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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, CO₂, CO and particulate matter were continuously monitored indoors and outdoors. Simultaneous PM₁₀ 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 (0.31-1.5 h⁻¹) and infiltration factors of 0.14, for both PM_{2.5} and PM₁₀, 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 PM₁₀ was estimated to be 32 µg m⁻³. Organic compounds in PM₁₀ 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. The inhalation cancer risks of metals and PAHs were found to be negligible.

Keywords: Cafeteria, Indoor air quality, PM₁₀, Metals, Organic compounds, Sources

38 1. Introduction

39

40 In recent decades, Indoor Air Quality (IAQ) has been the focus of attention by the scientific
41 community, governments and international organisations (Abdul-Wahab et al., 2015). It is a known fact
42 that we spend more than 90% of our time indoors and that the levels observed are often higher than
43 those recorded outdoors. Thus, the health risks from exposure to indoor air pollution may be larger than
44 those associated with outdoor pollution. Poor IAQ can be especially detrimental to vulnerable groups
45 such as children, young adults, the elderly, or those suffering from chronic respiratory and/or
46 cardiovascular diseases (Cincinelli and Martellini, 2017). Furthermore, IAQ in the workplace is
47 important because employees are occupationally exposed on a regular basis to a variety of pollutants
48 with potential impact on comfort, work-related health problems, sickness absence, and productivity (Al
49 horr et al., 2016; Carrer and Wolkoff 2018; Śmiełowska et al., 2017). IAQ in elderly care centres (e.g.
50 Almeida et al., 2016; Bentayeb et al., 2015; Buczyńska et al., 2014; Mendes et al., 2015; Mui et al.,
51 2008; Simoni et al., 2003), office buildings (e.g. Mandin et al., 2017; Śmiełowska et al., 2017; and
52 references therein) and schools (e.g. Alves et al., 2013; Barmparesos et al., 2018; Bennet et al., 2019;
53 Blondeau et al., 2005; Chatzidiakou et al., 2015; Fuoco et al., 2015; Kalimeri et al., 2016; Madureira et
54 al., 2016; Yang et al., 2015) has been extensively investigated. Some studies were also conducted to
55 evaluate the IAQ in restaurants with different cooking styles (Dai et al., 2018; Lee et al., 2001; Lee et
56 al., 2011; Elsharkawy and Javed, 2018; Mugica et al., 2001; Sofuoglu et al., 2015; Taner et al., 2013;
57 Wilson et al., 2011).

58 Restaurants not only have many of the same threats generally observed in office buildings, but they
59 also encompass several other pollutants given off by cookware, open flames, cooking appliances,
60 refrigeration units, strong chemical degreasers and aggressive cleaning solvents. Most of the published
61 studies report the monitoring of comfort parameters, ventilation and/or some regulated pollutants.
62 Gaseous compounds (CO, CO₂, NO_x and some volatile organic compounds), particles less than 2.5 and
63 10 µm in diameter (PM_{2.5} and PM₁₀, respectively), as well as some organic compounds in the particulate
64 phase, normally from the group of polycyclic aromatic compounds (PAHs) with proven carcinogenic
65 properties, are the most commonly monitored pollutants (Abdullahi et al., 2013; Maré et al., 2018; and
66 references therein).

67 University cafeterias, a specific type of eating establishment, are popular meeting places for students
68 and staff, providing space for leisure, relaxation and a room to have a quick lunch or snack. For this
69 category of restaurant, as far as we know, no exhaustive evaluation has been made. Using optical
70 counters, Zhao et al. (2010) measured PM concentrations in three different cafeterias located at
71 Tsinghua University, Beijing, China. However, no information is given on the chemical composition
72 of the particles and on the concentration of other pollutants. Hsu and Huang (2013) conducted a study
73 in the student cafeteria at Chia-Nan University in southern Taiwan. Measurements of relative humidity,
74 temperature, airflow velocity, CO₂ and total suspended particles were made, although the focus of the

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

77 This paper describes and discusses a short case study on a comprehensive air quality monitoring
78 programme carried out in a university cafeteria. Besides the measurement of traditional parameters,
79 such as regulated gaseous pollutants, the work had as main objective the detailed characterisation of
80 PM₁₀, including organic and inorganic components, in order to understand which sources and processes
81 contribute to the measured levels and to estimate the carcinogenic and non-carcinogenic risks associated
82 with inhalation of elements and PAHs by employees and customers. This preliminary study can serve
83 as a basis for improving future monitoring campaigns, especially as regards the definition of source-
84 marking chemical species, and for defining mitigation strategies to reduce exposure to pollutants in
85 similar facilities.

86

87 **2. Methodology**

88

89 **2.1. Sampling**

90

91 A one-week extensive sampling campaign was carried out from 27th November to 1st December 2017
92 at the León (Spain) university cafeteria. The commercial establishment is continuously open for 12
93 hours (7:00-19:00, UTC) from Monday to Friday. The cafeteria is one large open room (410 m²) with
94 fourteen tables, each capable of seating four to eight people, three standing round tables and stools along
95 the 21 m long bar (Fig. 1). A separate kitchen provides the space to prepare the daily special menus and
96 several dishes of fast food, such as burgers, sandwiches and French fries. The kitchen is equipped with
97 a diesel fired range cooker, electric grills and toasters. The extractor hood of the kitchen has 4.8 × 2.4
98 m. There is no information regarding the brand, model or power. The cafeteria has two entrance doors,
99 one opening to an adjacent alley and the other opening to an outdoor patio. A heat pump works as heater
100 during the colder months and as air conditioner during the warmer months. Attendance indoors was
101 highly variable, depending on the time of the day. Officially, the maximum occupancy allowed is 140
102 persons. People occupancy was logged manually throughout the campaign. In general, in the early
103 morning, there was a simultaneous presence of 10 to 20 people, reaching a peak at lunch time (60 to
104 100 people). During the afternoon, the frequency of attendance varied from 10 to 40 clients, with a
105 maximum around 16:00.

106 Temperature, relative humidity, CO₂ and CO were continuously monitored with air quality probes
107 inside (TSI, model 7545) and outside (Gray Wolf[®], WolfSense IQ-610) the cafeteria. Real-time laser
108 photometric instruments (TSI, DustTrak DRX 8533) were used to record particulate matter (PM₁, PM_{2.5}
109 and PM₁₀) concentration profiles over time in the indoor and outdoor environments, simultaneously.
110 All these continuous monitoring instruments were operated with 1-min resolution and were factory-
111 calibrated before use. Simultaneous sampling with two PM₁₀ high volume air MCV (model CAV-A/mb)

112 instruments for gravimetric quantification was carried out on pre-weighed 150 mm quartz fibre filters
113 (Pallflex®). The equipment was operated at a flow of 30 m³ h⁻¹. Samples were collected during the
114 opening hours, simultaneously indoors and outdoors. Night time samples were also taken in order to
115 compare the PM levels during occupancy and non-occupancy periods. Furthermore, PM₁₀ samples were
116 collected on Teflon membrane filters (47 mm diameter, Pall Corporation) with a TCR TECORA (model
117 2.004.01) operated at a flow of 2.3 m³ h⁻¹. The gravimetric quantification was performed with an
118 analytical balance (Mettler Toledo, XPE105 DeltaRange®). Due to the possibility of vandalism and
119 unfavourable weather conditions, with freezing temperatures, the outdoor equipment was shut down
120 and sheltered at night. Thus, outdoor measurements correspond to the occupancy period in the cafeteria.
121 All the data were registered in UTC.

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

129

130 **2.2. Chemical analyses**

131

132 After gravimetric determinations, thermal-optical analysis of PM₁₀ quartz filters was performed to
133 obtain the carbonaceous content (organic and elemental carbon, OC and EC). This method is based on
134 the CO₂ quantification, by a non-dispersive infrared (NDIR) analyser, which is released from the
135 volatilisation and oxidation of different carbon fractions under controlled heating. A laser beam and a
136 photodetector that measure the filter light transmittance allow separating the EC formed by OC
137 pyrolysis from the one that was initially in the filter (Alves et al., 2011).

138 Two circular punches of 47 mm from the quartz filters were sequentially extracted with
139 dichloromethane and methanol. The resulting extract was separated into five different organic fractions
140 by flash chromatography with silica gel and various solvents of increasing polarity. To meet the
141 detection limits required by the speciated organic compounds, the night sampling filters were combined
142 and extracted together. Thus, an average composition was obtained for non-occupancy periods at night.
143 After each elution, the different fractions were concentrated in a TurboVap system from Biotage and
144 evaporated to dryness by a nitrogen stream. Before chromatographic analysis, the fractionated extracts
145 containing polar compounds were derivatised to trimethylsilyl ethers. All the extracts were analysed by
146 gas chromatography-mass spectrometry (GC-MS). A detailed description of the whole procedure for
147 the speciation of organic compounds was previously reported by Alves et al. (2011).

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

152 The carbonyl-DNPH derivatives were extracted from the Radiello tubes with 2 mL of acetonitrile
 153 for 30 min, filtered through a micropore filter membranes with porosity of 0.45 μm , and analysed by
 154 high performance liquid chromatography. HPLC measurements were carried out with a Jasco PU-980
 155 pump, a Rheodyne 7125 six-port valve with a 20 μL loop and a Jasco MD-1510 diode array detector.
 156 An isocratic mixture of acetonitrile and water (60:40), at a flow rate of 1.5 mL min^{-1} , was used to
 157 separate the 2,4-DNPH on a Supelcosil™ LC-18 column (5 μm particle size; $L \times \text{I.D.}$: 250 \times 4.6 mm).
 158 The carbonyl concentrations were quantified with external calibrations curves constructed from
 159 standard solutions of carbonyl-DNPH derivatives in acetonitrile. The UV wavelength for detection was
 160 360 nm. VOCs in activated charcoal cartridges were chemically desorbed with 2 mL of carbon
 161 disulphide (CS_2) and 100 μL of internal standard (2-fluorotoluene) for 30 min and analysed by gas
 162 chromatography coupled to flame ionisation detector (GC-FID), following the conditions described in
 163 Pegas et al. (2010).

164

165 2.3. Air change rates

166

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

170

$$171 \quad C_t = C_{ext} + \frac{q_{\text{CO}_2} \times 10^6}{Q} - \left(C_{ext} - C_0 + \frac{q_{\text{CO}_2} \times 10^6}{Q} \right) e^{\left(\frac{-Q_t}{V} \right)} \quad (1)$$

172

173 where C_t is the indoor concentration of CO_2 at time t (ppm), C_{ext} is the outdoor concentration of CO_2
 174 (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
 175 entering the space ($\text{m}^3 \text{s}^{-1}$), q_{CO_2} is the volumetric indoor emission rate of CO_2 ($\text{m}^3 \text{s}^{-1}$), V is the volume
 176 of the room (m^3) and t is the interval since $t = 0$ (s). When the room is unoccupied there is no CO_2
 177 emission from the occupants, and $q_{\text{CO}_2} = 0$. Thus, Eq. (1) can be rearranged to give the following
 178 expression, which allows the ventilation rate (Q) to be estimated from measured concentration values
 179 time t apart:

180

$$181 \quad Q = -\frac{V}{t} \times \ln \left(\frac{C_t - C_{ext}}{C_0 - C_{ext}} \right) \quad (2)$$

182

183 The air change rate is defined as:

184

$$185 \quad ACR = Q/V \quad (3)$$

186

187 **3. Results and discussion**

188

189 ***3.1. Comfort parameters and gaseous pollutants***

190

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

207 CO₂ at the concentrations usually observed in buildings is not a direct health risk, but its levels can
208 be used as an indicator of occupant odours (bioeffluents). CO₂, primarily generated through human
209 metabolism, provides a good indication of air change rates. Its build up in indoor environments is
210 attributed to inefficient ventilation. ASHRAE (Standard 62.1-2016) reports that while not necessarily
211 hazardous, the amount of CO₂ in an indoor environment should remain under 1,000 ppm. The mean
212 daytime carbon dioxide concentration indoors was 891±226 ppm, decaying to values around 400 ppm
213 at night. A twofold difference in indoor concentration was observed between occupancy and non-
214 occupancy periods. Concentrations recorded throughout the weekend were similar to those observed in
215 nocturnal periods. The inter-day fluctuations depended on the occupancy rates, reaching a maximum
216 concentration of 3430 ppm in the early evening of Thursday (Fig. 2), the day the cafeteria closed later
217 due to a party attended by about 150 people.

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

225 Carbon monoxide levels were very low indoors. The maximum concentration ever recorded was 1.1
226 ppm. All the doors and windows were kept closed, due to the cold weather outside, preventing CO
227 infiltration into the cafeteria from the nearby low traffic street and doorstep smokers. Low
228 concentrations, always < 3 ppm, were also observed outdoors. ASHRAE (Standard 62.1-2016) lists a
229 maximum allowable short-term limit of 9 ppm for CO in indoor air, while the WHO (2010) recommends
230 CO concentrations based on exposure time ranging from 90 ppm (15-minute exposure) to 10 ppm (8-
231 hour exposure). The American Conference of Governmental Industrial Hygienists (ACGIH) has
232 assigned CO a threshold limit value (TLV) of 25 ppm as a time weighted average (TWA) for a normal
233 8-hour workday and a 40-hour workweek. The ACGIH limit is based on the risk of elevated
234 carboxyhemoglobin levels. The National Institute for Occupational Safety and Health (NIOSH) has
235 established a recommended exposure limit of 35 ppm as an 8-hour TWA and 200 ppm as a ceiling,
236 based on the risk of cardiovascular effects.

237 Formaldehyde and acetaldehyde, two ubiquitous compounds emitted by combustion processes, meat
238 cooking operations and consumer products, presented indoor mean concentrations of 8.8 and 17.8 µg
239 m⁻³, respectively, while the corresponding outdoor values were 1.3 µg m⁻³ and of the same order of the
240 blanks. The inter-week variations were always <10%. Formaldehyde was below the air quality guideline
241 of 100 µg m⁻³ (as a 30-min average value) recommended by the World Health Organisation (WHO) in
242 2010. However, short-time exceedance of this guideline level might have occurred in the indoor
243 environment. Acetaldehyde was included by the WHO in Group 2, which encompasses pollutants of
244 potential interest, but further investigation would be needed before it is clear whether there is enough
245 evidence to warrant their inclusion in the guidelines. However, based on studies of short- and long-term
246 exposure, countries such as Canada have set a maximum daily limit of 280 µg m⁻³.

247 The indoor levels of VOCs (Table 1) were higher than those observed outdoors. Among these,
248 ambient concentrations of benzene are regulated by law due to its toxicity (European Commission,
249 2009). In contrast, indoor levels of VOCs are not subject to any legislation. Of all the VOCs found in
250 buildings, BTEX (benzene, toluene, ethyl benzene, o-, m-, and p-xylene) are among the most common.
251 Indoors, these compounds can originate from multiple sources, including building materials (paints,
252 varnishes), consumer products (cleaning agents, fingernail polish, lacquers, adhesives), and human
253 activities (smoking and combustion). In outdoor air, traffic is pointed out as a major source. The outdoor
254 concentrations of benzene were far below the annual ambient air quality standard of 5 µg m⁻³ set by the

255 European Commission. Previous studies have reported the indoor BTEX levels at homes (Dai et al.,
256 2017; Esplugues et al., 2010; Fan et al., 2018; Liu et al., 2013), libraries (Cincinelli et al., 2016), schools
257 (Demirel et al., 2014; Pegas et al., 2011; Raysoni et al., 2017), printing shops and photocopy centres
258 (El-Hashemy and Ali, 2018), and a confined parking area (Castro et al., 2015). Highly variable
259 concentrations, up to hundreds of $\mu\text{g m}^{-3}$, have been obtained, depending on the source strengths.
260 Benzene levels of 18.4, 10.0, 9.3 and 3.7 $\mu\text{g m}^{-3}$ were measured in the dining areas of a Korean barbecue
261 style restaurant, a Chinese hot pot restaurant, a Chinese dim sum restaurant and a Western canteen,
262 respectively (Lee et al., 2001). The corresponding values reported for toluene were 156.1, 93.8, 81.6
263 and 17.6 $\mu\text{g m}^{-3}$. In the present study, *n*-butanol and toluene were the VOCs with the highest indoor air
264 concentrations. Butanol (or butyl alcohol) is present in many foods and beverages and is also used in a
265 wide range of consumer products, such as cleaning products or cosmetics. Toluene is commonly added
266 to gasoline and can enter the indoor spaces from outside. Toluene is also often used as solvent and can
267 be released from a variety of products found in homes, including paints and other finishes, adhesives,
268 and some personal care products. It is also found in tobacco smoke. Although detected at lower
269 concentrations compared to these two VOCs, *n*-hexane showed very high I/O ratios (up to 13),
270 suggesting the presence of active sources in the cafeteria. This aliphatic compound is primarily used as
271 a solvent and as a component of certain glues and adhesives, cleaning agents, biocidal products, ink and
272 toners, textile dyes, perfumes, fragrances and cosmetics (Federal Institute for Occupational Safety and
273 Health, 2017). It is also present in volatile fractions of various plant species, such as apples and orange
274 juice (Toxno, 2018). Moreover, Ahearn et al. (1996) reported that a variety of fungi found in buildings
275 are capable of releasing gases that include *n*-hexane.

276

277 **3.2. Particulate matter**

278

279 **3.2.1. Mass concentrations**

280

281 From the gravimetric measurements, average PM_{10} concentrations of 35.6 ± 5.6 , 12.4 ± 3.4 and 23.3
282 ± 10.0 and $\mu\text{g m}^{-3}$ were recorded indoors during the occupancy and non-occupancy hours, and in the
283 outdoor air, respectively, not exceeding the 24-h guideline of $50 \mu\text{g m}^{-3}$ set by the WHO. However, the
284 real-time data collected by the photometric monitors show strong variations throughout the day. The
285 highest values were reached at morning snack and lunch time, when the influx of students and staff was
286 typically higher. Throughout the week, the maximum $\text{PM}_{2.5}$ and PM_{10} values were in the ranges 44-92
287 and 62-136 $\mu\text{g m}^{-3}$, but peaks as high as 2560 and 2710 $\mu\text{g m}^{-3}$, respectively, were registered in late
288 afternoon and early evening of the day of a student party, during which even some firecrackers were
289 launched Fig. 3). Throughout the week, a peak was observed between 19:00 and 20:00, time at which
290 cleaning activities of the premises took place. Fine particles represented an overwhelming proportion
291 of PM_{10} indoors: $\text{PM}_{2.5}/\text{PM}_{10}=0.82-0.91$ (avg=0.88) and $\text{PM}_1/\text{PM}_{10}=0.81-0.91$ (avg=0.87). The

292 PM_{2.5}/PM₁₀ and PM₁/PM₁₀ ratios were even higher outdoors, denoting the dominance of fine aerosols:
293 0.92-0.97 (avg=0.95) and 0.91-0.97 (avg=0.94). Regardless of particle size, indoor concentrations were
294 1.5-2 times greater than those found in outside air. During the night, in the cafeteria, the concentrations
295 drop approximately 3 times.

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

298

$$299 C_{in} = F_{inf} \times C_{out} + C_{ig} \quad (4)$$

300

301 where C_{in} and C_{out} are the indoor and outdoor PM_{2.5} or PM₁₀ concentrations, F_{inf} is the infiltration factor,
302 and C_{ig} represent the concentrations of indoor-generated particles. The slopes of the correlations
303 between indoor and outdoor concentrations for the occupancy periods, i.e. the infiltration factors, were
304 0.14 for both PM_{2.5} and PM₁₀. As inferred from the CO₂ measurements, the low particle infiltration
305 factors denote air tightness. On average, concentrations of indoor-generated particles were 27 $\mu\text{g PM}_{2.5}$
306 m^{-3} and 32 $\mu\text{g PM}_{10} \text{m}^{-3}$. To evaluate the influence of outdoor particulate pollutants on indoor
307 environment, the concept of contribution rate (ρ) was applied, as suggested by Lv et al. (2017):

308

$$309 \rho = (F_{inf} \times C_{out})/C_{in} \times 100 \% \quad (5)$$

310

311 The contribution rates of outdoor sources were estimated to be, on average, 17.7 and 16.1 % for PM_{2.5}
312 and PM₁₀, respectively, whereas a major input from indoor sources was observed (82.3 and 83.9 %).

313

314 3.2.2. Chemical composition

315

316 To obtain a PM₁₀ mass balance (Fig. 4), the measured element concentrations were converted into
317 the respective mass concentrations of the most common oxides (SiO₂, Al₂O₃, CaO, MgO, MnO, Fe₂O₃,
318 TiO₂, K₂O, etc.). Because the GC-MS analyses indicated the presence of highly oxygenated compounds,
319 to derive the organic matter content in PM₁₀, a total organic mass to organic carbon ratio (OM/OC) of
320 1.8 was adopted (Polidori et al., 2008). Total carbon (TC = OC + EC) accounted for 36.0 \pm 5.8, 42.8 \pm
321 7.9 and 27.6 \pm 12.6 %wt. of the PM₁₀ mass indoors during the occupancy and non-occupancy hours,
322 and in the outdoor air, respectively (Fig. 5). Besides sampling and analysis artefacts affecting the
323 attainment of chemical mass balance, the unaccounted mass can partly be explained by the presence of
324 unanalysed constituents. Furthermore, part of the unaccounted PM mass is usually assigned to particle-
325 bound water (Tsyro, 2005). On average, I/O ratios of 1.5 and 0.7 were obtained for OC and EC,
326 respectively, suggesting the presence of organic matter emitting sources in the cafeteria, while
327 elemental carbon is predominantly generated from combustion processes in the outdoor environment.
328 Mean OC/EC ratios of 7.5, 3.5 and 3.6 were observed for the occupancy and night-time periods in the

329 cafeteria and for the outdoor air, respectively. A much higher ratio (21.0) was registered for the weekend
330 daytime period. Lower EC infiltration rates over the weekend due to the fact that the building is closed,
331 absence of indoor EC sources (e.g. toasting bread) and off-gassing of VOCs from surfaces leading to
332 secondary organic aerosol formation, may justify the higher proportion of OC compared to EC. It should
333 be noted that OC and EC concentrations decreased from 10.7 and 1.1 $\mu\text{g m}^{-3}$ in the periods of occupation
334 to 6.1 and 0.29 $\mu\text{g m}^{-3}$ in the weekend, respectively.

335 On average, water-soluble ions accounted for PM_{10} mass fractions of 6.0, 15.8, 13.4, and 1.3 %
336 during the occupancy of the cafeteria, at night-time indoors, in the daytime period on the weekend inside
337 the building, and at regular working hours outdoors, respectively. Except for Ca^{2+} and K^+ , the I/O ratios
338 for all water-soluble ions were less than 1. The higher I/O ratios for Ca^{2+} and K^+ may be related to
339 resuspension of fine dust from human movement. Concentration values in daytime were higher than at
340 night-time (Fig. 6). Together with chloride, the secondary ions sulphate, nitrate, and ammonium were
341 the dominant species. In addition to sea salt and biomass burning, chlorinated water and cleaning
342 products containing bleachers can release significant amounts of chloride. Ammonium may also
343 originate from cleaning products. For unknown reasons, in the outdoor sample of Tuesday,
344 concentrations increased approximately 4-fold compared to other days, contributing to a high standard
345 deviation. A good correlation between concentrations of total anions and total cations in PM_{10} was
346 observed, but an anion deficit was registered. Non-measured organic ions, such as formate and acetate,
347 could contribute to the ionic balance. However, the anion deficit is most likely due to the presence of
348 non-determined carbonates ($\text{CO}_3^{2-}/\text{HCO}_3^-$) from resuspended local soil. Outdoors, the molar ratio of
349 NH_4^+ to SO_4^{2-} was found to be greater than 2. This higher molar ratio suggests that in addition to
350 $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 was formed because of excess quantity of NH_4^+ . Ammonium nitrate is produced
351 in the presence of high NH_4^+ and HNO_3 concentrations, low temperatures and high relative humidity.
352 A mean $\text{NH}_4^+/\text{SO}_4^{2-}$ molar ratio of 1.6 was obtained indoors, indicating aerosols in the form of
353 $(\text{NH}_4)_2\text{SO}_4$ and NH_4HSO_4 . Due to the higher temperatures indoors, NH_4NO_3 may decompose to form
354 NH_3 and HNO_3 , which are then partially lost by deposition and sorption processes to indoor surfaces,
355 especially those with higher roughness, while another part reacts with NaCl and Ca -containing particles.
356 NaNO_3 and $\text{Ca}(\text{NO}_3)_2$ have low volatility compared to NH_4NO_3 .

357 Both indoors and outdoors, the elements with highest concentrations were Na, Mg, Al, Si, S, Cl, K,
358 Ca and Fe (Table 2). Elements detected at I/O much higher than 1, suggesting indoor sources, were Mg,
359 P, K, Ca, Cr, Ni and Zn. Some of these elements (e.g. Ca, Mg, K) are present in mineral matter and can
360 be associated with soil dust that is brought in from outdoors on shoes. It has been shown that
361 resuspended dust contributes to the airborne particulate matter in the “personal cloud” (i.e. personal
362 breathing zone). Resuspension rates of settled dust depend on factors such as amounts accumulated,
363 floor characteristics, walking style, shoe types, humidity and cleaning activities (Rasmussen et al.,
364 2018). Some elements may result from abrasion of metallic components from furniture and other
365 building materials. Cr, for example, is an important alloying element in stainless steel and is also present

366 as chromated copper arsenate (CCA), a preservative for furniture and wood building materials against
367 insect, bacterial, and fungal decline (Rivas et al., 2015; and references therein). P is likely associated
368 with chlorinated phosphorus-based flame retardants and plasticisers, which are ubiquitous in the indoor
369 environment (Araki et al., 2014). Many metals (e.g. Ni and Zn) are also in the list of ingredients of
370 numerous personal care products (Borowska and Brzóška, 2015; Omenka and Adeyi, 2016).

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

373

$$374 \quad EF = (X/Fe)_{\text{air}} / (X/Fe)_{\text{crust}} \quad (3)$$

375

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

391 The emissions from coal burning increase during coldest months due to the use of domestic heating
392 and cooking devices (Blanco-Alegre., 2019). In outdoor environments, many of these elements, in
393 particular Cu, Zn, Pb and Mo, are mostly associated with traffic emissions (Pant and Harrison, 2013).
394 In dwellings and in working and leisure places, in addition to infiltration from outdoors, these
395 constituents can originate from the multiplicity of materials and consumer products used indoors.
396 Attention is drawn to the fact that the concentration of some elements has increased sharply on the day
397 of the student party. Potassium reached a level of 2140 ng m⁻³, while in other occupancy periods
398 concentrations ranged from 305 to 367 ng m⁻³. Cl, Zn and Cu increased from usual levels in the ranges
399 214-396, 21-24 and 2.7-5.7 ng m⁻³ to values of 1870, 61 and 15 ng m⁻³, respectively, on the party day.
400 These elements are abundant constituents of tobacco smoke (Misha et al., 1986), reason why there are
401 suspicions that, although prohibited, this practice may have occurred during the event.

402 The particulate matter organic extracts encompassed several aliphatics, polycyclic aromatic
403 hydrocarbons (PAHs), alcohols, acids, sterols, glycerol derivatives, phenolic compounds, saccharides,
404 among others. Aliphatic compounds included the homologous series of *n*-alkanes from C₁₁ to C₃₂ and
405 some alkenes. These are ubiquitous compounds in the environment with both natural and anthropogenic
406 origin. Alkane homologues were detected at I/O ratios ranging from 1.0 (dodecane) to 54
407 (dotriacontane). The sum of their concentrations was as follows: 49.4 ng m⁻³ (indoor, occupancy), 15.9
408 ng m⁻³ (indoor, night-time), 16.5 ng m⁻³ (outdoor, daytime), and 6.0 ng m⁻³ (indoor, weekend, daytime).
409 The maximum indoor concentration (13.4 ng m⁻³) was observed for dotriacontane. Alkenes (C₁₄, C₁₆,
410 C₁₈, C₂₀ and C₂₃) were present at I/O ratios from 3 to 5 and very low concentrations, peaking in the
411 cafeteria for hexadecene (0.47 ng m⁻³). Excepting hexadecene, alkenes were not detected in samples
412 from non-occupancy periods.

413 Figure 7 depicts the mean concentrations of individual PAHs in the cafeteria and outdoors.
414 Excluding naphthalene and acenaphthene, two volatile PAHs consisting of two benzene rings, the I/O
415 ratios for all other polyaromatics were always < 1. Contrary to many other compounds, the average
416 outdoor concentrations (7.8 ng m⁻³) was 3.3 times higher than those measured indoors (2.4 ng m⁻³).
417 Levels in the cafeteria dropped to 0.49 ng m⁻³ on the weekend, whereas the night-time value (4.3 ng m⁻³)
418 was between the one measured during working hours in the cafeteria and that registered outdoors.
419 Although nocturnal measurements in the ambient air were not carried out in the present study, outdoor
420 concentrations usually higher at night may have contributed to the build-up of PAHs in the cafeteria
421 through infiltration processes. Due to residential heating emissions, decreased atmospheric mixing
422 height and nocturnal atmospheric stability, an appreciable increase in concentrations in the evening is
423 likely to occur. On the other hand, at daytime, after emission by combustion sources (e.g. traffic), PAHs
424 undergo chemical transformations. Better dispersion conditions and atmospheric reactivity during
425 daytime, in general, lead to lower diurnal concentrations compared to nocturnal values. PAH oxidative
426 transformations in ambient air are supposed to be started by gas-phase OH and NO₃ radicals, followed
427 by reactions with NO₂ and O₃ leading to ring-retaining quinones, nitro-PAHs and ring-opened phthalic
428 acids and their degradation products (Alam et al., 2015).

429 The homologous series of fatty alcohols from C₁₀ to C₃₀ was observed in the PM₁₀ samples (Table
430 3). 1-Hexacosanol and 1-octadecanol were the most abundant homologues. Their indoor concentrations
431 were more than 600 times higher than those measured outdoors. These *n*-alkanols are constituents of
432 many household and personal care products. Furthermore, octadecanol was identified as one of the
433 dominant compounds in fine organic aerosols from charbroilers and meat cooking operations (Rogge
434 et al., 1991). The homologous series of alkanolic acids (C₆-C₂₆) and some alkenolic acids were also
435 present. In general, the concentrations recorded during the occupancy period in the cafeteria far
436 exceeded those observed outside. Palmitic, stearic and oleic were the dominant acids, in agreement with
437 previous studies describing cooking emissions (Abdullahi et al., 2013; He et al., 2004; Rogge et al.,
438 1991; Zhao et al., 2007).

439 Some dicarboxylic acids from C₄ to C₁₆, at I/O ratios generally higher than 1, were present in PM₁₀,
440 but at lower concentrations compared to those of fatty acids. He et al. (2004), who also detected diacids
441 in emissions from cooking processes at lower levels in comparison with alkanolic and alkenolic acids,
442 suggested that these compounds are formed by oxidation of dialdehydes during the autoxidation process
443 of unsaturated lipids. Among diacids, azelaic (C₉) was the most abundant in the cafeteria, reaching
444 concentrations up to 67 ng m⁻³. The lab-engineered form of this acid is typically used in skincare
445 products. It is also an abundant compound in emissions from electronic cigarettes, heat-not-burn
446 tobacco products, and conventional cigarettes (Ruprecht et al., 2017). Biomass combustion is also a
447 source of diacids, in addition to emitting keto- (e.g. levulinic) and hydroxy-acids (e.g. glycolic, glyceric
448 and lactic) (Alves et al., 2011). All these acids were not detected in the outdoor samples or were present
449 in lower concentrations when compared to those observed inside the cafeteria. Liu et al. (2017)
450 measured a wide variety of carboxylic acids in a university classroom, indoors and outdoors, including
451 monoacids, diacids, hydroxy acids, carbonyl acids, and aromatic acids. Besides indoor sources such as
452 off-gassing of building materials, evidence for acid formation from indoor chemical reactions with
453 ozone was noticed. It was also suggested that human occupants may contribute to the abundance of
454 carboxylic acids either through direct emissions or surface reactions. Although generally present at very
455 low concentrations indoors, ozone may react with unsaturated organic compounds present in skin and
456 other materials. As observed in the university classroom, the elevated indoor concentrations of lactic
457 acid in the cafeteria during the occupancy period was likely human perspiration. A recent study
458 conducted in an art museum revealed the unexpected build-up of lactic acid from sweat (Pagonis et al.,
459 2019). It was found that this acid leaves the skin, travels through the air, and sticks to the walls and
460 other surfaces at high rates. It should be also noted that lactic and other short chain acids (levulinic,
461 glycolic, etc.) are frequently used in exfoliants, anti-wrinkle products and other cosmetics. Thus, both
462 the emission and partitioning mechanisms between the gas and the particulate phases of these and many
463 other compounds are complex and still far from being fully understood.

464 Some resin acids were detected as PM₁₀-bound organic components: dehydroabietic, isopimaric,
465 abietic and podocarpic. Among these, dehydroabietic acid dominated. Its concentrations outside the
466 cafeteria exceeded those observed indoors, pointing to the residential burning of coniferous wood
467 (Vicente and Alves, 2018). Although it is an unlikely source in the cafeteria, it should be noted that
468 very high concentrations of free resin acids were detected in depilatory wax strips (Nilsson et al., 2008).

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

476 Many other monosaccharides, disaccharides, polysaccharides and polyols, were detected in PM₁₀.
477 However, due to structural isomerism and identical fragmentation patterns in the mass spectra, their
478 individual quantification was not possible. Thus, a mean response factor of the calibrated compounds
479 with the same functional groups was applied. A much higher indoor concentration for these unidentified
480 saccharides and a strong nocturnal decay was observed, suggesting that many of these compounds were
481 produced by the day-to-day activities of the cafeteria.

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

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

492 Cholesterol and phytosterols were abundant compounds in PM₁₀. Concentrations in the cafeteria
493 during the occupancy period far exceeded those recorded outdoors. Cholesterol (C₂₇) is biosynthesised
494 by higher animals and found in body tissues, especially in oils and fats. Phytosterols, such as β -sitosterol
495 and stigmasterol (C₂₉), are present in plant lipid waxes and membranes. All these sterols were previously
496 observed in PM_{2.5} emitted from Chinese cooking (He et al., 2004; Zhao et al, 2007). However,
497 cholesterol is the only sterol that have been detected in meat cooking fumes (Rogge et al., 1991).

498 In the present study, many unusual compounds were detected in indoor aerosols, as far as we know,
499 for the first time. Some of the substances are part of the list of ingredients of various cosmetics. Ethylene
500 brassylate, for example, which was only present in indoor samples, is a macrocyclic compound mainly
501 used as a fragrance ingredient for its musk-like odour. Parsol MCX (ethylhexyl methoxycinnamate) is
502 used in the formulation of a wide variety of brands, including sunscreens and makeup products that
503 contain ingredients to protect the skin from the sun. A mean I/O ratio of 122 suggests its emission from
504 personal care products and concentration build-up in confined spaces, such as the cafeteria. Dibutyl
505 adipate is the diester of butyl alcohol and adipic acid. In cosmetics and personal care products, it is used
506 in nail polish and skin care products. Once again, much higher concentrations indoors than outdoors
507 (I/O=23) indicate a possible origin in the products used by the occupants. Because of its floral fragrance,
508 methyl dihydrojasmonate is an additive in a wide range of cleaning and personal care products. It was
509 absent from outdoor samples, but the indoor concentrations did not show appreciable differences
510 between occupancy and non-occupancy periods, suggesting the presence of a permanent source. Acetyl
511 tributyl citrate is an organic compound that is employed as a solvent in paints, inks, and nail enamel. It
512 may also be found in eye makeup and aerosol hair sprays. Moreover, it is used to make plastics more

513 flexible (e.g. plasticisers in packaging films for food), added as a flavour ingredient in non-alcoholic
514 beverages and applied in the manufacture of many pharmaceutical drugs. Acetyl tributyl citrate was
515 only detected in indoor aerosols. Pyroglutamic acid, known either as pyrrolidone-5-carboxylic acid, 5-
516 oxoproline, PCA or pidolic acid, is a ubiquitous natural amino acid derivative. It is used for dry skin
517 and hair products, owing to its humectant characteristics. Pidolic acid is found in large quantities in skin
518 and brain tissues. It is sold online as a nootropic dietary supplement to improve cognitive and executive
519 functions, and memory. The presence of this acid was only observed indoors, and at much higher
520 concentrations during the occupancy periods.

521 2-Propanol-1-chloro-phosphate, also known as Amgard TMCP or Hostaflam OP 820, is a chlorine
522 containing alkyl phosphate ester. It is mainly employed as an additive flame retardant, especially in
523 polyurethane foam, with some minor use in other plastics and as a back coating in textiles. It was only
524 present in indoor samples collected during daytime, including weekend. This suggests that the emission
525 of this compound can be promoted by sunlight, although its formation processes and detection in the
526 particulate phase deserve further investigation.

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

533 Some psychoactive substances, including drugs of abuse (e.g. tetrahydrocannabinol, THC), were
534 found in the particulate matter samples. THC is the principal psychoactive constituent of cannabis. THC
535 was only present in two indoor samples collected during the occupancy periods, reaching 21.7 ng m^{-3}
536 on Thursday, the day of the student party. THC was previously detected as a constituent of outdoor
537 airborne particles in 6 Spanish urban locations at concentrations ranging from 23 to 44 pg m^{-3} (Viana et
538 al., 2012). Another psychoactive substance in airborne particles was caffeine. However, this compound
539 was only detected in outdoor samples and during non-occupancy periods. Since caffeine is expected to
540 partition between the gas and the particulate matter (Viana et al., 2012), its absence during working
541 hours may be related to the higher temperatures in the cafeteria because of the use of heating. In the
542 outdoor air across Italy, caffeine reached gross average concentrations of $15 \pm 8 \text{ ng m}^{-3}$ in winter and
543 $1.2 \pm 0.6 \text{ ng m}^{-3}$ in summer, while in the metropolitan area of Rome the corresponding levels were
544 $1.5 \pm 0.5 \text{ ng m}^{-3}$ and $0.3 \pm 0.1 \text{ ng m}^{-3}$ (Viana et al., 2012; and references therein).

545 Nicotinic acid, also denominated niacin or vitamin B3, is present in all living cells and found in
546 whole and processed foods. It is used to make other chemicals and added to feeds and flours as a dietary
547 supplement. It is also used as a medication to treat dyslipidaemic states. Niacin was not detected in
548 outdoor air but reached concentrations up to 16.1 ng m^{-3} in the cafeteria during the occupancy period.

549 The mechanisms by which it is formed and remains airborne in the particulate phase are yet to be
550 clarified.

551 Irgafos 168 (tris(2,4-di-tert-butylphenyl)phosphite), as one of the common antioxidants, is
552 extensively used in polyolefin materials (e.g. polypropylene employed in food packaging), to protect
553 polymers from aging and oxidation. However, irgafos 168 may degrade into several products, such as
554 2,4-di-tert-butylphenol (DP1), and tris(2,4-di-tert-butylphenyl)phosphate (DP2), which is the oxidised
555 form of irgafos 168 (Yan et al., 2018). In the present study, whilst DP1 was detected at 5-fold higher
556 concentrations in the cafeteria than outside, DP2 was only present in two daytime indoor samples (both
557 with 19 ng m⁻³) and in just one outdoor sample (39 ng m⁻³).

558

559 3.2.3. Health risk analysis

560

561 Noncarcinogenic and carcinogenic risks associated with inhalation exposure to trace elements in
562 indoor PM₁₀ by the employees were estimated following the methodology proposed by the United States
563 Environmental Protection Agency (USEPA, 1989, 2009):

564

$$565 \text{THQ} = (\text{EF} \times \text{ED} \times \text{ET} \times \text{C}) / (\text{RfC} \times \text{AT}) \quad (6)$$

$$566 \text{TR} = (\text{EF} \times \text{ED} \times \text{ET} \times \text{C} \times \text{IUR}) / \text{AT} \quad (7)$$

567

568 The target hazard quotient (THQ) and target carcinogenic risk (TR) are dimensionless, EF is the
569 occupational exposure frequency (250 days per year, i.e. 5 days per 50 weeks), ED is the exposure
570 duration (25 years), ET is the exposure time (8 h per day), C is the metal concentration in the air (mg
571 m⁻³), and AT is the averaging time (365 days per year × 24 h per day × ED for noncarcinogenic effects
572 and 612,000 h, i.e., 70 years × 365 days per year × 24 h per day, for carcinogenic effects). RfC is the
573 USEPA reference concentration (mg m⁻³). Considering that for some elements, reference doses for oral
574 exposure (RfD, mg kg⁻¹ day⁻¹) are available instead of RfC values (USEPA, 2017, 2019), these latter
575 were derived using the following equation (USEPA, 2013):

576

$$577 \text{RfC} = (\text{RfD} \times \text{BW}) / \text{IR} \quad (8)$$

578

579 where IR and BW are the inhalation rate and body weight of an adult (20 m³ day⁻¹ and 70 kg). Chronic
580 inhalation unit risk (IUR) values for the carcinogenic elements found in the PM₁₀ samples were taken
581 from USEPA (2017): arsenic 4.3×10⁻³ (μg m⁻³)⁻¹, lead 1.2×10⁻⁵ (μg m⁻³)⁻¹, hexavalent chromium
582 8.4×10⁻² (μg m⁻³)⁻¹ and Ni oxide 2.6 × 10⁻⁵ (μg m⁻³)⁻¹. The IUR of Cr(VI) is based on a Cr(III):Cr(VI)
583 proportion of 1/6. Since in the present study total Cr was determined, one seventh of the concentration
584 obtained was used to estimate the risk.

585 The noncarcinogenic risks associated with inhalation exposure by employees to particulate trace
586 elements in the indoor air during the occupancy period are presented in Figure 8. A THQ < 1 indicates
587 no significant (acceptable) risk, a THQ > 1 suggests that the noncarcinogenic effect is likely to manifest,
588 whereas a high chronic risk is evident when THQ > 10 (Slezakova et al., 2014). The estimated mean
589 THQ for PM₁₀-bound trace elements ranged from null values for many elements 0.92 for Cl. The total
590 mean THQ of all eleven elements in PM₁₀ was 1.7. Cl and Mn were the most important contributors (55
591 and 38 %, respectively) to the total noncarcinogenic risk, followed by Si (6.0 %), and Al (1.3 %). If the
592 exposure time and frequency assumed for the working hours were considered for non-occupancy
593 periods and for outdoor air, ΣTHQ values of 0.17-0.26 and around 2.0 would be obtained, respectively.

594 For carcinogenic constituents, USEPA considers that targeting a 10⁻⁶ risk level for individual
595 chemicals and pathways will usually lead to negligible risks. However, caution is suggested to guarantee
596 that the cumulative cancer risk for all potential carcinogenic contaminants does not have a residual
597 cancer risk above 10⁻⁴ (Slezakova et al., 2014). In the present study, ΣTR was always < 2×10⁻⁶,
598 suggesting that the risk associated with the exposure to PM₁₀-bound carcinogenic elements is negligible.

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

603

$$604 \quad EC = (CA \times ET \times EF \times ED) / AT \quad (9)$$

605

606 where CA represent the benzo[a]pyrene equivalent concentrations (ng m⁻³). The inhalation unit risk
607 (IUR) of respiratory cancer for BaP_{eq} is 3.9 × 10⁻⁶ (ng kg day)⁻¹. This value is obtained multiplying the
608 inhalation cancer risk unit factor of BaP (1.1 × 10⁻⁶ (ng kg day)⁻¹) by the inhalation rate (20 m³ per day)
609 and dividing by the body weight (70 kg). The carcinogenic risk of the PAH mixture is then calculated
610 using the following equation:

611

$$612 \quad Risk = IUR \times EC \quad (10)$$

613

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

620

621 **4. Conclusions**

622

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

640 The total mean hazard quotient that represents the noncarcinogenic effects due to the inhalation of
641 particle-bound metals during the occupancy period in the cafeteria was higher than the acceptable level
642 (>1). The cumulative cancer risk for both carcinogenic metals and PAHs was lower than the acceptable
643 level (10⁻⁴). However, exposure to metals and PAHs occurs also via ingestion and dermal contact and
644 if these routes are considered, then the estimated risks might be higher.

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

654

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656

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674

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676

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Table 1Weekly concentrations of volatile organic compounds ($\mu\text{g m}^{-3}$) 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 2Minimum, maximum and average concentrations of elements (ng m^{-3}), 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	
Indoor Occupancy	Day																											
	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	
	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	
	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	
	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	
Indoor Night	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	-	-	-	-	-	-	
	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	
	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	
Outdoor Day	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	
	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	
	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	
	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	
Indoor Non-oc. Day	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	-	
	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	-	
I/O		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 3Concentrations (ng m⁻³) of oxygenated organic compounds in PM₁₀

	Indoor occupancy	Indoor non-occupancy (night-time)	Indoor non-occupancy (Sunday, daytime)	Outdoor daytime	I/O (occupancy)
<i>Saccharides</i>					
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
<i>Phenolics and alteration products</i>					
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±0.38	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±0.22	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
<i>Aliphatic alcohols</i>					
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
<i>Triterpenoid and steroid compounds</i>					
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
β-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
<i>Aliphatic acids</i>					
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±0.40	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	bdl	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
<i>Resin acids</i>					
Dehydroabiatic	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
<i>Other acids</i>					
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±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
<i>Glycerol derivatives</i>					
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
<i>Other compounds</i>					
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±0.40	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	-	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
Δ-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
<i>Plasticisers</i>					

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±0.008	bdl	bdl	0.05±0.10	0.08
Bis(2-ethylhexyl) adipate	0.006±0.009	bdl	bdl	0.09±0.19	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

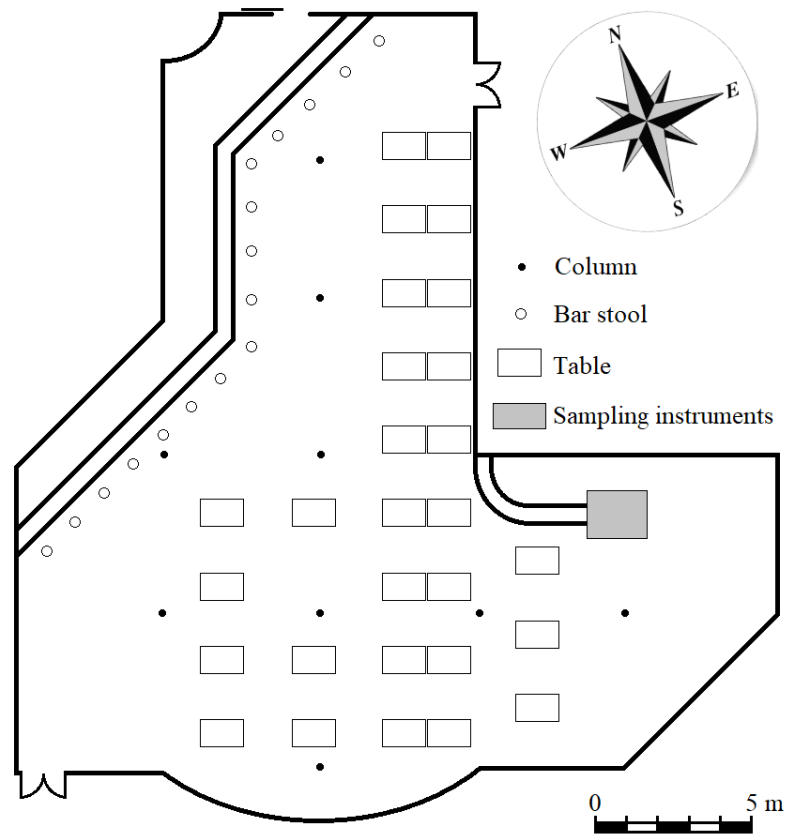


Fig 1. Floor plan of the cafeteria. Each table has four chairs.

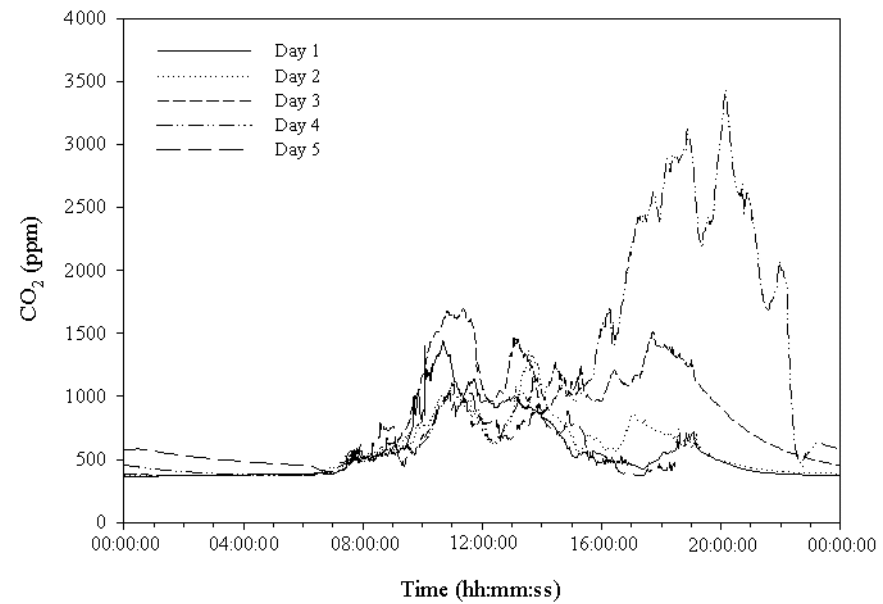
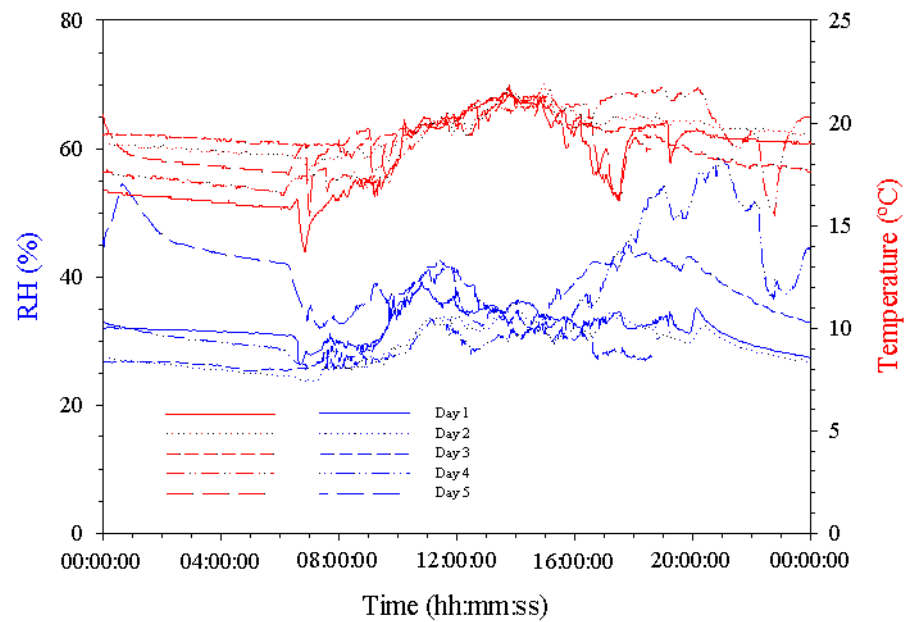


Fig 2. Variation of relative humidity, temperature and carbon dioxide levels on weekdays.

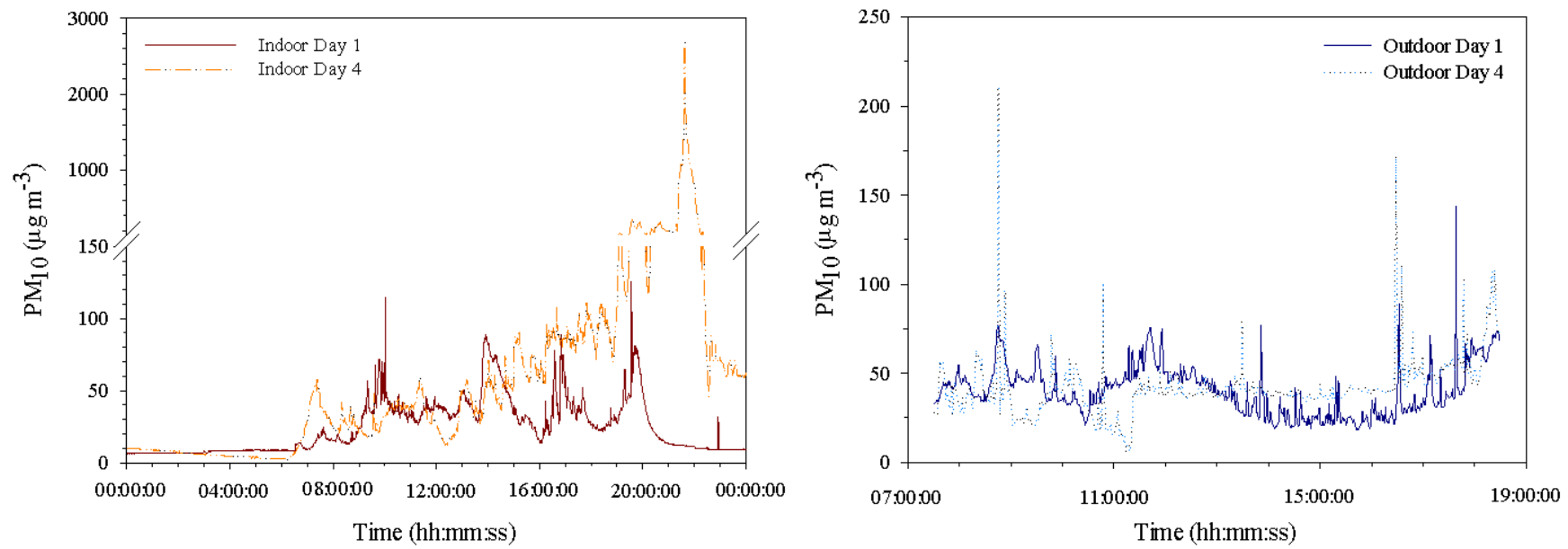


Fig. 3. Examples of PM₁₀ daily concentration profiles in the cafeteria and outdoors (Day 1 – Monday, Day 4 – Thursday).

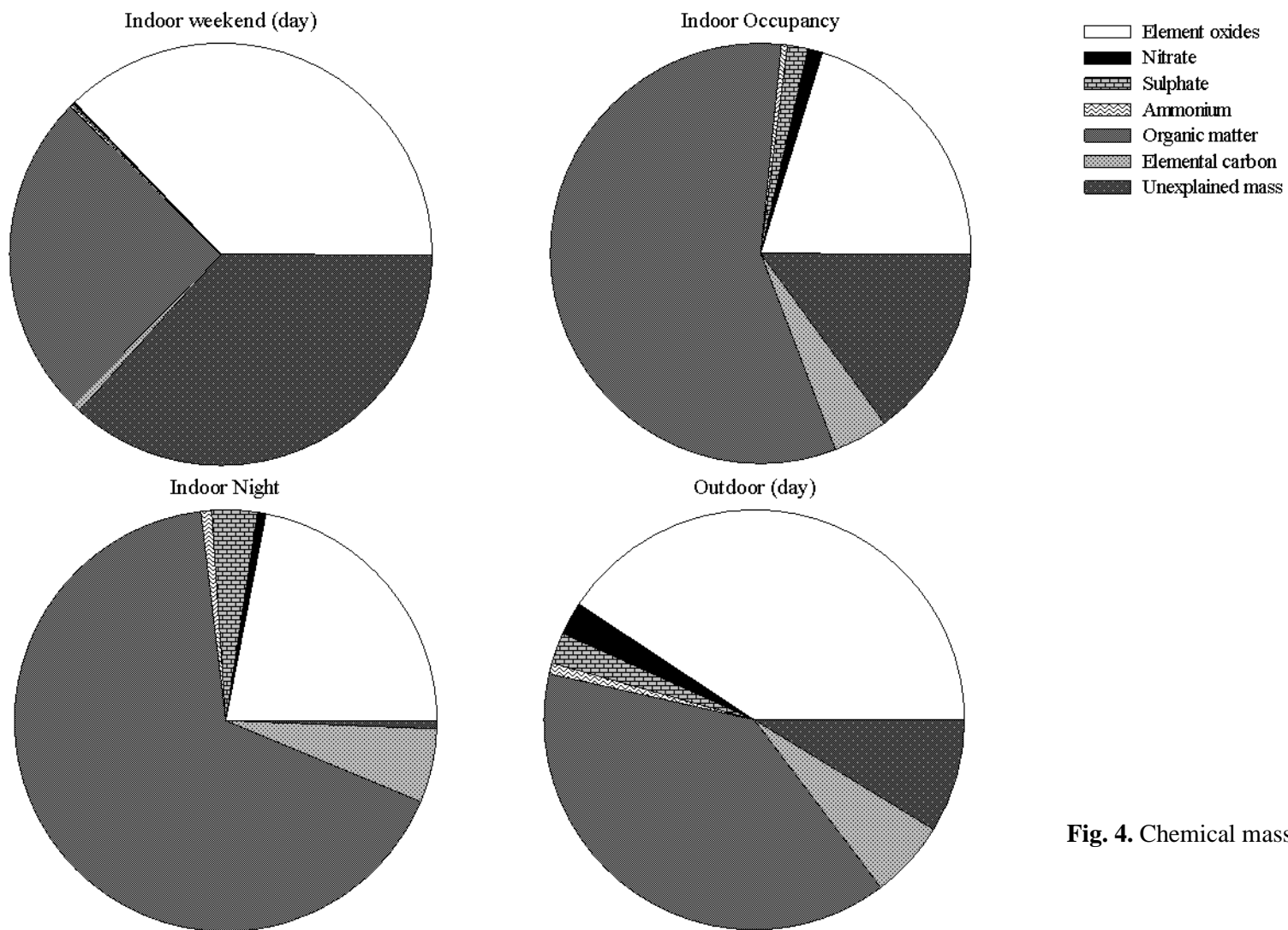


Fig. 4. Chemical mass closure of PM₁₀.

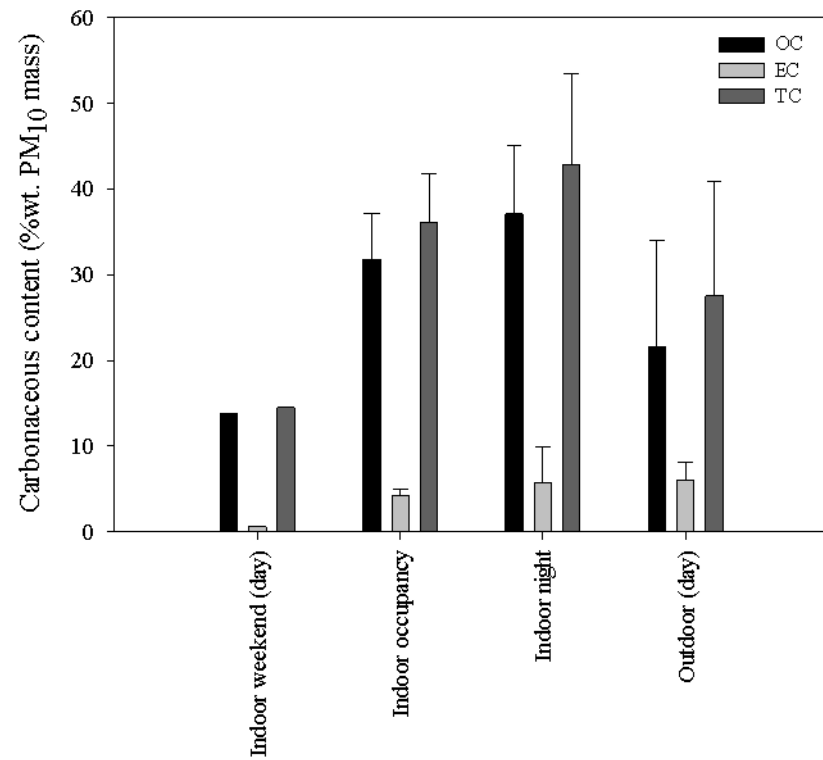
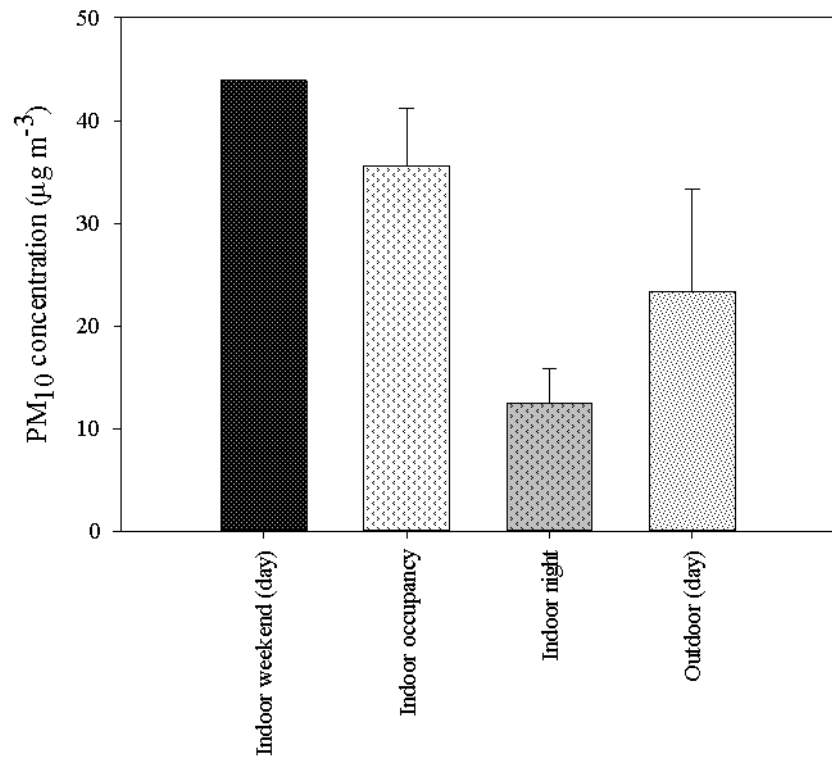


Fig 5. Means (\pm SD) of PM₁₀ concentrations and mass fractions of organic carbon, elemental carbon and total carbon (TC = OC + EC)

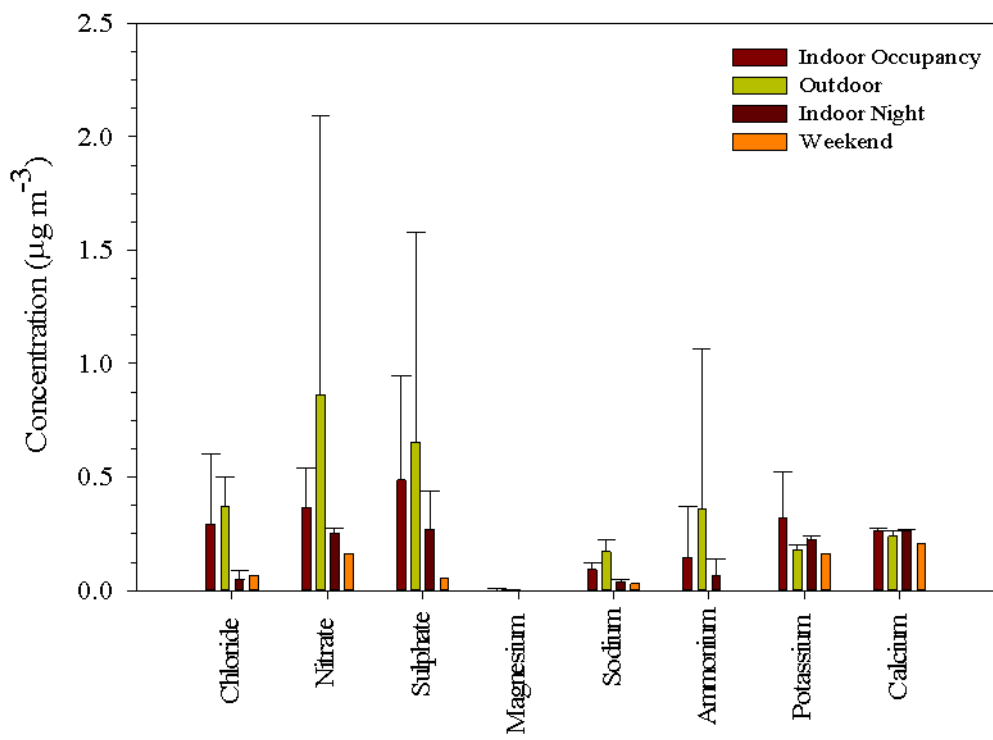


Fig. 6. Concentrations (mean \pm SD) of water-soluble ions in PM₁₀ collected indoors and outdoors.

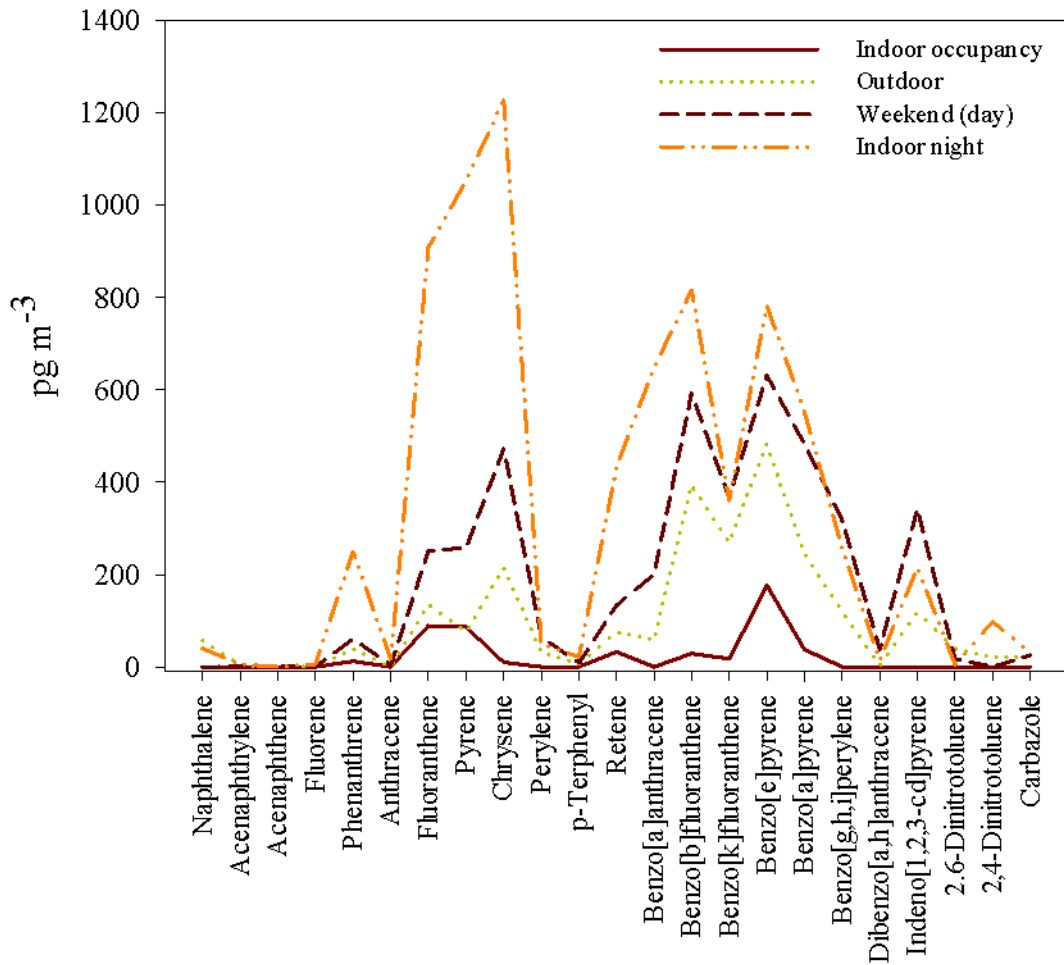


Fig. 7. Average concentrations of polycyclic aromatic hydrocarbons.

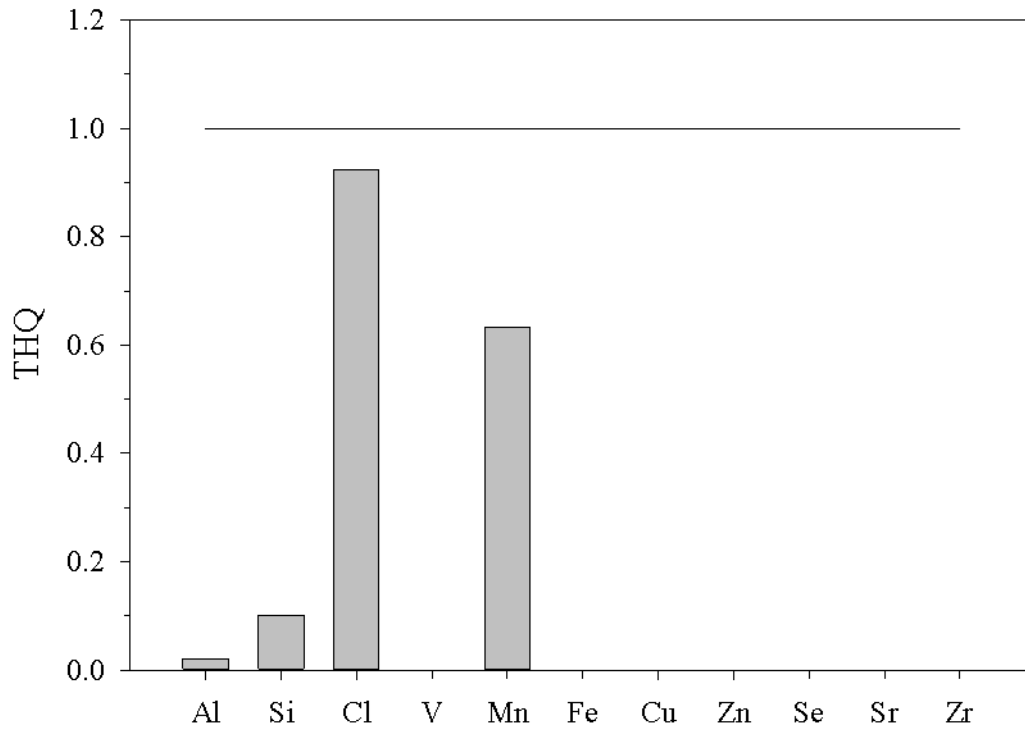


Fig. 8. Mean risks from inhalation exposure to noncarcinogenic PM₁₀-bound metals for the cafeteria workers during the occupancy period. The horizontal line represents the USEPA health-based guideline level.