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1 Seasonal fate and gas/particle partitioning of semi-volatile organic compounds
2 in indoor and outdoor air

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14 **Abstract:** Fifty-eight semi-volatile organic compounds (SVOCs) were investigated simultaneously in
15 three indoor (apartment, nursery and office building) and one outdoor environment in the centre
16 of Paris (France). All of these compounds except tetrabromobisphenol A were quantified in the
17 gaseous and particulate phases in all three environments, and at a frequency of 100% for the
18 predominant compounds of each SVOC class. Phthalic acid esters (PAEs) were the most abundant
19 group (di-iso-butyl phthalate: 29–661 ng.m⁻³, diethyl phthalate: 15–542 ng.m⁻³), followed by 4-
20 nonylphenol (1.4–81 ng.m⁻³), parabens (methylparaben: 0.03–2.5 ng.m⁻³), hexachlorobenzene
21 (HCB) (0.002–0.26 ng.m⁻³) and pentachlorobenzene (PeCB) (0.001–0.23 ng.m⁻³). Polycyclic
22 aromatic hydrocarbons (as Σ 8PAHs) ranged from 0.17 to 5.40 ng.m⁻³, polychlorinated biphenyls (as
23 Σ 7PCBi) from 0.06 to 4.70 ng.m³ and polybromodiphenyl ethers (as Σ 8PBDEs) from 0.002 to 0.40
24 ng.m⁻³. For most pollutants, significantly higher concentrations were observed in the nursery
25 compared to the apartment and office. Overall, the indoor air concentrations were up to ten times
26 higher than outdoor air concentrations. Seasonal variations were observed for PAEs, PCBs and
27 PAHs. SVOCs were predominantly identified in the gaseous phase (> 90%), except for some high-
28 molecular-weight PAEs, PAHs and PCBs.

29
30 **Key-Words:** indoor air, outdoor air, semi-volatile organic compounds, gaseous phase, particulate
31 phase, gas/particle partitioning.

32 33 **Highlights**

- 34 - Quantification of 58 semi-volatile organic compounds in indoor and outdoor air.
- 35 - Phthalates at highest concentrations in all environments whatever the season.
- 36 - Higher indoor air concentrations in a nursery than in an apartment or office.
- 37 - First determination of gas/particle partitioning for parabens.
- 38 - Most pollutants in gaseous phase whatever the environment.

39

40 **1. Introduction**

41 In recent decades, a wide variety of semi-volatile organic compounds (SVOCs) have been
42 detected in the environment and health damage associated with exposure to SVOCs has been
43 reported. Human exposure to these priority substances during foetal development or early life has
44 been associated with adverse birth outcomes and neurotoxic, immunotoxic and obesogenic
45 effects in children (Casas et al., 2013).

46 However, few studies have simultaneously quantified a large number of these compounds in
47 indoor air (Blanchard et al., 2014; Rudel and Perovich, 2009; Mandin et al., 2016). Some of the
48 studied compounds, such as diethylhexyl phthalate (DEHP), bisphenol A (BPA), 4-nonylphenol (4-
49 NP) and polybromodiphenyl ethers (PBDEs), have been registered on the European priority list of
50 the registration evaluation and authorization of chemicals system (Reach, 2015).

51 Given their widespread use in building materials and consumer products for many years,
52 SVOCs are ubiquitous in indoor environments (Blanchard et al., 2014; Laborie et al., 2016). Indeed,
53 synthetic flooring, plastic items, detergents and electronic appliances such as televisions and
54 computers have introduced new chemicals into indoor environments because of their particular
55 properties (flame retardants, biocides, refrigerants, surfactants, etc.) (Weschler, 2009). Moreover,
56 in densely urbanized areas, it appeared that indoor environments were a major source of
57 pollutants for outdoor ambient air (Rudel and Perovich, 2009).

58 SVOCs may be found adsorbed upon airborne particles, as well as in the gaseous phase. Most
59 literature data pertain to airborne particles, the gas phase, or the total air concentration for
60 several families of pollutants, but few studies have considered the gaseous and the particulate
61 phases simultaneously and individually (Blanchard et al., 2014; Fromme et al., 2009; Rudel et al.,

62 2010; Weschler et al., 2008). Few data are available for some compounds such as PBDEs,
63 particularly in the gas phase in indoor and outdoor environments (Blanchard et al., 2014).

64 Moreover, validation of models for the indoor environmental fate and human exposure such
65 as those described by Zhang et al. (2014) and prediction of gas/particle distribution from empirical
66 equations (Wei et al., 2016) require a great number of experimental data.

67 Thus, the overall objective of this study was to investigate SVOC occurrence in both the
68 gaseous and particulate phases of ambient air from different indoor environments. A wide variety
69 of SVOCs with distinct physico-chemical properties were targeted on the basis of their probable
70 occurrence in indoor air, their associated health concerns (Bonvallot et al., 2010) and the technical
71 feasibility of measurement simultaneously (Alliot et al., 2014). These compounds comprised seven
72 phthalic acid esters (PAEs), BPA, two alkylphenols (APs), four parabens, eight PBDEs,
73 tetrabromobisphenol-A (TBBPA), 18 PCBs, hexachlorobenzene (HCB), pentachlorobenzene (PeCB)
74 and 15 PAHs.

75 PAEs are ubiquitous environmental chemicals because they have been used as plasticizers for
76 more than 40 years and their applications are diverse, ranging from cosmetics or flooring to
77 pharmaceutical products and medical devices (Fromme et al., 2009). DEHP, with a high-production
78 volume of around 200 000 tons in the European Union, is mainly used in polyvinyl chloride (Gaudin
79 et al., 2011). Dimethyl (DMP) and diethyl (DEP) phthalates are commonly found in cosmetics as
80 carriers or solvents for synthetic musks (Sanchez-Prado et al., 2011). European legislation has
81 prohibited the use of DEHP for manufacturing single-use food packaging films. However, DnBP and
82 BBP are authorized in single-use packaging for fat-free food storage. In addition, in the cosmetic
83 industry, DEHP, BBP, n-pentyl, di-n-pentyl, iso-pentyl and di iso-pentyl phthalate have been
84 forbidden (Directive 2004/93/EC, 2004; Directive 2005/80/EC, 2005). DEHP, DBP and BBP are
85 prohibited in the manufacture of toys and childcare articles and the prohibition of DnOP, di-iso
86 nonyl phthalate (DiNP) and di-iso-decyl phthalate (DiDP) is limited to toys and childcare articles

87 (Directive 2005/84/EC, 2005). BPA is used in polycarbonate plastics in many consumer products
88 and in the epoxy resins lining food and beverage containers (WHO, 2011). APs, including
89 nonylphenol and octylphenol, are used as nonionic surfactants in detergents and cleaning
90 products and also in some pesticide formulations and industrial products (Ying et al., 2002).
91 Parabens are used as a preservative in personal care products (Błędzka et al., 2014). Among the
92 three major commercial PBDE mixtures, penta- and octa-BDEs have been banned in all
93 applications for the European Union market since 2004 (Directive2003/11/EC, 2003). Deca-BDEs
94 registered under the REACH regulation in 2010 have been restricted to applications such as
95 automotive equipment, textiles and construction and have been prohibited for electric and
96 electronic equipment (BSEF, 2012). PCBs, no longer produced or used, may still be found in indoor
97 air (Hazrati and Harrad, 2006; Heinzow et al., 2007). Finally, PAHs are combustion or industrial by-
98 products. However, in some cases, indoor air contamination by PAHs might originate from glues
99 containing coal tar (Heudorf and Angerer, 2001).

100 The main objectives of this study were to i) prioritize the concentrations of the 58 SVOCs in
101 indoor air for three different buildings in the centre of Paris and to compare them with outdoor air
102 levels, ii) evaluate the seasonal influence on indoor concentrations, iii) investigate SVOC
103 partitioning between the gaseous and particulate phases and iv) explore the relationships
104 between the different SVOCs.

105 **2. Material and methods**

106 **2.1. Sampling sites**

107 Two sampling campaigns were carried out, each over a 6-week period: the first was
108 performed during the non-heating period (from September 21st to November 3rd, 2011), and the
109 second was performed during the heating period (from January 23rd to March 5th, 2012). Each
110 sampling campaign included three successive 2-week periods. The outdoor air samples,
111 concomitant with the indoor samples, came out of a larger study on outdoor air pollution in Paris,

112 from 2009 to 2012 published in Teil et al. (2016). Three different indoor environments were
113 considered: a non-smoking apartment where two adults were living, a nursery (20 toddlers + six
114 adults) and an office building (two persons). Their occupants voluntarily participated in this study.
115 They exhibited no special equipment that might have impeded the results as there was no kitchen
116 or cafeteria in the office building. The three buildings were equipped with mechanical ventilation
117 systems.

118 All the sampling sites were located in densely urbanized areas. The nursery and the office
119 were located at a single site in the centre of Paris (5th arrondissement), in a group of buildings built
120 in 1965 and refurbished in 2003. The apartment was situated in a building constructed in 1993
121 located in the East of Paris (Les Lilas). The nursery and the office were located approximately 4 km
122 from the apartment and 1.7 km from the outdoor monitoring station (Airparif monitoring station,
123 Paris 13th arrondissement).

124 **2.2. Sampling procedure**

125 Both the gaseous and particulate phases of the air were collected at the outdoor and three indoor
126 sites. The sampler consisted of a filter holder (Ecomesure, Janvry, France), two serial cartridges for
127 the XAD-2 resin, a sampling pump (Busch, model SV1005GP, Maulburg, Germany) and a ball flow
128 metre (Hivolin, Germany). The sampling technique was derived from the method described by
129 Granier and Chevreuil (1997). The design of the sampling device and its implementation followed
130 the established recommendations for sampling a gas phase more or less charged with particles
131 (AFNOR, 2003). The efficiency of the adsorption was monitored by adding a second resin cartridge
132 which also allows one to increase the height of the adsorbent and the contact period. Using two
133 cartridges with a small section instead of one large cartridge prevents the emergence of a
134 preferential circulation pathway of air through the resin. Thus, according to the above-mentioned
135 standard, about 90% of the gaseous phase was quantified in the first cartridge (Alliot et al., 2014).

136 The sampling system is shown in SI-Appendix 1. The air flow rate was $0.6 \text{ m}^3 \cdot \text{h}^{-1}$ (medium
137 sampling volume), and the volume collected ranged from 129 to 175 m^3 . The particulate phase
138 was collected in total suspended particulate mode (TSP) onto a 47-mm quartz fibre filter (Q-MA,
139 supplied by Whatman), previously heated to 400°C for 4 h. These filters display an air retention
140 capacity greater than or equal to $\geq 99.95\%$ for particles $\geq 0.3 \mu\text{m}$. The gaseous phase was collected
141 onto XAD-2 resin (20-60 mesh, Sigma-Aldrich, Saint-Quentin Fallavier, France) cleaned by Soxhlet
142 extraction with methanol, acetone and hexane/ether (90/10, v/v) and stored at 4°C before use.
143 Thirteen grams of XAD-2 were used in two brass cartridges (13 mm in diameter; 10 cm long).

144 The indoor air sampling was conducted according to the recommendations of the Swiss
145 Federal Office of Public Health (Waeber et al., 2012). The samples from the apartment were
146 collected in a single room where people spent most of time, i.e., the living room. To minimize
147 artefacts due to air-flow variations close to the walls, the sampling locations in the three indoor
148 locations were set a minimum of 1 m from the wall and 1.50 m high to avoid the suspension of
149 settled particles. The sampling flow of $0.6 \text{ m}^3 \cdot \text{h}^{-1}$ corresponded to 2% per hour of the smallest
150 room volume (30 m^3) and was below the 10% value recommended by Waeber et al. (2012) to
151 avoid depletion of compounds in the room air and possible artefact consisting of particle phase
152 overestimation, and to minimize any change under steady state conditions. The sampling
153 conditions are detailed in SI-Appendix 2. The sampling air velocity through the system varied from
154 80 to $150 \text{ cm} \cdot \text{s}^{-1}$, i.e. lower than the maximum flow of $170 \text{ cm} \cdot \text{s}^{-1}$ recommended by the IEPA
155 method (IEPA/APC, 1985), to optimize the gaseous phase retention on the XAD-2 resin. The indoor
156 temperatures were continuously monitored during the sampling periods with Voltcraft devices
157 (DL-141TH) (Conrad Electronic, Poissy, France). The temperature remained stable in the three
158 indoor environments. Total suspended particulates (TSP) were determined by gravimetry and
159 expressed as $\mu\text{g} \cdot \text{m}^{-3}$ (SI-Appendix 2). Questionnaires were distributed to the occupants to i)
160 provide basic information for interpretation of the results (smoking, frying, use of sprays, cleaning

161 habits, window opening) and ii) record any particular event that might occur during the sampling
162 period (SI-Appendix 3).

163 **2.3. Analytical procedure**

164 Fifty-eight compounds from different chemical families were analyzed in the gaseous and
165 particulate phases. The list of molecules is presented in SI-Appendix 4. The analytical procedure is
166 described in Alliot et al. (2014). Internal standards were added to the gaseous and particulate
167 phase samples before extraction (SI-Appendix 5). The XAD-2 resins were extracted in a
168 polypropylene cartridge by percolation/depression with 100 mL of dichloromethane on a Visiprep
169 system (Supelco). The filters were extracted with 10 mL of hexane/acetone (1:1) by ultrasonic
170 treatment for 20 min. Then each extract was divided into two aliquots. The first aliquot (80%) was
171 retained for the quantification of PCBs, PAHs, PBDEs, PAEs and parabens, and the second aliquot
172 (20%) was retained for the quantification of APs. All experimental procedures (sampling + clean-up
173 + fractionation) and validations for sampling, extraction and analytical methods have been
174 published previously (Alliot et al., 2014).

175 The first aliquot was fractionated upon a Florisil cartridge in three fractions. The F1 fraction
176 was purified on a multi-layer column (neutral alumina activated at 150°C/silica activated at 150°C),
177 and PAHs were analyzed; then the extracts were purified by acidification before analysis for PCBs
178 and PBDEs. The two other fractions were not purified. F1, F2 and F3 were analyzed by gas
179 chromatography-mass spectrometry (GC-MS), gas chromatography-tandem mass spectrometry
180 (GC-MS/MS) or high-performance liquid chromatography tandem mass spectrometry (HPLC-
181 MS/MS) depending on the compounds sought. The second aliquot was analyzed by UPLC-MS/MS
182 using the method of Cladiere et al. (2013). The limits of detection (LODs) and the limits of
183 quantification (LOQs) are provided in SI-Appendix 5.

184 **2.4. QA/QC**

185 The compounds were quantified by internal calibration. Syringe standards were added to the
186 vials prior to analyses to estimate the internal standard recoveries ($^{13}\text{C}_{12}$ PCB194, CB209, BB209
187 and benzyl benzoate). Recoveries were previously assessed for all the target compounds. After
188 extraction and purification, recoveries for the gaseous phase ranged from 30% (TBBPA) to 136%
189 (CB-169), depending on the compound. The retention capacity of the XAD-2 cartridges was also
190 tested (Alliot et al., 2014). Almost all compounds were retained by the first cartridge (at least 90%
191 of the amount trapped). The resins from the two cartridges were mixed and extracted as a single
192 sample to avoid compound losses.

193 The concentrations were corrected by concomitant blank concentrations for all compounds.
194 With the exception of PAEs, all blanks were in accordance with analysis quality (SI, Appendix 5).
195 Peaks were integrated only when the signal-to-noise ratio was ≥ 3 , and peaks were quantified
196 when the signal-to-noise ratio was ≥ 9 , peak to peak.

197 **2.5. Data analysis**

198 ***Concentrations***

199 Concentrations were expressed as medians, minima (MIN), maxima (MAX) and means \pm
200 standard deviation (SD). The concentrations in both the gaseous and particulate phases were
201 expressed as $\text{ng}\cdot\text{m}^{-3}$. Total concentrations in air, expressed as $\text{ng}\cdot\text{m}^{-3}$, were the sums of the
202 gaseous and particulate phases. The results were expressed as the sum of all compounds in each
203 family (Σ compounds) and as individual compounds representative of each family (SI-Appendix 3).

204 The PAEs were expressed as $\Sigma 7\text{PAEs}$ (dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-
205 butyl phthalate (DnBP), di-iso-butyl phthalate (DiBP), butylbenzyl phthalate (BBzP), diethylhexyl
206 phthalate (DEHP) and di-n-octyl phthalate (DnOP)).

207 The PCBs were expressed as ΣPCBi (seven PCB indicators: CB28, 52, 101, 118, 138, 153 and
208 180), $\Sigma \text{PCB-DL}$ (CB77, 81, 126, 169 for non-ortho PCBs dioxin-like and CB105, 114, 118, 123, 156,
209 157, 167, 189 for mono-ortho PCBs dioxin-like) and $\Sigma 19\text{PCBs}$.

210 The PBDEs were expressed as $\Sigma 8$ PBDEs (BDE28, 47, 99, 100, 153, 154, 183 and 209).

211 The target PAHs were (i) fluorene (FLU) and phenanthrene (PHE), which are predominantly low-
212 molecular-weight compounds; (ii) chrysene (CHR) whose molecular weight = 228 mol g⁻¹; (iii)
213 benz[a]anthracene (BaA) and benzo[a]pyrene (BaP), which are carcinogens; and (iv)
214 benzo[ghi]perylene (BghiP), which has the highest molecular weight of the compounds studied.
215 CHR appeared to be the dividing line for the PAHs with a larger or smaller distribution in the
216 gaseous or particulate phase, as it had 48% in gaseous phase and 52% in particulate phase (Lu et
217 al., 2008). These compounds were expressed as $\Sigma 15$ PAHs and $\Sigma 8$ PAHs (AFNOR, 2003). This standard
218 used for the control of PAH emissions from stationary emission sources (BaP, BbF, BkF, IcdP, BaA,
219 DahA, BghiP, FTH) and includes the main carcinogenic compounds.

220 The parabens were expressed as methyl-4-hydroxybenzoate (MeP), ethyl-4-hydroxybenzoate
221 (EtP), propyl-4-hydroxybenzoate (PrP), butyl-4-hydroxybenzoate (BuP) and $\Sigma 4$ Parabens.

222 The APs were expressed as OP, 4-NP and $\Sigma OP+4-NP$.

223 **Statistical analysis**

224 Statistical analyses were performed with Friedman's non-parametric test for the comparison of
225 air concentrations from the three different environments ($\alpha = 0.05$, $n = 6$). Spearman's coefficient
226 was used to explore the correlations between the different SVOC families in the three indoor
227 environments ($\alpha = 0.001$, $n = 18$). The analyses were performed with XLSTAT PRO software
228 (Copyright holder CC-BY-SA-3.0, Addinsoft, Paris, France).

229 **3. Results and discussion**

230 **3.1. SVOC patterns in indoor air**

231 The SVOC concentrations in indoor air (gas + particles) are shown in Figure 1 and details are
232 given in Table 1 and SI-Appendix 6. All targeted compounds were quantified in the three indoor
233 environments, except TBBPA. PAEs were the most abundant SVOCs in indoor air with the following
234 sequence: PAEs > APs > PAHs \geq parabens \geq PCBs > PBDEs > HCB and PeCB. This sequence is in

235 accordance with the production, use, regulation and physico-chemical properties of the
236 compounds. The world production of PAEs and APs are estimated at 4.3 million tons (1 million for
237 Europe) and 500 000 tons per year, respectively (Peijnenburg and Struijs, 2006; Ying et al., 2002).
238 The annual global atmospheric emission of 16 PAHs in 2013 was estimated at 504 000 tons by
239 Chen et al. (2013).

240 For PAEs, parabens, APs and FLU, significantly higher concentrations were observed in the
241 nursery than in the apartment and the office ($p < 0.05$). These concentrations were 1.5 to two
242 times higher in the nursery than in the apartment and the office for PAEs, two to five times higher
243 for parabens and two to six times higher for APs. The highest concentrations were observed in the
244 nursery for all compounds associated with children's products such as toys or widely used care
245 products. Moreover, the apartment showed significantly lower PCB concentrations than the office
246 and the nursery ($p < 0.05$) (Table 2).

247 ***Plasticizers***

248 PAEs were quantified at a frequency of 100% in the three indoor environments, except BBzP
249 and DnOP, the levels of which were close to the LOQs (Table 1, SI-Appendix 5, SI-Appendix 6). As
250 previously reported in California (Rudel et al., 2010; Rudel and Perovich, 2009), PAEs were
251 ubiquitous in indoor air. The concentrations as $\sum 7$ PAEs ranged from 71 to 1362 $\text{ng}\cdot\text{m}^{-3}$.
252 Concentrations in the same range (from 143 to 2600 $\text{ng}\cdot\text{m}^{-3}$ as $\sum 9$ PAEs) were found in different
253 indoor environments including homes, offices and schools in the USA (Tran and Kannan, 2015).

254 DiBP, DnBP and DEP prevailed in the three indoor environments. This should be expected
255 because of their presence in many consumer products (Viñas et al., 2015) and their higher vapor
256 pressures ($3.6 \cdot 10^{-3}$ Pa for DiBP and DnBP and 0.130 Pa for DEP at 25°C) compared to other PAEs
257 (Staples et al., 1997). However, two different compound sequences – DiBP (29–661 $\text{ng}\cdot\text{m}^{-3}$),
258 followed by DEP (15–418 $\text{ng}\cdot\text{m}^{-3}$) and DnBP (11–136 $\text{ng}\cdot\text{m}^{-3}$) – prevailed in the office and the
259 apartment, while DEP (146 – 542 $\text{ng}\cdot\text{m}^{-3}$) was predominant in the nursery ($p < 0.05$, Table 2). This

260 can be due to differences in the number and type of sources such as daily cleaning inside the
261 nursery (SI-Appendix 3) and repeated use of body care products for children. Indeed,
262 determination of PAEs in cleaning and personal care products in Spain showed a preponderance of
263 DEP, particularly in body lotion and floor cleaner (Viñas et al., 2015). In addition, the higher indoor
264 temperature (SI-Appendix 2), which was 21.7 °C for the non-heating season and 23.6 °C for the
265 heating season in the nursery, compared with 19.3 °C and 20.3 °C in the apartment, and 20.3 °C
266 and 21.5 °C in the office, could contribute to higher air concentrations inside the nursery.

267 Higher orders of magnitude of DEP (130–4300 ng.m⁻³) and DnBP (52–1100 ng.m⁻³) were found
268 in dwellings at Cape Cod (USA), and the following sequence DEP > DnBP > DiBP was found in air
269 from apartments in California (USA) (Rudel et al., 2010, 2003). In the Stockholm area (Sweden),
270 the same predominant compounds were reported in indoor air from three different environments
271 i.e., homes, day care centres and workplaces. Their concentrations were higher than ours, except
272 for DMP. Offices appeared to be less contaminated by PAEs, with a Σ 6PAEs of 1500 ng.m⁻³,
273 followed by nurseries with 1900 ng.m⁻³ and then homes with 2700 ng.m⁻³ (Bergh et al., 2011). The
274 prevalence of these compounds might be related to their current use, given that they are present
275 in cosmetics and personal care products (Guo et al., 2013).

276 Overall, DEHP accounted for only 3–9% of the total PAE concentrations. A similar distribution
277 of compounds, with DiBP predominating and lower concentrations of DEP, was observed in French
278 dwellings (Blanchard et al., 2014). Furthermore, the materials used for furnishing might influence
279 these concentrations. DEHP can be emitted to the air from PVC flooring (Xu et al., 2009). Indeed,
280 in 16 types of vinyl flooring tested in the USA, DEHP was the most abundant (7 to 23 %) followed
281 by DiNP (20%) then DnBP (9%) (Liang and Xu, 2014a). The apartment, devoid of PVC flooring in
282 contrast to the office and the nursery, displayed the lowest DEHP concentrations. Moreover, the
283 lower mean temperature in the apartment could influence DEHP volatilization compared with the
284 other environments.

285 Whatever the indoor environment and the season BPA showed very low concentrations (< 1
286 $\text{ng}\cdot\text{m}^{-3}$). This compound is characterized by a vapour pressure of $5.3 \cdot 10^{-5}$ Pa at 25°C (Staples et al.,
287 1998) and is chemically bound to a polymer matrix (Brunelle, 2002), which limits passive emission
288 from materials. Among the three sites, the mean concentration trend was nursery > apartment >
289 office, possibly related to the abundance of equipment and plastic materials in the nursery relative
290 to other environments. Data in the literature concerning indoor BPA were scarce. However, our
291 concentrations were on the same order of magnitude as concentrations measured in apartments
292 (Rudel et al., 2010), houses and day care centres (Wilson et al., 2007) in the USA and dwellings and
293 offices in Japan (Inoue et al., 2006).

294 ***Surfactants***

295 APs (OP + 4-NP) were the second group by order of prevalence, showing a range of
296 concentrations from 1.65 to $90.9 \text{ ng}\cdot\text{m}^{-3}$. 4-NP is a relatively volatile compound (vapour pressure:
297 0.1 Pa at 25°C , (TOXNET, 2014)) and was the most common, with a contribution from 90 to 96% of
298 the total APs. This trend was similar in the three indoor environments. The results indicate higher
299 concentrations in the nursery compared to offices and the dwelling ($p < 0.05$, Table 2). Building
300 materials (concrete, cement, paint), which were similar in the nursery and the office, and the daily
301 use of detergents for surface cleaning in the nursery could be implicated in the indoor emissions
302 (SI-Appendix 4). Very few studies concerning APs in indoor air are available. In the USA, 4-NP was
303 detected in 95–100% of houses (Rudel et al., 2010). Our concentrations were on the same order of
304 magnitude as those measured in homes in the USA and Japan (Rudel et al., 2010; Saito et al.,
305 2004).

306 ***Preservatives***

307 Parabens were the group of SVOCs that showed the greatest discrepancies between buildings,
308 with concentration ratios from 1 to 5. Due to their primary use as antioxidants and preservatives,
309 the concentrations were higher in collective locations ($p < 0.05$) – the nursery ($3.70 \pm 0.89 \text{ ng}\cdot\text{m}^{-3}$)

310 and office ($1.70 \pm 0.73 \text{ ng.m}^{-3}$) – than in the apartment ($0.74 \pm 0.62 \text{ ng.m}^{-3}$) (Table 2). These
311 compounds are relatively volatile, and among the four compounds investigated, the two dominant
312 compounds were MeP (0.8 Pa at 25 °C) and PrP (4.73 Pa at 25 °C) (Ramirez et al., 2010). In the
313 USA, a similar compound distribution was observed, with MeP being the major compound
314 detected in indoor air in 67% of homes with a median concentration of 2.9 ng.m^{-3} (Rudel et al.,
315 2003).

316 ***PCBs and organochlorine compounds***

317 Despite the successive restrictions for the use of PCBs in 1976 and their prohibition in 1986 in
318 France, these organochlorine compounds remain omnipresent in all environmental
319 compartments. Indeed, new “non-aroclor” PCBs were detected as by-product of diarylide yellow
320 (Rodenburg et al., 2010) and other types of pigment production (Anezaki et al., 2015). The
321 concentrations found herein (Figure 1) were on the same order of magnitude as those observed in
322 homes and offices in the UK and Canada between 2003 and 2006 (Hazrati and Harrad, 2006; Zhang
323 et al., 2011). For the three living spaces, CB52, which is relatively volatile with a vapour pressure at
324 25 °C of 0.01 Pa (Burkhard et al., 1985), was the major compound, followed by CB28 and CB101 in
325 equal proportions (Table 1, SI-Appendix 6). This pattern is consistent with the literature, especially
326 in places situated away from combustion process emission sources (Granier and Chevreuril, 1991;
327 Hazrati and Harrad, 2006; Zhang et al., 2011). Our study showed similar PCB patterns among the
328 three indoor environments. It should be noted that a similar congener pattern was reported in the
329 air of apartments from urban areas in Italy (Menichini et al., 2007).

330 The nursery and office showed mean concentrations of $\sum\text{PCBi}$ higher than that in the
331 apartment (1.75 ± 1.82 and $1.48 \pm 0.63 \text{ ng.m}^{-3}$, respectively, versus $0.55 \pm 0.36 \text{ ng.m}^{-3}$; $p < 0.05$)
332 (Table 2). This difference could be explained by the fact that the buildings and the site had been
333 contaminated by PCBs for a long time before being refurbished. Additionally, slightly lower PCB
334 concentrations were found in the air of homes than in offices in Toronto (Canada) by Zhang et al.

335 (2011). PCB levels from 2 to 8.3 ng.m⁻³ were measured in the indoor air of apartments in urban
336 areas in Italy (Menichini et al., 2007). Similarly, a median concentration of 4 ng.m⁻³ was observed
337 in 181 public buildings in Germany. These buildings were mainly schools built between 1960 and
338 1975, which corresponds to the same time frame of the construction of our office and nursery
339 (Heinzow et al., 2007).

340 The average concentrations of HCB (office: 0.111 ± 0.035 ng.m⁻³; apartment: 0.120 ± 0.092
341 ng.m⁻³; nursery: 0.082 ± 0.084 ng.m⁻³) and PeCB (office: 0.132 ± 0.048 ng.m⁻³; apartment: 0.136 ±
342 0.082 ng.m⁻³; nursery: 0.051 ± 0.054 ng.m⁻³) remained low and varied slightly between buildings
343 and seasons, with the lowest values being observed in the nursery for the non-heating period. The
344 agricultural use of HCB as a fungicide has been banned in France since 1972. Recent sources of this
345 persistent organic pollutant and its degradation by-product, PeCB, are mainly related to transport
346 (50%) and accidental synthesis by industrial processes (29%) (CITEPA, 2014).

347 ***Flame retardants***

348 PBDEs were always detected, regardless of the season and indoor environment, and BDE209
349 and BDE47 were predominant. The highest Σ 8PBDEs concentrations were observed in the office
350 (0.132 ± 0.130 ng.m⁻³), while similar concentrations were found in the apartment (0.055 ± 0.060
351 ng.m⁻³) and the nursery (0.044 ± 0.052 ng.m⁻³). PBDE concentrations were close to those found in
352 buildings in Europe, on the order of magnitude of several hundred pg m⁻³, with BDE209 followed
353 by BDE47 present at higher concentrations in houses and apartments than in nurseries (Fromme
354 et al., 2009; Thuresson et al., 2012). BDE47 was the most volatile BDE detected (vapour pressure
355 2.15 10⁻⁴ Pa (Wania and Dugani, 2003)) and the most abundant in the office. TetraBDEs and
356 pentaBDEs have not been produced in the EU since 1997, but they were mainly used in
357 applications with a long lifetime, such as in automotive and upholstery applications, that are still in
358 use (European Commission, 2014). Moreover, formation of nona- to tri-BDE congeners by solar
359 photodecomposition of BDE209 has been reported (Bezares-Cruz et al., 2004). Our results are

360 consistent with those reported in Sweden by Thuresson et al. (2012) and de Wit et al. (2012), who
361 found maximum values in offices (4.0 ng.m^{-3}), followed by nurseries (0.3 ng.m^{-3}) and finally
362 apartments (0.06 ng.m^{-3}).

363 TBBPA remained below the LOQ of 0.14 ng.m^{-3} . Indeed, this compound has low volatility with
364 a vapour pressure at $25 \text{ }^{\circ}\text{C}$ of $1.8 \cdot 10^{-6} \text{ Pa}$ (TOXNET, 2014). Therefore, no particular trend could be
365 found for this compound, consistent with the literature. The concentrations measured in the
366 indoor air of houses and garages in the USA and of houses and offices in Japan and the UK are in
367 the range of $10\text{--}20 \text{ pg.m}^{-3}$ (Abdallah et al., 2008; Batterman et al., 2009; Inoue et al., 2006;
368 Takigami et al., 2009).

369 ***Compounds from combustion processes***

370 PAH concentrations, reported as $\Sigma 8\text{PAHs}$, showed no significant variations among the types
371 of building (office: $0.91 \pm 0.81 \text{ ng.m}^{-3}$, apartment: $1.05 \pm 2.14 \text{ ng.m}^{-3}$; nursery: $0.58 \pm 0.41 \text{ ng.m}^{-3}$).

372 Among PAHs, FLU and PHE were the most abundant in the three buildings, and the highest
373 concentrations were found in the nursery ($p < 0.05$). The nursery is located on the ground floor,
374 while the office and apartment are on the fourth and second floors, respectively. In Italy, indoor
375 PAH concentrations on the lower floors of buildings exceeded the concentrations at higher levels
376 when heating was off, outdoor vehicle exhaust being the dominant source. In contrast, the levels
377 were equal or even lower on the lower floors when the heating was on. The vertical gradient of
378 indoor PAHs between different floors was within a factor of 2 (Menichini et al., 2007). Moreover,
379 Krugly et al. (2014) reported a remarkably high PHE concentration in a kindergarten (up to 115
380 ng.m^{-3}) compared to school sites. They attributed this result to the site location in the city centre
381 with intensive road traffic in the vicinity and frequent opening of windows for ventilation.

382 Despite the possible emissions from cooking activities, the apartment and the nursery showed
383 BaP concentrations lower than the office, with mean values of 0.029 ± 0.019 and 0.006 ± 0.001
384 ng.m^{-3} , respectively, vs. $0.108 \pm 0.081 \text{ ng.m}^{-3}$ for the office ($p < 0.05$). In contrast, higher BaP

385 concentrations linked to cooking that ranged from 6.1 to 24 ng.m⁻³ were found in Chinese
386 dwellings (Zhu and Wang, 2003). The BaP concentrations in the nursery were lower than those
387 observed in the USA: from 0.054 to 0.156 ng.m⁻³ (Wilson et al., 2001). In this study, the TSP
388 concentrations could not explain the variability of BaP concentration between indoor sites.

389 In the UK, Delgado-Saborit et al. (2011) focused on the particulate phase of PAHs and found
390 no differences between a wide range of dwelling and work environments. The median PHE
391 concentrations in houses (n = 162) and offices (n = 30) were 0.34 and 0.22 ng.m⁻³, respectively.
392 Mean BaP concentrations of 0.09 ng.m⁻³ were reported for both environments.

393 The literature reports a wide range of environmental contexts and hypotheses on PAH occurrence
394 in indoor air, but it excludes all emissions other than combustion processes. However, other
395 modes should not be neglected given that PAHs may occur as traces in many products other than
396 fossil fuels. Some of the major companies manufacturing computer hardware mention on their
397 websites the accidental occurrence of PCBs, PBDEs, TBBPA and some PAHs in their products, at
398 levels below the limits prescribed by European environmental and health regulations (HP, 2015;
399 DELL, 2015). This is an example of potential emission sources at room temperature which might
400 contribute to differences in SVOC composition and concentration in indoor air, since most plastics
401 are derived from petroleum products.

402 **3.2. Indoor versus outdoor concentrations**

403 The indoor pattern was close to that observed in the outdoor environment by Teil et al. (2016)
404 (Figure 1, Table 1, SI-Appendix 6). The indoor air concentrations of PAE compounds, except DEHP,
405 were significantly higher (from 23 to 40 times) than the outdoor air concentrations (Table 1, SI-
406 Appendix 6). The opposite was found for outdoor concentrations of DEHP, which were similar to
407 those of the office and the apartment and only half that of the nursery. These discrepancies might
408 be explained by the type of sources of these compounds. DEHP originates from indoor and
409 outdoor passive volatilization, whereas other PAEs such as DEP and DiBP are emitted indoors by

410 materials and consumer products (Teil et al., 2006). Generally, it is agreed that outdoor sources of
411 particulate PAEs, such as tyre wear, are minor contributors to indoor contamination (Rakkestad et
412 al., 2007). AP concentrations in indoor air were eight to 80 times higher than the outdoor
413 concentrations (Table 1), indicating that indoor sources are important.

414 Indoor PCB concentrations were generally up to ten times higher than those measured in the
415 outdoor air in the same area (Teil et al., 2016). Studies of PCB contamination have shown indoor
416 air concentrations to be at least ten and up to 100 000 times higher than outdoor air
417 concentrations (Rudel and Perovich, 2009).

418 Indoor air concentrations of PAHs tend to exceed outdoor air concentrations for compounds
419 containing up to four rings. For molecules containing five rings and more, the trend indicates that
420 the greatest concentrations are outdoors. Similar results were found in homes in the USA;
421 however, acenaphthene (ACE), FLU, FTH and PHE appear to have significant outdoor sources and
422 suggest a combination of indoor and outdoor sources (Rudel et al., 2010).

423 Overall, the SVOCs showed higher indoor concentrations, except for the PAHs, suggesting
424 primarily indoor sources. The different SVOC sources were confirmed by Spearman coefficients,
425 which showed strong correlations between APs, parabens and PAEs. No correlation was found for
426 PAHs (Table 3).

427 **3.3. Indoor air concentrations depending on the season**

428 The variation of indoor concentrations (Table 1) was evaluated at two different outdoor
429 temperatures: a 15.4 °C mean daily value in the non-heating period and 4 °C in the heating period,
430 while the mean annual indoor temperature ranged from 19.3 to 23.6 °C (SI-Appendix 2).

431 The PBDE concentrations exhibited no seasonal variation, except for that of BDE209 inside the
432 office. For this compound, Hazrati and Harrad (2006) reported that although concentrations in
433 warmer months usually exceeded those in colder months, seasonal variability in buildings appears
434 less significant than that observed for outdoor air. Parabens remained steady in the office and the

435 nursery, related to a similar use of cleaning products, whereas the apartment displayed a 2.5-fold
436 lower concentration during the heating season compared to the non-heating season.

437 No seasonal variation was observed for PAEs except in the apartment. The low temperature
438 variation in the 3 indoor sites (maximum 2°C in the nursery) might explain this observation. The
439 significant increases of indoor PAE concentrations reported in the USA were observed for far
440 higher temperature increase (10 °C) (Bi et al.,2015 ; Liang and Xu ;2014b).

441 AP concentrations were slightly lower in the heating period in the office and the nursery, and
442 in the apartment, they were threefold lower in the non-heating season compared to the heating
443 season. For these three chemical families, considering a steady indoor temperature in the three
444 buildings and similar occupant behaviour in terms of window opening in the heating and non-
445 heating seasons, the absence of a statistically significant seasonal variation argues in favour of a
446 contaminant originating mainly from indoor sources rather than outdoor sources (Table 1).

447 A clear seasonal variation was observed for BPA, PCBs and PAHs whatever the indoor site
448 (Table 1). BPA displayed temporal variation, with a fourfold lower concentration in the heating
449 period compared to the non-heating period.

450 The PCB concentrations in the office showed the least seasonal variation, with a nearly
451 twofold lower concentration in the heating period (1.91 vs 1.05 ng.m⁻³). In the apartment and the
452 nursery, air concentrations decreased from 0.82 to 0.27 ng.m⁻³ and from 3.23 to 0.28 ng.m⁻³ in the
453 heating and non-heating periods, respectively. Thus, the seasonal variability of PCB indoor
454 concentrations seems higher than the variability between each site, whereas the temperature
455 inside the buildings remained steady, suggesting a potential outdoor source and higher
456 volatilization at ambient temperatures in the urban area. Indeed, seasonal variations of indoor
457 PCB concentrations were reported, the winter period presenting systematically lower
458 concentrations than the summer period (Waeber et al., 2012). However, in Birmingham (UK), the

459 seasonal variability of indoor contamination in offices, homes and public environments appeared
460 lower than that of outdoor air (Hazrati and Harrad, 2006).

461 In contrast to the other SVOCs, the seasonal variability of indoor PAH concentration was
462 relatively high. Thus, the indoor concentrations increased during the heating period for all PAHs.
463 The variations observed were mainly the result of PAH emissions from domestic heating activities
464 in winter. Indeed, outdoor air concentrations, measured as $\Sigma 8\text{PAHs}$, were twice as high in the
465 heating season as in the non-heating season. In 2012, domestic heating accounted for
466 approximately 74% of the emissions from combustion processes (CITEPA, 2014). Data from the
467 local ambient air monitoring network, Airparif, showed that from January 2011 to December 2012,
468 wide discrepancies occurred due to seasonal PAH variations, with only minor concentrations from
469 April to October. Moreover, the contribution of residential areas and domestic heating to PAH
470 levels in the urban centre in winter was shown, whereas during the non-heating period, only
471 background PAH levels corresponding to motorway traffic were observed. The seasonal pattern of
472 indoor air contamination results from that of outdoor contamination (Airparif, 2013). In Rome
473 (Italy), daily concentrations of BaP ranged from 0.5 (heating off) to 4.6 $\text{ng}\cdot\text{m}^{-3}$ (heating on) in
474 indoor (apartments) and from 0.6 (heating off) to 1.5 $\text{ng}\cdot\text{m}^{-3}$ (heating on) in outdoor (sample on
475 the roof) (Menichini et al., 2007). Thus, the indoor air exhibited higher concentrations than
476 outdoor air during the heating season.

477 **3.4. Gaseous/particulate phase partitioning**

478 APs and parabens were found almost exclusively in the gaseous phase, ranging from 85 to
479 97% and from 86 to 100% of air concentrations, respectively, regardless of the building and the
480 season. Additionally, PCBs were almost exclusively in the gaseous phase, from 73 to 100%, in the
481 non-heating period and from 33 to 100% in the heating period.

482 PBDEs were mainly in the particulate phase with a mean percentage ranging from 40 to 75%,
483 except BDE 28, which was exclusively in the gaseous phase. Unexpectedly, BDE 209 was detected

484 mainly in the gaseous phase, but its concentration was often around its LOQ. BDE-209 was only
485 rarely detected in the gaseous phase only and never in the particulate phase of indoor air in 30
486 French dwellings (Blanchard et al. 2014).

487 For PAEs, except for BBzP, DEHP and DnOP, the gaseous phase accounted for > 90% of air
488 concentrations. In contrast, DEHP was prevalent in the particulate phase, accounting for 63% in
489 the non-heating period and 84% in the heating period. BPA was mainly in the particulate phase
490 (65–72%), as previously observed by Blanchard et al. (2014).

491 Similarly, less volatile compounds such as the heavy PAHs, were almost exclusively in the
492 particulate phase in the heating period. In the indoor air of preschools in Lithuania, similar PAH
493 distributions were found with 100 and 80% of the two- and three-ring compounds, respectively, in
494 the gaseous phase, whereas 100 and 62% of the five- and six-ring compounds, respectively, were
495 in the particulate phase (Krugly et al., 2014). The high proportion of PAHs (85%) in the gaseous
496 phase of indoor air from a preschool in Portugal clearly showed that an adequate assessment of
497 indoor PAH concentrations and the associated exposure requires consideration of the gaseous
498 compounds, which are neglected in most studies (Oliveira et al., 2014).

499 Partitioning between the gaseous and particulate phases is shown on Figure 2 and in SI-
500 Appendix 7. Figure 2a represents the relationship between vapor pressure and gas/particle
501 partitioning during the non-heating period to avoid the effects associated with combustion
502 processes. The relationship was a non-linear regression ($r^2 = 0.596$, $p < 0.001$) with the following
503 equation (eq. 1):

$$504 \quad \% \text{ gaseous phase} = 103.97 / (1 + \text{Exp}(-4.08 - 0.91 * \log(\text{vapor pressure}))) \quad (\text{equation 1})$$

505 This equation is consistent with Junge-Pankow adsorption model (Harner and Bidleman,
506 1998).

507 For the heating period, the variation of gaseous phase vs. vapor pressure showed a similar
508 model equation than during the non heating period (fig. 2b and eq. 2).

509 % gaseous phase = $96/(1+\text{Exp}(-5,15-1,42*\log(\text{vapor pressure})))$. (equation2)

510 Compounds with vapor pressure higher than $1\ 10^{-3}$ Pa exhibited a percentage in the gaseous
511 phase greater than 80% except BDE 209 and hepta-hexa PCBs, which were mainly in the gaseous
512 phase while their vapor pressure was lower than $1\ 10^{-3}$ Pa. Indoor temperatures (20 °C) and low
513 indoor concentrations of TSP ($25\ \mu\text{g}\cdot\text{m}^{-3}$) favoured this general predominance of SVOCs in the
514 gaseous phase. Moreover, a seasonal influence on partitioning was observed, except for parabens,
515 PCBs and the lowest molecular-weight PBDEs and APs. The other compounds were higher in the
516 particulate phase during the heating period.

517 The K_p values showed a variation trend opposite to that of the gaseous phase concentrations
518 between the 2 periods ie an increase of the K_p during the heating period for the heavy compounds
519 with low vapor pressures ($< 10^{-3}$ Pa). However, the difference of temperature between the heating
520 and the non-heating periods was only 2 °C in the nursery and 1 °C for the 2 other sites.

521 The K_p values seemed rather related to the physico-chemical characteristics of the
522 compounds (molecular weight, vapor pressure), as discussed by Wei et al. (2016), than to the
523 particle origin (indoor/outdoor).

524

525 4. Conclusion

526 This study underlines the ubiquity of SVOCs in indoor air. A wide spectrum of 58 semi-volatile
527 organic compounds was quantified in air, even at trace levels, by a device using a medium-volume
528 sampler and multi-residue analysis.

529 Indoor air contamination by the different SVOC families was consistent with the sources,
530 production, uses and regulation of compounds. The concentrations and patterns found here were
531 consistent with literature. Among the contaminants studied, PAEs prevailed, whatever the location
532 and the time of year.

533 Our results highlighted, for the first time, the major occurrence of both APs and parabens in the
534 gaseous phase and confirmed the gas-phase prevalence of most PAEs, light PAHs and PCBs.

535 For all SVOCs the nursery displayed higher concentrations than the apartment and office. The
536 highest indoor air contamination in the nursery might be related to abundant synthetic materials
537 and extensive use of detergents and disinfectants, contributing to toddlers' high exposure through
538 inhalation.

539 For PBDEs, parabens and APs, the absence of temporal variation and the prevailing indoor as
540 compared to outdoor air concentrations, indicates that they predominate indoors. For PCBs,
541 markedly greater variability was found for season than for the indoor location, which is in line with
542 these compounds originating partially outdoors in urban areas. Moreover, the emission of PAEs
543 and PAHs by volatilization or combustion processes at ambient temperature in outdoor
544 environments might affect indoor air quality. The predominance of the gaseous phase in the
545 occurrence of pollutants indoor was shown, except for DEHP and BaP.

546 These data could also be useful for both inputs and validation of models for indoor environmental
547 fate and human exposure.

548

549

550

551 **Conflict of interest**

552 The authors declare that they have no conflict of interest.

553

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ACCEPTED MANUSCRIPT

Table 1. Mean SVOC concentrations (gas + particles; ng.m⁻³) for the non-heating and heating period, in indoor and outdoor air in centre of Paris. (LOQ: limit of quantification)

| ng m ⁻³ | Office (n=6) | | Apartment (n=6) | | Nursery (n=6) | | Outdoor (Teil et al., 2016) (n=6) | |
|--------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------------------------|-----------------|
| | Non-heating | Heating | Non-heating | Heating | Non-heating | Heating | Non-heating | Heating |
| DMP | 16 ± 11 | 7.4 ± 4.5 | 53 ± 37 | 44 ± 33 | 180 ± 69 | 52 ± 10 | 0.31 ± 0.28 | 0.21 ± 0.09 |
| DEP | 96 ± 54 | 49 ± 8 | 272 ± 163 | 63 ± 49 | 380 ± 180 | 160 ± 19 | 6.4 ± 3.8 | 6.8 ± 2.3 |
| DiBP | 248 ± 103 | 152 ± 24 | 454 ± 202 | 42 ± 22 | 116 ± 32 | 251 ± 24 | 7.8 ± 2.1 | 8.0 ± 1.1 |
| DnBP | 58 ± 22 | 63 ± 6 | 100 ± 31 | 32 ± 23 | 70 ± 30 | 75 ± 21 | 3.4 ± 0.89 | 1.3 ± 0.094 |
| BBzP | 0.70 ± 0.93 | 2.2 ± 0.000 | 8.6 ± 9.3 | 0.63 ± 0.000 | 50 ± 26 | 88 ± 21 | 0.21 ± 0.16 | 0.072 ± 0.061 |
| DEHP | 41 ± 21 | 23 ± 9.1 | 28 ± 22 | 6.2 ± 5.9 | 70 ± 37 | 67 ± 11 | 28 ± 15 | 19 ± 4.0 |
| DnOP | 0.093 ± 0.11 | 2.0 ± 2.3 | 0.23 ± 0.10 | 0.031 ± 0.053 | 0.008 ± 0.013 | 4.7 ± 7.8 | 0.029 ± 0.031 | < LOQ |
| Σ7PAEs | 462 ± 184 | 328 ± 36 | 918 ± 443 | 207 ± 150 | 868 ± 297 | 725 ± 74 | 46 ± 17 | 22 ± 3.4 |
| BPA | 0.72 ± 0.66 | 0.20 ± 0.17 | 0.68 ± 0.56 | 0.39 ± 0.53 | 1.1 ± 1.3 | 0.34 ± 0.31 | 0.14 ± 0.084 | 0.071 ± 0.057 |
| OP | 1.7 ± 0.66 | 1.3 ± 0.90 | 1.0 ± 0.35 | 0.43 ± 0.54 | 6.6 ± 3.1 | 4.8 ± 6.7 | 0.23 ± 0.099 | 0.06 ± 0.019 |
| 4-NP | 25 ± 12 | 15 ± 8.3 | 12 ± 3.4 | 4.2 ± 4.7 | 62 ± 16 | 36 ± 31 | 0.61 ± 0.33 | 0.56 ± 0.095 |
| ΣOP+4-NP | 26 ± 13 | 16 ± 9.2 | 14 ± 3.7 | 4.7 ± 5.2 | 69 ± 19 | 41 ± 38 | 0.84 ± 0.38 | 0.62 ± 0.10 |
| MeP | 0.80 ± 0.38 | 1.1 ± 0.54 | 0.81 ± 0.36 | 0.22 ± 0.25 | 2.21 ± 0.29 | 1.5 ± 0.29 | 0.038 ± 0.029 | 0.062 ± 0.035 |
| EtP | 0.14 ± 0.055 | 0.073 ± 0.002 | 0.074 ± 0.032 | 0.022 ± 0.024 | 0.36 ± 0.11 | 0.26 ± 0.045 | 0.012 ± 0.004 | 0.011 ± 0.009 |
| BuP | 0.36 ± 0.20 | 0.60 ± 0.25 | 0.23 ± 0.14 | 0.06 ± 0.068 | 1.4 ± 0.39 | 1.1 ± 0.23 | 0.012 ± 0.007 | 0.004 ± 0.003 |
| PrP | 0.079 ± 0.042 | 0.06 ± 0.035 | 0.034 ± 0.018 | 0.01 ± 0.009 | 0.17 ± 0.07 | 0.15 ± 0.045 | 0.004 ± 0.003 | 0.003 ± 0.001 |
| Σ4Parabens | 1.3 ± 0.64 | 1.9 ± 0.83 | 1.1 ± 0.55 | 0.32 ± 0.35 | 4.2 ± 0.85 | 3.1 ± 0.61 | 0.066 ± 0.027 | 0.08 ± 0.042 |
| CB28 | 0.23 ± 0.11 | 0.12 ± 0.038 | 0.12 ± 0.063 | 0.042 ± 0.042 | 0.57 ± 0.13 | 0.057 ± 0.012 | 0.022 ± 0.011 | 0.009 ± 0.002 |
| CB52 | 1.0 ± 0.52 | 0.65 ± 0.16 | 0.35 ± 0.15 | 0.12 ± 0.12 | 1.7 ± 0.33 | 0.15 ± 0.044 | 0.067 ± 0.039 | 0.035 ± 0.014 |
| CB101 | 0.22 ± 0.044 | 0.18 ± 0.082 | 0.10 ± 0.099 | 0.045 ± 0.05 | 0.91 ± 0.87 | 0.054 ± 0.031 | 0.041 ± 0.02 | 0.019 ± 0.004 |
| CB118 | 0.025 ± 0.015 | 0.041 ± 0.025 | 0.016 ± 0.012 | 0.013 ± 0.015 | 0.006 ± 0.009 | 0.006 ± 0.005 | 0.013 ± 0.007 | 0.009 ± 0.002 |
| CB138 | 0.076 ± 0.084 | 0.013 ± 0.011 | 0.053 ± 0.07 | 0.01 ± 0.005 | 0.007 ± 0.002 | 0.003 ± 0.003 | 0.015 ± 0.005 | 0.007 ± 0.001 |
| CB153 | 0.099 ± 0.097 | 0.03 ± 0.025 | 0.067 ± 0.079 | 0.019 ± 0.013 | 0.01 ± 0.005 | 0.004 ± 0.003 | 0.02 ± 0.005 | 0.008 ± 0.002 |
| CB180 | 0.17 ± 0.26 | 0.006 ± 0.005 | 0.10 ± 0.17 | 0.014 ± 0.013 | 0.01 ± 0.011 | 0.003 ± 0.005 | 0.02 ± 0.008 | 0.002 ± 0.001 |
| CB77 | < LOQ | < LOQ | 0.001 ± 0.001 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| CB81 | 0.003 ± 0.001 | < LOQ | 0.002 ± 0.002 | < LOQ | < LOQ | < LOQ | 0.002 ± 0.001 | < LOQ |
| CB126 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| CB169 | < LOQ | < LOQ | 0.001 ± 0 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| CB105 | 0.008 ± 0.005 | 0.012 ± 0.009 | 0.004 ± 0.004 | 0.003 ± 0.004 | 0.002 ± 0.003 | 0.001 ± 0.001 | 0.006 ± 0.003 | 0.003 ± 0.000 |
| CB114 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| CB123 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | 0.017 ± 0.007 | < LOQ |
| CB156 | 0.008 ± 0.011 | 0.001 ± 0.001 | 0.007 ± 0.01 | 0.001 ± 0.001 | 0.001 ± 0.001 | < LOQ | 0.001 ± 0.001 | < LOQ |
| CB157 | 0.001 ± 0.001 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| CB167 | 0.002 ± 0.004 | < LOQ | 0.002 ± 0.003 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| CB189 | 0.002 ± 0.003 | < LOQ | 0.001 ± 0.001 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| CB110 | 0.088 ± 0.023 | 0.081 ± 0.033 | 0.042 ± 0.043 | 0.02 ± 0.024 | 0.32 ± 0.22 | 0.018 ± 0.01 | 0.022 ± 0.01 | 0.011 ± 0.002 |
| Σ7PCBi | 1.9 ± 0.58 | 1.05 ± 0.34 | 0.82 ± 0.18 | 0.27 ± 0.24 | 3.2 ± 1.3 | 0.28 ± 0.07 | 0.19 ± 0.082 | 0.089 ± 0.021 |
| ΣPCB-DL | 0.052 ± 0.012 | 0.054 ± 0.036 | 0.034 ± 0.015 | 0.017 ± 0.02 | 0.009 ± 0.011 | 0.006 ± 0.006 | 0.041 ± 0.018 | 0.012 ± 0.002 |
| Σ19PCBs | 2.02 ± 0.56 | 1.1 ± 0.39 | 0.88 ± 0.21 | 0.29 ± 0.26 | 3.5 ± 1.5 | 0.3 ± 0.079 | 0.24 ± 0.10 | 0.10 ± 0.023 |
| HCB | 0.11 ± 0.048 | 0.10 ± 0.026 | 0.17 ± 0.09 | 0.067 ± 0.07 | 0.006 ± 0.003 | 0.15 ± 0.008 | 0.049 ± 0.037 | 0.046 ± 0.016 |
| PeCB | 0.14 ± 0.065 | 0.12 ± 0.034 | 0.16 ± 0.067 | 0.10 ± 0.099 | 0.002 ± 0.002 | 0.099 ± 0.021 | 0.013 ± 0.014 | 0.022 ± 0.008 |
| BDE28 | 0.0012 ± 0.0018 | 0.0021 ± 0.0004 | 0.0004 ± 0.0008 | < LOQ | 0.0031 ± 0.0042 | < LOQ | 0.0001 ± 0.0001 | 0.0001 ± 0.0000 |
| BDE47 | 0.041 ± 0.0478 | 0.043 ± 0.0307 | 0.0014 ± 0.0024 | 0.0009 ± 0.0015 | 0.026 ± 0.032 | 0.0057 ± 0.0051 | 0.0022 ± 0.0014 | 0.0035 ± 0.0027 |
| BDE100 | 0.0039 ± 0.0012 | 0.0031 ± 0.0015 | 0.0008 ± 0.0013 | < LOQ | 0.0007 ± 0.0013 | < LOQ | 0.0008 ± 0.0011 | 0.0012 ± 0.0012 |
| BDE99 | 0.016 ± 0.0044 | 0.012 ± 0.0027 | 0.0033 ± 0.0057 | < LOQ | 0.005 ± 0.0021 | 0.0041 ± 0.002 | 0.003 ± 0.003 | 0.0024 ± 0.0025 |
| BDE153 | < LOQ | < LOQ | 0.001 ± 0.002 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| BDE154 | 0.0011 ± 0.001 | 0.0007 ± 0.0006 | 0.0011 ± 0.0019 | < LOQ | 0 ± 0 | < LOQ | 0.0002 ± 0.0002 | < LOQ |
| BDE183 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| BDE209 | 0.13 ± 0.11 | 0.005 ± 0.01 | 0.043 ± 0.063 | 0.04 ± 0.052 | 0.013 ± 0.022 | 0.031 ± 0.053 | 0.0076 ± 0.0046 | < LOQ |
| Σ8PBDEs | 0.19 ± 0.172 | 0.067 ± 0.025 | 0.051 ± 0.077 | 0.041 ± 0.051 | 0.048 ± 0.062 | 0.041 ± 0.055 | 0.014 ± 0.011 | 0.0072 ± 0.0063 |
| ACY | 0.35 ± 0.065 | 0.012 ± 0.012 | 0.63 ± 0.16 | 0.002 ± 0.003 | 0.001 ± 0.001 | 0.01 ± 0.006 | 0.49 ± 0.85 | 0.34 ± 0.59 |
| ACE | 0.96 ± 1.6 | < LOQ | < LOQ | < LOQ | 3.6 ± 1.1 | < LOQ | 1.0 ± 0.76 | 0.34 ± 0.30 |
| FLU | 1.8 ± 0.70 | 4.6 ± 0.84 | 2.18 ± 0.63 | 1.6 ± 1.7 | 5.3 ± 1.8 | 0.008 ± 0.015 | 1.5 ± 0.70 | 1.0 ± 0.87 |
| PHE | 2.9 ± 1.2 | 6.2 ± 1.0 | 3.1 ± 1.1 | 2.1 ± 2.2 | 5.0 ± 1.9 | 5.7 ± 1.6 | 3.8 ± 1.2 | 2.7 ± 2.2 |
| ANT | < LOQ | 0.007 ± 0.004 | < LOQ | < LOQ | < LOQ | 0.007 ± 0.003 | < LOQ | 0.068 ± 0.057 |
| FTH | 0.21 ± 0.019 | 0.87 ± 0.32 | 0.12 ± 0.098 | 0.17 ± 0.15 | 0.21 ± 0.091 | 0.52 ± 0.22 | 0.61 ± 0.15 | 0.86 ± 0.55 |
| Pyr | 0.15 ± 0.046 | 0.77 ± 0.273 | 0.093 ± 0.066 | 0.14 ± 0.14 | 0.16 ± 0.084 | 0.40 ± 0.15 | 0.47 ± 0.15 | 0.65 ± 0.37 |
| BaA | 0.011 ± 0.01 | 0.12 ± 0.084 | 0.01 ± 0.008 | 1.3 ± 2.3 | 0.011 ± 0.009 | 0.07 ± 0.042 | 0.025 ± 0.012 | 0.15 ± 0.084 |
| CHR | 0.017 ± 0.014 | 0.17 ± 0.12 | 0.015 ± 0.013 | 1.3 ± 2.2 | 0.013 ± 0.01 | 0.11 ± 0.08 | 0.051 ± 0.023 | 0.26 ± 0.14 |
| BbF | 0.017 ± 0.008 | 0.23 ± 0.13 | 0.017 ± 0.006 | 0.016 ± 0.027 | 0.019 ± 0.006 | 0.115 ± 0.052 | 0.026 ± 0.011 | 0.21 ± 0.10 |
| BkF | < LOQ | 0.004 ± 0.007 | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | 0.006 ± 0.006 |
| BaP | 0.004 ± 0.006 | 0.141 ± 0.06 | < LOQ | 0.03 ± 0.019 | 0.002 ± 0.003 | 0.10 ± 0.041 | 0.013 ± 0.022 | 0.12 ± 0.084 |
| IcdP | < LOQ | < LOQ | < LOQ | < LOQ | 0.002 ± 0.003 | < LOQ | < LOQ | < LOQ |
| DahA | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ | < LOQ |
| BghiP | 0.013 ± 0.006 | 0.15 ± 0.045 | 0.013 ± 0.012 | 0.34 ± 0.55 | 0.01 ± 0.009 | 0.092 ± 0.036 | 0.023 ± 0.012 | 0.073 ± 0.023 |

| | | | | | | | | |
|------------------------|--------------|------------|-------------|-----------|--------------|-------------|-------------|------------|
| $\Sigma 8\text{PAHs}$ | 0.25 ± 0.009 | 1.5 ± 0.65 | 0.16 ± 0.11 | 1.9 ± 3.0 | 0.25 ± 0.083 | 0.90 ± 0.31 | 0.70 ± 0.20 | 1.4 ± 0.66 |
| $\Sigma 15\text{PAHs}$ | 6.5 ± 3.6 | 13.3 ± 2.8 | 6.2 ± 1.9 | 7.2 ± 9.3 | 14 ± 4.6 | 7.1 ± 1.7 | 8.2 ± 3.3 | 6.9 ± 4.7 |

Table 2. Differences in indoor air concentrations (gas + particles) according to the type of building (Friedman test, $p < 0.05$, $n = 6$).

| Compounds | | Significance $p < 0.05$ | Environment significantly different | |
|-----------|---------------------------|----------------------------|--|--------------------------|
| PAEs | DEP | + | Nursery | Higher concentrations |
| | DEHP | + | Nursery | |
| | $\Sigma 7\text{PAEs}$ | + | Nursery | |
| BPA | | - | | |
| APs | $\Sigma \text{OP+4-NP}$ | + | Nursery | Higher concentrations |
| Parabens | MeP | + | Nursery | Higher concentrations |
| | $\Sigma 4\text{parabens}$ | + | Nursery | |
| PCBs | CB-52 | + | Apartment | Lower concentrations |
| | $\Sigma 7\text{PCBi}$ | + | Apartment | |
| | $\Sigma 19\text{PCBs}$ | + | Apartment | |
| HCB | | - | | |
| PECB | | - | | |
| PBDEs | | - | | |
| PAHs | FLU | + | Nursery | Higher concentrations |
| | BaP | + | Nursery | |
| | $\Sigma 8\text{PAHs}$ | - | | |

Table 3. Relationships between SVOC concentrations in indoor air for all seasons and all buildings (Spearman coefficients; bold: significant r values for $p < 0.001$, $n = 18$).

| | $\Sigma 7\text{PAEs}$ | $\Sigma 4\text{Parabens}$ | $\Sigma \text{OP+4-NP}$ | $\Sigma 8\text{PAHs}$ |
|---------------------------|-----------------------|---------------------------|-------------------------|-----------------------|
| $\Sigma 4\text{Parabens}$ | 0.822 | 1 | | |
| $\Sigma \text{OP+4-NP}$ | 0.820 | 0.949 | 1 | |
| $\Sigma 8\text{PAHs}$ | -0.401 | -0.144 | -0.271 | 1 |
| $\Sigma 7\text{PCBi}$ | 0.643 | 0.727 | 0.799 | -0.265 |

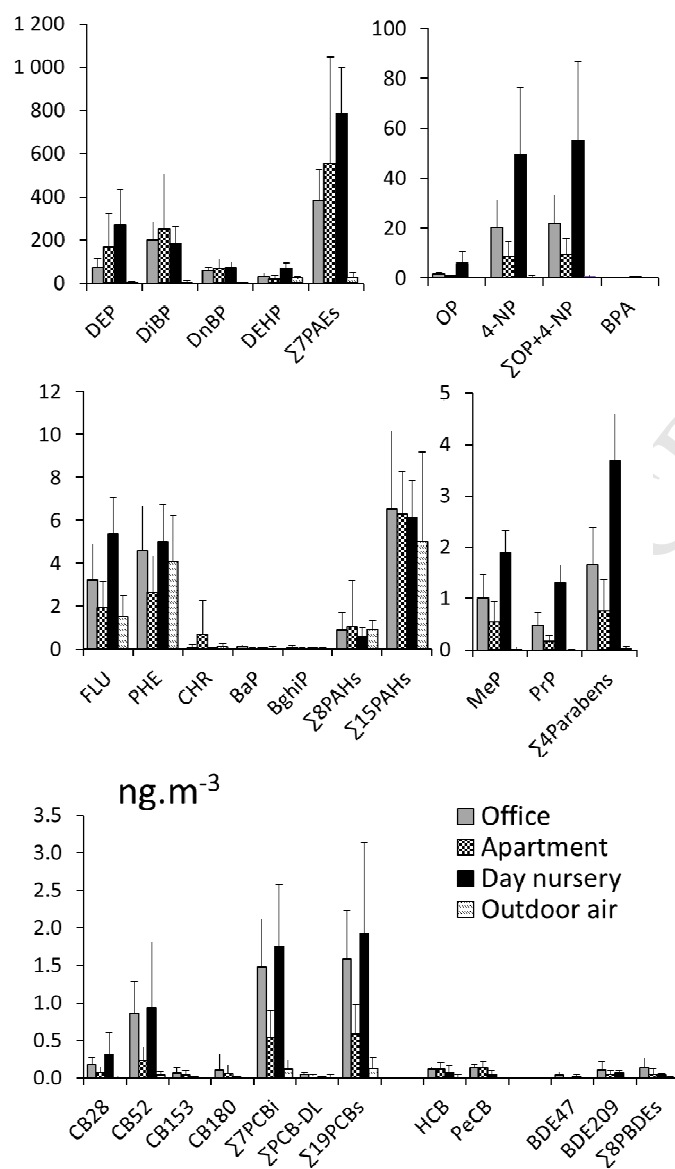


Figure 1: Individual SVOC concentrations (mean \pm SD, $\text{ng}\cdot\text{m}^{-3}$) in indoor air (gas + particles) and in outdoor air in the centre of Paris.

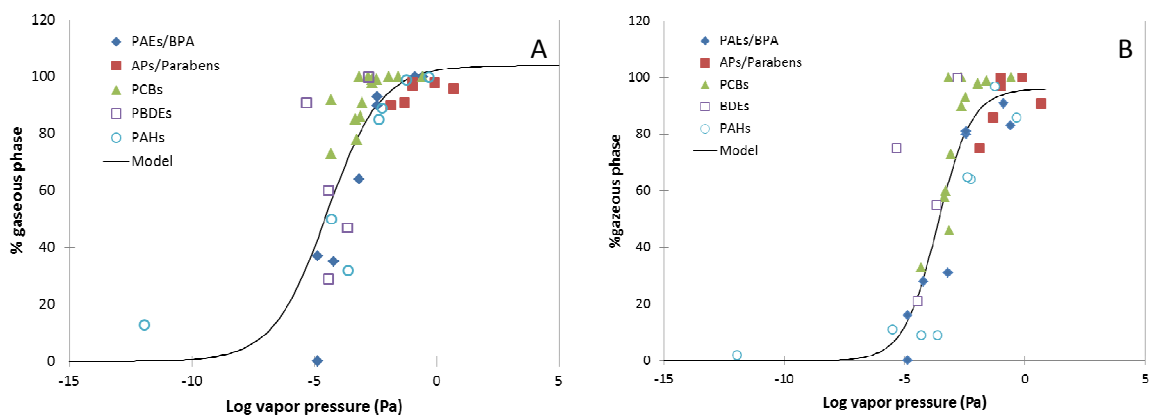


Figure 2: Vapor pressure vs. percentage in gaseous phase for the non-heating period (a) and heating period (b) (n=41)