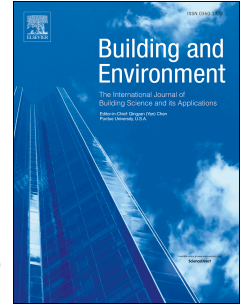


# Journal Pre-proof

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# Impact of vacuum cleaning on indoor air quality

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## Abstract

Vacuum cleaning can be a household source of particulate matter (PM) both from the vacuum motor and from settled dust resuspension. Despite the evidence of this contribution to PM levels indoors, the effect of this source on PM composition is still unknown. In this study, four vacuum cleaners (washable filter bag-less, wet, bagged and HEPA filter equipped robot) were tested for the emission rate of particulate mass and number. The detailed PM chemical characterisation included organic and elemental carbon, metals and organic speciation. PM<sub>10</sub> emission rates from bagged vacuum operation were much higher ( $207 \pm 99.0 \mu\text{g min}^{-1}$ ) compared with the ones obtained from wet ( $86.1 \pm 16.9 \mu\text{g min}^{-1}$ ) and washable filter bag less vacuums ( $75.4 \pm 7.89 \mu\text{g min}^{-1}$ ). Particle (8 to 322 nm) number emission rates ranged from  $5.29 \times 10^{11}$  (washable filter bag less vacuum) to  $21.2 \times 10^{11}$  (wet vacuum) particles min<sup>-1</sup>. Ratios of peak to background levels indicate that vacuuming can elevate the ultrafine particle number concentrations by a factor ranging from 4 to 61. No increase in PM mass or number concentrations was observed during the HEPA filter equipped vacuum operation. The increase in copper and elemental carbon PM<sub>10</sub> contents during vacuuming suggested motor emissions. Organic compounds in PM<sub>10</sub> included alkanes, PAHs, saccharides, phenolics, alcohols, acids, among others. However, it was not possible to establish a relationship between these compounds and vacuuming due to the vast array of possible household sources. The cancer risks associated with metals and PAH inhalation were negligible.

**Keywords:** Vacuum cleaners, Indoor air quality, Particles, Elemental composition, OC/EC, Organic compounds.

## 1. Introduction

People spend more than 90% of their daily life in indoor environments [1–3] and, for this reason, personal exposure to pollutants in these microenvironments is of great concern. Due to the

39 susceptibility of children and elderly to air pollution, numerous studies have been conducted to  
40 assess indoor air quality in schools [4–7], children [8–11] and elderly day care centres [12–14].  
41 Despite the importance of the above-mentioned microenvironments, most of people’s time is spent  
42 at home [1–3,15].

43 Indoors, particulate matter (PM) is one of the biggest health hazards [16]. Particulate matter is a  
44 heterogeneous mixture of different chemical components and physical characteristics, which are  
45 responsible for diverse health effects [17].

46 Household activities, such as cooking, smoking, hair spraying/drying, candle/incense burning or  
47 vacuuming, have been reported to generate considerable amounts of particulate matter indoors [18–  
48 20], which may have a strong influence on short-term exposure [21]. Isaxon et al. [18] evaluated  
49 the influence of household activities in 22 homes in Sweden on indoor airborne particles (number  
50 concentration and black carbon). The authors reported that despite the transient nature of indoor  
51 sources, they rapidly generate particulate peak concentrations. He et al. [20] quantified the effect of  
52 20 different household activities on indoor particle mass and number concentrations. The authors  
53 reported that depending on the type of source and housing characteristics, indoor particles  
54 increased distinctively. The influence of nine specific sources on particulate matter number size  
55 distribution and mass concentration was evaluated individually in an empty laboratory by Glytsos  
56 et al. [19]. High particle number concentrations during activation of the distinct sources and a great  
57 influence of the source type on particle number size distributions were observed. Studies carried  
58 out to assess household sources of PM reported that vacuuming can significantly elevate indoor PM  
59 concentrations [22–24] with a very high total lung deposition fraction by number [25].

60 According to a survey on time use patterns in Europe for woman and men aged 20 to 74, and across  
61 the whole year, cleaning and upkeep activities are among the most time consuming tasks,  
62 representing 13 to 28 % of the total time spent on domestic work [26]. An online survey (covering  
63 23 countries), aiming at assessing household’s cleaning habits and preferences, revealed that 33 %  
64 of respondents vacuum 2-5 times per week, while 46 % spend 1-2 hours vacuuming [27].

65 Some studies reported in the literature were focused on the operation of vacuum cleaners and their  
66 impacts on particle mass and number levels, both in laboratory chambers [21,28–31] and under real  
67 life conditions [18,20,32]. Additionally, a number of studies also included bioaerosol levels  
68 associated with vacuuming [28,30,31]. Although many studies have investigated particulate mass  
69 and number emissions during vacuum cleaning operations, an important gap in knowledge still  
70 exists with respect to the chemical characteristics of the released particles. The characterisation of  
71 the chemical composition of particles arising from specific indoor sources is of great interest due to  
72 the risk associated with specific PM components and the possibility of using certain compounds as  
73 tracers for source apportionment in indoor environments [33,34]. Regarding PM characterisation,  
74 Szymczak et al. [35] reported ultrafine particles from a commercial professional vacuum motor  
75 consisting almost entirely of copper. Vu et al. [25] suggested that particles released from the

76 vacuum cleaner motor were possibly carbon internal void aggregates. The authors' hypothesis was  
77 based on the finding that particles generated from vacuum cleaning were found to be nearly  
78 hydrophobic with an average growth factor around 0.98–1.10 for particle sizes of 50 and 100 nm.  
79 Isaxon et al. [18] reported an increase in black carbon levels during vacuum cleaning.

80 Despite the significant data provided by these and other studies, the impact of this source on  
81 household air quality is still uncertain due to the variability and complexity of vacuum cleaning and  
82 limited on-site experiments. Studies conducted in laboratory allow obtaining reproducible  
83 measurements with greater control of relevant factors that might influence the results and, thus,  
84 they can serve as a reference. However, particulate emission rates measured in laboratory chambers  
85 may substantially differ from those obtained in households since settled dust resuspension is not  
86 considered [36] and neither are the differences in dust loads in residential settings [20]. On the  
87 other hand, measurements conducted under real life conditions, in which concentration data is  
88 crossed with daily activity logs, can introduce some recall bias and misreporting.

89 The aim of the present study was to evaluate the impact of commercial vacuums on short-term  
90 particulate matter mass and number concentrations in indoor air. Since particle inhalation during  
91 vacuuming may adversely affect households, a detailed chemical characterisation of particulate  
92 matter was performed, which was the basis for a carcinogenic and noncarcinogenic risk  
93 assessment. The tests were carried out in a household under controlled conditions with respect to  
94 ventilation patterns and concurrent source events.

95

## 96 **2. Materials and methods**

### 97 **2.1. Sampling sites and strategy**

98 Three cylinder vacuum cleaners (washable filter bag less vacuum, wet vacuum, bagged vacuum)  
99 and a HEPA filter equipped robot were temporarily borrowed from Spanish homeowners for  
100 testing (Table 1). These devices were selected because cylinder vacuum cleaners are the prevalent  
101 type in the EU with a market share of 68% in 2016, whilst robotic cleaners have shown an  
102 increasing sales trend [37]. Measurements were performed in the living room (volume = 91.9 m<sup>3</sup>)  
103 of a suburban Spanish house in León from October to November 2017. Similarly to the approach  
104 described by Vu et al. [25], Wu et al. [36] and Corsi et al [32], during the monitoring campaign  
105 there were no other activities in the house and the measurements were carried out in a closed room  
106 (all the doors and windows were closed) to achieve minimum ventilation conditions. Ventilation  
107 rates, estimated by the CO<sub>2</sub> concentration decay method as described by Alves et al. [38], ranged  
108 between 0.24 and 0.62 h<sup>-1</sup>. The average estimates of ventilation rates are presented in Table 1. On  
109 average, 45 min measurements were conducted during vacuum cleaning. Only the person  
110 responsible for carrying out the activity was present in the room during the experiments. The living  
111 room tiled floor and rugs (two cut pile carpet/rug and one long threads shag rug) were vacuumed

112 twice with each vacuum cleaner at least one week apart. After the household activity ceased, the  
113 room was kept completely empty and closed until the restoration of particle concentration to the  
114 original level. Background indoor air measurements were also performed in the living room during  
115 which no activities were conducted in the house. The temperature (accuracy  $\pm 0.5^\circ\text{C}$ ), relative  
116 humidity (accuracy  $\pm 3.0\%$  with probe at  $25^\circ\text{C}$ ) and  $\text{CO}_2$  (accuracy  $\pm 3.0\%$  of reading with probe at  
117  $25^\circ\text{C}$ ) were continuously monitored with an indoor air quality probe (TSI, model 7545).  
118 Temperature and relative humidity ranged between  $19.7$  and  $26.3^\circ\text{C}$  and between  $30.6$  and  $45.7\%$ ,  
119 respectively, for the whole set of measurements (Table 1).

120 Real time size segregated particulate concentrations ( $\text{PM}_1$ ,  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ ) were recorded using a  
121 DustTrak monitor (TSI, DRX 8533). Real time particle size distributions and number  
122 concentrations in the range from  $8$  to  $322$  nm were measured using a Scanning Mobility Particle  
123 Spectrometer (SMPS, TSI Incorporated). The SMPS consists of an electrostatic classifier (TSI,  
124 Model 3071) and a condensation particle counter (TSI, Model 3022). The aerosol was sampled  
125 through polyethylene tubing. All reported data has been corrected for diffusion losses using  
126 equations described in Kulkarni et al. [39] for small particles and impaction/settling losses for larger  
127 particles as a function of size [40].

128 Simultaneous sampling with a  $\text{PM}_{10}$  high volume air (MCV, model CAV-A/mb) instrument was  
129 carried out. The equipment was operated at a flow of  $30\text{ m}^3\text{ h}^{-1}$ . Particulate samples were collected  
130 on pre-weighed  $150$  mm quartz fibre filters (Pallflex®).  $\text{PM}_{10}$  samples were also collected into  $47$   
131 mm Teflon filters using a low volume sampler (Echo TCR, Tecora) working at  $2.3\text{ m}^3\text{ h}^{-1}$ . To  
132 ensure the reliability of the measurements, the sampling devices were calibrated prior to sampling  
133 and maintenance was performed in a regular basis. The gravimetric quantification was performed  
134 following the specifications described in EN 12341:2014 [41], with a microbalance (XPE105  
135 DeltaRange®, Mettler Toledo, readability of  $0.01$  mg). The particulate mass was obtained from the  
136 average of six consecutive measurements (relative standard deviation  $< 0.02\%$ ), after conditioning  
137 the filters for  $24$  h in the weighing room. The high and low volume samplers and the real time  
138 monitoring instruments were placed in the middle of the room at a height of about  $1.5$  m [42].

139

## 140 **2.2. Analytical techniques**

141 The carbonaceous content in the  $\text{PM}_{10}$  samples (quartz filters) was analysed by a thermal optical  
142 transmission technique. The method includes controlled heating steps under inert ( $\text{N}_2$ ) and  
143 oxidising ( $\text{N}_2$  with  $4\%$  of  $\text{O}_2$ ) atmospheres. The carbonaceous content of the sample can be divided  
144 into organic carbon (OC), pyrolysed carbon (PC) and elemental carbon (EC). PC, which is  
145 produced from organic carbon during heating under inert atmosphere, was determined measuring  
146 the filter light transmittance through a laser beam and a photodetector. The OC/EC determination is  
147 based on the quantification of the  $\text{CO}_2$  released by a non-dispersive infrared (NDIR) analyser. The

148 latter was daily calibrated with standard CO<sub>2</sub> cylinders and the recovery was periodically verified  
149 by analysing filters impregnated with known amounts of potassium phthalate.

150 After weighing, Teflon filters were analysed by proton-induced X-ray emission (PIXE) to detect  
151 elements with atomic number above 10. Measurements were performed at the PIXE set-up fully  
152 dedicated to aerosol samples [43] at the 3 MV Tandetron accelerator of the INFN-LABEC  
153 laboratory, exploiting a 3 MeV proton beam. Further insight in the methods may be found in  
154 Lucarelli et al. [44].

155 Two 47 mm diameter punches of each quartz filter were extracted first with dichloromethane and  
156 then with methanol. The total organic extracts were fractionated by flash chromatography using  
157 eluents of increasing polarity through an activated silica-gel column. Four different fractions  
158 resulted from this process: (i) aliphatics, (ii) polycyclic aromatic hydrocarbons, (iii) n-alkanols,  
159 phenols, sterols and other hydroxyl compounds and (iv) acids and sugars. The different organic  
160 fractions were concentrated and dried by a gentle nitrogen stream before analysis. Extracts (i) and  
161 (ii) were analysed in a gas chromatograph-mass spectrometer (GC-MS) from Shimadzu. Extracts  
162 with oxygenated compounds were analysed in a GC-MS from Thermo Scientific. These latter  
163 fractions (iii and iv) include polar compounds, which require derivatisation before analysis. N,O-  
164 bis(trimethylsilyl)trifluoroacetamide (BSTFA): trimethylchlorosilane (TMCS) 99:1 (Supelco  
165 33149-U) was used as silylation reagent. The GC-MS calibrations were performed with injection of  
166 about 150 authentic standards (Sigma-Aldrich) at least at four different concentration levels.  
167 Standards and samples were both co-injected with internal standards: tetracosane-d50 and 1-  
168 chlorohexadecane. Additionally, for PAHs determination, a mixture of six deuterated compounds  
169 (1, 4-dichlorobenzene-d4, naphthalene- d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12,  
170 perylene-d12), was used. The organic extracts were injected in the single ion monitoring and total  
171 ion chromatogram modes and the compound identification was based on comparison of the mass  
172 spectra with the Wiley and NIST mass spectral libraries, comparison with authentic standards and  
173 analysis of fragmentation patterns[45]. A description of recovery efficiency tests for several  
174 compounds can be found in Oliveira et al. [46]. Field blanks were used to account for artefacts  
175 associated with transport, handling, and storage of filters, as described in the EN 12341:2014 [41].  
176 These filters were analysed in the same way as samples and the data obtained was subtracted from  
177 the samples in order to obtain corrected results.

178

### 179 **3. Results**

#### 180 **3.1. Particulate matter**

##### 181 **3.1.1. Mass concentrations**

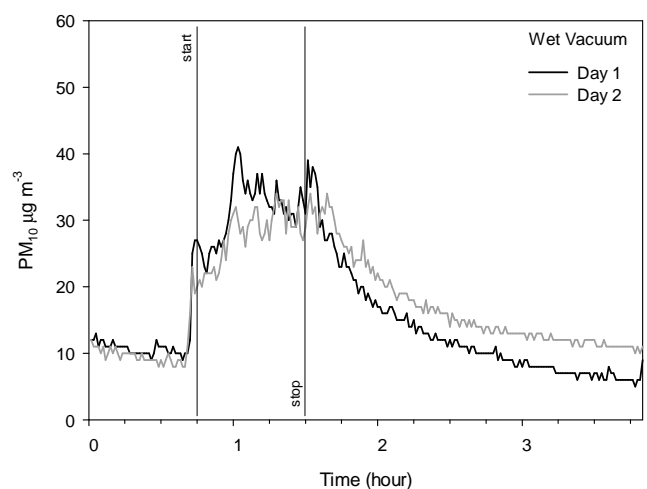
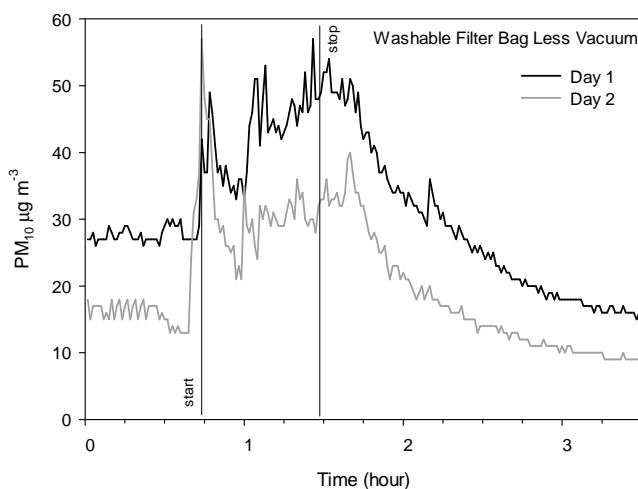
182 Figure 1 depicts the time resolved PM<sub>10</sub> mass concentrations during the operation of the four  
183 vacuum cleaners. An increase in PM<sub>10</sub> mass concentrations was observed close after the activation

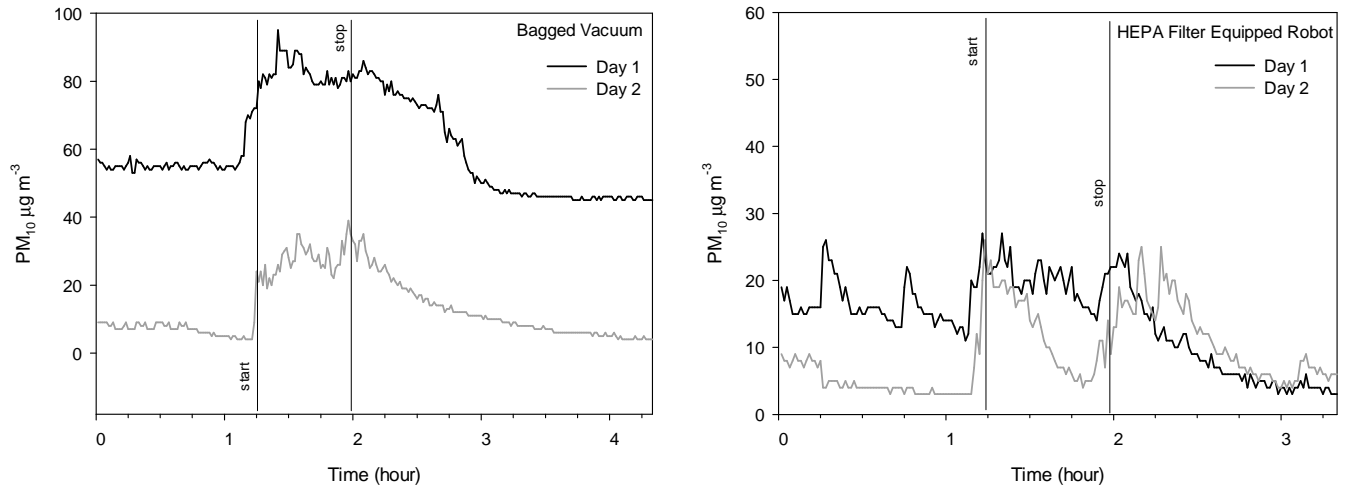


184 of the source. On average, increases over  $PM_{10}$  initial concentrations (before the activation of the  
 185 source) of  $1.61 \pm 0.636$ ,  $1.48 \pm 0.323$ ,  $1.22 \pm 0.035 \mu\text{g m}^{-3}$  were observed during the operation of  
 186 the bagged, wet and washable filter bag less vacuums, respectively. No increase in  $PM_{10}$  mass  
 187 concentrations was experienced during the operation with the HEPA filter vacuum cleaner. The  
 188 increase in PM concentrations during vacuuming is determined by the dust collection efficiency,  
 189 filtration elements employed and degree of reentrainment of already collected particles [47]. The  
 190 reentrainment of collected dust particles was found by Trakumas et al. [47] to be higher for  
 191 cyclonic and wet collectors. However, the authors highlighted that filter bag collectors also  
 192 reemitted particles after being loaded, depending on the particulate load and the type of filter  
 193 material used in the bag.

194 The ratios of peak to background values for  $PM_{10}$  concentrations indicate that vacuum cleaning  
 195 operations can elevate the indoor levels by a factor ranging from over 1.5 to over 2.5. Raaschou-  
 196 Nielson et al. [48] reported an increase in indoor  $PM_{2.5}$  by a factor of 1.3 in Danish infants'  
 197 bedrooms during vacuum cleaning. Fine particles dominate the  $PM_{10}$  mass as indicated by  
 198  $PM_{2.5}/PM_{10}$  and  $PM_1/PM_{10}$  ratios ranging from 0.74 to 0.81 and from 0.72 to 0.79 (except for the  
 199 HEPA filter equipped robot), respectively (Table 1). Despite the predominance of finer particles,  
 200 coarser particles were also recorded during the vacuuming tests. These coarser particles may result  
 201 from resuspension caused by direct contact of vacuum cleaner components with flooring and also  
 202 by the action of walking during vacuuming [32]. Corsi et al. [32] reported significant  $PM_{10}$  mass  
 203 resuspension during vacuuming with a mean time-averaged  $PM_{10}$  increase over  $17 \mu\text{g m}^{-3}$  above  
 204 background levels. Fine particle emissions during vacuum cleaning have been associated with  
 205 mechanical abrasion of the vacuum motor and spark discharging between the graphite brushes and  
 206 the commutator [19,21,29,35]. Vacuum motor emissions can be partly or totally removed with the  
 207 installation of a HEPA filter [47,49].

208





209 Figure 1.  $PM_{10}$  mass concentrations during vacuum cleaning.

210

211 The average particle emission rate ( $\overline{Q}_s$ ) was calculated as follows [20,50]:

212

$$213 \quad \overline{Q}_s = V \times \left[ \frac{C_{in} - C_{in0}}{\Delta t} + (\alpha + \kappa) \overline{C}_{in} - \alpha C_{in0} \right] \quad (1)$$

214

215 where  $V$  is the room volume,  $C_{in}$  and  $C_{in0}$  are the peak and initial indoor particle concentrations,  
 216 respectively,  $\alpha$  is the average air exchange rate,  $\alpha + \kappa$  is the average removal rate and  $\Delta t$  is the time  
 217 difference between the initial and peak particle concentration. The particle removal rate is the slope  
 218 obtained by plotting  $\ln(C_{in}/C_{in0})$  versus time [22,51,52].

219

220 Table 1. Characteristics of the vacuums tested, sampling conditions and  $PM_{10}$  mass and particle  
 221 number concentrations and emission rates during the operation of distinct vacuum cleaners.

	Washable filter bag less vacuum	Wet vacuum	Bagged vacuum	HEPA filter equipped robot
Vacuum characteristics				
Year of purchase	2010	2014	2016	2016
Motor power (W)	2200	750	1000	Battery powered model
Dust collection	Plastic chamber	Water tank	Disposable paper bag	Plastic chamber
Vacuum tests				
N	2	2	2	2
Air exchange rate ( $\alpha$ , $h^{-1}$ )	$0.29 \pm 0.06$	$0.42 \pm 0.05$	$0.50 \pm 0.16$	$0.47 \pm 0.07$
T ( $^{\circ}C$ )	$24.6 \pm 2.40$	$20.6 \pm 1.30$	$21.7 \pm 0.31$	$20.4 \pm 0.07$
HR (%)	$38.2 \pm 10.7$	$39.8 \pm 3.66$	$37.8 \pm 1.05$	$35.7 \pm 1.22$
$PM_{10}$ initial mass concentration ( $\mu g m^{-3}$ )	$30.5 \pm 9.19$	$18.0 \pm 9.90$	$42.0 \pm 41.0$	$23.5 \pm 0.707$



PM <sub>10</sub> peak mass concentration (µg m <sup>-3</sup> )	51.0 ± 8.49	37.5 ± 4.95	65.0 ± 42.4	26.0 ± 1.41
PM <sub>2.5</sub> /PM <sub>10</sub> (%)	74.2 ± 10.1	81.2 ± 4.45	81.1 ± 16.4	43.3 ± 3.61
PM <sub>1</sub> /PM <sub>10</sub> (%)	72.1 ± 10.6	79.4 ± 4.92	79.4 ± 17.7	39.2 ± 4.00
Particle number initial concentration (particles × 10 <sup>3</sup> cm <sup>3</sup> )	6.59 ± 4.16	3.57 ± 0.38	19.7 ± 16.9	4.88 ± 1.34
Particle peak number concentration (particles × 10 <sup>5</sup> cm <sup>3</sup> )	0.548 ± 0.014	2.10 ± 0.136	1.39 ± 0.102	0.050 ± 0.011
Emission rate PM <sub>10</sub> (µg s <sup>-1</sup> )	1.26 ± 0.131	1.44 ± 0.282	3.46 ± 1.65	-
Emission rate particle number (particles × 10 <sup>11</sup> min <sup>-1</sup> )	5.29 ± 1.48	21.2 ± 2.10	12.6 ± 4.54	-

222

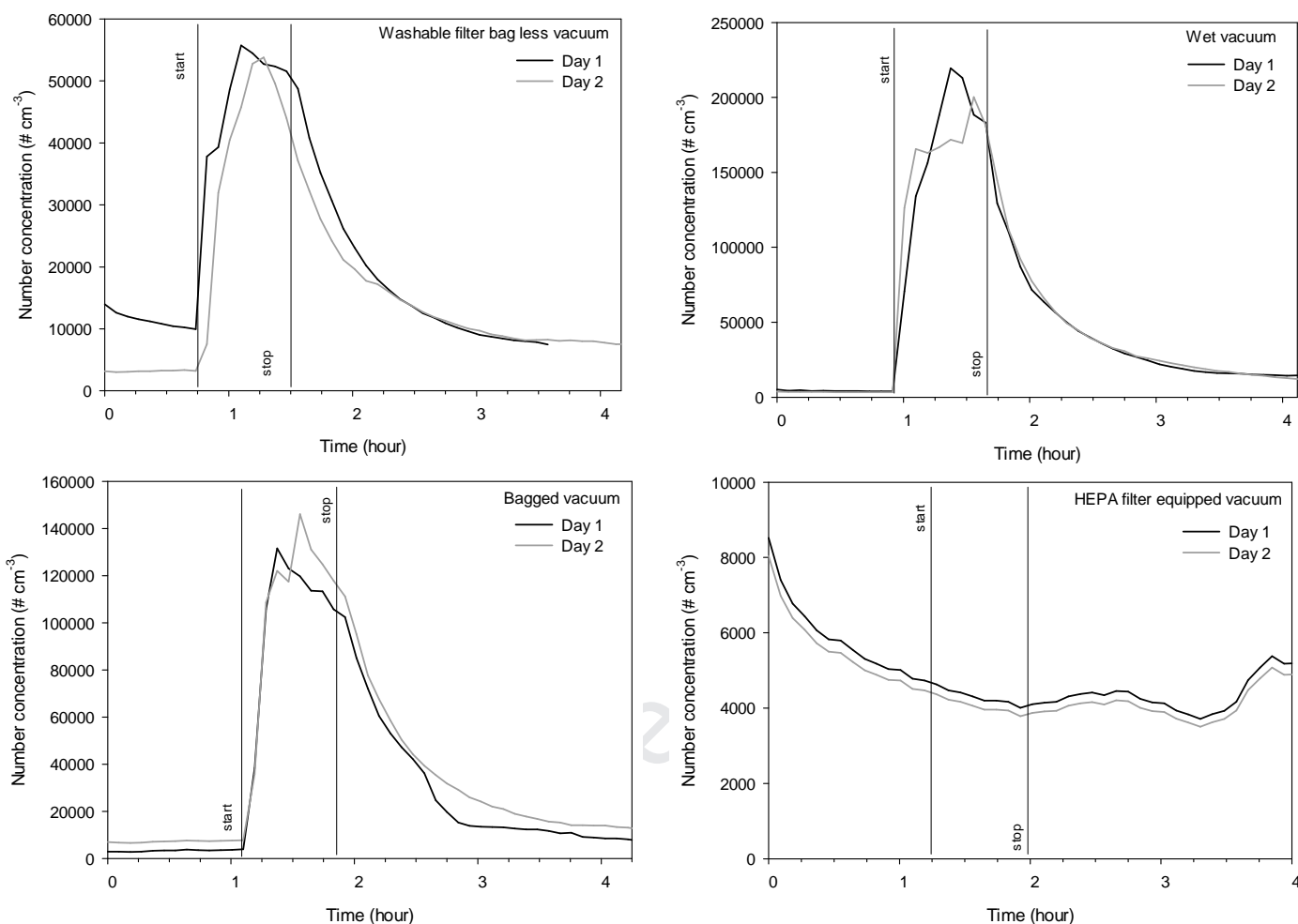
223 The estimated PM<sub>10</sub> emission rates from bagged vacuum operation were, on average, 2.4 and 2.8  
 224 times higher ( $207 \pm 99.0 \mu\text{g min}^{-1}$ ) than those from wet ( $86.1 \pm 16.9 \mu\text{g min}^{-1}$ ) and washable filter  
 225 bag less vacuum operation ( $75.4 \pm 7.89 \mu\text{g min}^{-1}$ ) (Table 1). The emission rates derived from this  
 226 study are in line with those presented in the literature. He et al. [20] reported a PM<sub>2.5</sub> emission rate  
 227 of  $70 \pm 40 \mu\text{g min}^{-1}$  for vacuuming. Higher vacuum emission rates ( $690 \pm 30 \mu\text{g min}^{-1}$ ) were  
 228 reported by Nasir and Colbeck [50] in a shared multi storey single room. The large variability in  
 229 vacuum emissions was highlighted in the study of Knibbs et al. [28]. The authors reported PM<sub>2.5</sub>  
 230 emission rates from 21 vacuum cleaners during warm and cold start tests in the ranges from 0.41 to  
 231 1962 and from 0.24 to 2870  $\mu\text{g min}^{-1}$ , respectively.

232

### 233 3.1.2. Number concentrations

234 Figure 2 illustrates the time evolution of the total particle number concentration during vacuuming.  
 235 The maximum particle number concentration was reached within a few minutes. The average  
 236 particle number concentration in the room was higher during the operation of the wet vacuum ( $1.69$   
 237  $\times 10^5 \pm 7.54 \times 10^2 \text{ particles cm}^{-3}$ ) and the bagged vacuum ( $1.09 \times 10^5 \pm 4.95 \times 10^3 \text{ particles cm}^{-3}$ ).  
 238 The HEPA filter equipped vacuum cleaner did not increase the number of particles in the room.  
 239 During its operation, the particle number concentration was  $4.53 \times 10^3 \pm 8.16 \times 10^2 \text{ particles cm}^{-3}$ ,  
 240 which was similar to the one recorded before the vacuum operation ( $5.86 \times 10^3$ ) and after the robot  
 241 was turned off ( $4.38 \times 10^3$ ).

242



243 Figure 2. Particle number concentrations during vacuum cleaning.

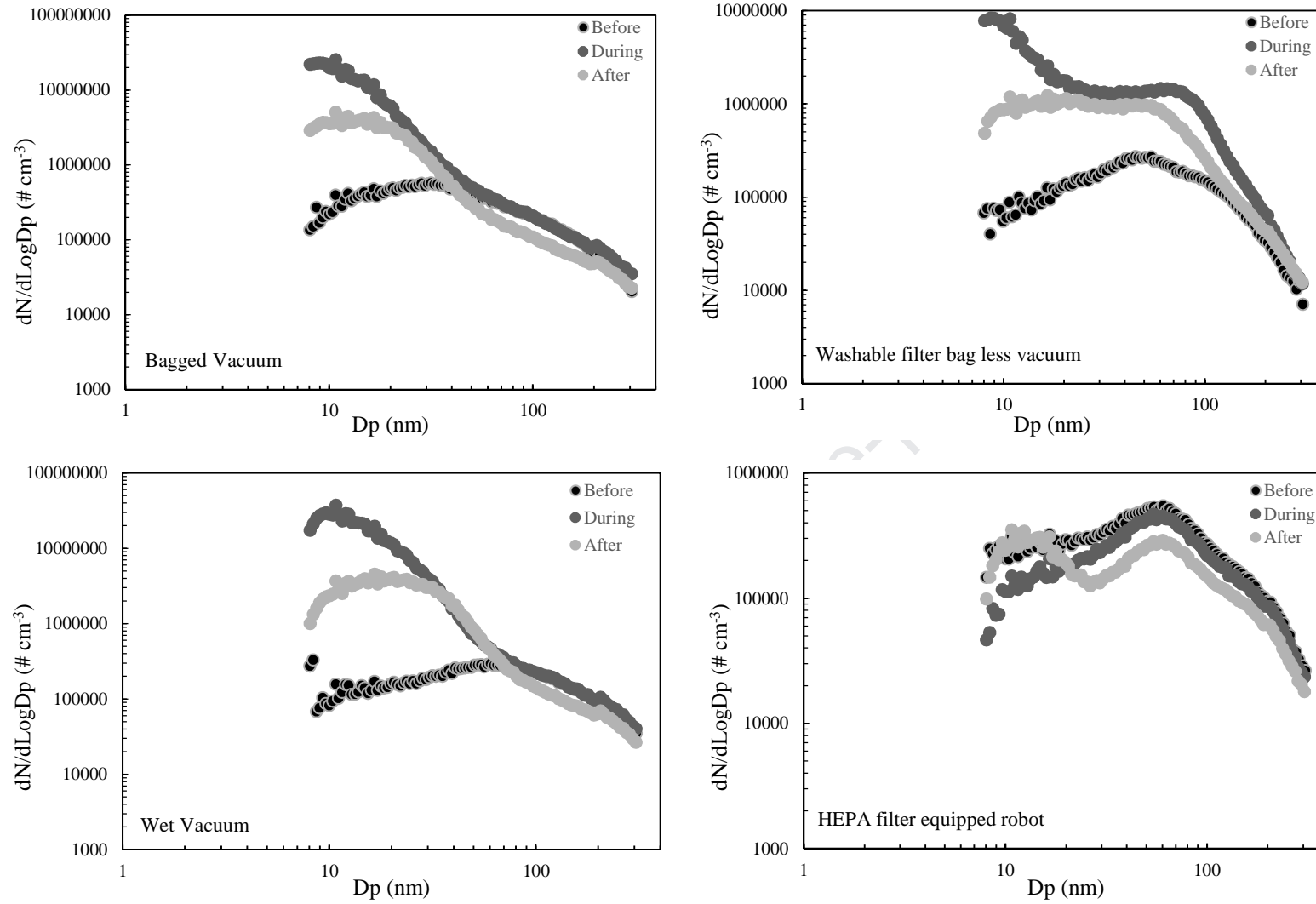
244

245 These results are in accordance with previous studies reporting very high (> 99%) fine particle  
 246 collection efficiency of vacuums equipped with a HEPA filter [29,53]. Manigrasso et al. [54]  
 247 documented a decrease in particle number concentration compared to background levels when  
 248 using a HEPA filter equipped vacuum cleaner, suggesting that the filter removes particles from the  
 249 ambient air to some extent. The ratios of peak to background levels for ultrafine particle number  
 250 concentrations presented in Table 1 indicate that vacuuming can elevate concentrations by a factor  
 251 ranging from 4 (bagged vacuum) to 61 (wet vacuum). Table 1 also provides estimates of particle  
 252 number emission rates for the different vacuums, which were calculated as described above for  
 253 particulate mass emissions (Equation 1). A previous study conducted by He et al. [20] in suburban  
 254 Brisbane households reported submicrometer particle emission rates of  $0.97 \pm 1.57 \times 10^{11}$  particles  
 255  $\text{min}^{-1}$  (particles from 0.007 to 0.808  $\mu\text{m}$ ). Knibbs et al. [28] measured particle number emission  
 256 rates from 21 vacuum cleaners in the range from 0.004 to  $108 \times 10^9$  particles  $\text{min}^{-1}$  (particles from  
 257 0.54 to 20  $\mu\text{m}$ ). The assessment of vacuum cleaning in a full-scale chamber carried out by Afshari  
 258 et al. [21] resulted in an emission rate of  $0.35 \times 10^{11}$  particles  $\text{min}^{-1}$  (particles from 0.02 and about  
 259 1.0  $\mu\text{m}$ ). Wu et al. [36] tested 3 different scenarios of vacuum cleaning in a closed living room,

260 including normal condition, filter removed, and filter and dust bag removed. The ultrafine particle  
261 number emission rates (from 0.0146 to 0.6612  $\mu\text{m}$ ) for the operation with no filter and without  
262 filter and dust bag was 2.2 and 2.5 times higher than that of the normal scenario ( $1.32 \pm 0.58 \times 10^{10}$   
263 particles  $\text{min}^{-1}$ ). In the present study, the average emission rates (particles from 0.008 to 0.322  $\mu\text{m}$ )  
264 were estimated to range from  $5.29 \times 10^{11}$  to  $21.2 \times 10^{11}$  particles  $\text{min}^{-1}$ .

265 Emissions should be compared with caution since the differences between vacuum cleaners  
266 (model, age, state of preservation, etc.), sampling conditions (real life monitoring campaigns vs  
267 laboratory chambers) and particle diameters may lead to non-generalisable results. In real life/on  
268 site monitoring campaigns, several factors, such as building characteristics, ventilation conditions,  
269 concurrent activities, cleaning routines, etc., should also be taken into account. In fact, He et al.  
270 [20] obtained variable results in different houses when vacuumed. In one of the houses, a doubling  
271 in  $\text{PM}_{2.5}$  concentrations was observed during vacuuming compared to background levels, while no  
272 increase was noticed in particle number concentrations. However, in a different house, the opposite  
273 behaviour was registered, with no increase in the  $\text{PM}_{2.5}$  mass concentrations, while the particle  
274 number concentration increased. The authors pointed out the differences in vacuums and in  
275 cleaning routines as possible reasons behind the observed results. House cleaning routines can  
276 affect both the dust resuspension and the dust loads available to vacuum, which, in turn, may affect  
277 the particle reemission. The effect of the vacuum cleaner or vacuum cleaner components on  
278 emissions can also be significant. Afshari et al. [21] investigated fine particle emissions when  
279 running a vacuum cleaner in a full-scale chamber. Two experiments were carried out: (i) vacuum  
280 cleaner operated with a dust bag and (ii) vacuum cleaner operated without dust bag, filters and hose  
281 in order to study the emissions from the motor only. The results revealed that the particle  
282 concentrations originating from the motor were higher than those from the vacuum cleaner with a  
283 bag.

284 Figure 3 displays the typical evolution of the distribution of the aerosol during the operation of the  
285 vacuum cleaners. While using the wet and bagged vacuum cleaners, more than 90 % (93 – 95 %) of  
286 the total particle number concentrations was found in the nucleation mode ( $N < 30 \text{ nm}$ ). This value  
287 dropped to 74 – 78 % when using the washable filter bag less vacuum cleaner. The high number of  
288 ultrafine particles emitted from vacuuming is consistent with previous studies [25,28,36,55]. The  
289 geometric mean diameter (GMD) of the particle size distribution ranged between 13.5 and 17.8 nm,  
290 while the source was active (excluding the HEPA filter equipped robot).



291

292

293

294 Figure 3. Typical average evolution of the mean particle size distribution before, during and after vacuum cleaning.

295 **3.2. Metals**

296 Trace and major elements were analysed in PM<sub>10</sub> samples (Table 2). Among them, Cu and Si  
 297 strongly dominated when the wet and bagged vacuum cleaners were run, while the washable filter  
 298 bag less vacuum and the HEPA filter equipped robot generated particles mainly containing Si and  
 299 Ca. Elements accounted for PM<sub>10</sub> mass fractions of 31.2, 20.1, 22.4 and 8.41 %wt., which  
 300 represented increases over background of 6.6, 4.3, 4.8 and 1.8 times for the washable bag less  
 301 filter, wet, bagged and HEPA filter equipped vacuum cleaners, respectively.

302

303 Table 2. PM<sub>10</sub> mass fractions (wt.%) of major and trace elements.

Element	Washable filter bag less vacuum	Wet vacuum	Bagged vacuum	HEPA filter equipped robot	Background
Na	0.100	bdl	bdl	0.331	1.05
Mg	0.751	0.762	0.416	0.152	0.152
Al	3.87	3.16	2.18	1.45	0.314
Si	9.74	4.675	4.39	2.73	0.678
P	0.106	0.167	0.060	0.041	0.013
S	1.30	0.419	0.801	0.370	0.648
Cl	1.27	0.515	0.599	0.319	0.749
K	1.71	0.435	0.585	0.359	0.336
Ca	5.94	2.68	2.41	1.77	0.396
Ti	0.884	0.418	0.248	0.133	0.006
V	0.002	bdl	bdl	bdl	bdl
Cr	0.005	bdl	bdl	bdl	0.005
Mn	0.049	bdl	0.019	0.021	0.007
Fe	1.88	0.867	0.843	0.635	0.263
Ni	0.004	0.013	0.007	0.001	0.001
Cu	2.92	5.78	8.89	0.012	0.012
Zn	0.246	0.130	0.116	0.062	0.049
As	bdl	0.010	0.004	bdl	0.004
Se	0.003	0.001	0.002	0.001	0.003
Br	0.009	bdl	0.003	bdl	0.002
Rb	0.017	bdl	0.015	0.001	bdl
Sr	0.040	0.021	0.037	0.011	bdl
Y	0.014	0.014	0.012	bdl	0.003
Zr	0.011	bdl	0.045	bdl	bdl
Mo	0.283	bdl	0.755	0.008	bdl
Pb	0.015	bdl	bdl	0.004	bdl
Σ Elements	31.2	20.1	22.4	8.41	4.69
Σ Element oxides	53.4	32.9	35.2	14.8	7.56

304 bdl – below the detection limit. The measured element concentrations were converted into the respective  
 305 mass concentrations of the most common oxides (SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, MgO, MnO, Fe<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, K<sub>2</sub>O, etc.)

306

307 The contribution of Cu to the PM<sub>10</sub> mass ranged from 0.01 %wt. (HEPA equipped robot) to 4.86  
308 %wt. (bagged vacuum), which represent an increase over background concentrations ranging from  
309 1.6 to 848 times. Metals contribute to 20 – 30% of the total weight of a vacuum cleaner. The  
310 metallic components are made of aluminum (motor and screws), stainless and galvanized steel  
311 (motor), brass (plug) and copper (plug, power cord, wire cables and motor) [56]. Cu concentrations  
312 up to 55 µg m<sup>-3</sup> were previously reported in particulate matter emissions from a professional  
313 vacuum cleaner in a test room [35].

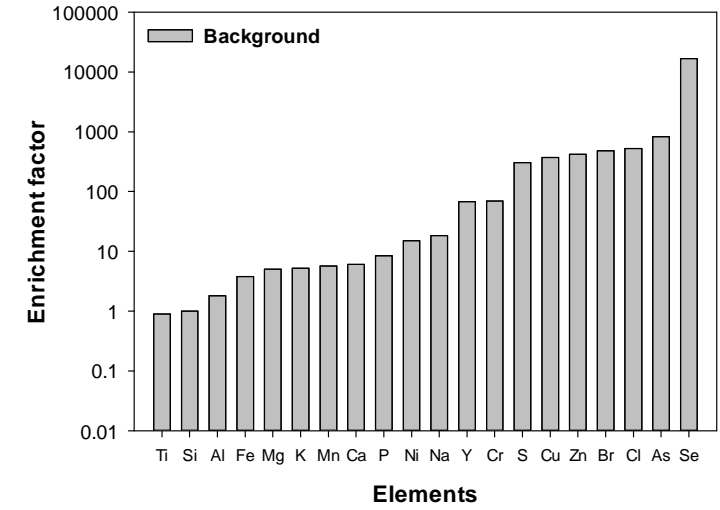
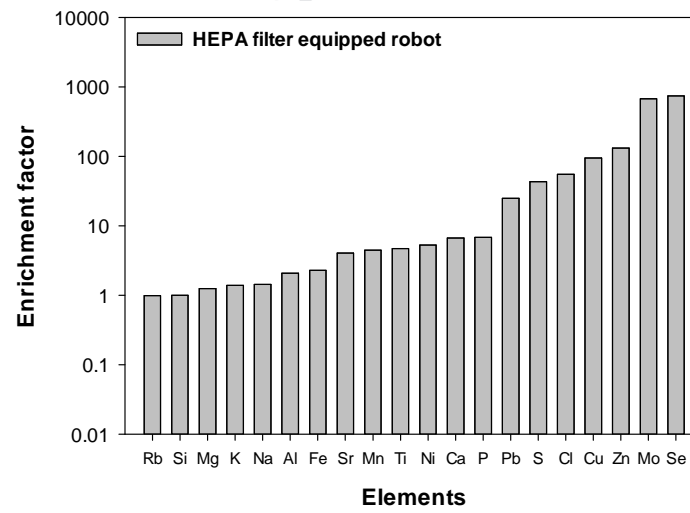
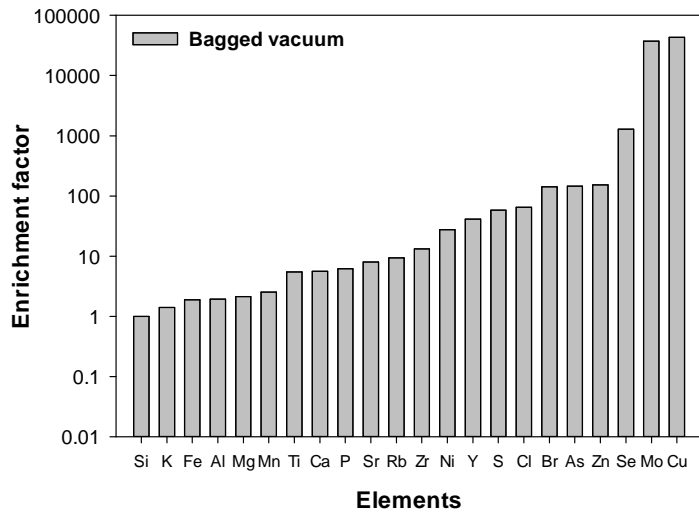
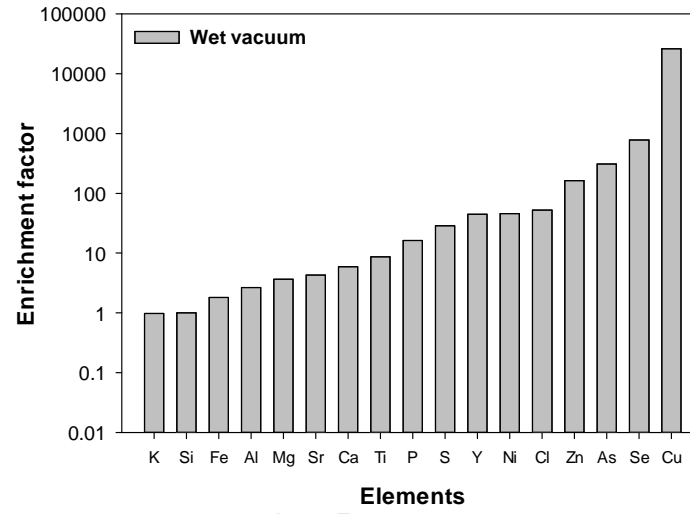
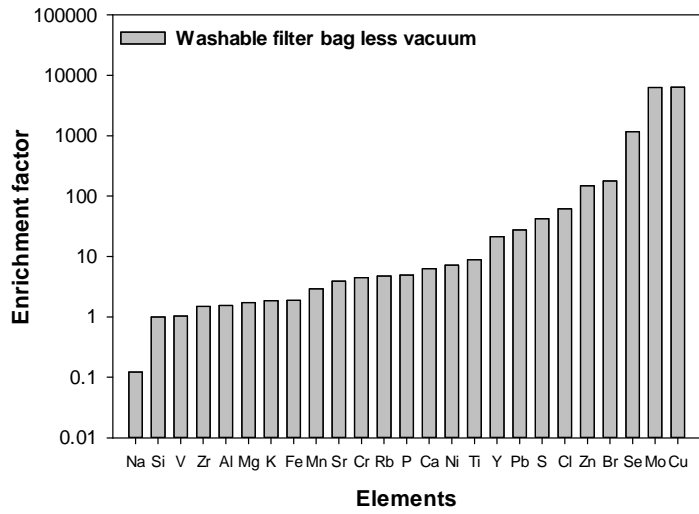
314 For each element, enrichment factors (EFs) were calculated according to equation 2, where E and R  
315 represent the concentrations of the element under analysis and the reference element, respectively:

316

$$317 \quad EF = (E/R)_{air}/(E/R)_{crust} \quad (2)$$

318

319 In the present study, Si was used as reference element due to its high abundance in the earth's crust.  
320 The average element concentrations in the upper continental crust were taken from Wedepohl [57].  
321 During vacuuming, minimal enrichments were obtained for some elements, such as Al, Mg, K, Fe,  
322 V, and Mn (EF < 5), indicating that these elements were mostly derived from soil dust. Rasmussen  
323 et al. [58] found significant relationships between concentrations in household settled dust and  
324 airborne particulate matter for several elements, namely Ag, Al, As, B, Cu, Fe, Mn, Pb, U, V and  
325 Zn. In the present study, other elements like P, Ca, Ni, S and Cl were enriched (Figure 4) during the  
326 operation of every vacuum cleaner tested and also in the background sample suggesting that the  
327 origin of the enrichment was not vacuuming. Zinc (132 < EF < 163) and selenium (743 < EF <  
328 1285) were also highly enriched elements in all vacuuming tests and background sample (EF = 419  
329 and EF = 16721 Zn and Se, respectively). Molybdenum was highly enriched (675 < EF < 37240) in  
330 all the vacuuming samples except in the one collected during the wet vacuum operation, whereas it  
331 was not enriched in the background sample. Copper EFs were very high when operating all the  
332 vacuum cleaners (over 6000, 25,000 and 41,000 for the washable filter bag less, wet and bagged  
333 vacuum cleaners, respectively), except for the HEPA equipped robot (EF < 100) (Figure 4).



334 Figure 4. Enrichment factors of elements in PM<sub>10</sub> sampled during vacuum cleaning.



335 A health risk assessment of exposure to major and trace elements by inhalation was carried out as  
 336 described by the United States Environmental Protection Agency (USEPA)[59], as follows:

337

$$338 \quad EC = (CA \times ET \times EF \times ED) / AT$$

339 (3)

340

341 where EC is the exposure concentration ( $\text{ng m}^{-3}$ ), CA is the element concentration ( $\text{ng m}^{-3}$ ), ET is  
 342 the exposure time ( $0.75 \text{ h d}^{-1}$ ), EF is the exposure frequency ( $144 \text{ d y}^{-1}$ ), ED is the exposure  
 343 duration (70 y) and AT is the averaging time ( $70 \text{ y} \times 365 \text{ d y}^{-1} \times 24 \text{ h d}^{-1}$ ). The exposure time used  
 344 in the calculations was based on the results of an online survey among 28,000 consumers from 23  
 345 countries on their vacuum cleaning habits [27]. According to the survey, 33% of the respondents  
 346 vacuum 2 to 5 times per week and 46% vacuum for half to one hour. Taking into account these  
 347 results, the exposure concentration was calculated assuming a vacuum frequency of three times per  
 348 week for 45 minutes.

349 The excess cancer risk posed by the individual metals associated with  $\text{PM}_{10}$  inhalation were  
 350 calculated following equation (4), where IUR is the inhalation unit risk ( $(\text{ng m}^{-3})^{-1}$ ).

$$351 \quad \text{Risk} = \text{IUR} \times \text{EC}$$

352 (4)

353 The IUR values were retrieved from the database provided by USEPA [60] for three elements (As,  
 354 Cr (VI), and Pb). In the present study, one seventh of total Cr concentration was used to estimate  
 355 the risk considering that the tabulated IUR is for Cr (VI), which is based on a Cr (III): Cr (VI)  
 356 proportion of 1/6. USEPA considers that a  $10^{-6}$  risk is below the level of apprehension, while risks  
 357 above  $1.0 \times 10^{-4}$  are of concern. In the present study, the cumulative cancer risk for all potential  
 358 carcinogenic elements was negligible (always  $< 5 \times 10^{-7}$ ).

359 Noncarcinogenic risks associated with inhalation exposure to trace elements in indoor  $\text{PM}_{10}$  were  
 360 estimated by the noncancer hazard quotient (HQ) following the methodology proposed by USEPA  
 361 [59,61]:

$$362 \quad HQ = EC / R_fC \quad (5)$$

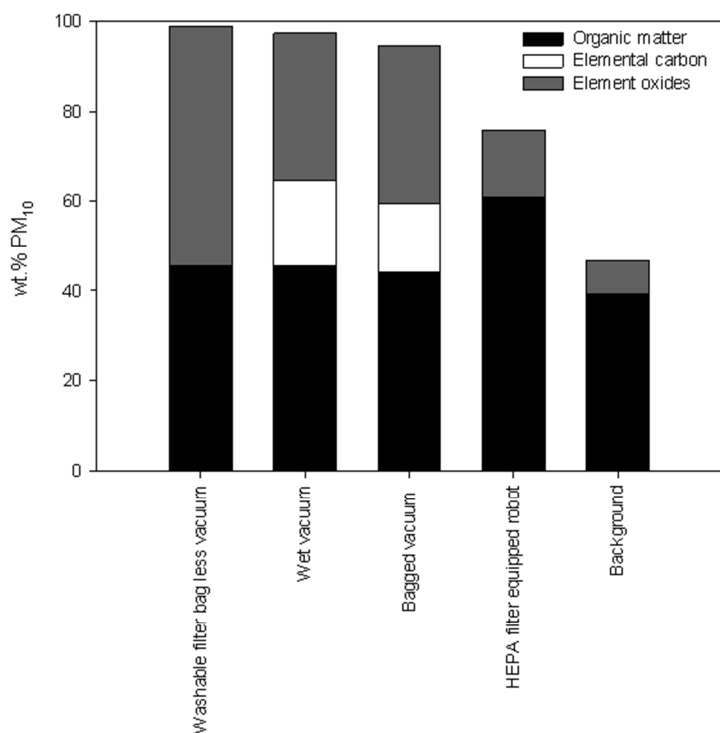
363  $R_fC$  is the USEPA reference concentration ( $\text{mg m}^{-3}$ ). Considering that, for some elements,  
 364 reference doses ( $R_fD$ ,  $\text{mg kg}^{-1} \text{ day}^{-1}$ ) are available instead of  $R_fC$  values [60,62], these latter were  
 365 calculated taking into consideration the inhalation rate and body weight of an adult following the  
 366 methodology described by USEPA [63]. The reference values were retrieved from the Integration  
 367 Risk Information System (IRIS) [62] and USEPA [60] databases. The HQ associated with  
 368 inhalation exposure to particulate trace elements in the indoor air during vacuuming were much  
 369 lower than the unity, indicating negligible risks.

370

371 **3.3. OC/EC**

372 Total carbon (TC) represented from  $32.7 \pm 2.68$  (washable filter bag less vacuum) to  $51.6 \pm 2.08$   
373 (wet vacuum) wt.% of the  $PM_{10}$  mass during vacuuming, corresponding to TC increases over  
374 background levels ranging from 1.2 to 1.8. EC levels were distinctively higher during the operation  
375 of the wet (19.0 wt.%  $PM_{10}$  mass) and bagged (15.4 wt.%  $PM_{10}$  mass) vacuum cleaners. For the  
376 HEPA filter equipped vacuum cleaner, as well as in background air samples, EC was not present at  
377 detectable levels (Figure 5). OC and EC concentrations in samples collected when the vacuum  
378 cleaners were run were not correlated with each other, indicating distinct sources. Contrarily, good  
379 correlations were found between particulate EC concentrations and both Cu ( $r^2 = 0.87$ ) and Ni  
380 concentrations ( $r^2 = 0.79$ ). Good correlations ( $r^2 > 0.75$ ) between OC and several elements,  
381 including Si, S, Cl, K, Ca, Fe, Zn and Se, were also recorded. Given that one of the main  
382 contributors to indoor particles is probably resuspended dust, some of which associated with soil,  
383 these correlations are not surprising. Household  $PM_{10}$  dust has been reported to contain appreciable  
384 amounts of carbonaceous particles, mainly OC, whereas in many samples EC was too low or  
385 undetectable [64]. Black carbon, on the other hand, has been associated with motor emissions from  
386 vacuum cleaners [18]. OC to EC ratios showed high variability, ranging from 1.7 (bagged vacuum)  
387 to 106 (washable filter bag less vacuum). Habre et al. [65] found that household  $PM_{2.5}$  OC fractions  
388 were mainly related to human activities, including vacuum cleaning, which leads to resuspension of  
389 dust and  $PM_{2.5}$  generation. Alves et al. [38] reported average OC/EC values ranging from 4.2 to 9.7  
390 in school classrooms. The researchers argued that these ratios were expected, since resuspended  
391 dust, some of which associated with soil, was found to be one of the main primary contributors to  
392 indoor particles.

393



394

395

396 Figure 5. Chemical mass closure of PM<sub>10</sub>.

397

398 In the present study, in order to convert the measured mass of OC to total organic matter (OM)  
 399 mass, the OC concentration was multiplied by a factor of 1.4 [66,67], which is an estimate of the  
 400 average molecular weight per carbon weight for the organic aerosol. The mass closure between  
 401 chemical and gravimetric measurements was nearly 100% for most samples, except for background  
 402 air and the sample collected during cleaning with the HEPA filter equipped robot (Figure 5). The  
 403 fraction of unidentified mass might be attributable to the selection of the multiplier factor to derive  
 404 the OM, particle-bound water, sampling artefacts, among others [67]. The presence of unanalysed  
 405 constituents might also be responsible for the unaccounted mass.

406

#### 407 3.4. Organics

408 The PM samples collected during vacuuming, as well as the background air samples, encompassed  
 409 several aliphatics, polycyclic aromatic hydrocarbons (PAHs), alcohols, acids, sterols, glycerol  
 410 derivatives, phenolic compounds, saccharides, among others.

411 The aliphatic fraction of particulate matter comprised n-alkanes from C<sub>11</sub> to C<sub>35</sub>. The maximum  
 412 concentrations were observed for the homologues in the range from C<sub>20</sub> to C<sub>22</sub>, which have been  
 413 described as characteristic of petrogenic sources. The carbon preference indices of n-alkanes were  
 414 in the range from 0.7 to 0.9 during the vacuuming operation and 1.1 for the background, suggesting  
 415 the contribution of petroleum derivatives [68]. The presence of these compounds may be related to

416 oil-based or petrochemical textiles such as nylon, polyester, acrylic and spandex, which are made  
 417 from natural gas or oil. Outgassing of lubricants applied to parts of the vacuum cleaners is another  
 418 likely source. The  $\Sigma_{25}$  n-alkane concentrations ranged from 22.4 to 39.3 ng m<sup>-3</sup> during vacuuming  
 419 and 17.3 ng m<sup>-3</sup> in the background air.

420 Discontinuous series of n-alkanols from C<sub>10</sub> to C<sub>30</sub> were detected in the PM<sub>10</sub> samples, maximising  
 421 at C<sub>16</sub>, during the operation of the tested vacuums, as well as in the background sample (Table 3).  
 422 Cetyl alcohol (C<sub>16</sub>H<sub>34</sub>O) is widely used in skin lotions and creams due to its water-binding  
 423 properties [69]. Other long-chain alcohols, such as myristyl (C<sub>14</sub>H<sub>30</sub>O) and stearyl alcohol  
 424 (C<sub>18</sub>H<sub>38</sub>O), were also found in all samples. Besides being used in a variety of cosmetic products as  
 425 emulsifier, emollient, antifoaming agent, and surfactant, stearyl alcohol has also been isolated from  
 426 human sebaceous lipids [69]. The  $\Sigma_{15}$  n-alkanol concentrations ranged from 362 to 858 ng m<sup>-3</sup>  
 427 during vacuuming and 172 ng m<sup>-3</sup> in the background air.

428

429 Table 3. Concentrations (ng m<sup>-3</sup>) of oxygenated organic compounds in PM<sub>10</sub>

	Wet vacuum	Bagged vacuum	HEPA filter equipped robot	Washable filter bag less vacuum	Background
<i>Saccharides</i>					
Galactosan	2.41	bdl	bdl	bdl	bdl
Mannosan	1.15	2.57	2.74	bdl	1.27
Levogluconan	8.27	9.69	23.6	5.93	22.1
Unidentified saccharides	45.2	47.4	46.5	278	4.50
<i>Phenolics and alteration products</i>					
Benzyl alcohol	1.96	bdl	6.77	bdl	bdl
Benzoic acid	0.643	0.683	0.858	1.26	0.187
4-Hydroxybenzoic acid	0.049	0.194	0.153	0.279	0.074
Trans-cinnamic acid	bdl	0.102	0.116	0.212	0.027
Pthalic acid	0.358	0.385	bdl	0.544	bdl
Vanillic acid	bdl	0.031	0.046	0.091	0.031
Syringic acid	bdl	bdl	bdl	bdl	0.054
Resorcinol	0.018	0.013	0.049	0.011	0.013
4-Methyl catechol	0.003	0.002	0.004	0.004	0.001
2,6-Dimethoxyphenol	0.005	0.003	0.021	0.005	0.001
Eugenol	0.006	0.006	0.016	0.005	0.001
Isoeugenol	bdl	2.42	3.92	1.78	bdl
4-Allyl-2,6-dimethoxyphenol (methoxy eugenol)	0.018	0.082	0.166	0.038	0.010
2,4-Di-tert-butylphenol	106	123	180	118	20.6
Pyrogallol	0.002	bdl	0.017	0.002	0.001
4-Phenylphenol	0.084	0.052	0.079	0.055	0.005
4-tert-butylphenol	11.9	3.03	24.8	13.3	1.71
4-Octylphenol	0.013	0.015	0.020	0.047	0.006

Thymol	2.236	0.063	4.74	1.634	0.199
Coniferyl alcohol	bdl	bdl	0.016	0.009	bdl
Sinapyl alcohol	0.006	0.003	0.025	bdl	0.001
Bisphenol F	bdl	bdl	bdl	3.99	bdl
Bisphenol A	57.8	8.902	bdl	46.9	0.543
<i>Aliphatic alcohols</i>					
1-Decanol	0.01	bdl	0.52	0.02	0.01
Dodecanol	26.2	53.0	243	38.3	7.02
Tetradecanol	79.9	139	204	122	8.78
1-Pentadecanol	29.8	57.7	87.2	68.2	32.9
Hexadecanol	137	201	220	189	82.7
Heptadecanol	5.50	5.27	9.87	4.48	5.90
1-Octadecanol	74.6	93.8	81.2	119	32.6
1-Eicosanol	1.82	1.23	2.98	2.58	1.05
1-Docosanol	1.70	0.40	3.72	bdl	bdl
1-Tricosanol	0.17	0.03	0.18	0.20	0.05
1-Pentacosanol	0.20	0.06	2.09	0.45	0.03
Hexacosanol	3.09	3.49	3.23	18.7	0.82
1-Heptacosanol	0.05	0.02	0.04	0.16	0.01
1-Octacosanol	0.82	0.41	0.57	5.23	0.09
1-Tricontanol	0.39	0.18	0.17	0.43	0.03
<i>Steroid compounds</i>					
Cholesterol	7.34	7.57	9.01	10.3	0.429
5-Cholesten-3-ol (epicholesterol)	bdl	0.099	bdl	bdl	bdl
β-Sitosterol	0.985	0.253	0.556	0.597	0.120
Lupeol	2.29	0.186	0.763	0.396	0.323
<i>Aliphatic acids</i>					
Octanoic acid	0.820	0.245	1.73	0.754	0.163
Nonanoic acid	0.246	0.453	3.79	1.18	0.194
Decanoic acid	27.9	4.15	6.26	4.01	0.323
Undecanoic acid	0.746	1.78	1.08	1.74	0.249
Dodecanoic acid	13.0	65.3	147	59.8	87.8
Tridecanoic acid	13.5	3.04	11.3	3.32	2.31
Tetradecanoic acid	33.7	71.5	144	92.3	165
Pentadecanoic acid	4.78	4.87	24.7	9.66	9.52
Hexadecanoic acid	bdl	49.5	504	332	141
Heptadecanoic acid	0.367	0.394	0.478	1.46	1.73
Octadecanoic acid	15.8	14.00	107	85.43	61.3
Nonadecanoic acid	0.089	0.097	0.096	0.20	0.109
Eicosanoic acid	0.229	0.124	bdl	0.84	0.443
Docosanoic acid	0.053	0.045	0.334	0.54	1.76
<i>Diacids</i>					
Butanedioic (succinic)	0.205	2.38	2.01	4.77	1.05
Hydroxybutanedioic (malic)	bdl	bdl	2.09	bdl	bdl

1,5-Pentanedioic (glutaric)	0.825	1.08	2.78	5.13	4.18
Hexanedioic (adipic)	0.090	2.61	3.54	6.85	2.05
Heptanedioic (pimelic)	0.711	0.608	0.530	0.978	0.248
Octanedioic (suberic)	0.324	1.18	0.853	1.44	0.251
Nonanedioic (azelaic)	0.840	3.10	2.72	6.57	1.03
<i>Other acids</i>					
Boric acid	bdl	bdl	8.00	1.32	6.05
2-Hydroxyethanoic (glycolic)	162	bdl	133	190	37.1
2,3-Dihydroxypropanoic (glyceric)	bdl	bdl	13.8	45.9	14.6
3-Hydroxybutanoic (3-hydroxybutyric)	0.491	0.497	0.335	0.807	0.166
9-Cis-Hexadecenoic (palmitoleic)	0.772	0.616	1.11	1.09	1.43
Cis,cis-9-12-octadecadienoic (linoleic)	0.821	bdl	bdl	bdl	0.147
Cis-9-Octadecenoic (oleic)	4.40	0.928	1.70	2.00	20.37
Cis-Pinonic	0.002	1.52	0.315	0.708	0.302
Citric acid	bdl	0.221	0.723	1.44	0.088
Adipic acid dioctyl ester	32.9	75.2	13.5	34.7	7.76
Abietic	bdl	bdl	0.378	bdl	bdl
Dehydroabietic	0.290	0.363	0.706	0.906	0.485
Isopimaric	bdl	bdl	bdl	0.034	0.009
Podocarpic acid	bdl	0.016	0.055	0.154	0.003
<i>Glycerol derivatives</i>					
Glycerol	0.917	136	237	671	144
Diethylene glycol	1.16	0.575	0.425	bdl	bdl
1-Monolauroyl-rac-glycerol	0.001	0.005	0.036	0.005	bdl
1-Monolinoleoylglycerol	4.86	bdl	86.0	4.97	1.08
Glycerol monostearate (monostearin)	13.4	19.9	26.3	93.2	4.59
1-Monopalmitate glycerol (1-monopalmitin)	15.3	19.4	16.2	71.6	3.50
<i>Other compounds</i>					
2,6-Di-tert-butyl-1,4-benzoquinone	0.933	1.33	0.52	1.34	0.214
(-)-Isopulegol	0.087	0.127	0.100	0.086	0.011
5-Isopropyl-3-Methylphenol	0.098	0.168	0.520	0.270	bdl
(1S, 2S, 3R, 5S)-2,3-Pinane-1,2-diol	0.091	0.300	0.628	0.255	0.06
Diethyltoluamide (DEET)	19.6	51.8	66.6	76.9	44.1
Tributyl phosphate (TBP)	bdl	118.42	bdl	bdl	0.922
Tetraacetylenediamine (TAED)	bdl	6.60	bdl	bdl	bdl
Parsol MCX	6.64	4.08	6.72	6.78	3.84
Fyrol FR-2 (tris(1,3-dichloroisopropyl)phosphate)	52.9	5.79	bdl	bdl	0.526
Acetyl tributyl citrate	2.63	144	3.98	4.99	3.28
Oxidised Irgafos 168	342	bdl	71.2	128	31.43
<i>Plasticisers</i>					
Benzyl butyl phthalate	nd	nd	nd	0.268	nd
Bis(2-ethylhexyl)adipate	0.109	nd	bdl	0.361	bdl
Bis(2-ethylhexyl)phthalate	bdl	0.419	bdl	0.603	bdl

Di-n-butyl phthalate	0.300	0.459	bdl	0.677	bdl
Diethyl phthalate	0.000	bdl	bdl	0.155	bdl
Dimethyl phthalate	0.100	nd	nd	0.100	0.010

430 bdl – below the detection limit; nd – not detected.

431

432 A series of n-alkanoic acids from C<sub>8</sub> to C<sub>22</sub>, maximising at C<sub>14</sub> or C<sub>16</sub>, were identified (Table 3).  
 433 Sources of alkanolic acids include cooking [70,71], emission from people's skin oils [72], incense  
 434 burning [73] and biomass combustion [74,75]. Other sources include biogenic contributors, such as  
 435 fungi, bacteria, spores, and pollen [76]. Lower molecular weight n-alkanoic acids (<C<sub>18</sub>) were  
 436 found in emissions from fossil fuel combustion, road dust and tyre wear debris [e.g. ,76,77]. Zhao  
 437 et al. [71] pointed out tetradecanoic (myristic) acid as a possible organic marker to distinguish  
 438 emissions from Chinese cooking. The Σ<sub>14</sub> n-alkanoic acid concentrations ranged from 111 to 951  
 439 ng m<sup>-3</sup> during vacuuming, while the background air sample presented a concentration of 472 ng m<sup>-3</sup>  
 440 <sup>3</sup>, indicating that these compounds are originated from household's activities.

441 Among diacids, the compound with highest concentrations was adipic acid (or C<sub>6</sub> diacid) (Table 3).  
 442 Dicarboxylic acids from C<sub>4</sub> to C<sub>8</sub> were recorded in fine organic aerosols from charbroilers and meat  
 443 cooking operations by Rogge et al. [78]. The authors identified hexanedioic acid as one of the  
 444 dominant compounds. Malic acid was only present in the sample collected when the HEPA filter  
 445 equipped robot was operated. Röhrl and Lammel [79] pointed out the influence of biogenic sources  
 446 on the occurrence of malic acid. The Σ<sub>8</sub> dicarboxylic acids concentrations ranged from 58.6 to 95.4  
 447 ng m<sup>-3</sup> during vacuum cleaning, while the background air sample presented a concentration of 21.0  
 448 ng m<sup>-3</sup>.

449 Several phenolic compounds were also detected in the samples; 2,4-di-tert-butylphenol, bisphenol  
 450 A and 4-tert-butylphenol were the most abundant (Table 3). Alkylphenols, such as 4-tert-  
 451 butylphenol and 2,4-di-tert-butylphenol, have a large variety of usages such as emulsifying agents  
 452 in latex paints, glue, and pesticide ingredients, in the preparation of antioxidants, curing agents, and  
 453 heat stabilisers for polymer resins, among others [80]. In the present study, concentrations ranging  
 454 from 106 to 180 ng m<sup>-3</sup> and from 3.03 to 24.8 ng m<sup>-3</sup> were registered for 2,4-di-tert-butylphenol and  
 455 4-tert-butylphenol, respectively. These compounds were also detected in background air samples.  
 456 Indoors, a source of alkylphenols is the biodegradation of alkylphenol ethoxylate, which is a widely  
 457 used surfactant in detergents. They can also be released from the surface of polymer resins, which  
 458 are used as antioxidant for wall or floor coverings [80]. Bisphenol A can act both as a plasticiser  
 459 and as a fungicide and is used in the production of polycarbonate and epoxy resins [81]. This  
 460 compound is ubiquitous in the atmosphere and its size distributions showed peaks in both fine and  
 461 coarse fractions. Soil resuspension has been suggested as a main source for bisphenol A in the  
 462 coarse fraction [82]. Bisphenol A is a decomposition product of polycarbonate, an ubiquitous  
 463 material indoors (e.g. hard plastic bottles, CDs, DVDs, etc.) [83]. Isoeugenol was another phenolic



464 compound detected in three of the four samples collected in the living room during vacuuming  
465 ( $1.78 - 2.42 \text{ ng m}^{-3}$ ) and was absent from the background air. Eugenol was detected in all samples  
466 although in much lower concentrations. Phenylpropenes, such as eugenol and isoeugenol, are  
467 produced by plants as defense compounds and as floral attractants of pollinators [84]. Isoeugenol is  
468 used in fragrance formulations which are incorporated into household laundry and cleaning  
469 products [85]. Eugenol is also common in consumer products, such as air fresheners [86]. This  
470 compound was also reported in samples collected in rooms from a Spanish household that had been  
471 treated with aerosols, electrical diffusion units, as well as with several cleaning products of  
472 domestic use [87]. Thymol was present in all samples and was among the phenolic compounds  
473 with the highest concentrations. Thyme essential oils are used in a variety of products in the food  
474 industry (preservatives and flavourings) and in cosmetics [88].  
475 Levoglucosan and its stereoisomers, mannosan and galactosan, were detected in samples (Table 3).  
476 These saccharides are formed from the thermal degradation of cellulose [89–91]. Although their  
477 individual quantification was not possible, many other saccharides were detected in  $\text{PM}_{10}$  samples.  
478 Cholesterol and  $\beta$ -sitosterol were also found in PM samples. Cholesterol was the most abundant. It  
479 is likely associated with cooking activities [92,93].  
480 Other hydroxyl compounds and phthalates were detected in the particulate matter organic extracts  
481 (Table 3). Among these, the most abundant were diethyltoluamide (DEET), tri(1,3-  
482 dichloroisopropyl)phosphate and Irgafos 168 (tris(2,4-di-tert-butylphenyl)phosphite). Tris(1,3-  
483 dichloro-2-propyl) phosphate was not detected in two of the five samples, reaching a concentration  
484 of  $52.9 \text{ ng m}^{-3}$  during the wet vacuum operation, which was up to 100 times higher than the  
485 background concentration. Flame retardants are used in many consumer and industrial products  
486 (e.g. electronics and electrical, building/construction, and textiles) to delay ignition and slow the  
487 spread of fire. Organic phosphorous containing flame retardants are mainly used in cellulosic  
488 materials, textiles, PVC-based products and polyurethane foam [94]. Air concentrations of this  
489 phosphate triester in European homes ranged from no detectable concentrations to  $21 \text{ ng m}^{-3}$  [95].  
490 Diethyltoluamide was present in all samples. It is used as insect repellent [96]. Irgafos 168  
491 (tris(2,4-di-tert-butylphenyl)phosphite) is a phosphite antioxidant used in several plastic packaging  
492 [97,98]. Parsol MCX (ethylhexyl methoxycinnamate) was also detected in all samples. It is  
493 frequently contained in personal care products as UV filter to protect human skin from UV  
494 radiation or as UV absorber to prevent light-induced product degradation [99].  
495 Several plasticisers were detected in PM samples. Di-n-butylphthalate (DBP) was the most  
496 abundant phthalate plasticiser present in the samples during vacuuming. In the background sample,  
497 only dimethyl phthalate was detected at quantifiable levels (Table 3). Plasticisers are widely used  
498 in the production of polyvinyl chloride (PVC) plastics, as well as in other applications such as  
499 glues, paints and cosmetics [100,101]. The vacuum body (external structure, dust container, power  
500 cord and wire cables) is made of several plastic components, including polypropylene (PP),

501 acrylonitrile butadiene styrene (ABS), high density polyethylene (HDPE) and PVC materials.  
 502 Other vacuum parts, such as the flexible hose, hose collaer and handle, floor brush and extension  
 503 tube are made of ethylene vinyl acetate (EVA), PP and HDPE [56]. Plasticisers can leach out from  
 504 PVC with materials aging and contaminate the environment [102].  
 505 The  $\Sigma_{19}$  PAHs concentrations ranged from 3.68 to 11.8 ng m<sup>-3</sup> during vacuuming and 3.32 ng m<sup>-3</sup> in  
 506 the background air (Table 4). The PAHs with highest concentrations were pyrene, chrysene and  
 507 benzo[b]fluoranthene. Delgado-Saborit et al. [103] measured sixteen PM-bound PAHs in 81  
 508 English households and reported concentrations ranging from undetectable levels to 25 ng m<sup>-3</sup>. In  
 509 Italy, Romagnoli et al. [104] documented concentrations of  $\Sigma_8$ PAHs in 10 private households in the  
 510 range from 0.4 to 8.4 ng m<sup>-3</sup>. Higher PAH concentrations were determined by Naumova et al. [105]  
 511 in 55 non-smoking urban residences in the USA. The indoor concentrations of  $\Sigma_{30}$  PAHs were 16-  
 512 220 ng m<sup>-3</sup> in Los Angeles, 21-310 ng m<sup>-3</sup> in Houston, and 22-350 ng m<sup>-3</sup> in Elizabeth.  
 513 Benzo[a]pyrene equivalent concentrations (BaP<sub>eq</sub>) were calculated (Table 4) multiplying the  
 514 measured levels of each PAH by the respective toxic equivalent factors (TEF), which were taken  
 515 from Bari et al. [106]. Dibenzo[a,h]anthracene was the compound that most contributed to the  
 516 carcinogenic potential of the PAH mixture for almost all the samples with values ranging from 33.8  
 517 % (washable filter bag less vacuum) to 48.3 % (bagged vacuum). During the wet vacuum operation  
 518 and in the background sample, the major contributor was benzo[a]pyrene accounting for 35.8 %  
 519 and 37.2 % to the carcinogenic potential, respectively.

520

521 Table 4. Concentrations of PAHs (ng m<sup>-3</sup>), carcinogenic potency of total PAHs (BaP<sub>eq</sub>, ng m<sup>-3</sup>) and  
 522 cancer risk.

PAHs	Wet vacuum	Bagged vacuum	HEPA filter equipped robot	Washable filter bag less vacuum	Background
Naphthalene	0.354	0.079	nd	nd	bdl
Acenaphthene	0.081	0.089	0.001	0.021	0.001
Fluorene	0.183	0.026	bdl	bdl	bdl
Phenanthrene	0.436	0.883	0.088	0.346	0.170
Anthracene	0.300	0.405	0.065	0.210	0.119
Acenaphthylene	nd	nd	nd	nd	nd
Retene	0.486	0.547	0.147	0.950	0.097
Fluoranthene	0.258	0.315	0.157	0.152	0.090
Pyrene	2.16	1.588	0.219	0.648	0.525
Chrysene	1.69	0.992	0.368	1.96	0.393
Benzo[a]anthracene	1.45	0.886	0.447	1.38	0.392

Benzo[b]fluoranthene	1.36	0.705	0.675	1.36	0.392
Benzo[k]fluoranthene	1.08	0.755	0.500	1.22	0.333
Benzo[e]pyrene	0.259	0.311	0.390	0.322	0.162
Benzo[a]pyrene	0.426	bdl	bdl	0.316	0.145
Perylene	0.080	nd	0.066	nd	nd
Dibenzo[a,h]anthracene	0.304	0.259	0.106	0.413	0.105
Indeno[1,2,3-cd]pyrene	0.453	0.226	0.255	0.677	0.194
Benzo[g,h,i]perylene	0.493	0.261	0.193	0.591	0.199
$\Sigma$ PAHs	11.8	8.33	3.68	10.6	3.32
BaP <sub>eq</sub>	1.19	0.54	0.30	1.22	0.39
Cancer Risk	$1.6 \times 10^{-8}$	$7.4 \times 10^{-9}$	$4.1 \times 10^{-9}$	$1.7 \times 10^{-8}$	$5.3 \times 10^{-9}$

523 bdl – below the detection limit; nd - not detected.

524

525 The inhalation exposure to PAHs was estimated following equation (3), where CA corresponds to  
 526 the BaP<sub>eq</sub> concentration (ng m<sup>-3</sup>). The excess cancer risk posed by PM-bound PAHs was  
 527 determined following equation (4) where IUR is calculated multiplying the cancer potency for  
 528 B[a]P of 3.9 ((mg kg day)<sup>-1</sup>) by the reference human inspiration rate per day (20 m<sup>3</sup>) and dividing  
 529 by the reference human body weight (70 kg). Table 4 displays the total carcinogenic risk calculated  
 530 from the particle-phase PAH mixture. The average carcinogenic risk was found to be negligible  
 531 ( $4.1 \times 10^{-9}$  to  $1.7 \times 10^{-8}$ ).

532

### 533 Conclusions

534 Cleaning activities are an important part of the household's daily routine and can contribute  
 535 significantly to personal exposure. Vacuuming is a recognised source of indoor particle generation,  
 536 however, there is still limited information on the impact of this particular source on indoor air  
 537 quality, especially concerning the PM composition, which is key to refine indoor source  
 538 apportionment and to improve estimates of residential human exposure.

539 This study presents indoor particulate mass and number emission rates and a comprehensive PM<sub>10</sub>  
 540 chemical characterisation during vacuuming cleaning with different devices (washable filter bag  
 541 less vacuum, wet vacuum, bagged vacuum) without any other active source. A sharp increase in  
 542 particle number concentrations was recorded when using most vacuum cleaners (4 to 61-fold in  
 543 relation to background air levels). The increase in the PM<sub>10</sub> mass concentrations due to vacuuming  
 544 was less pronounced, ranging from 1.2 to 1.6 in comparison with the initial concentrations (before  
 545 the activation of the source). While the bagged vacuum cleaner presented the highest PM<sub>10</sub>  
 546 emission rates, the particle (7.64-310.6 nm) number emission rates were highest during the wet

547 vacuum operation. No increase in  $PM_{10}$  mass concentrations or ultrafine particle number was  
548 observed when using the HEPA filter equipped vacuum cleaner.

549 When the wet and bagged vacuum cleaners were run, EC levels were substantially higher than  
550 those measured in the background air and while using other types of vacuum cleaners. The  
551 contribution of Cu to the  $PM_{10}$  mass ranged from 0.01 %wt. (HEPA equipped robot) to 4.86 %wt.  
552 (bag vacuum), which represented an increase over background concentrations ranging from 1.6 to  
553 848 times.

554 Wear of vacuum materials, grease and oils might be a source of particulate organic compounds.  
555 However, in the present study the organic speciation revealed the contribution of multiple sources,  
556 making it difficult to differentiate the possible input of vacuuming to the detected components.

557 Taking into account the numerous brands and models of vacuums available on the market, each  
558 possessing its own features (e.g. dust containers, bag materials, filtration systems etc), it is  
559 necessary to borne in mind that the findings of this study cannot be considered representative for  
560 each vacuum category (bag less, bagged, wet and robotic) and further investigations are necessary  
561 to consolidate the conclusions. Despite the limitations, the present study highlights the great  
562 variability in particle emission rates depending on the vacuum cleaner, suggesting that household  
563 exposure can be enhanced or reduced by proper selection of devices. Further investigation is  
564 needed to fully evaluate the potential health risk associated with this source.

565

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588

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Journal Pre-proof

**Highlights:**

- Vacuum cleaner type had a great effect on PM mass and number emissions
- HEPA filters can significantly reduce the PM emitted by the vacuum cleaner motor
- Elemental carbon increased markedly during the operation of wet and bagged vacuums
- Copper enrichment factors were high when using vacuum cleaners without HEPA filter
- The inhalation cancer risk for metals and PAHs was negligible

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**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Estela Domingos Vicente

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