Impact of vacuum cleaning on indoor air quality

Estela D. Vicente [a,*], Ana M. Vicente [a], Margarita Evtyugina [a], Ana I. Calvo [b], Fernanda Oduber [b], Carlos Blanco Alegre [b], Amaya Castro [b], Roberto Fraile [b], Teresa Nunes [a], Franco Lucarelli [c,d], Giulia Calzolai [d], Silvia Nava [c,d], Célia A. Alves [a]

[a] Centre for Environmental and Marine Studies (CESAM), Department of Environment and Planning, University of Aveiro, Aveiro, 3810-193, Portugal
[b] Department of Physics, IMARENAB University of León, 24071, León, Spain
[c] Department of Physics and Astronomy, Università di Firenze, 50019, Sesto Fiorentino, Italy
[d] INFN - Firenze, National Institute for Nuclear Physics - Florence Division, 50019, Sesto Fiorentino, Italy

ARTICLE INFO

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_{10} emission rates from bagged vacuum operation were much higher (207 ± 99.0 µg min^{-1}) compared with the ones obtained from wet (86.1 ± 16.9 µg min^{-1}) and washable filter bag less vacuums (75.4 ± 7.89 µg min^{-1}). Particle (8–322 nm) number emission rates ranged from 5.29×10^{11} (washable filter bag less vacuum) to 21.2×10^{11} (wet vacuum) particles min^{-1}. 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_{10} contents during vacuuming suggested motor emissions. Organic compounds in PM_{10} 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.

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

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

Household activities, such as cooking, smoking, hair spraying/drying, candle/incense burning or vacuuming, have been reported to generate considerable amounts of particulate matter indoors [18–20

], which may have a strong influence on short-term exposure [21]. Isaxon et al. [18] evaluated the influence of household activities in 22 homes in Sweden on indoor airborne particles (number concentration and black carbon). The authors reported that despite the transient nature of indoor sources, they rapidly generate particulate peak concentrations. He et al. [20] quantified the effect of 20 different household activities on indoor particle mass and number concentrations. The authors reported that depending on the type of source and housing characteristics, indoor particles increased distinctly. The influence of nine specific sources on particulate matter number size distribution and mass concentration was evaluated individually in an empty laboratory by Glytsos et al. [19]. High particle number concentrations during activation of the distinct sources and a great influence of the source type on particle number size distributions were observed. Studies carried out to assess household sources of PM reported that vacuuming can significantly elevate indoor PM concentrations [22–24] with a very high total lung deposition fraction by number [25].

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

Some studies reported in the literature were focused on the operation of vacuum cleaners and their impacts on particle mass and number levels, both in laboratory chambers [21,28–31] and under real life conditions [18,20,32]. Additionally, a number of studies also included bioaerosol levels associated with vacuuming [28,30,31]. Although many studies have investigated particulate mass and number emissions during vacuum cleaning operations, an important gap in knowledge still exists with respect to the chemical characteristics of the released particles. The characterisation of the chemical composition of particles arising from specific indoor sources is of great interest due to the risk associated with specific PM components and the possibility of using certain compounds as tracers for source apportionment in indoor environments [33,34]. Regarding PM characterisation, Szymczak et al. [35] reported ultrafine particles from a commercial professional vacuum motor consisting almost entirely of copper. Vu et al. [25] suggested that particles released from the vacuum cleaner motor were possibly carbon internal void aggregates. The authors' hypothesis was based on the finding that particles generated from vacuum cleaning were found to be nearly hydrophobic with an average growth factor around 0.98–1.10 for particle sizes of 50 and 100 nm. Isaxon et al. [18] reported an increase in black carbon levels during vacuum cleaning.

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

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

2. Materials and methods

2.1. Sampling sites and strategy

Three cylinder vacuum cleaners (washable filter bag less vacuum, wet vacuum, bagged vacuum) and a HEPA filter equipped robot were temporarily borrowed from Spanish homeowners for testing (Table 1). These devices were selected because cylinder vacuum cleaners are the prevalent type in the EU with a market share of 68% in 2016, whilst robotic cleaners have shown an increasing sales trend [37]. Measurements were performed in the living room (volume = 91.9 m^3) of a suburban Spanish house in León from October to November 2017. Similarly to the approach described by Vu et al. [25], Wu et al. [36] and Corsi et al. [32], during the monitoring campaign there were no other activities in the house and the measurements were carried out in a closed room (all the doors and windows were closed) to achieve minimum ventilation conditions. Ventilation rates, estimated by the CO_2 concentration decay method as described by Alves at al. [38], ranged between 0.24 and 0.62 h^{-1}. The average estimates of ventilation rates are presented in Table 1. On average, 45 min measurements were conducted during vacuum cleaning. Only the person responsible for carrying out the activity was present in the room during the experiments. The living room tiled floor and rugs (two cut pile carpet/rug and one long threads shag rug) were vacuumed twice with each vacuum cleaner at least one week apart. After the household activity ceased, the room was kept completely empty and closed until the restoration of particle concentration to the original level. Background indoor air measurements were also performed in the living room during which no activities were conducted in the house. The temperature (accuracy ± 0.5 °C), relative humidity (accuracy ± 3.0% with probe at 25 °C) and CO_2 (accuracy ± 3.0% of reading with probe at 25 °C) were continuously monitored with an indoor air quality probe (TSI, model 7545). Temperature and relative humidity ranged between 19.7 and 26.3 °C

Table 1
Characteristics of the vacuums tested, sampling conditions and PM_{10} mass and particle 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 (α, h^{-1})	0.29 ± 0.06	0.42 ± 0.05	0.50 ± 0.16	0.47 ± 0.07
T (°C)	24.6 ± 2.40	20.6 ± 1.30	21.7 ± 0.31	20.4 ± 0.07
HR (%)	38.2 ± 10.7	39.8 ± 3.66	37.8 ± 1.05	35.7 ± 1.22
PM_{10} initial mass concentration ($\mu g\ m^{-3}$)	30.5 ± 9.19	18.0 ± 9.90	42.0 ± 41.0	23.5 ± 0.707
PM_{10} peak mass concentration ($\mu g\ m^{-3}$)	51.0 ± 8.49	37.5 ± 4.95	65.0 ± 42.4	26.0 ± 1.41
$PM_{2.5}/PM_{10}$ (%)	74.2 ± 10.1	81.2 ± 4.45	81.1 ± 16.4	43.3 ± 3.61
PM_1/PM_{10} (%)	72.1 ± 10.6	79.4 ± 4.92	79.4 ± 17.7	39.2 ± 4.00
Particle number initial concentration (particles $\times\ 10^3\ cm^3$)	6.59 ± 4.16	3.57 ± 0.38	19.7 ± 16.9	4.88 ± 1.34
Particle peak number concentration (particles $\times\ 10^5\ cm^3$)	0.548 ± 0.014	2.10 ± 0.136	1.39 ± 0.102	0.050 ± 0.011
Emission rate PM_{10} ($\mu g\ s^{-1}$)	1.26 ± 0.131	1.44 ± 0.282	3.46 ± 1.65	–
Emission rate particle number (particles $\times\ 10^{11}\ min^{-1}$)	5.29 ± 1.48	21.2 ± 2.10	12.6 ± 4.54	–

and between 30.6 and 45.7%, respectively, for the whole set of measurements (Table 1).

Real time size segregated particulate concentrations (PM_1, $PM_{2.5}$, PM_{10}) were recorded using a DustTrak monitor (TSI, DRX 8533). Real time particle size distributions and number concentrations in the range from 8 to 322 nm were measured using a Scanning Mobility Particle Spectrometer (SMPS, TSI Incorporated). The SMPS consists of an electrostatic classifier (TSI, Model 3071) and a condensation particle counter (TSI, Model 3022). The aerosol was sampled through polyethylene tubing. All reported data has been corrected for diffusion losses using equations described in Kulkarni et al. [39]for small particles and impaction/settling losses for larger particles as a function of size [40].

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

2.2. Analytical techniques

The carbonaceous content in the PM_{10} samples (quartz filters) was analysed by a thermal optical transmission technique. The method includes controlled heating steps under inert (N_2) and oxidising (N_2 with 4% of O_2) atmospheres. The carbonaceous content of the sample can be divided into organic carbon (OC), pyrolysed carbon (PC) and elemental carbon (EC). PC, which is produced from organic carbon during heating under inert atmosphere, was determined measuring the filter light transmittance through a laser beam and a photodetector. The OC/EC determination is based on the quantification of the CO_2 released by a non-dispersive infrared (NDIR) analyser. The latter was daily calibrated with standard CO_2 cylinders and the recovery was periodically verified by analysing filters impregnated with known amounts of potassium phthalate.

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

Two 47 mm diameter punches of each quartz filter were extracted first with dichloromethane and then with methanol. The total organic extracts were fractionated by flash chromatography using eluents of increasing polarity through an activated silica-gel column. Four different fractions resulted from this process: (i) aliphatics, (ii) polycyclic aromatic hydrocarbons, (iii) n-alkanols, phenols, sterols and other hydroxyl compounds and (iv) acids and sugars. The different organic fractions were concentrated and dried by a gentle nitrogen stream before analysis. Extracts (i) and (ii) were analysed in a gas chromatograph-mass spectrometer (GC–MS) from Shimadzu. Extracts with oxygenated compounds were analysed in a GC-MS from Thermo Scientific. These latter fractions (iii and iv) included polar compounds, which required derivatisation before analysis. N,O-bis(trimethylsilyl)trifluoroacetamide (BSTFA): trimethylchlorosilane (TMCS) 99:1 (Supelco 33149-U)

was used as silylation reagent. The GC-MS calibrations were performed with injection of about 150 authentic standards (Sigma-Aldrich) at least at four different concentration levels. Standards and samples were both co-injected with internal standards: tetracosane-d50 and 1-chlorohexadecane. Additionally, for PAHs determination, a mixture of six deuterated compounds (1,4-dichlorobenzene-d4, naphthalene-d8, acenaphthene-d10, phenanthreme-d10, chrysene-d12, perylene-d12), was used. The organic extracts were injected in the single ion monitoring and total ion chromatogram modes and the compound identification was based on comparison of the mass spectra with the Wiley and NIST mass spectral libraries, comparison with authentic standards and analysis of fragmentation patterns [45]. A description of recovery efficiency tests for several compounds can be found in Oliveira et al. [46]. Field blanks were used to account for artefacts associated with transport, handling, and storage, as described in the EN 12341:2014 [41]. These filters were analysed in the same way as samples and the data obtained was subtracted from the samples in order to obtain corrected results.

3. Results

3.1. Particulate matter

3.1.1. Mass concentrations

Fig. 1 depicts the time resolved PM_{10} mass concentrations during the operation of the four vacuum cleaners. On average, increases of 1.61 ± 0.636, 1.48 ± 0.323 and 1.22 ± 0.035 times over the PM_{10} initial concentrations (before the activation of the source) were observed during the operation of the bagged, wet and washable filter bag less vacuums, respectively. No increase in PM_{10} mass concentrations was experienced during the HEPA filter vacuum cleaner operation. The increase in PM concentrations during vacuuming is determined by the dust collection efficiency, filtration elements employed and degree of reentrainment of already collected particles [47]. The reentrainment of collected dust particles was found by Trakumas et al. [47] to be higher for cyclonic and wet collectors. However, the authors highlighted that filter bag collectors also reemitted particles after being loaded, depending on the particulate load and the type of filter material used in the bag.

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

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

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

where V is the room volume, C_{in} and C_{in0} are the peak and initial indoor particle concentrations, respectively, α is the average air exchange rate, α + κ is the average removal rate and Δt is the time difference be-
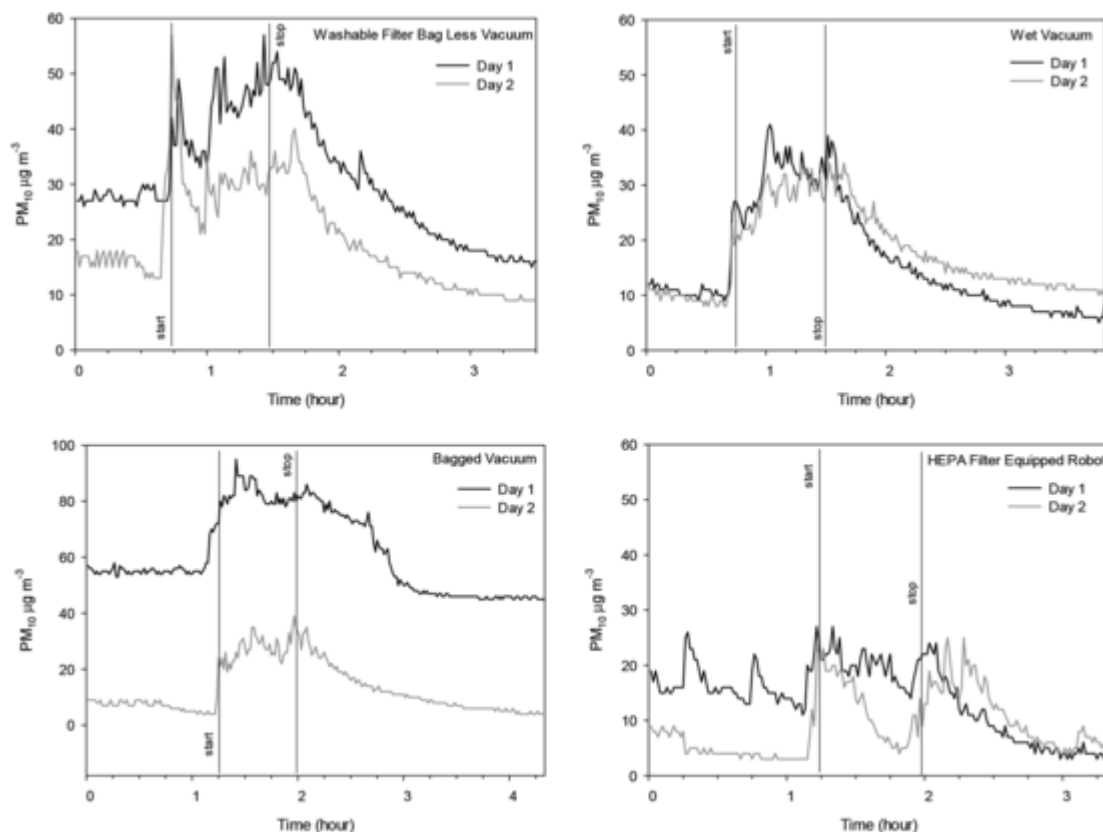
Fig. 1. PM_{10} mass concentrations during vacuum cleaning.

tween the initial and peak particle concentration. The particle removal rate is the slope obtained by plotting $\ln(C_{in}/C_{in0})$ versus time [22,51,52].

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

3.1.2. Number concentrations

Fig. 2 illustrates the time evolution of the total particle number concentration during vacuuming. An increase in particle number concentrations was observed close after the activation of the source. The average particle number concentration in the room was higher during the operation of the wet vacuum ($1.69 \times 10^5 \pm 7.54 \times 10^2$ particles cm^{-3}) and the bagged vacuum ($1.09 \times 10^5 \pm 4.95 \times 10^3$ particles cm^{-3}). The HEPA filter equipped vacuum cleaner did not increase the number of particles in the room. During its operation, the particle number concentration was $4.53 \times 10^3 \pm 8.16 \times 10^2$ particles cm^{-3}, which was similar to the one recorded before the vacuum operation (5.86×10^3) and after the robot was turned off (4.38×10^3).

These results are in accordance with previous studies reporting very high ($> 99\%$) fine particle collection efficiency of vacuums equipped with a HEPA filter [29,53]. Manigrasso et al. [54] documented a de-

crease in particle number concentration compared to background levels when using a HEPA filter equipped vacuum cleaner, suggesting that the filter removes particles from the ambient air to some extent. The ratios of peak to background levels for ultrafine particle number concentrations presented in Table 1 indicate that vacuuming can elevate concentrations by a factor ranging from 4 (bagged vacuum) to 61 (wet vacuum). Table 1 also provides estimates of particle number emission rates for the different vacuums, which were calculated as described above for particulate mass emissions (Equation (1)). A previous study conducted by He et al. [20] in suburban Brisbane households reported submicrometer particle emission rates of $0.97 \pm 1.57 \times 10^{11}$ particles min^{-1} (particles from 0.007 to 0.808 µm). Knibbs et al. [28] measured particle number emission rates from 21 vacuum cleaners in the range from 0.004 to 108×10^9 particles min^{-1} (particles from 0.54 to 20 µm). The assessment of vacuum cleaning in a full-scale chamber carried out by Afshari et al. [21] resulted in an emission rate of 0.35×10^{11} particles min^{-1} (particles from 0.02 and about 1.0 µm). Wu et al. [36] tested 3 different scenarios of vacuum cleaning in a closed living room, including normal condition, filter removed, and filter and dust bag removed. The ultrafine particle number emission rates (from 0.0146 to 0.6612 µm) for the operation with no filter and without filter and dust bag was 2.2 and 2.5 times higher than that of the normal scenario ($1.32 \pm 0.58 \times 10^{10}$ particles min^{-1}). In the present study, the average emission rates (particles from 0.008 to 0.322 µm) were estimated to range from 5.29×10^{11} to 21.2×10^{11} particles min^{-1}.

Emissions should be compared with caution since the differences between vacuum cleaners (model, age, state of preservation, etc.), sampling conditions (real life monitoring campaigns vs laboratory chambers) and particle diameters may lead to distinct results. In real life/ on site monitoring campaigns, several factors, such as building characteristics, ventilation conditions, concurrent activities, cleaning rou-
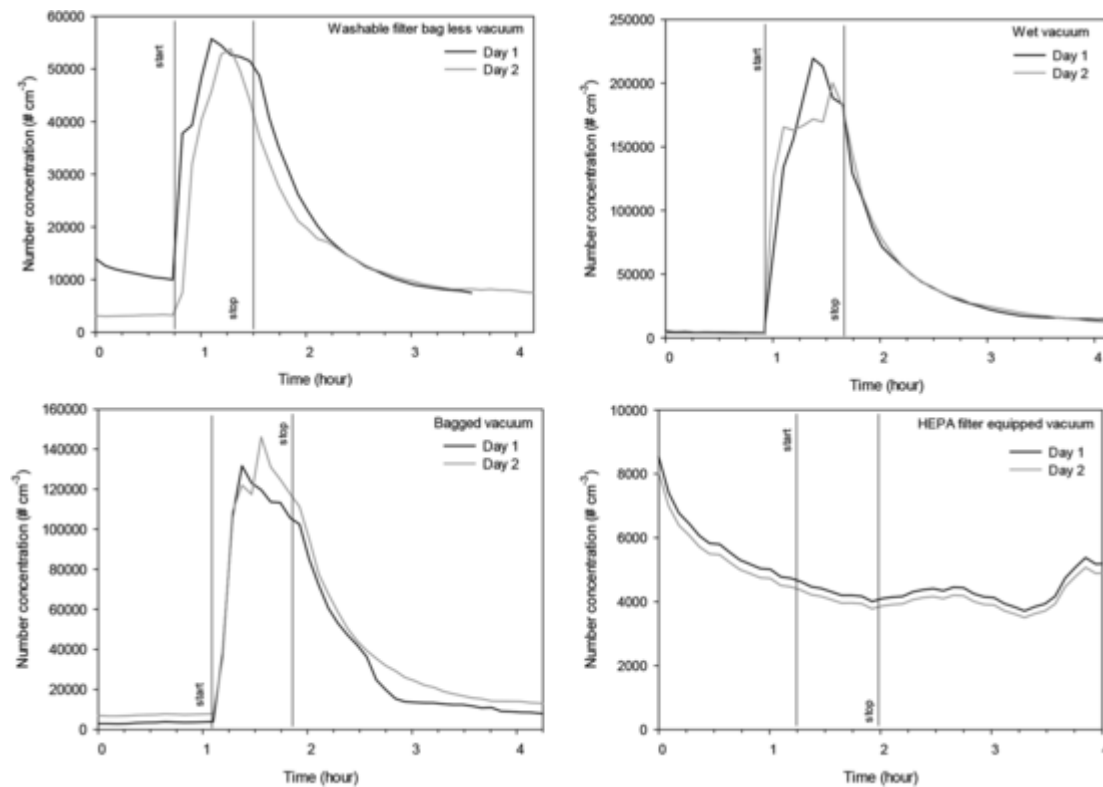
Fig. 2. Particle number concentrations during vacuum cleaning.

tines, etc., should also be taken into account. In fact, He et al. [20] obtained variable results in different houses when vacuumed. In one of the houses, a doubling in $PM_{2.5}$ concentrations was observed during vacuuming compared to background levels, while no increase was noticed in particle number concentrations. However, in a different house, the opposite behaviour was registered, with no increase in the $PM_{2.5}$ mass concentrations, while the particle number concentration increased. The authors pointed out the differences in vacuums and in cleaning routines as possible reasons behind the observed results. House cleaning routines can affect both the dust resuspension and the dust loads available to vacuum, which, in turn, may affect the particle reemission. The effect of the vacuum cleaner or vacuum cleaner components on emissions can also be significant. Afshari et al. [21] investigated fine particle emissions when running a vacuum cleaner in a full-scale chamber. Two experiments were carried out: (i) vacuum cleaner operated with a dust bag and (ii) vacuum cleaner operated without dust bag, filters and hose in order to study the emissions from the motor only. The results revealed that the particle concentrations originating from the motor were higher than those from the vacuum cleaner with a bag.

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

3.2. Metals

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

The contribution of Cu to the PM_{10} mass ranged from 0.01 wt.% (HEPA equipped robot) to 8.89 wt.% (bagged vacuum), which represented an increase over background concentrations ranging from 1.6 to 848 times. Metals contribute to 20–30% of the total weight of a vacuum cleaner. The metallic components are made of aluminum (motor and screws), stainless and galvanized steel (motor), brass (plug) and copper (plug, power cord, wire cables and motor) [56]. Cu concentrations up to 55 μg m^{-3} were previously reported in particulate matter emissions from a professional vacuum cleaner in a test room [35].

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

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

In the present study, Si was used as reference element due to its high abundance in the earth's crust. The average element concentrations in the upper continental crust were taken from Wedepohl [57]. During vacuuming, minimal enrichments were obtained for some elements, such as Al, Mg, K, Fe, V, and Mn (EF < 5), indicating that these elements were mostly derived from soil dust. Rasmussen et al. [58] found significant relationships between concentrations in household settled dust and airborne particulate matter for several elements, namely Ag, Al, As, B, Cu, Fe, Mn, Pb, U, V and Zn. In the present study, other elements like P, Ca, Ni, S and Cl were enriched (Fig. 4) during the operation of every vacuum cleaner tested and also in the background sample suggesting that the origin of the enrichment was not vacuuming. Zinc (132 < EF < 163) and selenium (743 < EF < 1285) were also highly enriched elements in all vacuuming tests and back-
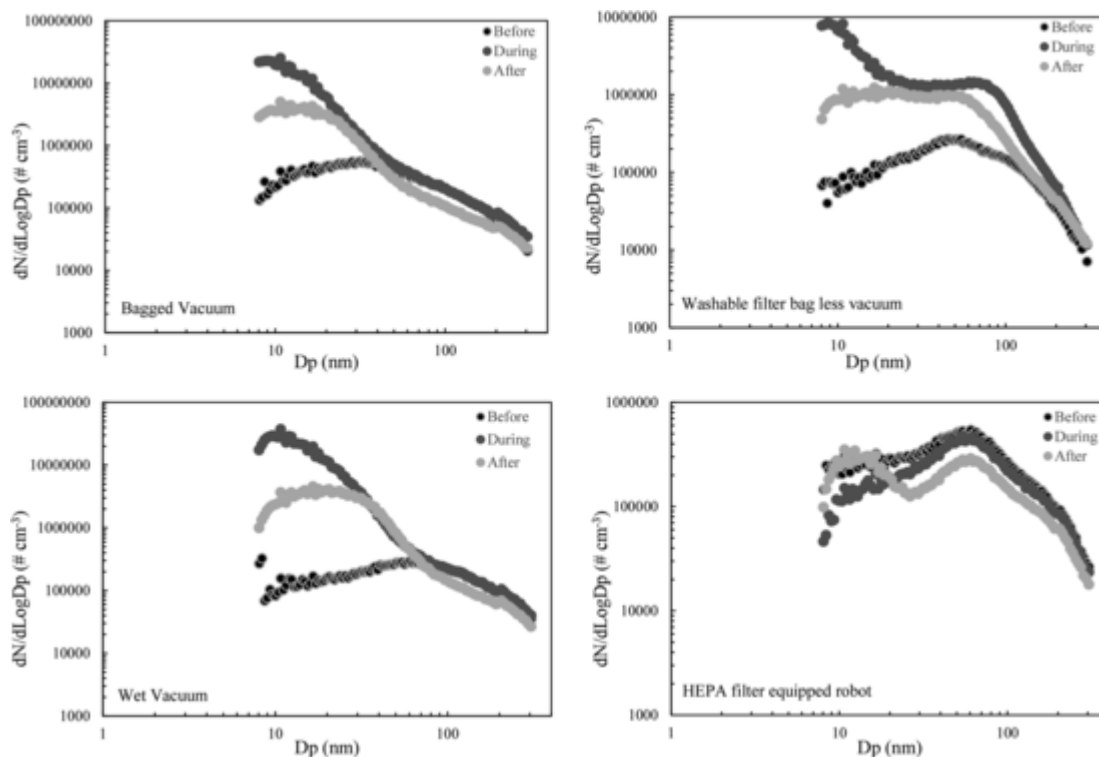
Fig. 3. Typical average evolution of the mean particle size distribution before, during and after vacuum cleaning.

ground sample (EF = 419 and EF = 16721 Zn and Se, respectively). Molybdenum was highly enriched (675 < EF < 37240) in all the vacuuming samples except in the one collected during the wet vacuum operation, whereas it was not enriched in the background sample. Copper EFs were very high when operating all the vacuum cleaners (over 6000, 25,000 and 41,000 for the washable filter bag less, wet and bagged vacuum cleaners, respectively), except for the HEPA equipped robot (EF < 100) (Fig. 4).

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

$$EC = (CA \times ET \times EF \times ED) / AT \qquad (3)$$

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

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

$$Risk = IUR \times EC \qquad (4)$$

The IUR values were retrieved from the database provided by USEPA [60] for three elements (As, Cr (VI), and Pb). In the present study, one seventh of total Cr concentration was used to estimate the risk considering that the tabulated IUR is for Cr (VI), which is based on a Cr (III): Cr (VI) proportion of 1/6. USEPA considers that a 10^{-6} risk is below the level of apprehension, while risks above 1.0×10^{-4} are

of concern. In the present study, the cumulative cancer risk for all potential carcinogenic elements was negligible (always < 5×10^{-7}).

Noncarcinogenic risks associated with inhalation exposure to trace elements in indoor PM_{10} were estimated by the noncancer hazard quotient (HQ) following the methodology proposed by USEPA [59,61]:

$$HQ = EC / R_fC \qquad (5)$$

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

3.3. OC/EC

Total carbon (TC) represented from 32.7 ± 2.68 (washable filter bag less vacuum) to 51.6 ± 2.08 (wet vacuum) wt.% of the PM_{10} mass during vacuuming, corresponding to TC increases over background levels ranging from 1.2 to 1.8. EC levels were distinctively higher during the operation of the wet (19.0 wt.% PM_{10} mass) and bagged (15.4 wt.% PM_{10} mass) vacuum cleaners. For the HEPA filter equipped vacuum cleaner, as well as in background air samples, EC was not present at detectable levels (Fig. 5). OC and EC concentrations in samples collected when the vacuum cleaners were run were not correlated with each other, indicating distinct sources. Contrarily, good correlations were found between particulate EC concentrations and both Cu ($r^2 = 0.87$) and Ni concentrations ($r^2 = 0.79$). Good correlations ($r^2 > 0.75$) between OC and several elements, including Si, S, Cl, K, Ca, Fe, Zn and Se, were also recorded. Given that one of the main contributors to indoor particles is probably resuspended dust, some

Table 2
PM$_{10}$ 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	4.68	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

bdl – below the detection limit. The measured element concentrations were converted into the respective mass concentrations of the most common oxides (SiO$_2$, Al$_2$O$_3$, MgO, MnO, Fe$_2$O$_3$, TiO$_2$, K$_2$O, etc.).

of which associated with soil, these correlations are not surprising. Household PM$_{10}$ dust has been reported to contain appreciable amounts of carbonaceous particles, mainly OC, whereas in many samples EC was too low or undetectable [64]. Black carbon, on the other hand,

has been associated with motor emissions from vacuum cleaners [18]. OC to EC ratios showed high variability, ranging from 1.7 (bagged vacuum) to 106 (washable filter bag less vacuum). Habre et al. [65] found that household PM$_{2.5}$ OC fractions were mainly related to human activities, including vacuum cleaning, which leads to resuspension of dust and PM$_{2.5}$ generation. Alves et al. [38] reported average OC/EC values ranging from 4.2 to 9.7 in school classrooms. The researchers argued that these ratios were expected, since resuspended dust, some of which associated with soil, was found to be one of the main primary contributors to indoor particles.

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

3.4. Organics

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

The aliphatic fraction of particulate matter comprised n-alkanes from C$_{11}$ to C$_{35}$. The maximum concentrations were observed for the homologues in the range from C$_{20}$ to C$_{22}$, which have been described as characteristic of petrogenic sources. The carbon preference indices of n-alkanes were in the range from 0.7 to 0.9 during the vacuuming operation and 1.1 for the background, suggesting the contribution of petroleum derivatives [68]. The presence of these compounds may be related to oil-based or petrochemical textiles such as nylon, polyester, acrylic and spandex, which are made from natural gas or oil. Outgassing of lubricants applied to parts of the vacuum cleaners is another likely source. The Σ$_{25}$ n-alkane concentrations ranged from 22.4 to 39.3 ng m^{-3} during vacuuming and 17.3 ng m^{-3} in the background air.

Discontinuous series of n-alkanols from C$_{10}$ to C$_{30}$ were detected in the PM$_{10}$ samples, maximising at C$_{16}$, during the operation of the
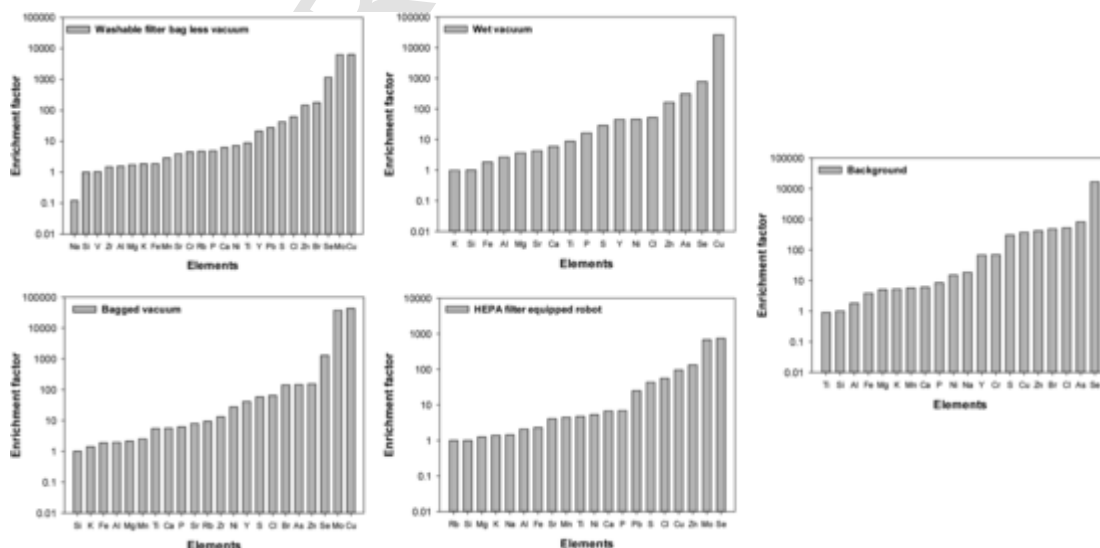

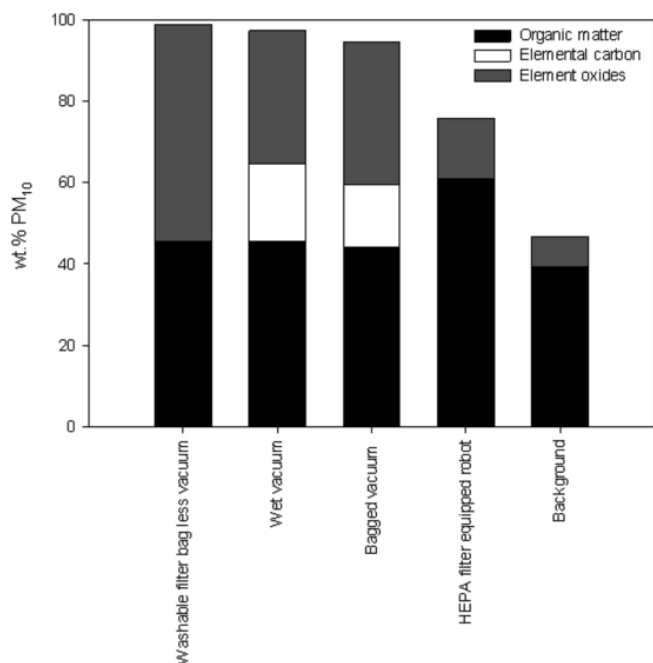**Fig. 4.** Enrichment factors of elements in PM$_{10}$ sampled during vacuum cleaning.

Fig. 5. Chemical mass closure of PM_{10}.

tested vacuums, as well as in the background sample (Table 3). Cetyl alcohol ($C_{16}H_{34}O$) is widely used in skin lotions and creams due to its water-binding properties [69]. Other long-chain alcohols, such as myristyl ($C_{14}H_{30}O$) and stearyl alcohol ($C_{18}H_{38}O$), were also found in all samples. Besides being used in a variety of cosmetic products as emulsifier, emollient, antifoaming agent, and surfactant, stearyl alcohol has also been isolated from human sebaceous lipids [69]. The Σ_{15} n-alkanol concentrations ranged from 362 to 858 ng m^{-3} during vacuuming and 172 ng m^{-3} in the background air.

A series of n-alkanoic acids from C_8 to C_{22}, maximising at C_{14} or C_{16}, were identified (Table 3). Sources of alkanoic acids include cooking [70,71], emission from people's skin oils [72], incense burning [73] and biomass combustion [74,75]. Other sources include biogenic contributors, such as fungi, bacteria, spores, and pollen [76]. Lower molecular weight n-alkanoic acids ($<C_{18}$) were found in emissions from fossil fuel combustion, road dust and tyre wear debris [e.g.,76,77]. Zhao et al. [71] pointed out tetradecanoic (myristic) acid as a possible organic marker to distinguish emissions from Chinese cooking. The Σ_{14} n-alkanoic acid concentrations ranged from 111 to 951 ng m^{-3} during vacuuming, while the background air sample presented a concentration of 472 ng m^{-3}, indicating that these compounds are originated from household's activities.

Among diacids, the compound with highest concentrations was adipic acid (or C_6 diacid) (Table 3). Dicarboxylic acids from C_4 to C_8 were recorded in fine organic aerosols from charbroilers and meat cooking operations by Rogge et al. [78]. The authors identified hexanedioic acid as one of the dominant compounds. Malic acid was only present in the sample collected when the HEPA filter equipped robot was operated. Röhrl and Lammel [79] pointed out the influence of biogenic sources on the occurrence of malic acid. The Σ_7 dicarboxylic acids concentrations ranged from 3.00 to 25.7 ng m^{-3} during vacuum cleaning, while the background air sample presented a concentration of 8.82 ng m^{-3}.

Several phenolic compounds were also detected in the samples; 2,4-di-tert-butylphenol, bisphenol A and 4-tert-butylphenol were the most abundant (Table 3). Alkylphenols, such as 4-tert-butylphenol and 2,4-di-tert-butylphenol, have a large variety of usages such as emulsifying agents in latex paints, glue, and pesticide ingredients, in the prepara-

tion of antioxidants, curing agents, and heat stabilisers for polymer resins, among others [80]. In the present study, concentrations ranging from 106 to 180 ng m^{-3} and from 3.03 to 24.8 ng m^{-3} were registered for 2,4-di-tert-butylphenol and 4-tert-butylphenol, respectively. These compounds were also detected in background air samples. Indoors, a source of alkylphenols is the biodegradation of alkylphenol ethoxylate, which is a widely used surfactant in detergents. They can also be released from the surface of polymer resins, which are used as antioxidant for wall or floor coverings [80]. Bisphenol A can act both as a plasticiser and as a fungicide and is used in the production of polycarbonate and epoxy resins [81]. This compound is ubiquitous in the atmosphere and its size distributions showed peaks in both fine and coarse fractions. Soil resuspension has been suggested as a main source for bisphenol A in the coarse fraction [82]. Bisphenol A is a decomposition product of polycarbonate, an ubiquitous material indoors (e.g. hard plastic bottles, CDs, DVDs, etc.) [83]. Isoeugenol was another phenolic compound detected in three of the four samples collected in the living room during vacuuming (1.78–3.92 ng m^{-3}) and was absent from the background air. Eugenol was detected in all samples although in much lower concentrations. Phenylpropenes, such as eugenol and isoeugenol, are produced by plants as defense compounds and as floral attractants of pollinators [84]. Isoeugenol is used in fragrance formulations which are incorporated into household laundry and cleaning products [85]. Eugenol is also common in consumer products, such as air fresheners [86]. This compound was also reported in samples collected in rooms from a Spanish household that had been treated with aerosols, electrical diffusion units, as well as with several cleaning products of domestic use [87]. Thymol was present in all samples and was among the phenolic compounds with the highest concentrations. Thyme essential oils are used in a variety of products in the food industry (preservatives and flavourings) and in cosmetics [88].

Levoglucosan and its stereoisomers, mannosan and galactosan, were detected in samples (Table 3). These saccharides are formed from the thermal degradation of cellulose [89–91]. Although their individual quantification was not possible, many other saccharides were detected in PM_{10} samples.

Cholesterol and β-sitosterol were also found in PM samples. Cholesterol was the most abundant. It is likely associated with cooking activities [92,93].

Other hydroxyl compounds and phthalates were detected in the particulate matter organic extracts (Table 3). Among these, the most abundant were diethyltoluamide (DEET), tris(1,3-dichloroisopropyl)phosphate and Irgafos 168 (tris(2,4-di-tert-butylphenyl)phosphite). Tris(1,3-dichloro-2-propyl)phosphate was not detected in two of the five samples, reaching a concentration of 52.9 ng m^{-3} during the wet vacuum operation, which was up to 100 times higher than the background concentration. Flame retardants are used in many consumer and industrial products (e.g. electronics and electrical, building/construction, and textiles) to delay ignition and slow the spread of fire. Organic phosphorous containing flame retardants are mainly used in cellulosic materials, textiles, PVC-based products and polyurethane foam [94]. Air concentrations of this phosphate triester in European homes ranged from no detectable concentrations to 21 ng m^{-3} [95]. Diethyltoluamide was present in all samples. It is used as insect repellent [96]. Irgafos 168 (tris(2,4-di-tert-butylphenyl)phosphite) is a phosphite antioxidant used in several plastic packaging [97,98]. Parsol MCX (ethylhexyl methoxycinnamate) was also detected in all samples. It is frequently contained in personal care products as UV filter to protect human skin from UV radiation or as UV absorber to prevent light-induced product degradation [99].

Several plasticisers were detected in PM samples. Di-n-butylphthalate (DBP) was the most abundant phthalate plasticiser present in the samples during vacuuming. In the background sample, only dimethyl phthalate was detected at quantifiable levels (Table 3). Plasticisers

Table 3
Concentrations (ng m^{-3}) of oxygenated organic compounds in PM$_{10}$.

	Wet vacuum	Bagged vacuum	HEPA filter equipped robot	Washable filter bag less vacuum	Background
Saccharides					
Galactosan	2.41	bdl	bdl	bdl	bdl
Mannosan	1.15	2.57	2.74	bdl	1.27
Levoglucosan	8.27	9.69	23.6	5.93	22.1
Unidentified saccharides	45.2	47.4	46.5	278	4.50
Phenolics and alteration products					
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
Phthalic 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	2.24	0.063	4.74	1.63	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	8.90	bdl	46.9	0.543
Aliphatic alcohols					
1-Decanol	0.014	bdl	0.517	0.018	0.014
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	0.403	3.72	bdl	bdl
1-Tricosanol	0.168	0.033	0.183	0.198	0.047
1-Pentacosanol	0.201	0.065	2.09	0.450	0.035
Hexacosanol	3.09	3.49	3.23	18.7	0.820
1-Heptacosanol	0.055	0.023	0.043	0.162	0.066
1-Octacosanol	0.823	0.410	0.565	5.23	0.094
1-Tricontanol	0.387	0.177	0.165	0.427	0.033
Steroid compounds					
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
Aliphatic acids					
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

Table 3 (*Continued*)

	Wet vacuum	Bagged vacuum	HEPA filter equipped robot	Washable filter bag less vacuum	Background
Octadecanoic acid	15.8	14.0	107	85.4	61.3
Nonadecanoic acid	0.089	0.097	0.096	0.198	0.109
Eicosanoic acid	0.229	0.124	bdl	0.844	0.443
Docosanoic acid	0.053	0.045	0.334	0.543	1.76
Diacids					
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
Other acids					
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	20.4
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	bdl	0.016	0.055	0.154	0.003
Glycerol derivatives					
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	bdl	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
Other compounds					
2,6-Di-tert-butyl-1,4-benzoquinone	0.933	1.33	0.520	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-Pinanediol	0.091	0.300	0.628	0.255	0.064
Diethyltoluamide (DEET)	19.6	51.8	66.6	76.9	44.1
Tributyl phosphate (TBP)	bdl	118	bdl	bdl	0.922
Tetraacetylethylenediamine (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	31.4
Plasticisers					
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	0.001	bdl	bdl	0.155	bdl
Dimethyl phthalate	0.100	nd	nd	0.100	0.010

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

are widely used in the production of polyvinyl chloride (PVC) plastics, as well as in other applications such as glues, paints and cosmetics [100,101]. The vacuum body (external structure, dust container, power cord and wire cables) is made of several plastic components, including polypropylene (PP), acrylonitrile butadiene styrene (ABS), high density polyethylene (HDPE) and PVC materials. Other vacuum parts, such as the flexible hose, hose collaer and handle, floor brush and extension tube are made of ethylene vinyl acetate (EVA), PP and

HDPE [56]. Plasticisers can leach out from PVC with materials aging and contaminate the environment [102].

The Σ_{19} PAHs concentrations ranged from 3.68 to 11.8 ng m^{-3} during vacuuming and 3.32 ng m^{-3} in the background air (Table 4). The PAHs with highest concentrations were pyrene, chrysene and benzo[b]fluoranthene. Delgado-Saborit et al. [103] measured sixteen PM-bound PAHs in 81 English households and reported concentrations ranging from undetectable levels to 25 ng m^{-3}. In Italy, Romagnoli et al. [104] documented concentrations of Σ_8 PAHs in 10 private house-

Table 4
Concentrations of PAHs (ng m^{-3}), carcinogenic potency of total PAHs (BaP$_{eq}$, ng m^{-3}) and 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
Σ PAHs	11.8	8.33	3.68	10.6	3.32
BaP$_{eq}$	1.19	0.535	0.300	1.22	0.388
Cancer Risk	1.6×10^{-8}	7.4×10^{-9}	4.1×10^{-9}	1.7×10^{-8}	5.3×10^{-9}

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

holds in the range from 0.4 to 8.4 ng m^{-3}. Higher PAH concentrations were determined by Naumova et al. [105] in 55 non-smoking urban residences in the USA. The indoor concentrations of Σ_{30} PAHs were 16–220 ng m^{-3} in Los Angeles, 21–310 ng m^{-3} in Houston, and 22–350 ng m^{-3} in Elizabeth.

Benzo[a]pyrene equivalent concentrations (BaP$_{eq}$) were calculated (Table 4) multiplying the measured levels of each PAH by the respective toxic equivalent factors (TEF), which were taken from Bari et al. [106]. Dibenzo[a,h]anthracene was the compound that most contributed to the carcinogenic potential of the PAH mixture for almost all the samples with values ranging from 33.8% (washable filter bag less vacuum) to 48.3% (bagged vacuum). During the wet vacuum operation and in the background sample, the major contributor was benzo[a]pyrene accounting for 35.8% and 37.2% to the carcinogenic potential, respectively.

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

4. Conclusions

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

This study presents indoor particulate mass and number emission rates and a comprehensive PM$_{10}$ chemical characterisation during vac-

uuming cleaning with different devices (washable filter bag less vacuum, wet vacuum, bagged vacuum) without any other active source. A sharp increase in particle number concentrations was recorded when using most vacuum cleaners (4–61-fold in relation to background air levels). The increase in the PM$_{10}$ mass concentrations due to vacuuming was less pronounced, ranging from 1.2 to 1.6 in comparison with the initial concentrations (before the activation of the source). While the bagged vacuum cleaner presented the highest PM$_{10}$ emission rates, the particle (8–322 nm) number emission rates were highest during the wet vacuum operation. No increase in PM$_{10}$ mass concentrations or ultrafine particle number was observed when using the HEPA filter equipped vacuum cleaner.

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

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

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

Declaration of competing interest

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.

References