-oc.	Non-o Day	EF	11	2.2	0.64	0.29	9.8	230	214	1.3	0.86	0.93	-	-	1.4	-	29	73	26	351	13370	615	-	-	-	-	1474	-
0/I			0.46	2.3	1.1	0.77	1.8	0.85	0.84	3.8	1.8	1.1	0.16	1.5	0.91	0.49	3.3	0.85	1.6	0.79	-	0.74	0.57	0.82	0.11	0.75	-	0.98

Table 3 Concentrations (ng  $\,\text{m}^{\text{-3}}$ ) of oxygenated organic compounds in  $PM_{10}$ 

	Indoor	Indoor	Indoor	Outdoor	I/O	
	occupancy	non-occupancy	non-occupancy	daytime	(occupancy)	
	occupancy	(night-time)	(Sunday, daytime)	daytime	(occupancy)	
Saccharides			C			
Galactosan	1.60±0.61	2.05	0.422	4.5±4.0	0.4	
Mannosan	3.72±2.12	4.49	0.904	8.4±6.2	0.4	
Levoglucosan	39.5±11.4	41.9	bdl	59±55	0.7	
Unidentified saccharides	90.6±46.6	0.84	2.00	10.6±4.9	8.5	
Phenolics and alteration products						
Pyrocatechol	0.006±0.002	0.004	bdl	0.005±0.004	1.0	
Resorcinol	0.019±0.012	0.073	0.003	0.10±0.12	0.2	
4-Methylcatechol	0.004±0.002	0.004	bdl	0.003±0.003	1.1	
5-Isopropyl-3-methylphenol	0.065±0.051	bdl	bdl	0.029±0.027	2.2	
2-Isopropyl-5-methylphenol (thymol)	0.64±0.36	0.070	0.242	$0.34\pm0.38$	1.9	
Eugenol	0.004±0.003	0.002	0.001	0.001±0.001	5.4	
2,4-Di-tert-butylphenol	26.4±19.8	bdl	4.03	5.4±4.7	4.9	
4-tert-Butylphenol + unknown	13.0±7.3	10.0	bdl	9±11	1.4	
Pyrogallol	0.004±0.004	0.004	0.001	0.005±0.003	0.8	
Isoeugenol	0.51±0.37	bdl	0.134	0.25±0.18	2.1	
2-Methoxy-4-propylphenol	0.002±0.001	0.005	bdl	0.003±0.003	0.5	
4-Phenylphenol	0.030±0.028	0.037	bdl	0.038±0.031	0.8	
4-Octylphenol	0.011±0.007	0.002	bdl	0.006±0.004	1.8	

Benzyl alcohol	10.0±4.5	0.050	3.24	18.0±20	0.6
Benzoic acid	0.38±0.43	0.063	bdl	0.024±0.029	15
3-Hydroxybenzoic acid	1.53±0.41	bdl	bdl	0.63±0.30	2.4
4-Hydroxybenzoic acid	0.65±0.23	0.587	0.097	0.82±0.67	0.8
Vanillic acid	0.35±0.23	0.172	0.118	0.56±0.36	0.6
Sinapyl alcohol	0.053±0.066	0.141	bdl	0.005±0.008	9.7
Syringic acid	$0.20\pm0.22$	0.452	0.073	0.24±0.25	0.8
trans-Cinnamic acid	0.22±0.23	bdl	0.071	0.080±0.065	2.8
4-Hydroxy-3-methoxycinnamic (ferulic) acid	0.032±0.028	0.004	bdl	0.013±0.009	2.4
3,4-Dihydroxycinnamic (caffeic) acid	0.013±0.017	bdl	bdl	0.001±0.001	9.0
4-Hydroxycinnamic (p-coumaric) acid	0.49±0.56	bdl	bdl	0.14±0.13	3.5
Aliphatic alcohols					
1-Decanol	0.043±0.038	0.001	0.004	0.005±0.007	9.3
1-Pentadecanol	11.8±7.6	8.63	13.2	bdl	-
iso- or anteiso-Pentadecanol	1.42±0.73	bdl	1.98	0.26±0.14	5.5
1-Hexadecanol	38.6±18.4	bld	72.2	0.06±0.13	681
2-Hexadecanol	4.8±2.1	1.60	bld	bdl	-
Iso- or anteiso-heptadecanol	0.56±0.64	bdl	0.860	bdl	-
1-Heptadecanol	2.2±2.0	1.62	bdl	bdl	-
1-Octadecanol	45±14	13.7	5.36	0.07±0.16	643
1-Eicosanol	2.05±0.80	1.27	0.399	1.2±1.1	1.7
1-Docosanol	2.25±0.87	2.14	0.320	2.0±1.3	1.2
1-Tricosanol	0.225±0.060	0.220	0.025	0.10±0.12	2.3
1-Tetracosanol	1.10±0.36	1.59	bdl	1.58±0.72	0.7

1-Pentacosanol	0.26±0.13	0.189	0.014	0.17±0.12	1.6
1-Hexacosanol	6.0±9.1	3.33	0.341	5.7±4.6	1.1
1-Heptacosanol	0.121±0.057	0.076	0.005	0.07±0.10	1.7
1-Octacosanol	1.5±0.21	1.76	0.240	2.4±3.3	0.6
1-Triacontanol	0.248±0.091	0.229	0.010	0.36±0.65	0.7
Triterpenoid and steroid compounds					
Tocopherol (Vit. E)	0.19±0.12	0.006	bdl	0.010±0.009	19
Cholesterol	20.0±8.5	0.170	0.019	0.14±0.15	139
5-Cholesten-3-ol (epicholesterol)	0.27±0.14	bdl	bdl	0.049±0.089	5.6
Stigmasterol	7.7±2.5	0.314	bdl	0.45±0.56	17
$\beta$ -Sitosterol	5.7±2.1	1.37	0.008	0.8±1.4	7.0
Lupeol	0.204±0.072	1.11	0.123	0.42±0.44	1.0
Aliphatic acids					
Hexanoic	1.07±0.44	0.053	bdl	0.43±0.35	2.5
Heptanoic	4.8±3.6	0.073	0.059	0.37±0.33	13
Octanoic	2.4±1.9	0.370	0.142	$0.46\pm0.40$	5.3
Decanoic	2.7±3.0	bdl	0.106	0.37±0.31	7.4
Undecanoic	0.20±0.29	bdl	0.081	bdl	-
Nonanoic	37±37	1.13	1.35	2.4±2.1	15
Dodecanoic	9±13	1.97	0.051	3.1±5.7	3.0
Tridecanoic	2.9±2.8	1.12	3.03	bdl	-
Isotridecanoic	3.2±2.3	3.00	4.21	bdl	-
Tetradecanoic (myristic)	72±28	30.3	66.2	1.0±1.3	73
Pentadecanoic	15.7±4.6	0.286	7.29	0.06±0.11	266

Isotetradecanoic	13.4±6.5	3.780	10.9	bdl	-
Hexadecanoic (palmitic)	117±69	bdl	bdl	10.6±9.9	11
Iso-hexadecanoic	4.5±2.6	0.143	2.06	bdl	-
5-Hexadecenoic	3.10±0.86	bdl	1.13	bdl	-
9-Hexadecenoic (palmitoleic)	1.43±0.85	0.0524	0.106	0.080±0.099	18
Heptadecanoic	14.8±8.2	bdl	0.523	0.38±0.34	39
Isoheptadecanoic	7.6±2.6	0.095	1.55	bdl	-
Octadecanoic (stearic)	116±62	bdl	bdl	1.9±2.6	61
9-Octadecenoic (oleic)	103±35	1.99	0.219	1.1±1.1	93
9,12-Octadecadienoic (linoleic)	30±14	0.272	0.017	0.47±0.74	64
9,12,15-Octadecatrienoic (linolenic)	17±25	bdl	bdl	0.35±0.77	49
Nonadecanoic	0.99±0.30	0.022	0.053	0.140±0.082	6.5
Eicosanoic	8.0±2.9	0.618	0.200	2.2±2.0	3.7
Heneicosanoic	bdl	bdl	0.221	0.22±0.23	-
Docosanoic	9.0±3.2	2.46	0.171	2.2±2.1	4.1
Pentacosanoic	2.0±1.7	bdl	bdl	0.10±0.23	20
Hexacosanoic	1.36±0.66	0.509	0.046	0.54±0.57	2.5
Diacids					
Butanedioic (succinic)	2.8±2.4	bdl	0.691	2.3±1.4	1.2
1,5-Pentanedioic (glutaric)	3.9±2.6	1.52	5.25	0.07±0.14	58
Hydroxybutanedioic (malic)	0.7±1.0	bdl	bld	0.5±1.2	1.2
Hexanedioic (adipic)	1.07±0.31	bdl	0.388	bdl	-
Heptanedioic (pimelic)	1.28±0.25	0.032	0.190	0.116±0.076	11
Octanedioic (suberic)	3.0±1.1	bdl	0.338	0.175±0.098	17

Nonanedioic (azelaic)	44±21	1.08	1.09	7.0±4.0	6.2
Decanedioic (sebacic)	0.111±0.031	bdl	bld	0.010±0.010	12
Hexanedecanoic (thapsic)	0.052±0.022	bdl	0.006	0.023±0.031	2.2
Resin acids					
Dehydroabietic	3.0±1.4	1.98	0.757	4.6±1.7	0.6
Isopimaric	0.14±0.31	0.130	0.057	0.12±0.16	1.1
Abietic	0.119±0.061	0.054	bdl	0.076±0.088	1.6
Podocarpic	0.038±0.027	bdl	bdl	0.001±0.002	1.1
Other acids					
Boric	15±15	2.42	11.8	13±12	1.1
Propionic (lactic)	130±120	bdl	35.9	bdl	-
4-Oxopentanoic (levulinic)	13.2±9.6	5.31	1.26	$0.64 \pm 0.88$	15
2-Hydroxyethanoic (glycolic)	32±42	bdl	6.45	5.3±8.6	6.0
2,3-Dihydroxypropanoic (glyceric)	9.4±7.0	bdl	1.41	1.0±1.1	9.7
3-Hydroxybutanoic (3-hydroxybutyric)	0.23±0.35	bdl	0.152	0.049±0.079	4.7
3,4-Dihydroxybutanoic	3.1±2.0	bdl	0.691	0.98±0.61	3.2
Nicotinic	8.6±5.3	4.67	6.84	bdl	-
cis-Pinonic	1.03±0.87	bdl	1.05	0.53±0.45	1.9
Pinic	1.10±0.80	0.933	0.424	1.5±1.4	0.7
2-Hydroxypropane-1,2,3-tricarboxylic (citric)	1.0±1.1	bdl	0.912	0.30±0.28	3.4
Glycerol derivatives					
Glycerol	80±35	17.7	57.2	32±30	2.5
Diethylene glycol	12.2±7.0	4.22	10.8	6.8±2.7	1.8
1-Monolauroyl-rac-glycerol	0.021±0.014	bdl	bdl	0.001±0.000	23

1-Glycerol monostearate (1-monostearin)	170±130	1.29	7.52	4.0±3.3	42
2-Glycerol monostearate (2-monostearin)	16±15	bdl	bdl	0.030±0.042	550
Heptadecanoic acid glycerin-(1)-monoester	6.0±4.9	bdl	bdl	bdl	-
1-Monolinoleoylglycerol	23±18	bdl	bdl	bdl	-
1-Monopalmitin	190±110	0.519	0.902	4.3±2.7	45
1-Monomyristin	33±22	0.776	0.616	1.350±0.066	25
Other compounds					
Myrtenol	0.005±0.005	bdl	bdl	-	-
Hydroquinone	0.142±0.022	0.112	bdl	0.15±0.12	0.9
Hydroxyacetophenone	0.19±0.17	0.382	bdl	1.40±0.91	0.1
2,6-Di-tert-butyl-1,4-benzoquinone	$0.44\pm0.40$	bdl	0.088	0.36±0.21	1.2
Methyl dihydrojasmonate	46±29	55.1	55.9	-	-
2-Propanol-1-chloro-phosphate (TCPP)	1.6±1.2	bdl	4.24	-	-
Ethylene brassylate	18.4±6.2	6.77	6.24	-	-
5-oxo-L-proline	19±14	bdl	1.24	-	-
Caffeine	10	6.31	9.85	1.4±3.0	-
Parsol MCX	12.6±6.6	2.93	11.2	0.10±0.14	122
Acetyl tributyl citrate	4.0±1.6	1.78	2.82	-	-
Diisooctyl adipate	10.0±3.5	2.55	5.40	2.7±5.9	3.8
Dibutyl adipate	14.2±7.4	6.33	4.82	0.6±1.3	23
$\Delta$ -1-Tetrahydrocannabinol	4.8±9.5	bdl	bdl	-	-
Piperonyl butoxide	9.3±3.7	4.25	6.68	-	-
Oxidised Irgafos 168	7±10	bdl	bdl	8±18	1.0

Plasticisers

Dimethyl phthalate	bld	0.002	bdl	bdl	-
Diethyl phthalate	0.064±0.092	bdl	bdl	0.24±0.27	0.26
Di-n-butyl phthalate	0.23±0.21	0.059	bdl	2.1±2.6	0.11
Benzyl butyl phthalate	$0.004 \pm 0.008$	bdl	bdl	$0.05\pm0.10$	0.08
Bis(2-ethylhexyl) adipate	0.006±0.009	bdl	bdl	$0.09\pm0.19$	0.06
Bis(2-ethylhexyl) phthalate	0.17±0.23	bdl	bdl	2.1±4.4	0.08
Unidentified phthalate	280±160	121	163	19±25	15

bld – below detection limit

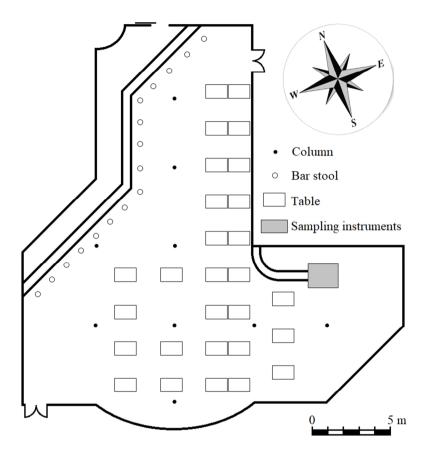


Fig 1. Floor plan of the cafeteria. Each table has four chairs.

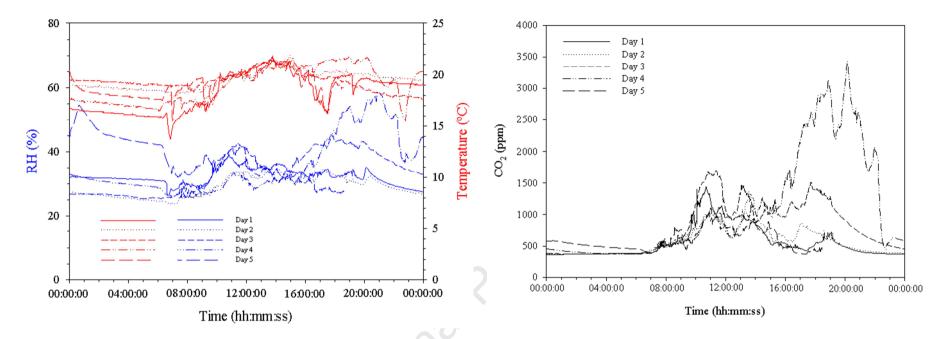
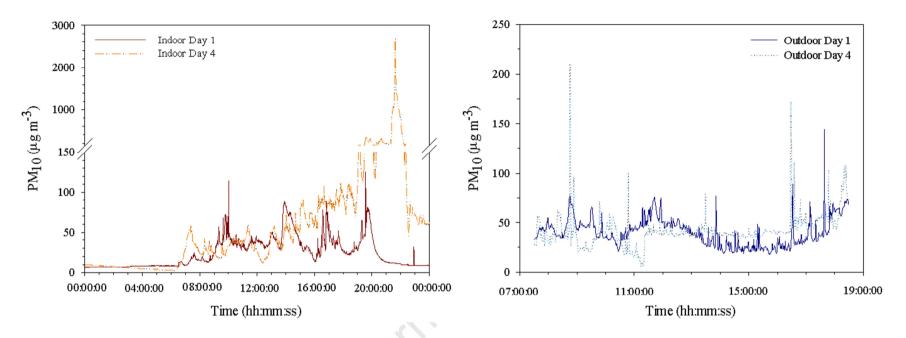
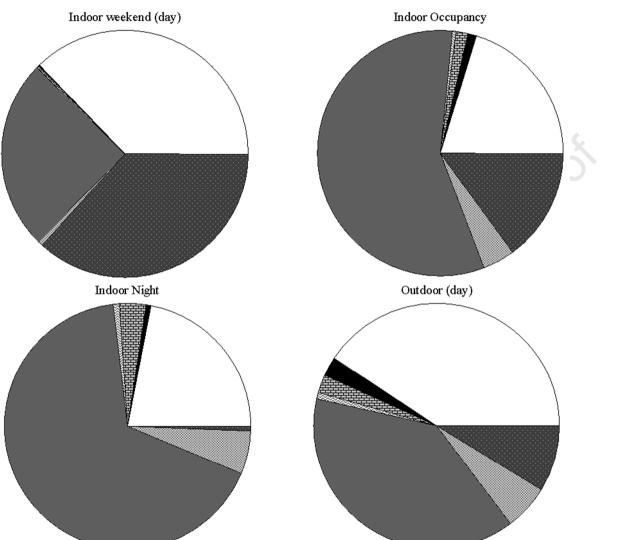


Fig 2. Variation of relative humidity, temperature and carbon dioxide levels on weekdays.



**Fig. 3.** Examples of  $PM_{10}$  daily concentration profiles in the cafeteria and outdoors (Day 1 – Monday, Day 4 – Thursday).



**Fig. 4.** Chemical mass closure of  $PM_{10}$ .

Element oxides
Nitrate
Sulphate
Ammonium

Organic matter
Elemental carbon
Unexplained mass

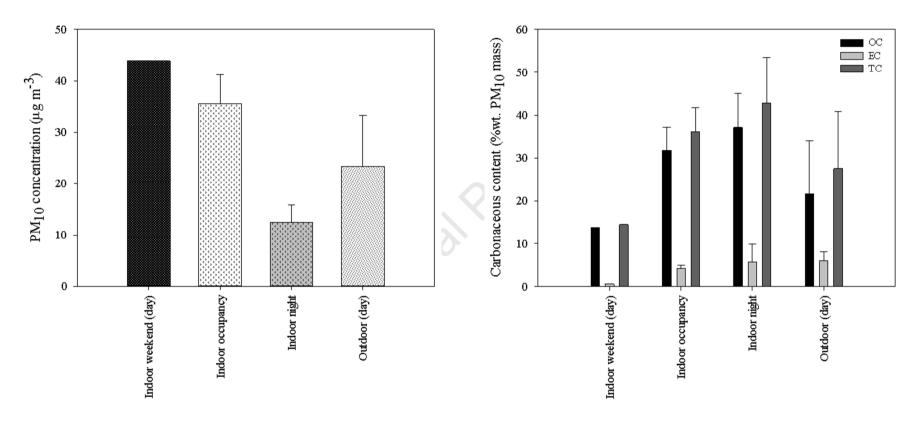


Fig 5. Means ( $\pm$ SD) of PM<sub>10</sub> concentrations and mass fractions of organic carbon, elemental carbon and total carbon (TC = OC + EC)

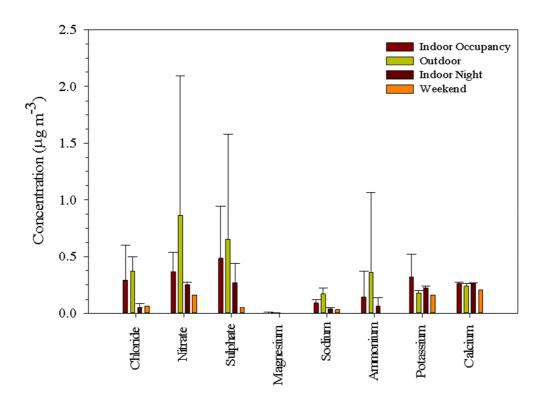


Fig. 6. Concentrations (mean $\pm$ SD) of water-soluble ions in PM $_{10}$  collected indoors and outdoors.

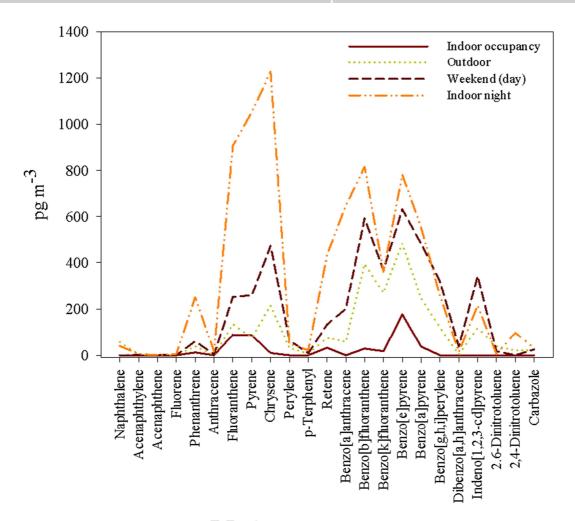
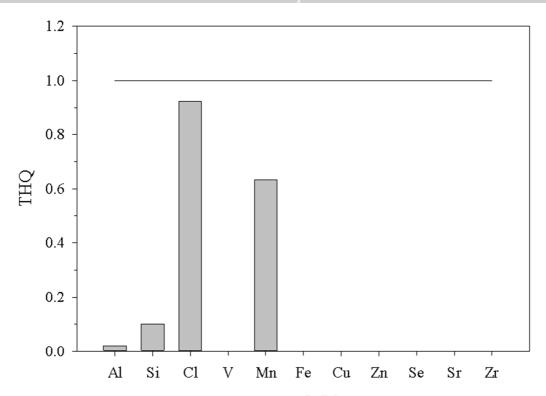


Fig. 7. Average concentrations of polycyclic aromatic hydrocarbons.



**Fig. 8**. Mean risks from inhalation exposure to noncarcinogenic  $PM_{10}$ -bound metals for the cafeteria workers during the occupancy period. The horizontal line represents the USEPA health-based guideline level.

#### **HIGHLIGHTS**

Indoor air quality in a cafeteria revealed poor ventilation conditions

Concentrations of most pollutants were much higher indoors than outdoors

More than 80% of the particles were generated indoors

PM<sub>10</sub> included components of personal care products, plasticisers and psychoactive drugs

Cancer risk associated with inhalation of metals and PAHs was found to be negligible

Conflicts of Interest: The authors declare no conflict of interest.

