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# 1 Impact of ironing on indoor particle levels and composition

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### 13 Abstract

Domestic chores are an important part of the household's daily routine and can contribute 14 15 significantly to personal exposure. In this study, the particulate mass and number concentrations were assessed when using two irons (steam iron and steam iron with 16 17 boiler) under distinct conditions (minimum ventilation and indoor doors open). The detailed PM<sub>10</sub> chemical characterisation included organic and elemental carbon, elements 18 and organic speciation. Particle number emission rates ranged from  $8.1 \pm 0.09 \times 10^{11}$  to 19  $15 \pm 3.4 \times 10^{11}$  particles min<sup>-1</sup>. Ratios of peak to background levels indicate that ironing 20 21 can elevate the ultrafine particle number concentrations by a factor ranging from 35 to 194.  $PM_{10}$  emission rates from steam iron, under minimum ventilation conditions (6.6 ± 22 1.4  $\mu$ g s<sup>-1</sup>), were higher than those from steam iron with the doors open (1.9 ± 1.6  $\mu$ g s<sup>-1</sup>). 23 24 The highest particle number and mass emission rates were recorded when the steam iron with boiler was used. Regarding the chemical composition of particles, elemental carbon 25 26 and strontium were only detected during ironing. Bromide concentrations increased noticeably over background levels (9 to 51 times) during ironing. PM<sub>10</sub> samples 27 28 encompassed a wide range of organic compounds, part of which can be attributed to the handling of textiles and the use of detergents, fabric softeners, cosmetics and personal 29 30 care products. Substances emitted by volatilisation or shedding of textile fibres, or due to 31 handling, can contribute to human exposure through inhalation. The cancer risks 32 associated with inhalation of metals and PAH were found to be negligible.

33

Keywords: Indoor air quality, Ironing, Particulate matter, Elemental composition,
OC/EC, Organic compounds.

37 **1. Introduction** 

Every year, ambient air pollution causes millions of premature deaths globally [1]. 38 Particulate matter (PM), classified by the International Agency for Research on Cancer 39 (IARC) as carcinogenic for humans (Group 1) [2], induces adverse health effects, 40 including respiratory symptoms, exacerbation of chronic respiratory and cardiovascular 41 42 diseases and decreased lung function [3,4,5,6]. Indoor exposure to PM is likely to be an 43 important contributor to the adverse health effects since most people spend nearly 90% 44 of their time in enclosed environments, especially in residential settings [7,8,9], meaning that much of the exposure to aerosols through inhalation occurs in built environments 45 [10,11]. Indoor particulate matter includes particles that infiltrate from outdoors, particles 46 from indoor sources and particles generated by indoor air chemistry [12,13,14,15]. 47

48 A substantial body of evidence shows that several household activities, such as cooking [16,17], use of biomass burning stoves for heating [18,19,20,21,22,23,24,25] and tobacco 49 50 smoking [26,27,28,29,30,31], are significant sources of particulate matter (mass and number). Additionally, other indoor sources such as vacuuming [30,32,33,34,35,36,37, 51 52 38], use of air fresheners [30,39,40], burning candles [31,37,40,41] and ironing [30, 31,42,43,44] were also investigated. Furthermore, the physicochemical characteristics of 53 some sources of indoor PM, such as cooking [17], cigarette smoking [45], incense burning 54 [46] and use of combustion appliances for heating purposes [47] have been characterised. 55 The interest in the PM chemical composition is driven by the risk associated with specific 56 PM components (e.g. PAHs, heavy metals, etc.) and also by the possibility of using 57 58 certain compounds as tracers for source apportionment in indoor environments [12, 48]. Despite the significant data provided by the above-mentioned studies, few researches, 59 however, have focused on the characterisation of particles from other indoor sources. 60

In the European Union, it is estimated that there are more than 300 million electric irons 61 in use with a sale of about 40 million units per year [49]. In the market, there are several 62 63 iron brands offering diverse solutions, such as dry irons (nowadays sparsely in-use), steam irons, travel irons and steam irons with boiler (steam stations), each with their own 64 65 features concerning power, flow of steam, sole plate, safety mechanisms and temperature 66 adjustment [50]. Ironing is one of the most time-consuming processes of laundry 67 treatment [51]. Regarding this household task, only particle mass [43] and number [30,31,42,44] have been assessed so far. Taking into account that identification and 68 69 characterisation of indoor emission sources is crucial to develop an effective exposure

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control, the aim of the present study is to assess the impact of ironing on particulate matter mass and number concentrations in indoor air. The detailed chemical characterisation of particulate matter was obtained in order to perform a health risk assessment (carcinogenic and noncarcinogenic) from the inhalation of  $PM_{10}$  and to identify compounds that may be used as specific source tracers in future work.

75

# 76 2. Materials and methods

# 77 2.1. Sampling sites and strategy

Considering that the use of steam during ironing provides a fast effective method to 78 transmit significant heat to the fabric and remove creases [52], two different types of 79 80 steam irons (steam iron and a steam iron with boiler) were selected to carry out the measurements in the present study. The set of items that were ironed represented 81 82 approximately the clothes accumulated over a week by a family of 4 people. The clothes comprised pieces of varied composition (e.g. cotton, polyester, etc.) and included freshly 83 84 washed linen (tablecloths, bedsheets and towels) and daily garment, such as shirts, tshirts, sweatshirts, blouses, jeans and trousers. The measurements were performed in the 85 living room (volume = 91.9  $m^3$ ) of a suburban Spanish house (León) under minimum 86 ventilation conditions with both the ironing systems (all the room doors and windows 87 were kept closed - condition I). Additionally, the particulate levels generated during 88 ironing with the steam iron were also characterised while keeping the living room door 89 opened and the windows closed (condition II). On average, measurements were 90 91 conducted for three and a half hours and each test condition was repeated twice in different weeks. During the execution of the task, no other activities took place in the 92 house. However, daily life activities were conducted before the activity started since the 93 94 house was occupied. After ceasing the activity, sampling continued until the particle 95 concentration decreased to levels of the same order as the ones recorded before the activation of the source. Background indoor air measurements were also performed, in 96 97 the absence of indoor activities, during a weekend when the homeowners were away. Background levels were monitored for periods of time similar to those recorded during 98 99 sampling with ironing.

An air quality probe (model 7545, TSI) was used to monitor continuously the temperature,
relative humidity and CO<sub>2</sub> (1-min resolution). Ventilation rates were estimated by the
CO<sub>2</sub> concentration decay method as described by Alves et al. [53].

103 A real-time laser photometric instrument (DustTrak DRX 8533, TSI) was used to record 104 particulate matter mass concentrations over time ( $PM_1$ ,  $PM_{2.5}$  and  $PM_{10}$ , 1-min 105 resolution). Particle number concentrations and size distribution from 8 to 322 nm were 106 measured with a scanning mobility particle sizer (SMPS; Classifier model 3071, CPC 107 model 3022, TSI Incorporated).

108  $PM_{10}$  sampling for gravimetric and chemical analysis was carried out from the activation of the source until the iron was switched off (about 3.5 h) using simultaneously a high-109 volume sampler (MCV, model CAV-A/mb) operated at 30 m<sup>3</sup> h<sup>-1</sup> and a low volume 110 sampler (TCR TECORA, model 2.004.01) at a flow of 2.3 m<sup>3</sup> h<sup>-1</sup>. The first sampling 111 instrument was equipped with pre-weighed quartz fibre filters (150 mm diameter, 112 113 Pallflex®) while the second collected the samples on Teflon membrane filters (47 mm 114 diameter, Pall Corporation). The collected mass of particles on filters was gravimetrically 115 determined at an accuracy of 0.01 mg (XPE105 DeltaRange®, Mettler Toledo). The inlet of the samplers was placed at the breathing zone and in close proximity to the iron board. 116 117 The comparison of  $PM_{10}$  concentrations obtained from the simultaneous measurements with MCV and Tecora presented a strong linear correlation ( $r^2 = 0.96$ ) with a slope close 118 119 to the unity (0.94). The comparison between the PM<sub>10</sub> concentrations determined with the 120 gravimetric and photometric instruments also displayed strong correlations coefficients  $(r^2 = 0.92 \text{ and } r^2 = 0.96 \text{ with the MCV and Tecora, respectively}).$ 121

122

# 123 **2.2. Analytical techniques**

The organic (OC) and elemental (EC) carbonaceous content in the  $PM_{10}$  samples (quartz filters) was analysed by a thermal optical transmission technique described in detail in previous studies [54]. For mass balance purposes, OC was converted into organic matter (OM) to account for oxygen, nitrogen, hydrogen and other atoms not determined by the thermo-optical method. The multiplier factor adopted was 1.9 taking into account the presence of highly oxygenated compounds determined by gas chromatograph-mass spectrometer (GC-MS) [55].

131 Concentrations of elements with Z > 10 were measured in the PM<sub>10</sub> samples (Teflon 132 filters) by proton-induced X-ray analysis (PIXE, proton beam energy 3 MeV) at the 133 INFN-LABEC laboratory [56]. A detailed description of the method can be found 134 elsewhere [57]. For mass balance purposes, the measured element concentrations were 135 converted into the respective mass concentrations of the most common oxides (SiO<sub>2</sub>, 136 Al<sub>2</sub>O3, MgO, MnO, Fe<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, K<sub>2</sub>O, etc.).

To characterise the organic fraction, portions of the quartz filters were extracted first by 137 138 refluxing dichloromethane for 24 h and then for 10 min intervals with methanol (three times) using an ultrasonic bath. The extracts were filtered, concentrated and then 139 140 evaporated to dryness under high pure nitrogen gas. The total organic extracts were 141 fractionated by flash chromatography and analysed by GC-MS. A detailed description of the method and equipment can be found elsewhere [58]. The organic compounds were 142 identified by comparing the spectra of the samples with those in the Wiley mass spectral 143 reference libraries. Further confirmation was carried out with authentic standards. 144

Field blanks were prepared, handled and analysed by the same procedures as the samplesand the data was subtracted to obtain corrected results.

147

# 148 **2.3. Estimation of particulate emission rates**

The calculation of emission rates was carried out based on the mass-balance equation
[21,36,59]. The rate of change in indoor aerosol concentrations with respect to time is
expressed as follows (equation (1)):

152 
$$\frac{dC_{in}}{dt} = P\alpha C_{out} + \frac{Q_s}{V} - (\alpha + \kappa)C_{in}$$
(1)

where  $C_{in}$  and  $C_{out}$  are the indoor and outdoor particle concentrations, respectively, P is the penetration efficiency, k is the deposition rate,  $\alpha$  is the air exchange rate,  $Q_s$  is the indoor particle generation rate, t is time and V is the room volume.

The average emission rates were calculated assuming a penetration efficiency equal to
one, well-mixed conditions and using average values instead of functions (equation (2)).
Additional assumptions about the experimental conditions were made as described by He
et al. [60].

160 
$$\overline{Q_s} = V \times \left[ \frac{C_{in} - C_{in0}}{\Delta t} + \overline{(\alpha + \kappa)} \overline{C_{in}} - \alpha C_{in0} \right]$$
 (2)

In equation (2),  $\overline{Q_s}$  is the average particle emission rate (mass and number),  $C_{in}$  and  $C_{in0}$ are the peak and initial indoor particle concentrations (mass and number), respectively,  $\Delta t$  is the time difference between the initial and peak particle concentration,  $\overline{(\alpha + \kappa)}$  is the average removal rate and  $\alpha$  is the average air exchange rate. This equation ignores the aerosol dynamic processes such as condensation, evaporation and coagulation that contribute to formation and removal of particles [36]. 167 The average removal rate was determined as described in previous studies [61, 62, 63, 168 64]. Briefly, assuming that when an indoor source is active the contribution of outdoor 169 penetration to indoor concentrations is negligible, the decay of particles indoors after the 170 source stopped is described by equation (3):

$$171 \quad \frac{dC_{in}}{dt} = -(\alpha + \kappa)C_{in} \tag{3}$$

172 Integrating equation (3), the following expression is obtained (equation (4)):

173 
$$\ln\left(\frac{c_{in_t}}{c_{in_0}}\right) = -(\alpha + \kappa)t \tag{4}$$

174 The slope obtained from the plot of  $\ln(C_{in}/Ci_{n0})$  versus time is equal to  $-(\alpha + \kappa)$  [61, 62, 175 63, 64].

176

# 177 **2.4. Enrichment factors**

178 Enrichment factors (EFRs) were calculated to determine whether the indoor PM179 originated from crustal or non-crustal sources following equation (5):

180 
$$EFR = \frac{(E/R)_{air}}{(E/R)_{crust}}$$
 (5)

where E is the concentration of the element under analysis and R is the concentration of
the reference element. Silicon was used in this study as a reference because it is a major
constituent of the earth's crust. The average element concentrations in soil were obtained
from Wedepohl [65].

185

# 186 2.5. Health risk assessment

187 In this study, a health risk analysis for estimating the occurrence of adverse health effects 188 resulting from the inhalation of  $PM_{10}$  was performed by using inorganic elements and 189 polycyclic aromatic hydrocarbon (PAH) concentrations.

190 The reference concentration ( $R_fC$ , ng m<sup>-3</sup>) was used for toxic elements, while the 191 inhalation unit risk (IUR, ng m<sup>-3</sup>) was adopted for carcinogenic compounds (As, Cr (VI), 192 Pb, Co, Cd, Ni and benzo[a]pyrene). These values were taken from databases provided 193 by USEPA [66,67]. In the case of unavailability of reference concentrations, reference 194 doses for oral exposure ( $R_fD$ ) were used to derive  $R_fC$  values as described by USEPA 195 [68]. Regarding chromium, the total concentration was determined in this study. Taking 196 into account that it was established that the concentration ratio of Cr(VI) to Cr(III) in the

- air is about 1 to 6 [68], the excess cancer risk of Cr(VI) was calculated as one seventh oftotal Cr concentration.
- 199 The exposure concentration  $(EC_X)$  of the selected non-carcinogenic elements and 200 carcinogenic elements was calculated as follows [69]:

$$201 \quad EC_{X} = (CA \times ET \times EF \times ED) / AT$$
(6)

where  $EC_X$  is the exposure concentration (ng m<sup>-3</sup>), CA is the compound concentration 202 (ng m<sup>-3</sup>), ET is the exposure time, EF is the exposure frequency, ED is the exposure 203 204 duration and AT is the averaging time. The exposure time used in the calculations was 205 based on the results of a survey on consumer behaviour regarding laundry treatment in 206 Germany [51]. Taking into account the survey results, the exposure concentration was calculated assuming an ironing frequency of once a week for 2 h over 70 years of activity 207 208 in housekeeping during human life; therefore, these values were assumed for ET, EF, ED and AT, respectively: 2 h d<sup>-1</sup>, 48 d y<sup>-1</sup>, 70 y, 70 y  $\times$  365 d y<sup>-1</sup>  $\times$  24 h d<sup>-1</sup>. 209

- The cancer risk from inhalation exposure to PAHs was estimated using the benzo[a]pyrene equivalent concentrations (BaP<sub>eq</sub>, ng m<sup>-3</sup>) as CA. The BaP<sub>eq</sub> concentrations were calculated multiplying the particle-phase concentrations of PAHs (EPA 16 priority PAHs) by the respective toxic equivalent factors (TEF), which were taken from Bari et al. [70].
- For non-carcinogenic elements, the hazard quotient (HQ) was calculated as follows [69]:
- $HQ = EC / R_fC$

(8)

The excess cancer risk associated with the carcinogenic elements was calculated asfollows [69]:

- 219 Risk =  $IUR \times EC$
- 220
- 221 **3. Results**

### **3.1. Comfort parameters**

The mean temperature and relative humidity ranged between 21.8 and 25.7 °C and between 40.5 and 60.6%, respectively, for the whole set of measurements (Table 1). The increase in the relative humidity over background levels (before the activation of the source) was, on average, from 12 to 14% and from 13 to 17% during steam iron and steam iron with boiler (condition I), respectively. When the door of the living room was closed, condition I (steam iron and steam iron with boiler), the ventilation rate varied between 229 0.16 and 0.42  $h^{-1}$ . During condition II, when the door of the living room was open, the

230 average ventilation rate was  $1.8 \pm 0.39 \text{ h}^{-1}$ .

- 231 Table 1. Sampling conditions,  $PM_{10}$  mass and particle number concentrations and
- emission rates during ironing.

	Steam iron I	Steam iron II	Steam iron with boiler
Ν	2	2	2
Air exchange rate $(\alpha, h^{-1})$	$0.25 \pm 0.13$	$1.8 \pm 0.39$	$0.38 \pm 0.06$
PM <sub>10</sub> initial mass concentration (µg m <sup>-3</sup> )	$51 \pm 2.8$	$16 \pm 1.4$	$21 \pm 2.8$
$PM_{10}$ peak mass concentration (µg m <sup>-3</sup> )	$340 \pm 62$	$98 \pm 16$	$444 \pm 16$
PM <sub>2.5</sub> /PM <sub>10</sub> (%)	$94 \pm 0.2$	$87 \pm 5.7$	$94 \pm 2.3$
PM <sub>1</sub> /PM <sub>10</sub> (%)	$93 \pm 0.4$	$85 \pm 6.3$	$93 \pm 2.6$
Particle number initial concentration (particles $\times 10^3$ cm <sup>-3</sup> )	$3.6 \pm 1.3$	$6.0 \pm 1.6$	$7.2 \pm 3.4$
Particle peak number concentration (particles $\times 10^5$ cm <sup>-3</sup> )	$4.4 \pm 1.2$	$2.6 \pm 0.03$	$3.6 \pm 0.30$
Emission rate PM <sub>10</sub> (µg s <sup>-1</sup> )	$6.6 \pm 1.4$	$1.9 \pm 1.6$	$8.3 \pm 3.1$
Emission rate particle number (particles $\times 10^{11} \text{ min}^{-1}$ )	$12 \pm 2.3$	$8.1 \pm 0.09$	$15 \pm 3.5$

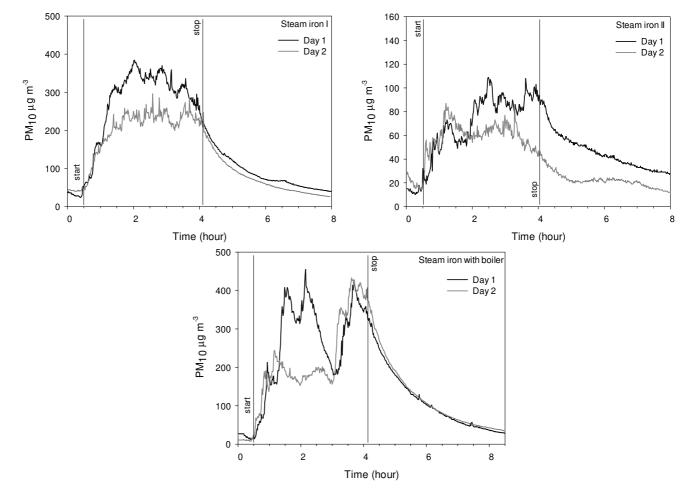
233

# 234 **3.2. Particulate matter**

# 235 **3.2.1. Mass concentrations**

236 Fig. 1 shows the time resolved  $PM_{10}$  mass concentrations during ironing. On average, 237 increases over PM<sub>10</sub> initial concentrations (before the activation of the source) of 4.8, 4.2, 12 times were observed when the steam iron (condition I), steam iron (condition II) and 238 steam ironing with boiler (condition I) were in use, respectively. The PM<sub>10</sub> mass 239 concentration profiles during steam iron (condition I) were similar on both measurement 240 days, although the values were generally lower on the second day of experiments. During 241 the first hour and a half using the steam iron (condition II), slightly higher PM<sub>10</sub> 242 243 concentrations were recorded on the second day of measurements, while the opposite, with more marked differences in the  $PM_{10}$  levels, was recorded for the remaining period 244 245 of activity. For the steam iron with boiler, during the first two and a half hours, concentrations were much higher on the first day of measurements in comparison with 246 247 those recorded on the second day, while in the last hour the profiles and concentrations were similar. The discrepancies observed, both for levels and profiles, may result from 248 differences in the number, size and composition of ironed garments. Peak mass 249 concentrations ranging from 87  $\mu$ g m<sup>-3</sup> (steam iron II) to 455  $\mu$ g m<sup>-3</sup> (steam iron with 250 boiler) were recorded. Steam ironing in an enclosed space (condition I) generated peak 251 252 concentrations three to four times higher than those observed with the room door open 253 (condition II). Higher peak concentrations during steam ironing with boiler can result

from the generation of steam at high pressure, which likely enhances the shedding of 254 fibres from clothing and consequently the mass of particles generated in comparison with 255 256 the conventional steam iron. Lower PM<sub>10</sub> peak concentrations were recorded by Schiavon et al. [43], ranging from 86.9 to 119.8  $\mu$ g m<sup>-3</sup>, during ironing (steam iron with boiler) in 257 a closed and poorly ventilated room. In addition of being released during ironing, particles 258 259 can also be emitted from other actions that are closely connected to the studied activity such as handling clothes before and after ironing. In fact, previous studies reported that 260 manipulation of clothing can be a strong source of indoor aerosol particles [71,72]. 261



262

263 Fig. 1.  $PM_{10}$  mass concentrations during ironing.

264

During the manipulation of a cotton shirt, Licina et al. [71] reported sharp PM<sub>10</sub> concentration peaks in the breathing zone, at times exceeding 40  $\mu$ g m<sup>-3</sup>. Ferro et al. [72] also reported that folding clothes (mean PM<sub>10</sub> personal exposure from 15-min over 250  $\mu$ g m<sup>-3</sup>) and blankets (mean PM<sub>10</sub> personal exposure from 15-min over 200  $\mu$ g m<sup>-3</sup>) resulted in high PM exposure.

In the present study, PM<sub>10</sub> concentrations decreased to background levels (concentration
in the room before the activity started) about 3 (steam iron I and II) to 5 h (steam iron
with boiler) after turning off the iron. Schiavon et al. [43] observed that one hour after
ceasing ironing the concentrations were still three times higher than the ones recorded
before switching the iron on.
Fine particles dominated the PM<sub>10</sub> mass as indicated by PM<sub>2.5</sub>/PM<sub>10</sub> and PM<sub>1</sub>/PM<sub>10</sub>

average ratios ranging from 85 to 94% (Table 1). Contrarily to the results obtained here, Schiavon et al. [43] reported  $PM_{2.5}/PM_{10}$  and  $PM_1/PM_{10}$  ranging from 28 to 56% and from 19 to 46%, respectively.

The estimated PM<sub>10</sub> emission rates from steam iron under minimum ventilation (condition I) were, on average, over three times higher  $(6.6 \pm 1.4 \ \mu g \ s^{-1})$  than those from steam iron with the living room door open (condition II)  $(1.9 \pm 1.6 \ \mu g \ s^{-1})$ . The highest emission rates were recorded when using the steam iron with boiler  $(8.3 \pm 3.1 \ \mu g \ s^{-1})$ (Table 1).

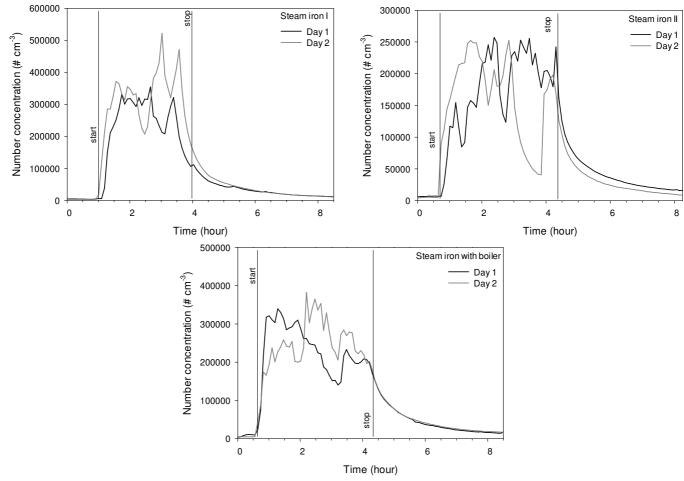
284

# 285 **3.2.2. Number concentrations**

286 Fig. 2 illustrates the time evolution of the total particle number concentration during 287 ironing. As observed for PM<sub>10</sub> mass concentrations, differences in the levels and profiles of number concentrations were also recorded. When ironing started, the number 288 concentration of particles increased sharply. Although a rapid increase was observed 289 when the activity started, it took about 1-2 h to reach peak concentrations depending on 290 291 the iron and condition tested. When the living door was open (steam iron II) the decay 292 rate was faster than that under minimum ventilation conditions. The average particle number concentration in the room was lower during the operation of the steam iron with 293 the living room door open  $(1.7 \pm 0.092 \times 105 \text{ particles cm}^{-3})$ . Steam ironing  $(2.6 \pm 0.22)$ 294 × 105 particles cm<sup>-3</sup>) and steam ironing with boiler (2.4  $\pm$  0.069 × 105 particles cm<sup>-3</sup>) 295 296 with the doors closed led to similar average particulate number concentrations in the 297 room. The ratios of peak to background levels for ultrafine particle number concentrations indicate that ironing can elevate levels by a factor ranging from 35 to 194. The higher 298 299 ratios for particulate number concentrations in comparison with particulate mass concentrations in similar conditions could be due to the increase of ultrafine particles 300 301 released during the ironing activity. The peak concentrations obtained in the present study are much higher than those reported by Ciuzas et al. [42]. The researchers conducted the 302 source assessment in a test chamber representing a typical room (volume =  $35.8 \text{ m}^3$ ) and 303

reported peak concentrations ranging from  $3 \pm 0.58 \times 10^3$  to  $36 \pm 6 \times 10^3$  particles cm<sup>-3</sup> 304 and from  $19 \pm 2.4 \times 10^3$  to  $28 \pm 4 \times 10^3$  particles cm<sup>-3</sup> during ironing with and without 305 steam, respectively (particles from 0.01 to 0.3 µm). Lower peak concentrations were 306 reported by Afshari et al. [30], ranging from  $0.55 \times 10^3$  particles cm<sup>-3</sup> (flat iron without 307 steam) to  $7.2 \times 10^3$  particles cm<sup>-3</sup> (flat iron with steam) (particles from 0.02 to 1.0 µm). 308 A previous study conducted by Wallace and Ott et al. [31] in the basement of an American 309 household documented peak concentrations in the range from  $21 \times 10^3$  to  $148 \times 10^3$ 310 particles  $cm^{-3}$  (particles from 0.01 to 1.0 µm). 311





313 Fig. 2. Particle number concentrations during ironing.

314

The maximum particle number concentrations detected by Ciuzas et al. [42] and Afshari

et al. [30] were much lower than the ones obtained in the present study. The discrepancy

317 is probably because only one cotton sheet was ironed in their studies in comparison with

the vast array of clothing and linen ironed in this work.

In addition to the fact that only a small number of studies assessed the impact of this 319 320 source on indoor air quality, it should be borne in mind that differences in sampling 321 conditions (laboratory chambers vs on site measurements), devices employed to measure 322 the particle concentrations, size ranges, test conditions, as well as the chosen irons and 323 the operation mode (e.g. steam ironing versus ironing without steam) make the 324 comparison between studies difficult. In fact, Afshari et al. [30] observed that ironing a cotton sheet with steam generated 10 times more particles than ironing without steam. 325 326 The authors hypothesised that the higher generation of particles during steam ironing 327 could be ascribed to (i) the expulsion of burned fibres from the cotton sheet due to the 328 steam or (ii) the formation of new particles by homogeneous nucleation during the cooling 329 of the water vapour from the steam.

In the present study, the average emission rates (particles from 0.008 to 0.322  $\mu$ m) were estimated to range from 8.1 × 10<sup>11</sup> to 15 × 10<sup>11</sup> particles min<sup>-1</sup> (Table 1). Wallace and Ott et al. [31] reported emission rates from steam ironing ranging from 0.4 × 10<sup>11</sup> to 3.5 × 10<sup>11</sup> particles min<sup>-1</sup> (particles from 0.01 to 1.0  $\mu$ m). The assessment of ironing a cotton sheet in a full-scale chamber carried out by Afshari et al. [30] resulted in lower emission rates ranging from 0.007 × 10<sup>11</sup> particles min<sup>-1</sup> (flat iron without steam) to 0.06 × 10<sup>11</sup> particles min<sup>-1</sup> (flat iron with steam) (particles from 0.02 to 1.0  $\mu$ m).

When ironing clothes using the steam iron, keeping the living room door closed, more 337 than 60% (64–69%) of the total particle number concentrations was found in the Aitken 338 mode ( $30 \le N \le 100$  nm). This value dropped to 59–63% when the doors were opened 339 (condition II). The use of the steam iron with boiler also generated the highest number of 340 particles in the Aitken mode (61-62%). The geometric mean diameter (GMD) of the 341 342 particle size distribution ranged between 41.1 and 62.6 nm, while the source was active. After switching off the iron, an increase of the GMD of the ultrafine particle mode was 343 344 observed in all experiments.

345

## 346 **3.3. Elements**

The  $PM_{10}$  mass fractions of 26 elements analysed in samples collected during ironing, and in the background air sample, are shown in Table 2. Cl, Si and Na were the most abundant inorganic elements found in the samples from steam ironing (conditions I and II), while during the operation of the steam iron with boiler the dominant elements were Si, Al and Cl. Elements accounted for  $PM_{10}$  mass fractions of 12, 14 and 5.5 %wt., which

- represented increases over background levels of 2.6, 3.0 and 1.2 times for steam iron I,
- 353 steam iron II and steam iron with boiler, respectively.
- 354

Element	Steam Iron - I	Steam iron - II	Steam iron with boiler	Background
Na	1.9	$2.6 \pm 0.036$	$0.48 \pm 0.14$	1.05
Mg	0.81	$0.94 \pm 0.030$	$0.19 \pm 0.014$	0.15
Al	0.92	$1.0 \pm 0.50$	$0.71 \pm 0.21$	0.31
Si	2.3	$2.5 \pm 1.2$	$2.4 \pm 0.55$	0.68
Р	0.021	$0.028 \pm 0.016$	$0.012 \pm 0.006$	0.013
S	0.73	0.89 ± 0.16	$0.22 \pm 0.003$	0.65
Cl	3.0	$3.2 \pm 0.11$	$0.61 \pm 0.099$	0.75
K	0.61	$0.78 \pm 0.056$	$0.15 \pm 0.014$	0.34
Ca	1.4	$1.4 \pm 0.62$	$0.60 \pm 0.12$	0.40
Ti	0.14	$0.13 \pm 0.047$	$0.030 \pm 0.004$	0.006
V	0.0003	$0.001 \pm 0.000$	bdl	bdl
Cr	0.002	bdl	$0.001 \pm 0.001$	0.005
Mn	0.006	$0.010 \pm 0.006$	$0.001 \pm 0.001$	0.007
Fe	0.30	$0.39 \pm 0.24$	$0.11 \pm 0.004$	0.26
Ni	0.001	$0.002 \pm 0.000$	bdl	0.001
Cu	0.006	$0.014 \pm 0.003$	$0.009 \pm 0.009$	0.012
Zn	0.031	$0.043 \pm 0.011$	$0.013 \pm 0.001$	0.049
As	0.001	bdl	bdl	0.004
Se	0.0001	bdl	bdl	0.003
Br	0.021	$0.011 \pm 0.005$	$0.016 \pm 0.000$	0.002
Rb	0.002	$0.002 \pm 0.003$	bdl	bdl
Sr	0.011	$0.014 \pm 0.009$	$0.007 \pm 0.001$	bdl
Y	0.0003	bdl	bdl	0.003
Zr	0.002	bdl	$0.003 \pm 0.003$	bdl
Мо	bdl	bdl	bdl	bdl
Pb	bdl	$0.014 \pm 0.009$	bdl	bdl
Σ Elements	12	$14 \pm 2.6$	5.5 ± 1.1	4.7

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

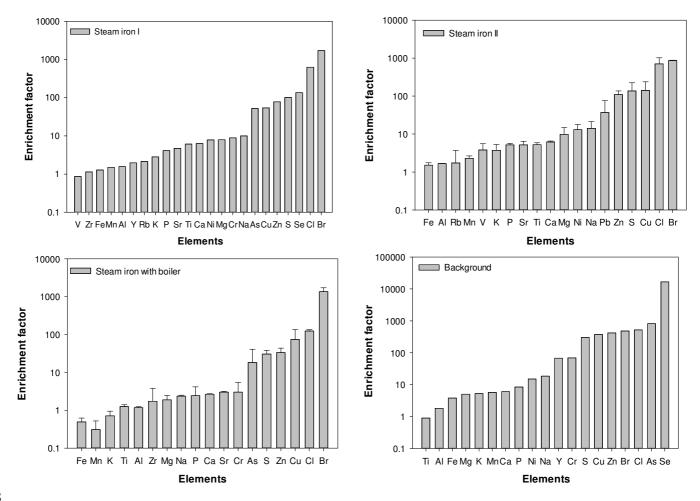
356 bdl – below the detection limit.

357

Although several elements made up a small part of the  $PM_{10}$  mass, the increase in concentration over the background level was noticeable. This was observed for Br (9–51 times) and Ti (18–98 times). Additionally, Sr was only detected in the samples collected when the source was active.

As reviewed by Licina et al. [73], trace elements, including heavy metals, are among the chemicals found in clothing. These can result from the manufacturing process (e.g., dyeing, bleaching, finishing) or have been added to achieve a specific function (e.g.

- control microbial activity, shield against UV radiation, enhance water repellence). 365 366 Clothing additives include nanomaterials, such as titanium dioxide (TiO<sub>2</sub>), zinc oxide (ZnO), copper and carbon nanotubes [73]. The type of material and colour are important 367 factors regarding the elemental content in clothing [74,75]. Rovira et al. [75] reported 368 369 high levels of Cr in polyamide dark clothes, high Sb concentrations in polyester clothes, and high Cu levels in some green cotton fabrics. Turner [74] detected Br in clothing and 370 laundry dryer lint from natural and synthetic garments, suggesting that this element was 371 derived from clothing fibres. Brominated compounds are used in textiles as flame 372 373 retardants and used as disperse dyes [74].
- Enrichment factors close to the unity (Fig. 3) for elements such as V, Zr, Fe, Mn, Al, Rb 374 and K, indicate a predominant natural contribution. Additionally, low EFs ( $5 \le EF \le 10$ ) 375 were registered for Ca, Mg, Ti and P. Several of these elements (e.g. Al, Ca, Fe, insoluble 376 377 fractions of K, Mg and Na) have been generally associated with mineral dust [76,77]. In the present study, very good correlations were obtained between K. Mg and Na ( $r^2 >$ 378 0.96). These elements also correlated well with Ti ( $r^2 > 0.83$ ) and Zn ( $r^2 > 0.84$ ) and 379 showed moderate correlations with Mn ( $r^2 > 0.51$ ), Fe ( $r^2 > 0.58$ ), Ca ( $r^2 > 0.59$ ) and Ni 380 381  $(r^2 > 0.67)$ . These strong relationships suggest the existence of common sources for these elements, probably resuspension of previously deposited dust (e.g. soil dust brought in 382 the shoes or dust particles deposited on clothing). 383
- In the present study, other elements like Cu ( $54 \le EF \le 141$ ), S ( $31 \le EF \le 136$ ) and Zn 384 (313 < EF < 109) were enriched in PM10 from all ironing tests. Bromide (862 < EF < 109)385 1712) and Cl ( $124 \le EF \le 702$ ) were highly enriched in all the ironing samples (Fig. 3). 386 A strong correlation was found between the concentrations of Cl and S ( $r^2 > 0.95$ ). The 387 occurrence of these elements in the PM<sub>10</sub> samples may derive from cleaning products 388 since they are part of the composition of these consumer items. In addition to the presence 389 390 in the background air, the contamination of the clothes during cleaning activities and the 391 ineffectiveness of washing in removing these metallic constituents, may justify the higher 392 concentrations in the particles resulting from ironing. Regarding Br, the only correlations were found with Cr ( $r^2 = 0.54$ ), Al ( $r^2 = 0.68$ ) and Si ( $r^2 = 0.76$ ). Moreover, only three of 393 the analysed elements displayed strong correlation with  $PM_{10}$  concentrations: Al ( $r^2 =$ 394 0.70), Si ( $r^2 = 0.87$ ) and Br ( $r^2 = 0.96$ ). As far as we know, the elemental composition of 395 particles released during ironing has never been examined. 396
- 397



398

Fig. 3. Enrichment factors of elements in PM<sub>10</sub> sampled during ironing.

400

401 In the present study, we hypothesised that the occurrence of these elements in the  $PM_{10}$ 402 samples might be associated with their evaporation from clothing when subjected to high temperatures during ironing and subsequent condensation on the surface of pre-existing 403 particles. Garment handling during the activity can also release fibres, which contributes 404 to the migration of elements into the air. In addition to the use of brominated compounds 405 as discussed above, Cr, Al and Si are also employed in the textile industry with different 406 purposes. For example, silicon based compounds (flame retardants, antimicrobial agents, 407 surfactants, water and oil proofing agents) are used in textile industry due to their unique 408 surface properties [78,79]. Metal salts (e.g. chromium and aluminium) are used as 409 mordant to improve the affinity between dyes and fibres; they can also produce different 410 colours and improve the fastness of a dye [80]. Additionally, several metals, including 411 chromium, are widely used in dyes [81]. 412

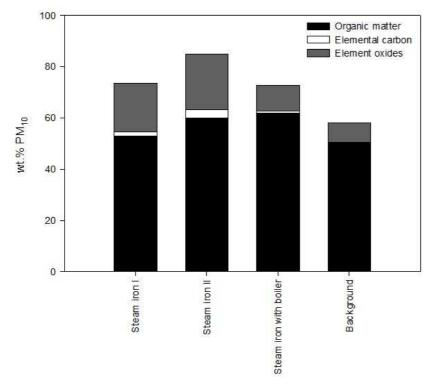
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#### 414 **3.4. OC/EC**

415 Total carbon (TC) represented from  $30 \pm 0.91$  (steam iron I) to  $34 \pm 2.4$  (steam iron with boiler) %wt. of the PM<sub>10</sub> mass during ironing and  $28 \pm 2.1$  %wt. of the PM<sub>10</sub> mass in the 416 background air. Elemental carbon (EC), not detected in the background sample, 417 418 comprised from  $1.1 \pm 0.2$  to  $3.1 \pm 0.9$  %wt. of the PM<sub>10</sub> mass during ironing. The EC 419 increase over background may be attributable to burned fabric fibres released during ironing. OC and EC concentrations in samples collected during ironing were not 420 correlated with each other, suggesting distinct sources. On the contrary, good correlations 421 were found between particulate OC concentrations and both Si ( $r^2 = 0.81$ ) and Br 422 concentrations ( $r^2 = 0.91$ ). 423

424 The OC particulate fractions in indoor settings can arise from a wide variety of sources. 425 In schools, several researchers have partially ascribed the PM organic content to clothing 426 fibres [82,83]. Additionally, household  $PM_{10}$  dust has been reported to contain appreciable amounts of carbonaceous particles, mainly OC [84]. Although the handling 427 428 and ironing of clothes may constitute the major source of indoor carbonaceous compounds found in PM<sub>10</sub> samples, the possibility of other contributing sources cannot 429 430 be ruled out. Indoors, there are several sources of carbonaceous particles, both OC and 431 EC, such as cooking [17], vacuum cleaning [37,85], biomass burning for heating purposes [22,47], cigarette smoking [45,86] and incense burning [46]. Despite the vast array of 432 possible sources, during the sampling campaign, the only activity that might have 433 contributed to the measured levels was cooking since none of the other activities took 434 435 place (carried out before ironing). In fact, as observed in the present study, despite the temporary nature of indoor sources, they can release particles that remain for hours inside 436 437 the household [e.g. 36,37,87,88]. In cooking generated particles, organic carbon is the major constituent and the release of OC and EC is closely related to the cooking method. 438 439 As reviewed by Abdullahi [17], indoor OC/EC ratios can vary between 4.3 and 7.7 depending on the method of food preparation. In the present study, higher OC to EC ratios 440 441 were recorded, ranging from  $11 \pm 4$  (steam iron II) to  $31 \pm 7$  (steam iron with boiler). The mass closure between chemical and gravimetric measurements ranged from 58% 442 443 (background) to 85% (ironing) (Fig. 4). The unaccounted mass might be ascribed to sampling and analysis artefacts, to PM<sub>10</sub>-bound water and to unanalysed constituents. 444

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- 447



- 449 Fig. 4. Chemical mass closure of  $PM_{10}$ .
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#### 451 **3.5. Organic compounds**

The detailed organic composition of  $PM_{10}$  samples is presented in Table 3. The  $PM_{10}$ 452 samples encompassed n-alkanes from C<sub>11</sub> to C<sub>35</sub>, maximising at C<sub>25</sub> and C<sub>27</sub> for samples 453 collected during ironing under minimum ventilation conditions. The carbon preference 454 455 index (CPI), calculated for the whole range of n-alkanes, oscillated from 0.9 to 1.4 denoting an input of n-alkanes derived from petroleum derivatives [89]. The presence of 456 these compounds seems to be closely related to ironing, decreasing noticeably with the 457 458 increase in the ventilation rates (steam iron II). In fact, opening the living room door decreased the  $\Sigma_{25}$  n-alkane concentrations more than 3000 times. Steam ironing with 459 boiler led to higher n-alkane concentrations ( $\Sigma 25 2478 \text{ ng m}^{-3}$ ) in comparison with steam 460 iron ( $\Sigma_{25}$  1427 ng m<sup>-3</sup>) under similar ventilation conditions (minimum), representing an 461 462 increase over background concentrations ranging from 82 to 143 times. In a previous study, assessing the impact of vacuuming on PM<sub>10</sub>-bound organic compounds in the same 463 464 room, much lower concentrations of n-alkanes were recorded ( $\Sigma_{25}$  from 22.4 to 39.3 ng  $m^{-3}$  during vacuuming). The presence of these compounds was hypothesised to partially 465 466 derive from oil-based or petrochemical textiles such as nylon, polyester, acrylic and 467 spandex [85].

468

	Steam iron I	Steam iron II	Steam iron with boiler	Background
Aliphatics				
Undecane	5.98	nd	95.4	0.309
Dodecane	0.646	0.193	1.43	0.026
Tridecane	0.281	0.183	1.09	0.012
Tetradecane	0.132	nd	1.48	bdl
Pentadecane	1.08	nd	2.87	0.525
Hexadecane	0.733	nd	3.62	bdl
Heptadecane	bdl	nd	bdl	bdl
Octadecane	6.95	bdl	17.0	bdl
Nonadecane	14.4	bdl	32.2	bdl
Eicosane	25.6	bdl	58.4	1.39
Heneicosane	47.6	bdl	98.9	2.94
Docosane	70.2	bdl	138	5.96
Tricosane	111	bdl	211	5.15
Tetracosane	142	bdl	251	1.03
Pentacosane	180	bdl	311	bdl
Hexacosane	155	bdl	212	bdl
Heptacosane	174	bdl	317	bdl
Octacosane	134	nd	170	bdl
Nonacosane	131	nd	204	bdl
Triacontane	73.1	nd	88.2	bdl
Hentriacontane	79.8	nd	129	bdl
Dotriacontane	36.7	nd	57.8	bdl
Tritriacontane	13.4	nd	29.7	nd
Tetratriacontane	12.0	nd	32.0	nd
Pentatriacontane	11.1	nd	14.8	nd
Dodecene	2.07	nd	5.23	nd
Tetradecene	0.240	nd	4.38	bdl
Hexadecene	0.369	nd	0.947	0.429
Octadecene	nd	nd	nd	bdl
Eicosene	nd	nd	nd	bdl
Tricosene	18.7	0.050	18.5	0.291
Squalene	21.0	nd	33.2	0.351
Saccharides and polyols				
Levoglucosan	15.1	23.1	5.59	22.1
Mannosan	nd	nd	nd	1.27
Ribitol	2.97	6.31	1.51	nd
D-glucuronic acid lactone	5.94	4.08	nd	nd
Quebrachitol	nd	nd	0.018	nd
Meso-Erythritol	nd	nd	nd	0.280
Other saccharides and polyols	135	82.9	14.4	3.82
Triterpenoid and steroid compounds				
Lupeol	nd	nd	nd	0.323

# 469 Table 3. Concentrations (ng $m^{-3}$ ) of organic compounds in PM<sub>10</sub>.

Cholesterol	734	766	2682	4.29
5-Cholesten-3-ol	9.57	48.3	94.5	nd
Stigmasterol	nd	nd	14.9	nd
β-Sitosterol	30.2	33.8	30.2	1.20
Aliphatic alcohols				
1-Nonanol	1.41	1.98	0.914	nd
1-Decanol	0.970	1.29	3.09	0.014
1-Dodecanol	29.7	115	122	7.02
1-Tetradecanol	114	348	431	8.78
1-Pentadecanol	165	407	298	32.9
C <sub>15</sub> Alkanol isomers	7.20	31.3	7.95	3.65
1-Hexadecanol	691	675	909	82.9
C <sub>16</sub> Alkanol isomers	3.13	16.1	7.44	1.39
1-Octadecanol	1046	689	2496	32.6
1-Nonadecanol	5.41	32.8	90.8	1.06
1-Eicosanol	nd	nd	115	nd
C <sub>20</sub> Alkanol isomers	193	10.4	275	1.48
1-Heneicosanol	11.7	10.6	26.4	nd
1-Docosanol	64.4	43.4	137	bdl
1-Tricosanol	384	201	613	0.470
C <sub>23</sub> Alkanol isomers	11.0	13.0	37.2	0.000
1-Tetracosanol	29.6	41.6	54.5	nd
1-Pentacosanol	4.89	4.87	9.34	0.035
C <sub>25</sub> Alkanol isomers	23.4	22.2	51.7	nd
1-Hexacosanol	21.1	18.5	17.1	0.820
1-Heptacosanol	6.65	5.72	4.10	0.018
C <sub>27</sub> Alkanol isomers	23.7	47.8	1.36	3.83
1-Octacosanol	667	578	382	0.094
1-Nonacosanol	0.394	5.76	nd	nd
1-Tricontanol	164	201	102	0.047
Alkanoic acids				
1-Heptanoic acid	nd	nd	5.64	nd
1-Octanoic acid	8.36	8.27	46.5	0.163
1-Nonanoic acid	31.8	29.1	225	0.194
1-Decanoic acid	44.4	34.2	137	0.323
1-Undecanoic acid	3.11	2.96	9.36	0.249
1-Dodecanoic acid	167	298	843	87.8
1-Tridecanoic acid	24.7	14.6	83.6	2.31
C <sub>13</sub> Alkanoic acid isomers	19.7	nd	78.3	nd
1-Tetradecanoic acid	1636	1438	1524	165
C <sub>14</sub> Alkanoic acid isomers	32.8	nd	62.1	nd
1-Pentadecanoic acid	592	351	1674	9.52
C <sub>15</sub> Alkanoic acid isomers	254	5.74	678	nd
1-Hexadecanoic acid	3851	2075	5415	127
C <sub>16</sub> Alkanoic acid isomers	149	9.12	381	nd
1-Heptadecanoic acid	282	230	434	1.73

C <sub>17</sub> Alkanoic acid isomers	334	20.0	904	nd
1-Octadecanoic acid	1637	1263	4180	60.9
1-Nonadecanoic acid	9.74	6.35	6.70	0.109
1-Eicosanoic acid	123	51.6	248	0.443
C <sub>20</sub> Alkanoic acid isomers	18.1	nd	46.7	nd
1-Heneicosanoic acid	8.83	nd	34.7	nd
$C_{21}$ Alkanoic acid isomers	31.9	nd	59.8	nd
1-Docosanoic acid	65.3	25.4	111	1.76
1-Tricosanoic acid	23.7	5.06	38.8	nd
1-Tetracosanoic acid	120	54.8	261	0.285
1-Pentacosanoic acid	19.3	3.82	34.1	nd
C <sub>25</sub> Alkanoic acid isomers	16.7	4.01	28.6	nd
1-Hexacosanoic acid	28.2	9.41	45.1	nd
C <sub>26</sub> Alkanoic acid isomers	4.33	nd	4.63	nd
1-Octacosanoic acid	4. <i>33</i> 8.07	2.85	10.7	nd
1-Triacontanoic acid	2.10	nd	3.33	nd
Alkenoic acids	2.10	lia	5.55	nu
9-Tetradecenoic acid	27.0	nd	80.1	nd
Pentadecenoic acid	28.7	nd	80.6	nd
9-Cis-hexadecenoic (palmitoleic) acid	701	416	1925	1.43
10-Heptadecenoic acid	150	nd	364	nd
Cis-9-octadecenoic (oleic) acid	1821	1661	4795	20.4
Cis,cis-9-12-octadecadienoic (linoleic) acid	282	127	447	1.43
10-Nonadecenoic acid	7.11	nd	24.3	nd
Eicosenoic acid	27.1	nd	53.8	nd
Alkanedioic acids	27.1	na	55.0	na
1,4-Butanedioic (succinic) acid	1.26	2.79	6.99	1.05
1,5-Pentanedioic (glutaric) acid	7.51	20.8	bdl	4.18
Hexanedioic (adipic) acid	6.33	6.69	59.1	2.05
Heptanedioic (pimelic) acid	0.496	0.546	1.08	0.248
Octanedioic (suberic) acid	2.23	1.28	7.42	0.251
Nonanedioic (azelaic) acid	9.67	5.22	57.8	0.885
Decanedioic (sebacic) acid	1.23	1.05	nd	nd
Other acids		1100		
4-Oxopentanoic (levulinic) acid	21.0	27.7	89.1	nd
2,3-Dihydroxypropanoic (glyceric) acid	6.85	8.34	nd	14.6
3-Hydroxybutanoic (3-hydroxybutyric) acid	0.274	0.455	0.478	0.166
2-Hydroxy-butanedioic (L-(-)-malic) acid	0.433	1.51	nd	nd
Hexanedioic (adipic) acid dibutyl ester	16.4	45.7	14.5	12.2
Cis-Pinonic acid	0.238	0.356	nd	0.302
Pinic acid	nd	1.42	nd	nd
Citric acid	0.082	0.12	nd	0.088
Dehydroabietic acid	0.194	0.980	5.06	0.485
Isopimaric acid	2.85	1.33	nd	0.009
Abietic acid	0.234	0.152	nd	nd
Podocarpic acid	nd	nd	nd	0.003
-				

Alkyl esters of fatty acids				
Tetradecanoic acid 1-methylester	45.3	27.3	147	25.2
Hexadecanoic acid, methyl ester	65.9	46.3	280	63.2
Hexadecanoic acid, ethyl ester	4.45	2.75	15.2	1.32
Hexadecanoic acid, isopropyl ester	247	144	652	60.8
Hexadecanoic acid, hexadecyl ester	19.5	18.6	89.8	nd
Dodecanoic acid, isooctyl ester	20.4	20.1	48.6	7.42
Octadecanoic acid, methyl ester	62.9	22.9	238	12.7
Hexanoic acid 2-ethyl-, hexadecyl ester	25.8	23.1	198	1.96
Hexanedioic acid, dioctyl ester (dioctyl adipate)	nd	181	206	7.23
Octanoic acid, hexadecyl ester	10.0	12.8	98.6	0.173
Phenolics and alteration products				
Pyrocatechol	0.011	0.009	0.028	bdl
5-Isopropyl-3-methylphenol	0.107	0.235	0.579	bdl
Resorcinol	0.024	0.028	0.323	0.013
4-Methyl catechol	0.016	0.007	0.038	0.001
2,6-Dimethoxyphenol	0.029	nd	nd	0.001
Eugenol	0.030	0.017	0.066	0.001
4-Allyl-2,6-dimethoxyphenol (methoxy eugenol)	nd	nd	nd	0.010
Pyrogallol	0.007	0.009	0.012	0.001
Isoeugenol	0.941	1.83	2.14	bdl
2-Methoxy-4-propylphenol	0.441	0.716	0.833	nd
4-Phenylphenol	nd	nd	nd	0.005
4-Octylphenol	0.303	0.024	0.464	0.006
2,4-Di-tert-buthylphenol	69.8	112	108	20.6
Sinapyl alcohol	0.012	0.012	nd	0.001
Benzyl alcohol	16.7	43.4	55.1	bdl
Benzoic acid	0.234	0.797	13.8	0.187
4-Hydroxybenzoic acid	0.263	0.360	1.09	0.074
Benzoic acid alkyl esters	239	126	1160	12.6
Trans-cinnamic acid	0.224	0.316	bdl	0.027
Vanillic acid	0.021	0.016	bdl	0.031
Syringic acid	0.023	0.040	bdl	0.054
Sinapic acid	nd	0.112	nd	nd
4-Hydroxy-3-methoxycinnamic (ferulic) acid	nd	nd	nd	nd
4-Hydroxycinnamic (p-coumaric) acid	0.198	0.086	nd	nd
4-Tert-butylphenol	4.12	8.29	14.3	1.71
Bisphenol A	90.5	17.4	60.9	5.43
Glycerol derivatives				
Glycerol	804	788	783	143
1-Monolauroyl-rac-glycerol	nd	0.028	nd	nd
Ethylene glycol palmitate	76.1	36.2	151	nd
1-Monomyristin	323	61.8	528	nd
Glyceryl pentadecanoate	164	51.0	496	nd
1-Monopalmitin	709	635	1896	3.50
1-Hexadecyl glycerol	8.09	nd	nd	nd

Glycerol, mono-heptadecanoate	10.9	nd	21.3	nd
2-Monolinolein	10.9 51.7	9.39	170	nd
2-Oleoylglycerol (2-monoolein)	50.4	9.39 nd	103	nd
1-Monolinoleoylglycerol	50.4 741	652	2074	nd
1-Glycerol monostearate (1-monostearin)	339	032 197	944	4.59
Glycerol tricaprylate	32.3	42.1	36.2	4.39 nd
Plasticisers	52.5	42.1	50.2	nu
Benzyl butyl phthalate	36.0	67.1	150	5.68
Bis(2-ethylhexyl)adipate	0.230	nd	1.83	bdl
Bis(2-ethylhexyl)phthalate	bdl	bdl	1.65	bdl
Di-n-butyl phthalate	bdl	0.095	0.030	bdl
Dimethyl phthalate				
Diethyl phthalate	0.004	0.016	0.033	0.010
Unidentified phthalates	15.1	5.08	41.8	49.6
Other compounds	2437	3435	8763	253
÷	20.2		22.0	1
Diethylene glycol	28.3	56.9	22.0	nd
1,2,3-Hexanetriol	0.102	nd	0.223	nd
2,6-Di-tert-butyl-p-benzoquinone	0.994	0.089	2.53	0.214
(S)-(-)b-Citronellol	nd	nd	11.4	nd
α-Hexylcinnamaldehyde	14.3	1.80	17.2	0.211
2-Propanol-1-chloro-phosphate (TCPP)	nd	42.3	58.7	12.5
Acetyl tributyl citrate	17.5	30.9	27.1	3.25
(1S, 2S, 3R, 5S)-2,3-Pinanediol	nd	nd	0.373	0.064
Tocopheryl acetate	1913	98.0	493	nd
Diethyltoluamide (DEET)	47.5	164	69.6	43.3
Parsol MCX	207	91.7	351	5.43
Oxidised Irgafos 168	21.4	bdl	nd	29.4
Irganox 1076	nd	65.3	12.1	nd
Methyl dihydrojasmonate	5.93	nd	nd	nd
7,9-Di-tert-butyl-1-oxaspiro[4.5]deca-6,9-diene-				
2,8-dione Benzothiazole	5.76	nd	nd	nd
	0.052	0.030	0.094	nd
Butylphenyl methylpropional (Lilial)	1.20	0.460	6.13	0.067
PAHs				
Naphthalene	0.288	0.236	0.643	bdl
Acenaphthene	0.007	0.009	0.036	0.001
Fluorene	bdl	bdl	0.197	bdl
Phenanthrene	0.183	0.067	0.513	0.170
Anthracene	bdl	bdl	0.048	0.119
Acenaphthylene	0.405	0.093	0.355	nd
Retene	0.534	0.192	1.03	0.097
Fluoranthene	0.236	0.124	0.592	0.090
Pyrene	0.274	0.111	0.774	0.525
Chrysene	0.117	0.050	1.56	0.393
Benzo[a]anthracene	bdl	bdl	1.70	0.392
Benzo[b]fluoranthene	0.496	bdl	0.929	0.392

Benzo[k]fluoranthene	nd	0.173	0.607	0.333
Benzo[e]pyrene	0.120	0.283	0.151	0.162
Benzo[a]pyrene	0.021	bdl	0.970	0.145
Perylene	nd	0.031	0.026	nd
Dibenzo[a,h]anthracene	bdl	0.010	0.096	0.105
Indeno[1,2,3-cd]pyrene	0.039	0.200	2.51	0.194
Benzo[g,h,i]perylene	0.051	0.257	0.389	0.199

470 bld – below the detection limit; nd - not detected.

471

472 Alkenes were also detected in some samples at low concentrations. Squalene, a 473 constituent of skin oil, was detected in the  $PM_{10}$  samples at concentrations ranging from 474 21.0 to 33.2 ng m<sup>-3</sup> in the samples collected under minimum ventilation conditions, while 475 it was not detected during steam ironing with the living door open (condition II).

In the present study, several saccharides and polyols were identified in samples.
Levoglucosan was the most abundant compound in this organic class and was present
both during ironing (5.59–23.1 ng m<sup>-3</sup>) and in the background air (22.1 ng m<sup>-3</sup>).
Levoglucosan derives from the thermal decomposition of cellulose [90].

480 Within terpenoids and sterols, cholesterol was the most abundant compound. It was detected at higher concentrations during ironing (734–2682 ng  $m^{-3}$ ) in comparison with 481 background (4.29 ng m<sup>-3</sup>) (Table 3). Cholesterol has been reported in emissions from 482 cooking activities [17] and it is also present as epidermal lipid [91]. In fact, a previous 483 study carried out in children bedrooms and day care centres documented the presence of 484 cholesterol in dust, pointing out desquamated skin cells, cooking, skin care products and 485 486 outdoor-to-indoor transport of cholesterol-containing particles as probable sources [92].  $\beta$ -sitosterol, the second major sterol found in the present study during ironing (30.2–33.8 487 ng  $m^{-3}$ ) and in the background sample (1.20 ng  $m^{-3}$ ), has also been detected in Chinese 488 489 cooking (vegetables) emissions [93].

Alcohols from C<sub>9</sub> to C<sub>30</sub> were detected in the PM<sub>10</sub> samples (Table 3). The  $\Sigma_{19}$  alkanol 490 concentrations ranged from 3521 to 6192 ng m<sup>-3</sup> during ironing, whereas a much lower 491 level (177 ng m<sup>-3</sup>) was observed in the background air. 1-Octadecanol and 1-hexadecanol 492 493 were the most abundant alcohols, followed by 1-octacosanol and 1-tricosanol. Their 494 concentrations during ironing were from 8 to over 1000 times higher than those measured in background air. Alkanols, typically with a chain length of  $C_{10}$  and higher, are widely 495 employed in personal care and cosmetic products, as well as in household products, 496 497 including laundry detergents and fabric softeners [94].

Twenty-two saturated fatty acids from C<sub>7</sub> to C<sub>30</sub>, maximising at C<sub>14</sub> (myristic), C<sub>16</sub> 498 (palmitic) or  $C_{18}$  (stearic) were identified in PM<sub>10</sub> samples (Table 3). The  $\Sigma_{22}$  alkanoic 499 acid concentrations ranged from 5942 to 17,614 ng m<sup>-3</sup> during ironing, while the 500 background air sample presented a concentration of 458 ng m<sup>-3</sup>. Alkanoic acids are 501 502 ubiquitous compounds from multiple sources [17,95]. In the present study, alkenoic acids 503 were also found in PM<sub>10</sub> samples. The highest concentrations were recorded for palmitoleic acid and oleic acid. Unsaturated fatty acids, such as oleic, linoleic and 504 palmitoleic acids, have been described in cooking emissions [17]. Fatty acids, such as 505 506 oleic, palmitic, stearic, linoleic and myristic acids, are also included in the list of chemical 507 formulations of cosmetic products [96]. n-Alkanedioic acids were also identified in the 508  $PM_{10}$  samples (Table 3). Dicarboxylic acids may have various origins, including 509 photochemical reactions [17,95,97,98].

510 Other acids were also detected in the  $PM_{10}$  samples. The most abundant was levulinic 511 acid (21.0–89.1 ng m<sup>-3</sup>), which has been described as an oxidant product of skin oils [99]. 512 This keto acid is also used in a variety of applications, including personal care products 513 [100]. This compound was previously detected indoors in a university cafeteria during 514 the working hours (occupancy period) with an average concentration of  $13.2 \pm 9.6$  ng m<sup>-3</sup> 515 [101].

516 Glycerol and several derivatives were present in the  $PM_{10}$  samples. Glycerol is a major 517 breakdown product of triglycerides during meat cooking [17]. It was present in samples 518 collected during ironing (783–804 ng m<sup>-3</sup>) and in background (143 ng m<sup>-3</sup>). In cosmetics, 519 this polyol is widely employed as solvent and humectant [96].

520 Alkyl esters of fatty acids were also present in the PM<sub>10</sub> samples. The highest 521 concentrations were recorded for isopropyl palmitate (hexadecanoic acid, isopropyl ester), methyl palmitate (hexadecanoic acid, methyl ester) and dioctyl adipate 522 523 (hexanedioic acid, dioctyl ester). Among other possible origins, isopropyl palmitate and methyl palmitate can be released indoors from the use of laundry detergents, fragrances 524 525 and air fresheners [102,103]. Dioctyl adipate is used as a plasticiser and, among many other applications, it is employed in textile spin finishes and pigment wetting agent [104]. 526 527 The most abundant phenolic compounds detected in the samples were benzoic acid alkyl esters, bisphenol A, 2,4-di-tert-butylphenol, benzyl alcohol and 4-tert-butylphenol. 528 Benzoic acid and a wide range of derivatives, such as alkyl esters, are widely used in 529 530 different industrial sectors (e.g. flavouring agents in food, cosmetic and hygiene products) 531 [105]. Bisphenol A (BPA) has been detected in newly purchased infant clothing [106]

and also in washed clothes [107]. Wang et al. [107] investigated the occurrence of 532 533 bisphenol chemicals in daily clothes. The maximum concentrations of BPA were detected 534 in new garments. It was also demonstrated that laundry cannot remove BPA efficiently 535 but cause cross contamination in clothes. BPA is not used directly in textile production 536 but is employed as an intermediate chemical in the manufacture of antioxidants and dyes [106, and references therein]. In the present study, this compound was found in all 537 samples during ironing  $(17.4-90.5 \text{ ng m}^{-3})$  and in the background air  $(5.43 \text{ ng m}^{-3})$ . As 538 reviewed by Licina et al. [73], a wide range of organic compounds have been detected in 539 540 clothing, including alkylphenols and alkylphenol ethoxylates. In fact, an indoor source of 541 alkylphenols might be attributed to the biodegradation of alkylphenol ethoxylate, which 542 is a widely used surfactant in laundry detergents [108]. Phenylpropenes, such as eugenol 543 and isoeugenol, were also detected in the samples, although at low concentrations. These 544 compounds are fragrance materials commonly used in household products, such as detergents [109], consumer products (e.g. air fresheners) [110] and also in a wide range 545 546 of cosmetics and toiletries [111].

547 Fragrance compounds are likely to be found in clothes and other textiles, especially after 548 laundering, due to the use of detergents and fabric softeners [111]. Butylphenyl 549 methylpropional (lilial) and  $\alpha$ -hexylcinnamaldehyde, compounds used as fragrance ingredients in laundry detergents and softeners [112,113], were detected in all samples. 550 Their concentrations were from 7 to 91 times higher than in background air. Other 551 552 fragrance components were detected in some of the analysed samples such as citronellol, 553 which has been found in the composition of cosmetics and toiletries [111], and also 554 methyl dihydrojasmonate, widely used in cleaning and personal care products [114]. The 555 use of cosmetic and personal care products was also evidenced by the presence of Parsol MCX, a common ingredient in sunscreen personal care products [115]. During ironing, 556 the concentrations of this compound oscillated from 5.43 to  $351 \text{ ng m}^{-3}$ . 557

Other hydroxyl compounds were detected in the particulate matter organic extracts. 558 559 Among these, diethyltoluamide (DEET) was found in all samples, including in background air, in concentrations ranging from 43.3 to 164 ng m<sup>-3</sup>. This compound is 560 used as insect repellent in various topical forms containing between 10 and 95% of DEET 561 [111]. 2-Propanol-1-chloro-phosphate (TCPP), a common phosphorous organic flame 562 retardant [116], was found in the background sample and in two of the samples collected 563 during ironing  $(12.5-58.7 \text{ ng m}^{-3})$ . Irganox 1076 and Irgafos 168, which are employed as 564 antioxidants in the manufacture of plastics [117], were also detected in some of the 565

analysed samples. Diethylene glycol is a toxic compound used in some dyes [118]. This 566 567 compound was only found in samples collected during ironing in concentrations ranging from 22.0 to 56.9 ng m<sup>-3</sup>. Benzothiazole was found in PM<sub>10</sub> samples collected during 568 ironing in concentrations ranging from 0.03 to 0.09 ng m<sup>-3</sup>, whereas it was absent from 569 570 the background air sample. This compound has been found previously in clothing textiles (present in 23 of 26 investigated garments) in concentrations ranging from 0.45 to 51 µg 571  $g^{-1}$  textile [119]. Acetyl tributyl citrate, a citric acid ester, is widely used as plasticiser in 572 cosmetics and also in non-cosmetic applications, such as vinyl, coatings and adhesives, 573 574 including the ones intended to be taken as components of articles for packaging, transporting or holding food [120]. This compound was detected in the PM<sub>10</sub> samples 575 collected during ironing (17.5–30.9 ng m<sup>-3</sup>) and in background air (3.25 ng m<sup>-3</sup>). Other 576 plasticisers were present in PM<sub>10</sub> samples. Benzyl butyl phthalate, diethyl phthalate and 577 578 dimethyl phthalate were detected in all samples, including background. Benzyl butyl phthalate had the highest concentrations showing an increase over background 579 580 concentrations ranging from 6 to 26 times. Additionally, during steam ironing with boiler 581 bis(2-ethylhexyl)adipate, bis(2-ethylhexyl)phthalate and di-n-butyl phthalate were also 582 detected in concentrations ranging from 0.033 to 1.83 ng m<sup>-3</sup>.

The  $\Sigma_{19}$  PAHs concentrations ranged from 1.84 (steam iron II) to 13.1 ng m<sup>-3</sup> (steam iron with boiler) during ironing and 3.32 ng m<sup>-3</sup> in the background air (Table 3). The measured PAH concentrations are comparable to those observed in previous studies conducted in European residential settings [121,122]. The congeners with highest concentrations during ironing were retene, benzo[e]pyrene and indeno[1,2,3-cd]pyrene.

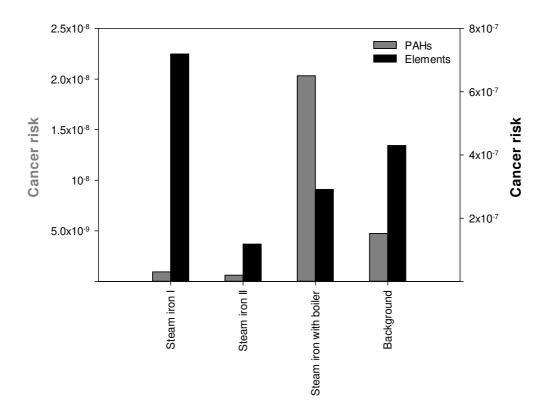
588

# 589 **3.6. Health risk assessment**

Household chores, such as ironing, make up an important part of everyday life and can 590 591 be responsible for human exposure to hazardous pollutants, such as particulate matter and 592 its constituents. Toxicity is related to constituents that are chemically bound to particles 593 in the respirable size range, some of which classified by the IARC as Group 1 594 carcinogens, after establishing causal relationships between exposure to these agents and human cancer. Among the PM-bound constituents, As, Cr (VI), Pb, Co, Cd, Ni and 595 596 benzo[a]pyrene can pose a threat to human health since they have been classified by the IARC as Group 1 based on "sufficient evidence" of carcinogenicity in humans. 597

598 The additional risks of developing cancer due to inhalation exposure during ironing to 599 carcinogenic elements (As, Cr (VI), Pb, Co, Cd and Ni) over the lifetime of an individual 600 (assumed to be 70 years) were found to be negligible (always  $< 7.2 \times 10^{-7}$ ) with respect 601 to the acceptable risk (lower than  $1 \times 10^{-6}$ ) (Fig. 5). The noncancer hazard quotients (HQ) 602 associated with inhalation exposure by households to particulate trace elements in the 603 indoor air during ironing were much lower than unity (safe limit), indicating negligible 604 risks.

605



606

607 Fig. 5. Excess cancer risk for inhalation exposure to PAHs and elements.

608

In the present study, the average concentrations of PAH (expressed as BaP equivalent concentration) ranged from 0.05 to 1.66 ng m<sup>-3</sup> for the steam iron (condition II) and steam iron with boiler, respectively. The resulting excess lifetime lung cancer risks for the exposed person were  $6.2 \times 10^{-10}$  and  $2.0 \times 10^{-8}$  (Fig. 5), i.e. much lower than the acceptable level of  $10^{-6}$ .

Although the cancer risk and non-cancer hazard quotient associated with exposure to PM<sub>10</sub> from ironing thorough the inhalation pathway were found to be negligible, it is necessary to bear in mind that, in contrast to the short period of exposure to this domestic task, laundry/ironing workers are occupationally exposed during long working hours, so this activity can represent a danger for these professionals.

619

# 620 **4.** Conclusions

621 The present study aimed to obtain number and mass concentrations, as well as to detail the chemical composition, of particulate matter emitted during ironing with different 622 623 appliances (steam iron and steam iron with boiler) and under distinct conditions 624 (condition I: steam iron and steam iron with boiler with the room door closed, and 625 condition II: steam iron with indoor living room door open). Despite the transient nature of this source, it generated high peaks of particle number and mass concentrations. The 626 627 greatest effect was observed for particle number concentrations (8–322 nm) with average 628 increases from 33 to 72 times over background levels. The increase in PM<sub>10</sub> mass 629 concentrations ranged, on average, from 4 to 10 times over background levels. The 630 highest particulate mass and number emission rates were observed during steam ironing 631 with boiler, indicating that the steam pressure may play a role on particle generation. The 632 results highlighted the great variability in particle emission rates depending not only on 633 the equipment but also on the operator behaviour, suggesting that household exposure 634 can be enhanced or reduced by proper space ventilation and iron selection.

635 In the present study, a vast array of compounds was identified and quantified in the PM<sub>10</sub> 636 samples, some of which may derive from shedding of fibres and volatilisation followed 637 by condensation of constituents of textile fabrics. Elemental carbon, not detected in the background air, encompassed from  $1.1 \pm 0.2$  to  $3.1 \pm 0.9$  %wt. of the PM<sub>10</sub> mass during 638 639 ironing. Bromide was highly enriched in all the ironing samples. Although the contribution of this element to the PM<sub>10</sub> mass was small, the increase in Br concentrations 640 641 over background values ranged from 9 to 51 times during ironing. The good correlations 642 between Br and Cr, Al and Si and between OC and both Si and Br concentrations suggest 643 a common source for these constituents. Many of the PM<sub>10</sub>-bound organic compounds 644 detected are likely to originate in laundry detergents and softeners, as well as in 645 components incorporated during the textile manufacturing process (e.g. flame retardants). 646 Additionally, multiple compounds with other possible sources were also detected, such 647 as markers of cooking, ingredients of personal care products and plasticisers. The cancer risk and non-cancer hazard quotient associated with household exposure to PM<sub>10</sub> from 648 649 ironing thorough the inhalation pathway were found to be negligible.

Although it needs to be supplemented with additional measurements, the database
obtained in this study is potentially useful to determine the contribution of ironing to the
indoor particulate matter levels through source apportionment models. Further

653 investigations, carried out under controlled laboratory environment, are necessary to654 consolidate the conclusions.

## 655 **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.

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