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1 **Semi-volatile organic compounds in the particulate phase in dwellings: A**
2 **nationwide survey in France**

3
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21

22 **Abstract**

23 Sixty-six semi-volatile organic compounds (SVOCs)—phthalates, polybrominated diphenyl
24 ethers (PBDEs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons
25 (PAHs), one pyrethroid, organochlorine and organophosphorous pesticides, alkylphenols,

26 synthetic musks, tri-n-butylphosphate and triclosan—were measured on PM₁₀ filters collected
27 over 7 days during a nationwide survey of 285 French dwellings, representative of nearly 25
28 million housing units. Thirty-five compounds were detected in more than half of the
29 dwellings. PAHs, phthalates and triclosan were the major particle-bound SVOCs, with a
30 median concentration greater than 1 ng m⁻³ for butylbenzyl phthalate (BBP) (median: 1.6 ng
31 m⁻³), di(2-ethylhexyl) phthalate (DEHP) (46 ng m⁻³) and di-iso-nonyl phthalate (DiNP) (7.9
32 ng m⁻³), and greater than 0.1 ng m⁻³ for triclosan (114 pg m⁻³), benzo(a)anthracene (44.4 pg m⁻³)
33 ³), benzo(a)pyrene (138 pg m⁻³), benzo(b)fluoranthene (306 pg m⁻³), benzo(g,h,i)perylene
34 (229 pg m⁻³), and indeno(1,2,3-c,d)pyrene (178 pg m⁻³). For most of the SVOCs, higher
35 concentrations were found in the dwellings of smokers and during the heating season. The
36 concentrations of banned SVOCs—namely, PCBs and organochlorine pesticides—were
37 correlated. Permethrin, 4-tert-butylphenol and bisphenol-A showed no correlation with the
38 other SVOCs and seemed to have their own specific sources. Most SVOCs were positively
39 associated with PM₁₀ concentration, suggesting that any factor that raises the mass of indoor
40 airborne particles also increases the exposure to SVOCs through inhalation.

41

42 **Keywords:** Indoor air quality, indoor environment, airborne particles, PM₁₀, particulate
43 matter, endocrine disruptors

44

45 **Highlights**

- 46 ► 66 SVOCs were measured in airborne particles (PM₁₀) in dwellings.
- 47 ► PAHs, phthalates and triclosan have the highest concentrations.
- 48 ► Concentrations are higher in smokers' dwellings and during the heating season.
- 49 ► Correlations between SVOCs provide trends regarding common determinants.
- 50 ► High indoor PM₁₀ concentrations promote SVOCs in the particulate phase.

51 **1. Introduction**

52 Semi-volatile organic compounds (SVOCs) are a broad spectrum of molecules from different
53 chemical families that have numerous properties (Weschler and Nazaroff, 2008). They can be
54 used as pesticides, biocides, plasticizers, flame retardants, surfactants, and lubricants.
55 Consequently, they are introduced in numerous applications in buildings or used daily by the
56 entire population. Some SVOCs are also emitted by combustion processes, such as polycyclic
57 aromatic hydrocarbons (PAHs) or polybrominated dibenzo-p-dioxins/furans. Once emitted
58 through evaporation or abrasion in the indoor environment or introduced from the outdoors,
59 their chemical or biological degradation is limited, and they persist indoors (Weschler and
60 Nazaroff, 2008). This persistence also explains why some SVOCs that were banned a few
61 years ago, such as polychlorinated biphenyls (PCBs), are still detected in the air and dust
62 inside buildings (Lehmann et al., 2015).

63 SVOCs are of concern due to their widespread exposure through different environmental
64 media and pathways and their established or suspected health effects, e.g., effects on the
65 reproductive system (Hauser and Calafat, 2005; Saillenfait et al., 2015), lung cancer (IARC,
66 2010), leukemia (Ward et al., 2009), effects on the cognitive and behavioral development
67 (Lyche et al., 2015), and autism (Messer, 2010). Moreover some SVOCs have the potential to
68 disrupt endocrine functions while interfering with hormones (Hwang et al., 2008; Rudel and
69 Perovich, 2009).

70 In indoor environments, SVOCs are divided between the gas phase, airborne particles, and
71 dust settled on floors and surfaces. Of the extensive literature published on SVOCs in
72 buildings, few studies have focused on indoor air compared to settled dust. Moreover, most
73 studies on indoor air have considered PAHs (Ma and Harrad, 2015).

74 The objective of this study was to assess the concentrations of SVOCs bound to indoor PM₁₀
75 at a nationwide level. Sixty-six SVOCs—including phthalates, polybrominated diphenyl

76 ethers (PBDEs), PCBs, PAHs, one pyrethroid, organochlorine and organophosphorous
77 pesticides, alkylphenols, synthetic musks, tri-n-butylphosphate and triclosan—were studied.
78 Correlations between SVOCs were analyzed, and the influence of the season, smoking in the
79 dwelling, and indoor PM_{10} concentration on particle-bound SVOCs was assessed.

80

81 **2. Materials and methods**

82 **2.1. Sample design**

83 The samples were collected via a nationwide survey carried out by the French Observatory of
84 indoor air quality (2003–2005) in a representative sample of the housing stock (Kirchner et
85 al., 2007). Occupied main residences were randomly selected from the 24,672,136
86 permanently occupied housing units in mainland France, excluding oversea territories. The
87 dwelling selection method for the survey was a three-stage process using a probability
88 proportional to size sampling design to ensure that each housing unit had an equal probability
89 of being selected (Golliot et al., 2003). The first stage of the design consisted of randomly
90 selecting primary sampling units (PSUs) among the smallest territorial divisions of France.
91 The second stage corresponded to the sampling of segments within each PSU. In the third
92 stage, housing units were randomly selected within each segment. At the end, 6,268 addresses
93 were drawn at random, and 4,165 households were contacted. The final sample comprises 567
94 dwellings representing the French housing stock.

95

96 **2.2. Sampling of PM_{10}**

97 The measurements were conducted from October 2003 to December 2005. Approximately
98 70% of the measurements were performed during the heating season (October–April), and the
99 remaining dwellings were visited during the non-heating season (May–September). The
100 sampling period was one week (7 days) in each dwelling. The sampling was activated during

101 predefined occupation hours—i.e., in the evening from 5 pm to 8 am the next day (Monday to
102 Friday) and in continuous mode throughout the weekend—, to characterize the indoor
103 concentrations to which the occupants are exposed over one week.

104 The PM₁₀ were collected in the living room through a 2100 Mini-Partisol air sampler
105 (Rüpprecht & Patashnick, Albany, NY, USA), coupled to a ChemPass model 3400 sampling
106 system integrating both PM_{2.5} and PM₁₀ PEMS impactors operating at 1.8 ± 0.2 L min⁻¹. The
107 flow rate was checked onsite with a flow rate calibrator DryCal DC-Lite (Bios International,
108 Butler, NJ). The total sampled volume was 12.6 ± 0.6 m³. Particles were collected on pre-
109 weighed 37 mm diameter PTFE membranes (polytetrafluoroethylene, 2 µm porosity, Gelman
110 Sciences, Ann Arbor, MI, USA). The filters were weighted using a microbalance with a
111 precision of 1 µg (Mettler MT5, Sartorius AG, Goettingen, Germany) in a temperature- and
112 humidity-controlled room. After the gravimetric analysis, the filters were stored at -18 °C
113 until SVOC analysis in 2014. Conservation tests showed that storage at -18 °C does not lead
114 to any loss of the target molecules (Blanchard et al., 2014a). Damaged filters, equipment
115 failure and invalid sampling flow rate reduced the number of valid filters for analysis to 285
116 (Ramalho et al., 2006).

117

118 **2.3. Selection of compounds**

119 A health-based ranking was the starting point for the selection of the compounds (Bonvallot et
120 al., 2010). Briefly, compounds were ranked based on published concentrations in home settled
121 dust and toxicity indicators. In addition, the technical feasibility was considered: some
122 compounds were dropped because they could not be analyzed simultaneously with the others
123 through a multi-residue analytical method (e.g., perfluorinated compounds), whereas other
124 compounds were added, such as triclosan and alkylphenols. Triclosan is an antibacterial agent
125 used in a broad range of household and personal care products (Bedoux et al., 2011), but no

126 data exist on indoor air concentrations. Alkylphenols are used in numerous and various
127 products. 4-tert-butylphenol has been classified as an endocrine disruptor by the European
128 Commission (EC, 2015). However, few studies have reported indoor air concentrations. In the
129 US, 4-tert-butylphenol was recently detected in all 50 homes investigated in California (Rudel
130 et al., 2010) (median: 12 ng m⁻³). In Japan, 4-tert-butylphenol was detected in 99% of the 45
131 homes studied by Saito et al. (2004), and 4-tert-octylphenol was detected in 52%. The median
132 concentrations of 4-tert-butylphenol and 4-tert-octylphenol were 36 and 3.2 ng m⁻³,
133 respectively. Sixty-six SVOCs were ultimately considered for analysis. Table S1 in the
134 supporting information provides the target SVOC names, abbreviations, CAS numbers,
135 molecular weights, and boiling points.

136

137 ***2.4. Sample preparation and SVOC analysis***

138 The SVOCs were simultaneously analyzed in PM₁₀ samples using a simple and efficient
139 multi-residue method based on thermal extraction (TE) and gas chromatography/tandem mass
140 spectrometry (GC/MS/MS). A detailed description of the analytical method is already
141 available elsewhere (Mercier et al., 2012; Blanchard et al., 2014b). This method has been
142 slightly modified for the purposes of this study, as described below (and see Mercier et al.,
143 2014); namely, internal standards and tandem mass spectrometry instead of mass
144 spectrometry were used to minimize matrix interference and background noise.

145 ***Reagents and chemicals***

146 Acetone Pestipur[®] was purchased from Carlo Erba Reagents (Val de Reuil, France). Certified
147 standards of aldrin, cis- and trans-chlordane, 4,4'-DDE, 4,4'-DDT, dieldrin, α -endosulfan,
148 endrin, heptachlor, α -HCH, γ -HCH, metolachlor, chlorpyrifos, diazinon, fenchlorphos,
149 atrazine, methoprotrolyne, oxadiazon, fenprothrin, permethrin, tri-n-butylphosphate,
150 anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(g,h,i)perylene,

151 benzo(k)fluoranthene, chrysene, dibenzo(a,h)anthracene, fluoranthene, fluorene, indeno(1,2,3-
152 c,d)pyrene, phenanthrene, pyrene, PCB 77, PCB 105, PCB 126, butylbenzyl phthalate (BBP),
153 di-n-butyl phthalate (DBP), di(2-ethylhexyl) phthalate (DEHP), diethyl phthalate (DEP), di-
154 isobutyl phthalate (DiBP), DiBP D₄, di-isononyl phthalate (DiNP), di-(2-methoxyethyl)
155 phthalate (DMEP), dimethyl phthalate (DMP), di-n-octyl phthalate (DOP), 4-n-amyphenol,
156 4-tert-butylphenol, 4-n-nonylphenol, 4-tert-octylphenol, bisphenol-A and triclosan were
157 purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). The standard of parathion D₁₀
158 was purchased from Cambridge Isotope Laboratories, Inc. (Andover, MA, USA). The purity
159 of the certified standards was above 95%. Individual standard stock solutions (1 g L⁻¹) were
160 prepared in acetone by accurately weighing 25 mg (± 0.1 mg) of standards into 25 mL
161 volumetric flasks and stored at -18 °C.

162 Acetone solutions (100 mg L⁻¹) of 4,4'-DDT ¹³C₁₂, metolachlor D₆, chlorpyrifos D₁₀ and
163 trans-cypermethrin D₆, acetonitrile solution (100 mg L⁻¹) of trans-cyfluthrin D₆, cyclohexane
164 solutions (10 mg L⁻¹) of chrysene D₁₂, pyrene D₁₀, galaxolide and tonalide, cyclohexane
165 solution (100 mg L⁻¹) of BBP D₄ and cyclohexane mixture (10 mg L⁻¹) of 8 PCBs (PCB 28,
166 31, 52, 101, 118, 138, 153 and 180) were purchased from Dr. Ehrenstorfer GmbH (Augsburg,
167 Germany). Nonane solutions (50 mg L⁻¹) of BDE 28, 47, 85, 99, 100, 119, 153 and 154,
168 nonane/toluene (3%) mixture (5 mg L⁻¹) of ¹³C₁₂-PBDE 47, 99 and 153, nonane/toluene
169 (10%) solution (50 mg L⁻¹) of ¹³C₁₂-triclosan and toluene solution (50 mg L⁻¹) of tri-n-
170 butylphosphate-d₂₇ were purchased from Wellington Laboratories (Guelph, ON, Canada).
171 Acetonitrile solution (100 mg L⁻¹) of bisphenol-A ¹³C₁₂ and isooctane solution (100 mg L⁻¹) of
172 pentachlorobenzene ¹³C₆ were purchased from Cambridge Isotope Laboratories, Inc.
173 (Andover, MA, USA).
174 Calibration solutions were prepared in acetone by the appropriate dilution of individual
175 standard stock solutions and commercial solutions.

176 Pesticide-grade glass wool was purchased from Supelco (Bellefonte, PA, USA). Glass
177 desorption tubes (17.8 cm length \times 6 mm O.D. \times 4 mm I.D.) were purchased from Gerstel
178 GmbH & Co. KG (Mülheim an der Ruhr, Germany). Prior to use, glass wool plugs and glass
179 desorption tubes were heated at 350 °C for 5 min to remove trace organic compounds and
180 minimize background peaks.

181 *Sample and calibration preparation*

182 Each PTFE membrane was cut into four quarters using a scalpel on a glass support. The first
183 quarter of the membrane was used for the analysis of trace compounds, and the remaining
184 three quarters were dedicated to the analysis of highly concentrated compounds, such as
185 PAHs or phthalates. Each quarter was then inserted into a glass desorption tube fitted with a
186 glass wool plug to prevent system contamination due to particles pulled by the helium flow.

187 Each calibration solution in acetone was spiked (1 μ L) on a glass wool plug inside a glass
188 desorption tube also fitted with one-quarter of an unused PTFE membrane.

189 After the addition of internal standards (ISTDs) by spiking 1 μ L of the ISTD solution on the
190 glass wool plug, each glass desorption tube was then immediately transferred to the
191 autosampler for analysis.

192 *Thermal extraction*

193 Thermal extraction of the analytes was performed using a Gerstel TDS3/TDSA2 automatic
194 thermal desorption device (Gerstel GmbH & Co. KG, Mülheim an der Ruhr, Germany)
195 coupled by a transfer line to a Cooled Injection System (CIS, Gerstel GmbH & Co. KG). The
196 thermal extraction process can be divided into two main steps. In the first step, target
197 compounds are thermally extracted (30 to 325 °C (hold 8 min) at 60 °C min⁻¹ in the TDS
198 splitless mode) and carried by the helium flow (100 mL min⁻¹) through the heated transfer line
199 (325 °C) to the cold CIS equipped with a baffled glass liner and cooled with liquid carbon
200 dioxide (-40 °C) during the thermal extraction process to cryo-focus and concentrate the

201 analytes prior to transfer to the capillary column. Following the thermal extraction step, the
202 trapped compounds are rapidly transferred to the capillary column for analysis by heating the
203 CIS (-40 °C to 325 °C (hold 15 min) at 12 °C s⁻¹ in the solvent vent mode). Several methods,
204 differing only in the amount of sample introduced into the capillary column (from 2 to 12.5%
205 of the sample), were used to cover a wide dynamic range from ultra-trace to highly
206 concentrated compounds.

207 *GC/MS analysis*

208 The thermal desorption device was interfaced to a 7890 GC system coupled to a 7000B
209 GC/MS Triple Quad (Agilent Technologies) operated in electron impact ionization (EI) mode
210 (70 eV). Chromatographic separation was performed on a Rtx[®]-PCB capillary column (30 m
211 length × 0.25 mm I.D., 0.25 µm film thickness) supplied by Restek. Helium was used as the
212 column carrier gas at a constant flow rate of 2.0 mL min⁻¹. Chromatographic separation was
213 carried out with the following oven temperature program: 50 °C (hold 2 min), first ramp at 30
214 °C min⁻¹ to 140 °C, and second ramp at 10 °C min⁻¹ to 320 °C (hold 7 min to reach an analysis
215 time of 30 min). The transfer line, ion source and quadrupole temperatures were fixed at 325,
216 300 and 180 °C, respectively. The mass spectrometer was operated in multiple reaction
217 monitoring (MRM) mode. The two most intense and specific MRM transitions for each
218 compound (quantifier and qualifier transitions) were monitored for identification,
219 confirmation and quantification.

220 *Quality Assurance and Quality Control (QA/QC)*

221 This TE-GC/MS/MS method was previously validated in terms of accuracy and precision
222 (Mercier et al., 2012; 2014) via a replicated analysis of the National Institute of Standards and
223 Technology (NIST, Gaithersburg, MD, USA) standard reference materials SRM 1649b (urban
224 dust) and 2585 (house dust). The limits of detection (LODs) and limits of quantification
225 (LOQs) are reported in Table 1.

226 Because of the diversity of the compounds with different chemical and physical properties,
227 labeled and unlabeled ISTDs were selected to match the physical and chemical properties of
228 the analytes as closely as possible, covering volatility differences in particular. 4,4'-DDT
229 $^{13}\text{C}_{12}$, metolachlor D_6 , pentachlorobenzene $^{13}\text{C}_6$, chlorpyrifos D_{10} , fenchlorphos, parathion
230 D_{10} , trans-cyfluthrin D_6 , trans-cypermethrin D_6 , methoprotetryne, fenpropathrin, tri-n-
231 butylphosphate- d_{27} , chrysene D_{12} , pyrene D_{10} , $^{13}\text{C}_{12}$ -PBDE 47, 99 and 153, BBP D_4 , DiBP D_4 ,
232 4-n-amyphenol, bisphenol-A $^{13}\text{C}_{12}$ and $^{13}\text{C}_{12}$ -triclosan were added prior to the thermal
233 extraction step and used as ISTDs. All compounds were quantified with the appropriate ISTD
234 to compensate for the variability associated with the TE-GC/MS/MS analysis from quadratic
235 calibration curves generated for each compound by analyzing at least five different calibration
236 solutions.

237 Each batch of 20 samples included: i) several instrumental (a glass wool plug) and procedural
238 (one-quarter of a PTFE membrane and a glass wool plug) blank samples analyzed as regular
239 samples to assess whether the samples may have been contaminated during analysis, ii)
240 several calibration samples analyzed at least at the beginning and end of the batch to check for
241 the stability of the detector response, and iii) one calibration sample prepared from
242 commercial solutions provided by other suppliers to validate the preparation of the calibration
243 solutions.

244 Positive values for each substance were confirmed by comparing retention times and MRM
245 transition ratios between the calibration solutions and the samples.

246 Eleven field blank samples from different dwellings were treated identically to the samples
247 except that no air was drawn through the sampler. For 59 of the 66 SVOCs, the quantities on
248 the field blank filters were not detectable. For DEHP, DEP, DiBP, DiNP,
249 benzo(k)fluoranthene, permethrin and triclosan, the amounts on the field blank filter were

250 below the LOQ. On one filter, the mass of both benzo(k)fluoranthene and permethrin reached
251 the LOQ.

252

253 *2.5. Statistical analysis*

254 For the statistical analyses, all concentrations below the LOD were set at LOD/2.
255 Concentrations below the LOQ were substituted with the raw output values provided by the
256 laboratory to maintain variability despite higher uncertainties in this range. The geometric
257 mean was estimated only when more than 75% of the concentrations were above the LOQ.

258 Each dwelling ($n = 567$) is associated with a sampling weight, which is the inverse of the
259 probability of inclusion in the sample, which enables the calculation of national estimates for
260 all SVOCs. Because only 285 filters were available for SVOC analysis, the sampling weights
261 needed to be adjusted for nonresponse (i.e., dwellings without SVOC measurements) to avoid
262 bias in the estimates. The weights of the 285 respondents were corrected to compensate for
263 the elimination of the non-respondents. The weights were increased based on the response
264 probability, p , within a group of dwellings. p was estimated by the rate of the number of
265 respondents divided by the number of dwellings belonging to the group. Each sampling
266 weight of the group was then multiplied by the estimated $1/p$ to provide the adjusted weight of
267 each dwelling. A logistic regression model was used to estimate p , and nine groups of
268 dwellings were built by the cross-classification method. The number of groups was defined to
269 avoid being too low p ($p > 0.10$), which would have resulted in unstable estimators. It was
270 then possible to express the results obtained from the sample in terms of national estimates of
271 SVOC concentration on airborne particles. The ‘survey’ package from the R software
272 application (<http://www.R-project.org>) was used (Lumley, 2004, 2010a, 2010b). Quantiles
273 were estimated with the function ‘svyquantile’ of the ‘survey’ package based on the

274 cumulative distribution function (CDF). Details about these calculations and computations of
275 quantiles and their standard error are available in (Lumley, 2010a; Mandin et al., 2014).
276 A non-parametric Kruskal-Wallis test was used to compare ranked SVOC concentrations
277 between dwellings with or without smokers and between dwellings investigated during the
278 heating or the non-heating season. A Dunn test was performed in the case of a significant
279 Kruskal-Wallis test to identify the pairs of categories that differ. Because the study was not
280 originally designed to be representative of smoking habits inside dwellings or the heating
281 season at a nationwide level, sampling weights were not used in these analyses. The statistical
282 level for significance was assigned as $P < 0.05$, and the Bonferroni correction was applied to
283 account for multiple comparisons. The Spearman correlations between the ranked SVOC
284 concentrations were calculated with XLSTAT 2015.4.01 (Addinsoft, Paris, France). This
285 statistical analysis was applied to the 35 SVOCs detected in more than half of the dwellings.
286 Five SVOCs detected in nearly 50% of the dwellings or of particular concern were also
287 considered: bisphenol-A, oxadiazon, BDE 100, 4-tert-octylphenol and galaxolide.

288

289 **3. Results and discussion**

290 **3.1. SVOC concentrations in airborne PM_{10}**

291 The concentrations of the 66 particle-bound SVOCs at the nationwide level ($N = 24,672,136$)
292 are reported in Table 1.

293 [Table 1]

294 Thirty-five compounds (53%) were detected in more than half of the dwellings. The
295 compounds detected in more than 98% of the dwellings were the thirteen PAHs, DEHP, DiNP
296 and triclosan. The other phthalates were also frequently detected, except for the more volatile
297 ones, DMP and DMEP, which were only detected in 39% and 8% of the dwellings,
298 respectively. BDE 47 and BDE 99 were the more concentrated PBDEs on PM_{10} . Among

299 PCBs, the tri- and tetra-congeners were less frequently detected on PM₁₀ because they are
300 more volatile; PCBs 101, 118, 138, and 153 (penta- and hexa-) were the major ones (detected
301 in > 80% of the dwellings). 4-n-nonylphenol was scarcely detected, whereas 4-tert-
302 butylphenol and 4-tert-octylphenol were detected in approximately one of every two
303 dwellings. Except lindane, oxadiazon, 4,4'-DDT and 4,4'-DDE, the organochlorine and
304 organophosphorous pesticides were detected on PM₁₀ in less than 20% of the dwellings. Four
305 compounds were never detected on PM₁₀, namely BDE 119, PCB 126, heptachlore, and cis-
306 chlordane.

307 Twenty-seven SVOCs (41%) were quantified in more than half of the dwellings. Several
308 orders of magnitude were observed between the concentration ranges. The highest median
309 concentrations were measured for the phthalates with values around 1 ng m⁻³ up to 46 ng m⁻³
310 for DEHP. Some PAHs followed with median values around several hundred pg m⁻³, with
311 benzo(b)fluoranthene having the highest median concentration (306 pg m⁻³) on PM₁₀.
312 Triclosan, permethrin, and the remaining PAHs stayed within an order of magnitude lower—
313 i.e., medians of approximately 10–100 pg m⁻³. The lowest particle-bound PAH concentrations
314 were observed for 3-ring congeners (anthracene and fluorene), and the highest were measured
315 for 5- or 6-ring PAHs—namely, benzo(b)fluoranthene and benzo(g,h,i)perylene. Since the 3-
316 ring PAHs are mainly found in the gas phase, low concentrations in the particulate phase were
317 expected. Overall, the lowest particle-bound concentrations were observed for BDE 47 >
318 BDE 99 > 4,4'-DDT > PCBs.

319 The results from studies targeting SVOCs exclusively in the particulate phase or separately
320 from the gas phase concentration were considered for an adequate comparison. Only studies
321 targeting dwellings were considered. The median values are reported in Table 2a.

322 [Table 2a]

323 The PAH concentrations in the French dwellings are largely consistent with the other studies
324 worldwide. The concentrations observed in China (Wang et al., 2013, 2014a) appear to be
325 higher but remain on the same order of those measured during winter in Rome, Italy
326 (Romagnoli et al., 2014). The median PBDE concentrations are considerably lower in France
327 than in North America (Batterman et al., 2009), which has already been reported when
328 comparing PBDE use and population exposure over the world: the higher PBDE
329 concentrations encountered in the United Kingdom and North America indoor environments
330 are due to stricter fire safety standards (Birnbaum and Staskal, 2004; Fromme et al., 2009).
331 BDE 47 and BDE 99 reported in Hong Kong on PM_{2.5} (Wang et al., 2013, 2014a) are lower
332 than the medians measured in the present study. Regarding phthalates, the median
333 concentrations are of the same order of magnitude as the ones measured elsewhere except for
334 DiBP. Compared to Blanchard et al. (2014b) and other countries, DiBP median
335 concentrations are lower because the sampling was performed earlier (2003–2005) and
336 because DiBP has been used as a substitute for DBP in Europe according to the European
337 Council for Plasticisers and Intermediates (ECPI) over the past decade. The phthalate median
338 concentrations reported in one Chinese study are rather high (Wang et al., 2014b). Another
339 study carried out in China also reported high BBP and DEHP concentrations in ten newly
340 decorated apartments, with median concentrations of 1.9 and 1.3 $\mu\text{g m}^{-3}$, respectively (Pei et
341 al., 2013).

342 For the SVOCs that were not quantified in more than half of the dwellings (no median
343 reported), the ranges of concentrations were compared, considering studies reporting particle-
344 bound SVOCs in indoor air of dwellings. They are presented in Table 2b.

345 [Table 2b]

346 Regarding particle-bound PBDEs (28, 85, 100, 153, and 154), the concentration ranges
347 measured in France were lower than those measured in the US by Batterman et al. (2009);

348 they were of the same order of magnitude as those observed in Hong-Kong (Wang et al.,
349 2014a). With regard to phthalates, the DBP and DEP concentration ranges were lower than in
350 Norway (Rakkestad et al., 2007) and the US (Tran and Kannan, 2015) and much lower than in
351 China (Wang et al., 2014a; Zhang et al., 2014). Finally, in the frame of the RIOPA Study
352 (Relationship of Indoor, Outdoor and Personal Air), chlordane was measured in PM_{2.5} in 104
353 non-smoker dwellings from three large cities in the US (Zhu and Jia, 2012). The maximum
354 and median concentrations for trans- and cis-chlordane were high (medians: 17 and 7 pg m⁻³,
355 respectively). In the French housing stock, trans-chlordane was detected in only 1% of the
356 dwellings (*vs.* 29% in RIOPA) and never quantified, and cis-chlordane was never detected
357 (*vs.* 22% in RIOPA). Chlordane was banned in Europe and the US in 1981 and 1983,
358 respectively, but its use as termicide in building construction remained until it was later
359 banned in 1988 (Offenberg et al., 2004). The longer use in the US and a sampling period
360 nearer to the banning date in RIOPA (1999–2000) could explain the higher chlordane
361 concentrations observed in the US compared to France.

362 No large studies were found in the literature to compare the concentrations of 4,4'-DDE,
363 permethrin, PCBs or triclosan to our results.

364

365 **3.2. Particle-bound SVOC concentrations according to season and smoking in the dwelling**

366 The particle-bound SVOC concentrations were compared between dwellings with no smokers
367 vs. dwellings with at least one indoor smoker. In addition, the sampling season may have an
368 influence considering that the dwellings were not investigated during the same period. The
369 season was defined by the heating system turned on or off during the week of measurement.
370 The season appeared to have a strong influence on the indoor concentrations for the other
371 parameters measured during the housing survey, i.e., volatile organic compounds and
372 aldehydes (Langer et al., 2016). To determine whether this relation with the heating period is
373 also observed for SVOCs, their concentrations were stratified over the heating and non-
374 heating seasons. The median concentrations in the heating season with or without smoking,
375 along with the concentration in the non-heating season with or without smoking, are presented
376 in Table 3.

377 [Table 3]

378 The resulting *P*-values of the Kruskal-Wallis test for these four categories were <0.0001 for
379 all SVOCs except permethrin (*P*=0.032), indicating that the concentration in at least one
380 category is globally higher or lower than that of one or the other groups. This result confirmed
381 the major influence of smoking and the season on the particle-bound concentration of all
382 SVOCs.

383 According to the results from the multiple comparisons, SVOCs on PM₁₀ can be categorized
384 into four groups. Half of the SVOCs—i.e., 20 of the 40 for which the statistical test was
385 performed—were associated with both the season and tobacco smoking. Their concentrations
386 were significantly higher during the heating season and in smokers' dwellings. Most of the
387 PAHs can be found in this group. Synthetic musks, most of the organochlorine pesticides and
388 PCBs, and two phthalates also belong to this group. The second group included 15 SVOCs
389 that are more influenced by smoking than by the season: PBDEs, oxadiazon, permethrin,

390 triclosan, two PAHs, four phthalates and PCBs. In contrast, benzo(g,h,i)perylene and
391 bisphenol-A are more influenced by the season than by smoking. In these two groups, the
392 trend remains similar, with higher concentrations during the heating season or in smokers'
393 dwellings. Finally, anthracene and 4-tert-octylphenol cannot be classified in any of the three
394 previous groups.

395 Indoor sources of PAHs are related to combustion processes, such as tobacco smoking,
396 cooking, and wood burning (Fromme et al., 2004; Ma and Harrad, 2015). Additionally,
397 outdoor PAHs from traffic exhausts, industries, and residential heating penetrate into the
398 buildings. In the winter, outdoor PAH concentrations are higher due to residential heating
399 combined with less favorable atmospheric dispersion conditions, which could explain the
400 higher indoor concentrations observed during the heating season. Romagnoli et al. (2014) in
401 Italy, Zhu and Jia (2012) in the American RIOPA study, Ohura et al. (2004) in Japan, and Zhu
402 et al. (2009) in China also observed significantly higher concentrations of particle-bound
403 PAHs in the winter compared to the summer. Zhu and Jia (2012) determined that $55 \pm 9\%$ of
404 the variance of particulate PAH concentrations were attributable to the season. To study the
405 influence of season and smoking on the PAH profile, the average relative contribution of
406 every PAH to the total PAH concentrations on PM_{10} was calculated in the dwellings
407 according to the season and smoking status. The profiles are presented in Figure 1.

408 [Figure 1]

409 Overall, the PAHs show similar trends, with a higher relative contribution of high-molecular-
410 weight congeners. Nevertheless, the contribution of 3- and 4-ring PAHs (here, fluorene,
411 benzo(a)anthracene, and chrysene) was higher in smokers' dwellings, whereas the
412 contribution of 5- and 6-ring PAHs (e.g., benzo(b)fluoranthene, benzo(k)fluoranthene, and
413 benzo(g,h,i)perylene) was lower, as observed by previous authors (Slezakova et al., 2009).

414 The opposite trend was observed when comparing the heating season to the non-heating
415 season.

416 In contrast to our observations, Zhang et al. (2014) found significantly higher concentrations
417 of phthalates (BBP, DBP, DEP, DEHP, DMP, and DOP) on PM_{10} and $PM_{2.5}$ in summer
418 compared to winter in 13 dwellings in Tianjin, China. This result may be explained by
419 different climatic conditions—e.g., a hotter and more humid summer in China than in France.
420 In summer, with higher temperatures, the equilibrium between the gas phase and particulate
421 phase is expected to favor the gas phase, leading to lower SVOC concentrations in the
422 particulate phase. Additionally, in our study different dwellings were investigated at the two
423 seasons. Therefore the seasonal effect was only assessed as an overall factor and not for a
424 particular dwelling.

425 Finally, one of the more important differences between dwellings was associated with
426 triclosan concentrations, which were 4 to 5 times higher in smokers' dwellings than in those
427 of non-smokers. The use of triclosan is not associated with tobacco products. Nevertheless,
428 Kim et al. (2011) found a significant association between cigarette smoking and the urine
429 concentration of triclosan in the Korean adult population aged 18–69.

430

431 **3.3. Correlations between SVOC and PM_{10} concentrations**

432 The correlations between ranked concentrations of SVOCs are presented in Figure 2. With the
433 exception of five pairs, all the SVOCs were significantly and positively correlated.

434

[Figure 2]

435 The strongest correlations were observed within chemical families. All PAHs were highly
436 correlated ($\rho > 0.8$) except the 3-ring PAHs, i.e., anthracene, fluorene and phenanthrene. This
437 result suggests that lower- and higher-molecular-weight PAHs share different sources, which
438 was already observed in previous studies (Delgado-Saborit et al., 2011). The organochlorine

pesticides were moderately correlated with each other ($\rho > 0.4$). The strong correlation between 4,4'-DDT and 4,4'-DDE ($\rho=0.8$) is expected because 4,4'-DDE is a metabolite of 4,4'-DDT. The correlations with and between the other organochlorine, lindane and oxadiazon, suggest common sources or uses of these pesticides, which are now all banned in France. They were substituted in North America and Europe by pyrethroids, such as permethrin (Williams et al., 2008), which displayed a low correlation with lindane and oxadiazon ($\rho < 0.2$) in our study. PBDEs were highly correlated with each other ($\rho > 0.8$) as well as with PCBs ($\rho > 0.8$). Among phthalates, two groups could be distinguished: BBP, DBP and DEHP on one side (cross-correlated with $\rho > 0.6$), with DEP, DiBP and DiNP slightly less correlated with each other. The strong correlation between DBP and DiBP ($\rho=0.86$) suggests common sources for these two compounds and could be explained by the use of DiBP as a substitute for DBP. Overall, the weaker correlations observed between phthalates could indicate that they share different sources. Phthalates are used in a large number of domestic and consumer products, as diverse as building products (vinyl floorings, cables), domestic items (shower curtains, tablecloths, toys, food packages), care products (nail polishes) and perfumes (Hauser and Calafat, 2010). Galaxolide and tonalide were strongly correlated ($\rho=0.81$), which is consistent with the fact that they are used commonly in personal care products and perfumes in Europe (Roosens et al., 2007). Finally, the two alkylphenols are poorly correlated ($\rho < 0.2$), illustrating that they do not share similar sources.

When comparing correlations between chemical families, positive correlations ($\rho > 0.6$) between PCBs and organochlorine pesticides were found, particularly between 4,4'-DDE and PCB 101 ($\rho=0.81$). PCBs were largely used in buildings in the 1970s—e.g., as sealants—and are still measured in dust and indoor air in dwellings constructed during this period (Lehmann et al., 2015). Organochlorine pesticides were used contemporarily before being phased out. The correlations between these two groups suggest that they were both used during the same

464 period in the dwellings. Whitehead et al. (2014) observed this trend in the US for settled dust
465 concentrations. Moderate correlations ($\rho > 0.6$) were also observed between PCBs and some
466 phthalates, particularly DBP and DEHP. Strong correlations ($\rho > 0.8$) between tonalide and
467 PCB 52, and tonalide and DEHP were also found, but no appropriate assumption could
468 explain these associations. Three compounds displayed low correlations or none at all with
469 the other SVOCs, namely, permethrin, 4-tert-butylphenol and bisphenol-A. This result
470 confirms that they have their own specific uses and sources indoors.

471 The median PM_{10} concentration measured in the French housing stock was $31 \mu\text{g m}^{-3}$, and the
472 95th percentile was $182 \mu\text{g m}^{-3}$. The concentrations of the 40 most detected particle-bound
473 SVOCs were significantly correlated with PM_{10} concentration, with a correlation coefficient
474 ranging from 0.29 (permethrin) to 0.77 (DBP; Figure 2). A higher available surface in a given
475 room, including suspended particulate matter, is able to adsorb more SVOCs. Thus,
476 increasing PM_{10} indoor concentrations should lead to higher particle-bound SVOC
477 concentrations: in addition to their adsorption on surface coatings, such as walls and furniture,
478 SVOCs will also adsorb on newly available airborne particles. This finding is consistent with
479 the influence of tobacco smoking, a major source of particles indoors, on the concentrations
480 of nearly all SVOCs. If this increase is particularly noticeable for smoking, the emissions of
481 particles from other indoor sources—including cooking, incense or candle burning, and
482 resuspension—should similarly lead to higher particle-bound SVOC concentrations as well as
483 an increase in the exposure to SVOCs through inhalation. Because ultrafine particles are
484 associated with a larger specific surface, adsorbed SVOCs are likely to be found in the most
485 inhalable fraction of the PM_{10} mass fraction. The influence of the particle dynamics as well as
486 the size fraction collecting the most SVOCs was shown by Liu et al. (2010), based on a
487 modeling approach.

488

489 **4. Conclusion**

490 To our knowledge, this is the first time that indoor concentrations of such a wide range of
491 SVOCs have been assessed simultaneously at a national level. The survey was initially
492 designed for the sampling of particles and thus the analysis of particle-bound SVOCs. To
493 obtain a full understanding of indoor air concentrations in homes for the studied 66 SVOCs,
494 their respective gas phase concentration is currently being modeled from their measured
495 concentration on PM₁₀. The nationwide total concentrations of SVOCs will enable an
496 exposure assessment for the general population in France. The significant correlations
497 observed among all major SVOCs and PM₁₀ imply that all sources of indoor particles may
498 increase exposure to SVOCs through inhalation.

499

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505

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LIST OF TABLES

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Table 2b. Ranges of particle-bound SVOC concentrations in residential indoor air measured worldwide

Table 3. SVOC median concentrations (pg m^{-3}) according to the heating season and smoking in the dwelling

Table 1. Frequency of detection, frequency of quantification and national concentration estimates of 66 particle-bound SVOCs (N = 24,672,136)

SVOC	Unit	LOD	% > LOD	LOQ	% > LOQ	P5	P25	P50	P75	P95	GM	GSD
Polycyclic aromatic hydrocarbons												
Anthracene	pg m ⁻³	0.8	98	2.1	97	4.1	7.8	12.6	19.5	51.2	12.9	2.5
Benzo(a)anthracene	pg m ⁻³	2.1	100	4.2	99	6.5	18.0	44.4	143	655	53.4	4.4
Benzo(a)pyrene	pg m ⁻³	2.1	100	4.2	99	7.5	31.2	138	327	1,097	109	4.9
Benzo(b)fluoranthene	pg m ⁻³	4.2	100	8.4	100	42.7	117	306	636	1,806	299	3.3
Benzo(g,h,i)perylene	pg m ⁻³	4.2	100	8.4	100	27.3	92.2	229	432	1,153	210	3.1
Benzo(k)fluoranthene	pg m ⁻³	2.1	100	4.2	100	9.2	24.7	64	142	337	60.9	3.2
Chrysene	pg m ⁻³	2.1	100	4.2	100	13.6	34.0	75.9	187	1,403	91.5	3.8
Dibenzo(a,h)anthracene	pg m ⁻³	2.1	95	4.2	87	<2.1	7.8	21.6	45.4	126	19.2	3.6
Fluoranthene	pg m ⁻³	4.2	100	8.4	100	19.1	38.6	68.2	128	570	75.9	2.8
Fluorene	pg m ⁻³	2.1	99	4.2	99	6.0	13.4	19.5	44.1	213	25.7	3.0
Indeno(1,2,3-c,d)pyrene	pg m ⁻³	8.4	99	20.9	94	<8.4	70.6	178	312	959	155	3.2
Phenanthrene	pg m ⁻³	4.2	100	8.4	98	16.8	33.9	54.1	104	300	60.7	2.5
Pyrene	pg m ⁻³	4.2	100	8.4	99	14.6	30.7	53.9	103	463	61.4	2.9
Organochlorine pesticides												
Aldrin	pg m ⁻³	2.1	0.5	4.2	0.5	<2.1	<2.1	<2.1	<2.1	<2.1	NA	NA
α -endosulfan	pg m ⁻³	10.4	19	20.9	8	<10.4	<10.4	<10.4	<10.4	28.2	NA	NA
α -HCH	pg m ⁻³	2.1	9	4.2	2	<2.1	<2.1	<2.1	<2.1	<2.1	NA	NA
Atrazine	pg m ⁻³	4.2	0.3	10.4	0.3	<4.2	<4.2	<4.2	<4.2	<4.2	NA	NA
Cis-chlordane	pg m ⁻³	2.1	0	4.2	0	NA	NA	NA	NA	NA	NA	NA
Dieldrin	pg m ⁻³	4.2	23	8.4	13	<4.2	<4.2	<4.2	<4.2	32.0	NA	NA
Endrin	pg m ⁻³	10.4	1	20.9	1	<10.4	<10.4	<10.4	<10.4	<10.4	NA	NA
Heptachlor	pg m ⁻³	10.4	0	20.9	0	<10.4	<10.4	<10.4	<10.4	<10.4	NA	NA
γ -HCH / Lindane	pg m ⁻³	2.1	56	4.2	33	<2.1	<2.1	<4.2	6.4	39.7	NA	NA
Metolachlor	pg m ⁻³	1.0	1	2.1	1	<1.0	<1.0	<1.0	<1.0	<1.0	NA	NA
Oxadiazon	pg m ⁻³	0.4	47	1.0	25	<0.4	<0.4	<0.4	<1.0	4.3	NA	NA
Trans-chlordane	pg m ⁻³	2.1	1	4.2	0	<2.1	<2.1	<2.1	<2.1	<2.1	NA	NA
4,4'-DDE	pg m ⁻³	0.4	53	1.0	33	<0.4	<0.4	<1.0	1.9	27.8	NA	NA
4,4'-DDT	pg m ⁻³	1.0	84	2.1	75	<1.0	<2.1	5.1	24.6	142	NA	NA

SVOC	Unit	LOD	% > LOD	LOQ	P5	P25	P50	P75	P95	GM	GSD
Organophosphorous pesticides											
Chlorpyrifos	pg m ⁻³	41.8	10	83.5	< 41.8	< 41.8	< 41.8	< 41.8	< 83.5	NA	NA
Diazinon	pg m ⁻³	41.8	2	83.5	< 41.8	< 41.8	< 41.8	< 41.8	< 41.8	NA	NA
Pyrethroids											
Permethrin	pg m ⁻³	20.9	76	41.8	< 20.9	< 41.8	55.9	162	1,836	NA	NA
PBDEs											
BDE 28	pg m ⁻³	0.4	22	1.0	< 0.4	< 0.4	< 0.4	< 0.4	1.9	NA	NA
BDE 47	pg m ⁻³	2.1	88	4.2	< 2.1	< 4.2	11.0	33.2	126	NA	NA
BDE 85	pg m ⁻³	4.2	1	8.4	< 4.2	< 4.2	< 4.2	< 4.2	< 4.2	NA	NA
BDE 99	pg m ⁻³	2.1	87	4.2	< 2.1	< 4.2	8.9	19.5	60.3	NA	NA
BDE 100	pg m ⁻³	2.1	47	4.2	< 2.1	< 2.1	< 2.1	4.3	13.6	NA	NA
BDE 119	pg m ⁻³	4.2	0	8.4	NA	NA	NA	NA	NA	NA	NA
BDE 153	pg m ⁻³	4.2	7	8.4	< 4.2	< 4.2	< 4.2	< 4.2	< 4.2	NA	NA
BDE 154	pg m ⁻³	4.2	3	8.4	< 4.2	< 4.2	< 4.2	< 4.2	< 4.2	NA	NA
PCBs											
PCB 28	pg m ⁻³	0.4	18	1.0	< 0.4	< 0.4	< 0.4	< 0.4	2.2	NA	NA
PCB 31	pg m ⁻³	0.4	23	1.0	< 0.4	< 0.4	< 0.4	< 0.4	2.2	NA	NA
PCB 52	pg m ⁻³	0.4	58	1.0	< 0.4	< 0.4	< 1.0	1.9	11.5	NA	NA
PCB 77	pg m ⁻³	0.4	11	1.0	< 0.4	< 0.4	< 0.4	< 0.4	1.5	NA	NA
PCB 101	pg m ⁻³	0.4	89	1.0	< 0.4	< 1.0	1.5	5.2	29.9	NA	NA
PCB 105	pg m ⁻³	0.4	61	1.0	< 0.4	< 0.4	< 1.0	2.6	18.5	NA	NA
PCB 118	pg m ⁻³	0.4	86	1.0	< 0.4	< 1.0	1.5	5.7	47.9	NA	NA
PCB 126	pg m ⁻³	1.0	0	2.1	NA	NA	NA	NA	NA	NA	NA
PCB 138	pg m ⁻³	0.4	89	1.0	< 0.4	< 1.0	2.3	8.4	53.6	NA	NA
PCB 153	pg m ⁻³	0.4	89	1.0	< 0.4	< 1.0	2.4	7.5	40.2	NA	NA
PCB 180	pg m ⁻³	0.4	77	1.0	< 0.4	< 1.0	1.3	4.4	25.1	NA	NA
Phthalates											
BBP	ng m ⁻³	0.17	98	0.42	< 0.42	0.6	1.6	5.3	31.9	2.0	5.0
DEHP	ng m ⁻³	1.7	100	4.2	9.1	22.3	46.2	110	389	52.6	3.3
DBP	ng m ⁻³	0.42	72	0.84	< 0.42	< 0.42	< 0.84	3.2	25.2	NA	NA

SVOC	Unit	LOD	% > LOD	LOQ	% > LOQ	P5	P25	P50	P75	P95	GM	GSD
DEP	ng m ⁻³	0.42	62	0.84	45	< 0.42	< 0.42	< 0.84	2.6	9.8	NA	NA
DiBP	ng m ⁻³	0.17	96	0.42	70	< 0.42	< 0.42	0.861	2.8	22.9	NA	NA
DiNP	ng m ⁻³	0.84	99	1.7	97	2.0	4.0	7.9	17.6	50.1	8.7	3.0
DMEP	ng m ⁻³	0.04	8	0.08	5	< 0.04	< 0.04	< 0.04	< 0.04	< 0.08	NA	NA
DMP	ng m ⁻³	0.02	39	0.04	13	< 0.02	< 0.02	< 0.02	< 0.04	0.070	NA	NA
DOP	ng m ⁻³	0.42	4	0.84	2	< 0.42	< 0.42	< 0.42	< 0.42	< 0.42	NA	NA
Synthetic musks												
Galaxolide	pg m ⁻³	41.8	47	83.5	31	< 41.8	< 41.8	< 41.8	122	759	NA	NA
Tonalide	pg m ⁻³	10.4	58	20.9	41	< 10.4	< 10.4	< 20.9	40.8	377	NA	NA
Alkylphenols												
4-n-nonylphenol	pg m ⁻³	10.4	2	20.9	1	< 10.4	< 10.4	< 10.4	< 10.4	< 10.4	NA	NA
4-tert-butylphenol	pg m ⁻³	10.4	56	20.9	28	< 10.4	< 10.4	< 20.9	23.7	52.8	NA	NA
4-tert-octylphenol	pg m ⁻³	10.4	46	20.9	26	< 10.4	< 10.4	< 10.4	22.5	108	NA	NA
Other SVOCs												
Bisphenol-A	ng m ⁻³	1.0	38	2.6	13	< 1.0	< 1.0	< 1.0	< 2.6	6.0	NA	NA
Triclosan	pg m ⁻³	10.4	98	20.9	95	< 20.9	42.9	114	359	1,855	138	4.4
Tri-n-butylphosphate	pg m ⁻³	41.8	12	83.5	7	< 41.8	< 41.8	< 41.8	< 41.8	107	NA	NA

LOD: limit of detection; LOQ: limit of quantification; Px: centile x; GM: geometric mean; GSD: standard deviation of the geometric mean (dimensionless); NA: non-applicable

LOD and LOQ represent the limits of detection and quantification for the lowest sampled volume (9.6 m³).

Table 2a. Particle-bound SVOC median concentrations in residential indoor air measured worldwide

Country	France	UK	Italy	US	Hong-K.	China	USA	Norway	USA	China
Size fraction	PM ₁₀	TSP	PM _{2.5}	PM _{2.5}	PM _{2.5}	PM _{2.5} /TSP	TSP	PM ₁₀	TSP	TSP
Period of measurements	Dec. 2010 - April 2011	May 2005 - May 2007	Winter 11-12+ Spring 12+ Summer 12	June 1999- May 2000	April - Dec. 2010	April - Dec. 2010	March 2006 - Aug. 2007	NA	Jan. - May 2014	Sept. 2012 - Jan. 2013
Estimate	Median	GM	AM	Median	Median	Median	Median	AM	Median	Median
n Dwellings	n=30	n=162	n=10	n=115	n=6	n=7	n=12	n=2	n=20	n=28*
This study	Blanchard et al., 2014b	Delgado-S. et al., 2011	Romagnoli et al., 2014#	Zhu and Jia, 2012	Wang et al., 13,14a	Wang et al., 13,14a	Batterman et al., 2009	Rakkestad et al., 2007	Tran and K., 2015	Wang et al., 2014b
Anthracene	6	50		5.7	10	80/190				
Benzo(a)anthracene		60	208/50/36#	22	60	270/730				
Benzo(a)pyrene	65	90	980/124/64#	52	100	600/970				
Benzo(b)fluoranthene		180		3.6						
Benzo(g,h,i)perylene		150	1,780/224/142#	170	170	1,200/1,560				
Benzo(k)fluoranthene		110								
Chrysene		140			100	520/1,050				
Dibenzo(a,h)anthracene		30	170/28/18#	7.4	20	190/340				
Fluoranthene	22	360		57	340	2,200/5,260				
Fluorene		130			90	870/2,950				
Indeno(1,2,3-c,d)pyrene		110	1,390/180/94#	150	180	1,150/1,500				
Phenanthrene	25	340		58	470	2,880/10,200				
Pyrene	20	290		69	290	980/3,400				
4,4'-DDT		5.1								
Permethrin		5.9								
BDE 47	7				1.0	34.9/102				
BDE 99	8				0.03	13.8/34.3				
PCB 101		1.5								
PCB 118		1.5								
PCB 138		2.3								
PCB 153		2.4								
PCB 180		1.3								
BBP	2.4							5.5	1.2	0.138
DEHP	41.5	46.2						13.5	22.9	560*
DiBP	30.2	0.861							33.9	720*
DiNP	15.7	7.9								
Triclosan		114								

TSP: total suspended particles; NA: not available; AM: arithmetic mean; GM: geometric mean; *: 14 dwellings and 14 offices; #: the reported means correspond to the average of the means measured in Winter (4 homes), Spring (5 homes) and Summer (9 homes) respectively.

Bold highlights studies with the same size fraction sampled (PM₁₀). Case studies conducted in one or two locations were excluded for this comparison. Studies targeting only new or newly decorated dwellings were not used for the comparison. For PAHs, studies performed in the context of solid biomass combustion were not considered.

Table 2b. Ranges of particle-bound SVOC concentrations in residential indoor air measured worldwide

Country	France	USA	China	Hong-Kong	Norway	USA	China	China	USA
Size fraction	PM₁₀	TSP	PM _{2.5} /TSP	PM _{2.5}	PM₁₀	TSP	TSP	PM₁₀	PM _{2.5}
Period of measurements	Dec. 2010 - April 2011	March 2006 - Aug. 2007	April - Dec. 2010	April - Dec. 2010	NA	Jan. - May 2014	Sept. 2012 - Jan. 2013	Dec. 2010 + June 2011	June 1999- May 2000
n Dwellings	n=30	n=12	n=7	n=6	n=2	n=20	n=28*	n=13	n=104
Unit	This study (min-P95)	Batterman et al., 2009	Wang et al., 2014a	Wang et al., 2014a	Rakkestad et al., 2007	Tran and Kannan, 2015	Wang et al., 2014b	Zhang et al., 2014	Zhu and Jia, 2012
BDE 28	pg m ⁻³	nd - 1.9	nd - 45 / nd - 52	nd - 1.8		0.9 - 451	nd - 4,920*	7.3 - 1,466	nd - 1,700
BDE 85	pg m ⁻³	nd - <4.2	nd - 7.7 / nd - 10.2	nd - 2.1		3.4 - 361		0.08 - 7.4	nd - 670
BDE 100	pg m ⁻³	nd - 13.6	0.5 - 57.9 / 1.8 - 141	nd - 22.1		nq - 2.4	nd - 1,750*	0.3 - 47.6	
BDE 153	pg m ⁻³	nd - <4.2	nd - 20.6 / nd - 58.6	nd - 1.0					
BDE 154	pg m ⁻³	nd - <4.2	nd - 14.5 / nd - 192	nd - 0.6					
DBP	ng m ⁻³	nd - 25.2			74 - 85				
DEP	ng m ⁻³	nd - 9.8							
DMP	ng m ⁻³	nd - 0.07							
t-chlordane	pg m ⁻³	nd - <2.1							
c-chlordane	pg m ⁻³	<2.1							

TSP: total suspended particles; NA: not available; *: 14 dwellings and 14 offices; P95: 95th percentile; nd: not detected; nq: not quantified; t- and c-chlordane: trans- and cis-chlordane

Bold highlights studies with the same size fraction sampled (PM₁₀). Case studies conducted in one or two locations were excluded for this comparison. Studies targeting only new or newly decorated dwellings were not used for the comparison.

Table 3. SVOC median concentrations (pg m^{-3}) according to the heating season and smoking in the dwelling

SVOC	Heating season		Non heating season		Paired comparison
	Smoker	Non-smoker	Smoker	Non-smoker	
	H+S	H+NS	NH+S	NH+NS	
%	43	25	11	21	-
SVOCs showing influence of both the season and the smoking					
Benzo(a)anthracene	170	53	42	13	} H+S > H+NS > NH+NS*** H+S > NH+S > NH+NS***
Chrysene	259	85	80	30	
Benzo(a)pyrene	380	170	93	17	} H+S > H+NS > NH+NS*** H+S > NH+S > NH+NS**
Benzo(b)fluoranthene	724	374	243	94	
Pyrene	123	59	52	23	
Benzo(k)fluoranthene	145	88	46	18	H+S > H+NS** > NH+S* NH+S > NH+NS*
Dibenzo(a,h)anthracene	58	29	13	< LOQ	H+S > H+NS** > NH+S** NH+S > NH+NS**
Fluoranthene	171	75	74	34	H+S > H/NH+NS*** H+S > NH+S** > NH+NS***
Indeno(1,2,3-cd)pyrene	324	234	120	48	H+S/NS > NH+NS/S*** NH+S > NH+NS*
Lindane	7.6	< LOD	< LOQ	< LOD	H+S > NH+S* > H/NH+NS**
4,4'-DDE	2.0	< LOD	< LOQ	< LOD	H+S > NH+S* > NH+NS* H+S > H+NS***
4,4'-DDT	16	3.7	9.2	2.4	H+S > H/NH+NS***
PCB 138	9.9	1.6	3.6	< LOQ	H/NH+S > NH+NS** H+S > H+NS***
PCB 118	7.0	1.1	2.1	< LOQ	} H+S > NH+S* > NH+NS** H+S > H+NS***
PCB 153	9.3	1.5	3.1	< LOQ	
PCB 180	4.5	< LOQ	1.6	< LOQ	
BBP	6,222	1,290	1,708	736	H+S > H+NS*** H+S > NH+S* > NH+NS**
DEHP #	143	43	83	22	H+S > H+NS > NH+NS*** H+S > NH+S** > NH+NS***
Galaxolide	158	< LOD	50	< LOD	H+S > H/NH+NS*** H+S > NH+S** > NH+NS*
Tonalide	65	< LOQ	24	< LOD	H+S > NH+S* > H/NH+NS*

SVOCs influenced predominantly by smoking

Fluorene	56	17	38	14	H/NH+S > H/NH+NS**
Phenanthrene	95	51	63	43	H/NH+S > NH+NS** H+S > H+NS***
Oxadiazon	< LOQ	< LOD	1.0	< LOD	H/NH+S > H/NH+NS***
Permethrin	90	78	124	53	NH+S > NH+NS*
BDE 47	45	12	23	4.8	H/NH+S > NH+NS*** H+S > H+NS*** > NH+NS*
BDE 99	19	8.8	15	5.5	H/NH+S > NH+NS** H+S > H+NS***
BDE 100	5.0	< LOD	< LOQ	< LOD	H/NH+S > H/NH+NS*
PCB 52	2.6	< LOQ	1.0	< LOD	H/NH+S > H/NH+NS**
PCB 101	6.8	1.2	2.6	< LOQ	H/NH+S > H/NH+NS*
PCB 105	2.7	< LOQ	1.0	< LOD	H/NH+S > H/NH+NS*
DBP	5,298	< LOQ	1,161	< LOD	H+S > NH+S*** > H+NS* H+NS > NH+NS*
DEP	1,795	< LOQ	< LOQ	< LOD	H+S > H/NH+NS*** NH+S > NH+NS*
DiBP	3,370	613	1,329	454	H/NH+S > H/NH+NS**
DiNP	16,570	7,940	8,803	5,606	H+S > H/NH+NS*** NH+S > NH+NS*
4-tert-butylphenol	< LOQ	< LOD	< LOQ	< LOD	H/NH+S > H/NH+NS*
Triclosan	305	81	313	67	H/NH+S > H/NH+NS***

SVOCs influenced predominantly by the season

Benzo(g,h,i)perylene	487	333	159	69	H+S/NS > NH+S/NS***
Bisphenol-A	1,091	< LOD	< LOD	< LOD	H+S > NH+NS*** H+NS > NH+NS**

SVOCs with no trend regarding influence of season and smoking

Anthracene	16	14	15	9.1	NH+NS < other **
4-tert-octylphenol	28	< LOD	< LOQ	< LOD	H+S > other ***

#: percentage of dwellings in each group of season and smoking status; LOD: limit of detection;

LOQ: limit of quantification; *: $P < 0.05$, **: $P < 0.01$ and *** $P < 0.001$ adjusted with the Bonferroni correction to account for the multiple comparisons; # concentrations expressed in ng m^{-3} .

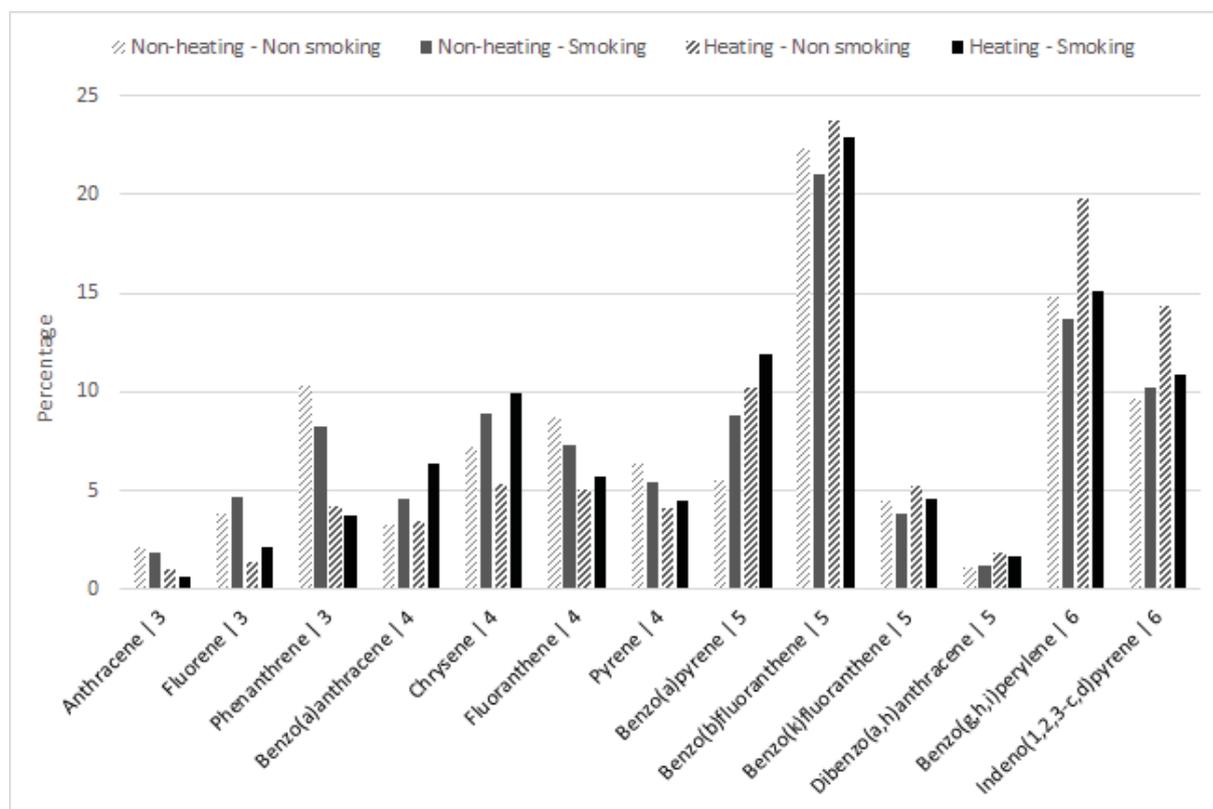
The SVOCs with concentrations < LOQ in each of the four categories are not reported.

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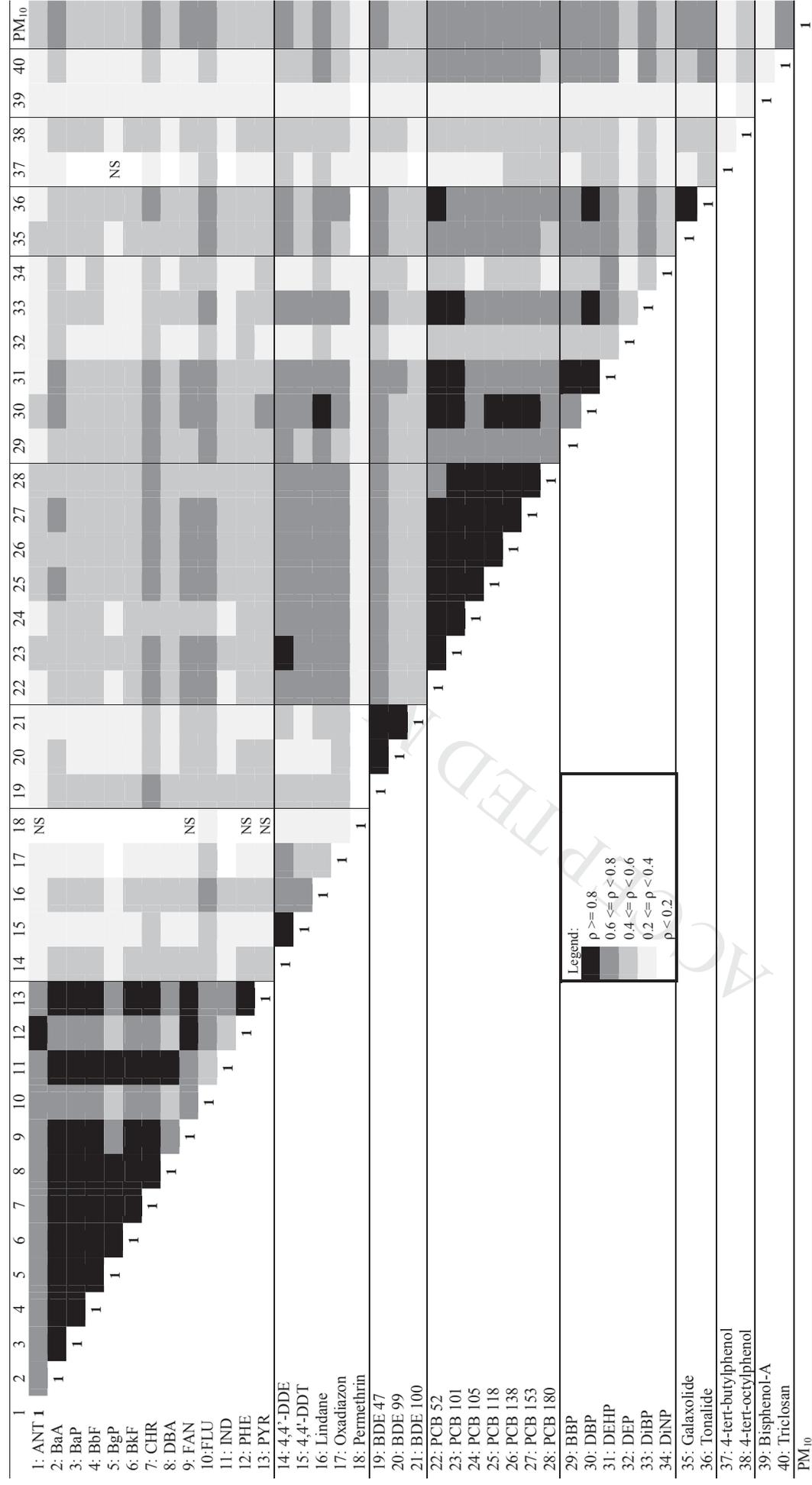
Figure 1. Average relative contribution (%) of each PAH to the total concentrations of PAHs in PM₁₀ according to season and smoking in the dwelling

Figure 2. Spearman rank correlation coefficients between concentrations of 40 particle-bound SVOCs and PM₁₀ concentration

Figure 1. Average relative contribution (%) of each PAH to the total concentrations of PAHs in PM₁₀ according to season and smoking in the dwelling



The figure next to each PAH name indicates the number of rings.

Figure 2. Spearman rank correlation coefficients between concentrations of 40 particle-bound SVOCs and PM₁₀ concentration

1: ANT=Anthracene; 2: BaA=Benzo(a)anthracene; 3: BaP=Benzo(a)pyrene; 4: BbF=Benzo(b)fluoranthene; 5: BgP=Benzo(ghi)perylene; 6: BkF=Benzo(k)fluoranthene; 7: CHR=Chrysene; 8: DBA=Dibenzo(a,h)anthracene; 9: FAN=Fluoranthene; 10: FLU=Fluorene; 11: IND=Indeno(1,2,3-c,d)pyrene; 12: PHE=Phenanthrene; 13: PYR=Pyrene; NS: non-significant (P > 0.05)

Highlights

- ▶ 66 SVOCs were measured in airborne particles (PM₁₀) in dwellings.
- ▶ PAHs, phthalates and triclosan have the highest concentrations.
- ▶ Concentrations are higher in smokers' dwellings and during the heating season.
- ▶ Correlations between SVOCs provide trends regarding common determinants.
- ▶ High indoor PM₁₀ concentrations promote SVOCs in the particulate phase.