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Human exposure to PCBs, PBDEs and bisphenols revealed by hair analysis: A comparison between two adult female populations in China and France

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► To cite this version:

F.-J. Peng, E.M. Hardy, R. Beranger, S. Mezzache, N. Bourokba, et al.. Human exposure to PCBs, PBDEs and bisphenols revealed by hair analysis: A comparison between two adult female populations in China and France. *Environmental Pollution*, 2020, 267, pp.115425. 10.1016/j.envpol.2020.115425 . hal-02960983

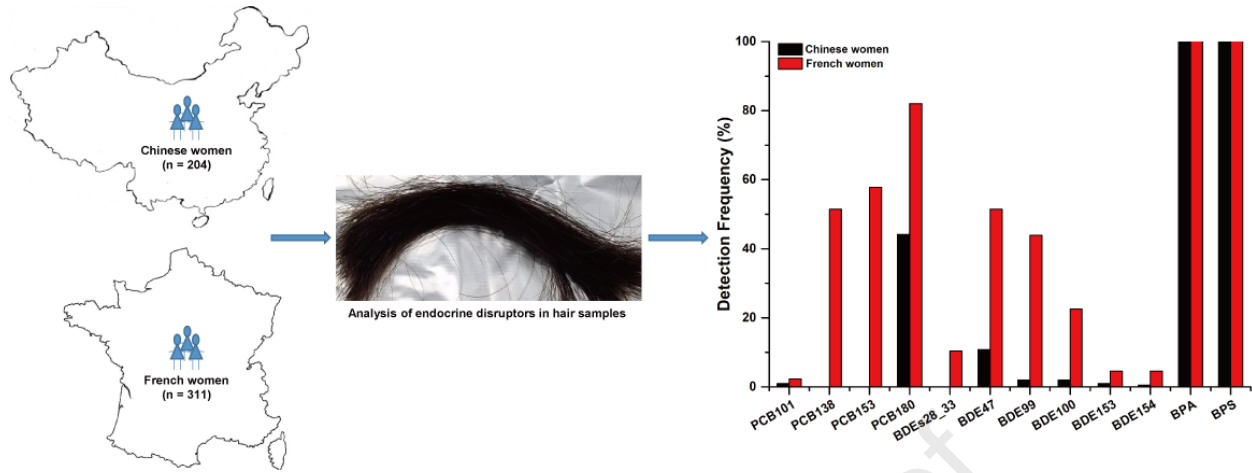
HAL Id: hal-02960983

<https://hal.science/hal-02960983v1>

Submitted on 8 Oct 2020

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1 **Human exposure to PCBs, PBDEs and bisphenols revealed by hair**
2 **analysis: a comparison between two adult female populations in**
3 **China and France**

4

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19

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22 **Abstract:** Humans are exposed to various anthropogenic chemicals in daily life,
23 including endocrine-disrupting chemicals (EDCs). However, there are limited data on
24 chronic, low-level exposure to such contaminants among the general population. Here
25 hair analysis was used to investigate the occurrence of four polychlorinated biphenyls
26 (PCBs), seven polybrominated diphenyl ethers (PBDEs) and two bisphenols (BPs) in
27 204 Chinese women living in the urban areas of Baoding and Dalian and 311 pregnant
28 French women. All the PCBs and PBDEs tested here were more frequently detected
29 in the hair samples of the French women than in those of the Chinese women. In both
30 cohorts, PCB 180 and BDE 47 were the dominant PCB and PBDE congener,
31 respectively. PCB 180 was found in 82% of the French women and 44% of the
32 Chinese women, while the corresponding values of BDE 47 were 54% and 11%,
33 respectively. A discriminant analysis further demonstrated the difference in PCBs and
34 PBDEs exposure profile between the two cohorts. These results demonstrate that hair
35 analysis is sufficiently sensitive to detect exposure to these pollutants and highlight
36 differences in exposure between populations even at environmental levels. Although
37 BPA and BPS were found in 100% of the hair samples in both cohorts, the French
38 women had significantly higher levels of BPA and BPS than the Chinese women. The
39 median concentrations of BPA were one order of magnitude higher than BPS in both
40 the Chinese (34.9 versus 2.84 pg/mg) and the French women (118 versus 8.01 pg/mg)
41 respectively. Our results suggest that both French and Chinese populations were
42 extensively exposed to BPA and BPS.

43 **Capsule:** In the present study, French women were more frequently exposed to all
44 PCBs and PBDEs analyzed and more extensively exposed to BPA and BPS than
45 Chinese women.

46

47 **Key words:** Human biomonitoring; PCBs; PBDEs; BPs; Hair analysis; Women

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Journal Pre-proof

49 **1. Introduction**

50 Synthetic chemicals have become a part of our daily life. Some of these chemicals are
51 endocrine disrupting chemicals (EDCs) that can interfere with some important
52 developmental processes in humans and wildlife (WHO/UNEP, 2013).
53 Polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and
54 bisphenol A (BPA) are ubiquitous endocrine disruptors related to adverse effects on
55 reproduction and development in humans (Rudel and Perovich, 2009; Pinson et al.,
56 2016).

57
58 PCBs are a group of industrial compounds formerly used in electrical capacitors,
59 transformers, paint and rubber sealant (Safe, 1984). With regard to their long-range
60 transport capability, persistence, bioaccumulation and toxicity, the production and use
61 of PCBs were globally banned since decades and these chemicals were listed as
62 persistent organic pollutants (POPs) under the Stockholm Convention in 2001 (Xing
63 et al., 2005). PBDEs are another class of POPs, used as flame-retardant additives in a
64 variety of consumer products, e.g. plastics, polyurethane foams, textiles and electronic
65 products (WHO, 1994). PBDEs show similar properties to PCBs, e.g. lipophilic,
66 persistent, and bioaccumulative in biological tissues (Hooper and McDonald, 2000).
67 As a result, the two PBDEs pentabromodiphenyl ether and octabromodiphenyl ether
68 were phased out worldwide between 2004 and 2010 (Gentes et al., 2012). However,
69 they are still frequently detected in human samples, especially in cases of
70 occupational exposure (Hurley et al., 2017; Zota et al., 2018; Qiao et al., 2019). BPA
71 is a high production volume (HPV) chemical used in the synthesis of polycarbonate
72 plastics and epoxy resins (USEPA, 2010). Despite non-persistence, BPA exposure is
73 widespread among general population because of its ubiquity (Meeker and Ferguson,

74 2011). Considering the endocrine-disruptive property and potential effects on human
75 health of BPA, North America and the European Union have banned its use in certain
76 products, e.g. baby bottles and toys (Chen et al., 2016). As a result, in many consumer
77 products BPA has been replaced by chemicals with similar properties, such as
78 bisphenol S (BPS). However, in vitro and in vivo studies have demonstrated that BPS
79 is as hormonally active as BPA and has also endocrine-disrupting effects (Rochester
80 and Bolden, 2015). Due to the increasing production and usage, BPS has been found
81 in 89.4% of urine samples (n = 2676) from U.S. adults and children participating in
82 the National Health and Nutrition Examination Survey 2013-2014 (Lehmler et al.,
83 2018).

84

85 Even though biological fluids have been used extensively for the assessment of
86 human exposure to PCBs, PBDEs (blood) and bisphenols (urine), some studies also
87 demonstrated the possibility to detect these pollutants in hair (Covaci et al., 2008;
88 Barbounis et al., 2012; Behrooz et al., 2012; Wen et al., 2008; Tzatzarakis et a., 2015;
89 Nehring et al., 2017). As compared to other matrices used in biomonitoring (e.g.,
90 blood, breast milk, urine or tissues), hair analysis owns several advantages. For
91 instance, hair collection is non-invasive and transport and storage of the samples do
92 not require refrigeration. More importantly, pollutants in hair can be retained for long
93 time intervals, since active processes (metabolism or excretion) within the hair are
94 absent (Tsatsakis et al., 2008). These advantages make hair a biological matrix highly
95 used in legal medicine (Barroso et al., 2011). The concentration of biomarkers in hair
96 is considered to be representative of the chronic exposure of the individual, covering
97 from weeks to months depending on the length of the sample analyzed. The research
98 work conducted over the past decade helped to increasingly consider hair as a suitable

99 matrix to monitor human exposure to various pollutants (e.g., Appenzeller et al., 2012;
100 Zheng et al., 2014). In animal models, strong correlation between level of exposure
101 and concentration of chemicals in hair has been demonstrated for many pollutants,
102 particularly for POPs (Appenzeller et al., 2017; Grova et al., 2018). Other studies
103 conducted on human demonstrated that the concentration of PCB and PBDEs in hair
104 was significantly correlated with concentration in blood (Zheng et al., 2014; Zheng et
105 al., 2016) and in tissues (Poon et al., 2014), and could thus be considered a reliable
106 surrogate of the internal dose of pollutants. The previous studies that demonstrated the
107 possibility to detect endocrine disruptors such as PCBs, PBDEs and bisphenols in hair
108 usually focused on a single chemical family. These were mostly conducted on a
109 limited number of subjects selected from the same group (e.g. same living area,
110 occupation), and provided relevant information on local exposures. In order to further
111 explore the relevance of hair analysis in epidemiology, larger studies involving
112 subjects from different geographical areas are still needed to demonstrate the
113 possibility to highlight regional “clusters” of exposure, based on the concentration of
114 pollutants detected in hair.

115

116 To assess cumulative exposure to EDCs of female adult populations and to explore
117 the possible regional variability in exposure, the concentration levels of four PCBs,
118 seven PBDEs and two BPs were here determined in hair samples collected from 311
119 pregnant French women (18-43 years old) belonging to the ELFE cohort and 204
120 Chinese women (25-45 years old) living in the urban areas of Baoding and Dalian.
121 The specific objectives of the present study were i) to examine exposure profiles of
122 selected EDCs among women in different geographical regions, ii) to compare the
123 exposure patterns between Chinese and French women studied, iii) to investigate

124 correlations between the different biomarkers of exposure, and iv) to explore
125 associations between population characteristics and hair concentrations of dominant
126 pollutants.

127

128 **2. Materials and methods**

129 2.1. Chemicals and reagents

130 Thirteen common EDCs were selected as target compounds in the present study:
131 PCBs 101, 138, 153 and 180, BDEs 28, 33, 47, 99, 100, 153 and 154, BPA and BPS,
132 based on the analytical feasibilities and on their usage, exposure levels in the
133 environment, and results from previous bio-monitoring studies (e.g., Kang et al., 2011;
134 Carnevale et al., 2014; Tzatzarakis et al., 2015). Standards of the four PCBs (i.e.,
135 PCBs 101, 138, 153 and 180) and seven BDEs (i.e., BDEs 28, 33, 47, 99, 100, 153
136 and 154) were purchased from Dr. Ehrenstorfer (Augsburg, Germany) and
137 AccuStandard (New Haven, CT, USA), respectively. BPA and BPS were obtained
138 from Sigma-Aldrich (Diegem, Belgium). Abbreviations and other details (e.g., CAS
139 no. and Log *K_{ow}*) are provided in the supporting information (Table S1). Internal
140 standards gamma-hexachlorocyclohexane-d₆ (γ -HCH-d₆), β -endosulfan-d₄ and
141 trans-permethrin-d₆ were purchased from Dr. Ehrenstorfer (Germany); BPA-d₁₆ from
142 Sigma-Aldrich (Belgium) and thiabendazole-¹³C₆ from TRC (Canada). The purity of
143 each standard was higher than 95 %. Methanol (analytical grade), acetonitrile (HPLC
144 grade), ethyl acetate (pesticide grade), formic acid (HPLC grade) and ammonium
145 acetate (HPLC grade) were acquired from Biosolve (Germany). Sodium phosphate
146 monobasic monohydrate (NaH₂PO₄, H₂O), sodium dodecyl sulfate (SDS), sodium
147 metabisulfite (Na₂S₂O₅) and 2, 3, 4, 5, 6-pentafluorobenzyl bromide (PFBBBr) were
148 purchased from Sigma-Aldrich (Diegem, Belgium). Potassium carbonate (K₂CO₃),

149 sodium hydroxide (NaOH) and sodium phosphate dibasic dihydrate ($\text{Na}_2\text{HPO}_4 \cdot 2\text{H}_2\text{O}$)
150 were bought from Merk (Germany).

151

152 2.2. Sample collection

153 Hair samples were collected from 204 Chinese women in 2016 and 311 pregnant
154 French women in 2011. The selection of these two populations was based on the
155 difference in historical consumption of PCBs and penta-BDEs between China and
156 France and on the difference in estimated BPA daily intake between Chinese adults
157 and French adults (e.g., Breivik et al., 2002; Mai et al., 2005; Huang et al., 2017;
158 Abbasi et al., 2019). Details on the study design, populations, recruitment, and
159 sampling procedures have been previously reported (Béranger et al., 2018; Palazzi et
160 al., 2018; Charles et al., 2019; Peng et al., 2020). In summary, the Chinese women
161 were recruited in the framework of a clinical study investigating the effect of chronic
162 pollution exposure on skin, scalp and hair (Naudin et al., 2019). Inclusion criteria for
163 selection of Chinese women were: presenting natural hair from root to tip and a length
164 of hair ≥ 30 cm, non-smoking, non-drinking alcohol, healthy, not being pregnant, age
165 between 25 and 45 years, and living in the cities of Baoding and Dalian (Northern
166 China) for at least 15 years with an indoor occupation (Palazzi et al., 2018).
167 Socio-demographic data (age and education) and frequency of consuming red meat or
168 fish/seafood were collected through self-reported questionnaires (Table S2). The
169 pregnant French women were selected from the ELFE cohort designed to evaluate the
170 influence of environment on child development, health and socialization (Charles et
171 al., 2019). Selected pregnant French women in the current study met the following
172 inclusion criteria: presenting a sufficient amount of hair for analysis, giving birth to
173 singleton infants without congenital malformation, age over 18 years, and resident in

174 either northeastern (Champagne-Ardenne, Bourgogne, and Lorraine) or southwestern
175 (Aquitaine, Midi-Pyrénées, and Poitou-Charentes) French regions (Béranger et al.,
176 2018). Most of these French women were office, sales, and service workers (52.7%)
177 or involved in intermediate occupations (25.1%). Self-reported questionnaires were
178 administered to collect information on sociodemographic (e.g., age and education;
179 Table S2) and medical characteristics during the maternity stay (before discharge).
180 For both Chinese women and French women, height and weight were measured to
181 calculate body mass index (BMI). Hair samples were collected from the scalp in the
182 posterior vertex region of the head. Each hair sample (~ 100 fibers) was wrapped in
183 aluminum foil, sealed in a kraft envelope and stored at room temperature. All the
184 participants signed an informed consent. The protocol of the Chinese women study
185 was approved by the Ethics Committee of the Chinese Academy of Inspection and
186 Quarantine Cosmetics Tech Center (protocol n°: 2015-033-DY-024), and the protocol
187 of French women study was approved by the French Consulting Committee for the
188 Treatment of Information in Medical Research (n°10.623), the French National
189 Commission for the Confidentiality of Computerized Data (n°910504), and the
190 Committee for the Protection of Persons (n° CPP-IDF IX-11024).

191

192 2.3. Sample preparation and analysis

193 For the Chinese women, only the 0-12 cm (proximal part = starting from the skin) of
194 hair strands were used for analyses, which corresponds to the 12 months before hair
195 sampling (Kintz et al., 2015). For the French women, only the 0-9 cm (proximal part)
196 of hair strand was used for analysis, which covers the period of pregnancy by
197 assuming an average hair growth rate of 1 cm per month (Béranger et al., 2018). Hair
198 samples were successively washed with a sodium dodecyl sulfate solution and

199 methanol before extraction, to remove any possible externally deposited chemicals on
200 the hair surface, according to a protocol previously developed (Duca et al., 2014).
201 Hair samples were then powdered using a Retsch-MM 200 vibrant-ball mill (5 min at
202 25 Hz).

203

204 The protocol for the analysis of target EDCs in hair was the same as that reported for
205 pesticides in hair (Hardy et al., 2015; Béranger et al., 2018). Briefly, EDCs were
206 extracted from approximately 50 mg of hair powder using 1 mL of acetonitrile/water
207 mixture (80/20, v/v) incubated overnight at 40 °C. An aliquot of 0.3 mL was used to
208 analyze PCBs 101, 138, 153, BDEs 28 and 33 by solid phase microextraction (SPME)
209 analysis coupled to gas chromatography-tandem mass spectrometry (GC-MS/MS).
210 Another aliquot of 0.3 mL was used to analyze PCB 180, BDEs 47, 99, 100, 153, 154
211 and BPA by derivatization with 30 mg of K₂CO₃, 1 mL of acetonitrile and 100 µL of
212 PFBBBr-acetonitrile mixture (75/25, v/v) and analysis by GC-MS/MS; a final portion
213 of 0.2 mL was used to analyze BPS by ultra-performance liquid
214 chromatography-tandem mass spectrometry (UPLCMS/MS).

215

216 Procedure blanks and quality control (QC) samples were determined per set of 30
217 samples. QC samples were a pool of hair spiked with standards at eight different
218 concentrations (0.5, 1, 2, 5, 10, 20, 50 and 100 pg/mg). They were analyzed
219 simultaneously and in the same way as field samples. The recoveries of each
220 compound at concentrations of 1, 10 and 100 pg/mg ranged 64%-141%. Limit of
221 detection (LOD) was defined as the lowest concentration of a compound detected in
222 the samples analyzed in the current study, as previously detailed (Appenzeller et al.,
223 2017). LODs ranged from 0.02 pg/mg for PCB 180 to 5.47 pg/mg for BPA. Because

224 of the co-elution, BDE 28 and BDE 33 could not be differentiated and were marked as
225 BDE 28_33. Method validation parameters are presented in Table S3.

226

227 2.4. Statistical analysis

228 Statistical analyses were performed in R program version 3.5.2. Statistical
229 significance was defined at a p value < 0.05 . For descriptive statistics, if the percentile
230 estimate was below LOD, the value was reported as “ $< LOD$ ”. In further statistical
231 analyses, concentrations below the LOD were assigned a value of $LOD/2$.
232 Mann-Whitney U tests were performed to compare concentrations of each compound
233 between the two countries or between the two sub-regions within a country.
234 Kruskal-Wallis tests were conducted to compare concentrations of dominant
235 compounds (detection frequency (DF) $\geq 75\%$) among the four sub-regions. Wilcoxon
236 signed rank tests were performed to examine the difference in concentrations between
237 chemicals. Partial least square-discriminant analyses (PLS-DA) were conducted on
238 POPs profile data using the mixOmics R package (Rohart et al., 2017), to estimate
239 discrimination between Chinese and French hair samples. Spearman rank correlation
240 coefficients (r) between compounds were calculated for each data set (China and
241 France separately). Only chemicals detected in more than ten samples were included
242 in the correlation analyses, and the results were presented in a heatmap. Multivariate
243 linear regression analyses were conducted on each data set (China and France
244 separately) to evaluate associations of hair concentrations of dominant chemicals (DF
245 $\geq 75\%$) with population characteristics (Table S2). In these models, response
246 variables were concentrations of dominant pesticides while explanatory variables
247 were population characteristics. For the Chinese women, the specific population
248 characteristics were age, BMI, education, living cities (Baoding and Dalian), and

249 frequency of consuming red meat or fish/seafood, which were reported in our
250 previous studies (Palazzi et al., 2018; Peng et al., 2020). For the French women, the
251 specific population characteristics were age, BMI, education, living areas (northeast
252 and southwest), and smoking habit (smoking and non-smoking), which were partly
253 reported in previous studies (Béranger et al., 2018; 2020). Age and BMI were
254 considered as continuous variables, and the remaining characteristics were considered
255 as categorical variables (Table S2). Concentration data were log₁₀ transformed in
256 these analyses as their distributions were highly skewed. Backward stepwise
257 elimination was used to identify optimal models (Zuur et al., 2009). Residuals were
258 visually inspected for heterogeneity and independence by plotting residuals against
259 fitted values and each explanatory variable, respectively. Additionally, a histogram of
260 the residuals was used to verify normality.

261

262 **3. Results**

263 3.1. Hair concentrations of EDCs

264 3.1.1. Chinese women

265 Table 1 presents the descriptive results of all the compounds measured in this study.
266 Of the four PCB congeners, PCB 180 was the most frequently detected (DF = 44%;
267 Fig. 1); PCB 138 and 153 were not detected in any samples; PCB101 was found in
268 two hair samples from women in Dalian (Table S4). All the seven PBDEs were
269 seldom detected (Fig. 1). BDE 47 was the predominant congener and detected in 11%
270 of the hair samples. The median concentrations of all PCB and BDE congeners were
271 below the LODs. BPA and BPS were detected at measurable concentrations in all the
272 hair samples. The 25th, 50th and 75th percentile concentrations of BPA were 23.4, 34.9
273 and 57.2 pg/mg, respectively, and the corresponding levels of BPS were 1.43, 2.84

274 and 6.34 pg/mg, respectively. The BPA concentration was significantly higher than
275 BPS (Wilcoxon signed rank test, $p < 0.05$). Comparing women in Baoding with those
276 in Dalian, concentrations of PCB 180 and BPA were significantly higher in hair
277 samples from the women in Baoding, but the BPS concentration was significantly
278 higher in hair samples from the women in Dalian (Mann-Whitney U tests; Table S4).
279 Given the low DFs of most PCBs and BDEs, only PCB 180, BDE 47, BPA and BPS
280 were included in the correlation analysis. There was a statistically significant positive
281 correlation between the concentration levels of BPA and BPS ($r = 0.31$; $p < 0.001$).

282

283 3.1.2. French women

284 Similar to the Chinese women, PCB 180 was the most frequently detected PCB
285 congener in the French women, with DF of 82% and a median concentration of 0.25
286 pg/mg. While PCB 101 was detected in only 2% of the hair samples, PCB 138 and
287 PCB 153 were detected in 45 and 49% of the hair samples, respectively. Again, BDE
288 47 (DF = 57%) was more frequently detected than the remaining six BDE congeners
289 (Fig. 1), with concentrations ranging from <LOD to 152 pg/mg and a median
290 concentration of 1.37 pg/mg. BDE 99 and 100 were detected in 49 and 20% of the
291 hair samples, with concentrations in the ranges of <LOD-396 and <LOD-73.7 pg/mg,
292 respectively. BDEs 28_33, 153 and 154 were detected at similar frequencies (DF = 7,
293 5 and 4%, respectively). BPA and BPS were detected in 100% of hair samples, but the
294 concentration of BPA was significantly higher than BPS (Wilcoxon signed rank test,
295 $p < 0.05$). The 25th, 50th and 75th percentile concentrations of BPA were 63.4, 118 and
296 213 pg/mg, respectively, and the corresponding levels of BPS were 2.11, 8.32 and
297 27.9 pg/mg, respectively. Comparing pregnant women in northeastern France with
298 those in southwestern France, the concentrations of BDEs 28_33, 100, 153 and 154

299 were significantly higher in the hair samples collected from women in southwestern
300 France ($p \leq 0.015$; Table S4). There were no statistically significant differences in
301 concentrations of the remaining compounds between the two regions.

302

303 Because PCB 101 was found in only 2% of the hair samples, it was excluded from the
304 correlation analysis. Within the remaining three PCB congeners, there was a strong
305 positive correlation between PCBs 138 and 153 ($r = 0.80$, $p < 0.001$). These two
306 congeners were significantly positively correlated with PCB 180 as well (Fig. 2), with
307 correlation coefficient values of 0.33 and 0.35, respectively. Of PBDEs, the strongest
308 correlation was found between BDEs 153 and 154 ($r = 0.65$, $p < 0.001$), followed by
309 correlations between BDEs 99 and 100 ($r = 0.53$, $p < 0.001$), between BDEs 99 and
310 47 ($r = 0.49$, $p < 0.001$), between BDEs 100 and 153 ($r = 0.44$, $p < 0.001$), and
311 between BDEs 100 and 47 ($r = 0.43$, $p < 0.001$) (Fig. 2). BPA and BPS were strongly
312 positively correlated ($r = 0.67$, $p < 0.001$). Additionally, there were weaker but
313 statistically significant correlations between compounds from different classes (Fig. 2).
314 For example, BPA and BPS were significantly correlated with PCB 180, BDEs 47, 99
315 and 100 ($r = 0.14-0.36$, $p \leq 0.014$).

316

317 3.2. Comparing EDCs in Chinese women and French women

318 All the target PCBs and PBDEs were more frequently detected in the French women
319 (DFs = 2-82%) than in the Chinese women (DFs = 0%-44%; Fig. 1). French women
320 had significantly higher levels of all PCBs and PBDEs than Chinese women
321 (Mann-Whitney U tests, $p \leq 0.017$; Table 1), except PCB 101 that was seldom found
322 in both populations. The PLS-DA plot illustrates the discrimination between Chinese
323 and French women on the basis of their PCBs and PBDEs concentration in hair (Fig.

324 3a). Additionally, hair samples from the women in northeastern France were
325 separated from those in southeastern France, but there was no clear separation
326 between samples from the Chinese women in Baoding and Dalian (Fig. 3b).

327

328 Although BPA and BPS were found in all the hair samples from both cohorts, the
329 French women had significantly higher levels of BPA and BPS than the Chinese
330 women (Fig. 4). Concentrations of BPA and BPS in French women from both
331 sub-regions (northeast and southwest) were significantly higher than those from the
332 two Chinese cities (Kruskal-Wallis tests, $p < 0.05$, Fig. S1).

333

334 3.3. Associations between population characteristics and hair concentrations of
335 dominant EDCs

336 Results from the Multivariate linear regression are presented in Table S5. Chinese
337 women consuming red meat 4-7 times/week had 93% (95% confidence interval (CI):
338 22 to 205%) higher level of BPA in hair compared to those never consuming red meat
339 or less than 1 time/week. Women living in Dalian had 62% (95% CI: 0.7 to 121%)
340 higher BPS level compared to those in Baoding. Age, BMI, education, and frequency
341 of fish/seafood consumption were not statistically significantly associated with either
342 BPA or BPS level in hair.

343

344 Regarding the French women, the PCB 180 concentration in hair was positively
345 associated with age of the women ($p = 0.045$) but negatively associated with the
346 education levels ($p = 0.006$). A one-year increase in age was related to 3.6% (95% CI:
347 0 to 7.2%) increase in PCB 180 level. Women with university-level education had 38%
348 (95% CI: -56 to -13%) lower PCB level in hair compared to those with lower

349 education levels. BPA concentration was also negatively associated with the
350 education levels (both $p < 0.001$). Women who reported university-level education
351 had 28% (95% CI: -40 to -13%) lower BPA level than those who reported lower
352 education levels. Additionally, a one-unit increase in BMI was associated with 3.6%
353 (95% CI: 1.6 to 5.7%) increase in BPA level. The BPS concentration was also
354 positively associated with BMI ($p = 0.036$). A one-unit increase in BMI was
355 associated with 4.9% (95% CI: 0.2 to 9.6%) increase in BPS level. In addition,
356 smoking women had 74% (95% CI: 8.9 to 177%) higher BPS level compared to
357 non-smoking women. There were no statistically significant associations between the
358 living areas and concentrations of the above three compounds.

359

360 **4. Discussion**

361 We reported hair concentrations of four PCBs, seven PBDEs, and two BPs in 204
362 Chinese and 311 French women. We found higher detection frequencies (DF) and
363 concentrations for all the target PCBs and PBDEs in the French women as compared
364 to the Chinese women (Fig. 1 and Table 1). The discriminant analysis further
365 demonstrated the difference in the PCBs and PBDEs exposure profile between the
366 two populations (Fig. 3). Notably, PCB 180 was detected in 82% of the French
367 women, suggesting the widespread exposure to PCB 180 in this population.
368 Additionally, the DF values of PCBs 138, 153 and 180 determined in the French
369 women (45, 50, and 82%, respectively) are greater than those (23%, 25% and 59%)
370 reported from a recent study conducted on 151 French children using hair analysis
371 (Iglesias-González et al., 2020). The children studied by the latter authors and the
372 women described in the present study were not necessarily living in the same places,
373 but both studies included a large number of subjects and covered several areas of

374 France. Although the comparison between the two populations has to be conducted
375 with caution, the higher DF observed in the women compared to the children could
376 suggest lifelong bioaccumulation of PCBs in the French women. BPA and BPS were
377 detected in 100% of the hair samples from both the Chinese and the French women,
378 but the Chinese women had significantly lower concentrations of both BPA and BPS
379 than the French women (Fig. 4). These results suggest that both cohorts were widely
380 exposed to BPA and BPS. These results also confirm that hair analysis can effectively
381 distinguish different exposure patterns of organic contaminants among general
382 populations.

383

384 The higher DF and concentration of PCBs in the French women is likely due to the
385 higher use of PCBs in France than in China in the past, as it has been reported that
386 France and China contributed to approximately 4.1% and 0.6% of the total historical
387 global PCB consumption, respectively (Breivik et al., 2002). Although the sampling
388 time was slightly different between the French and the Chinese women (2011 versus
389 2016), this time difference is unlikely to be responsible for the differences reported
390 here because of the well-known low degradation of PCBs in the environment.
391 Similarly, the different life stages (pregnancy vs non-pregnancy) between the French
392 and the Chinese women is not considered to have biased the comparison between the
393 two cohorts. This assumption is supported by a previous work conducted on the same
394 French women, which demonstrated that the concentration of pesticides (both
395 persistent and non-persistent) in hair present very low variability between the
396 different trimester of pregnancy (Béranger et al., 2018). Similarly, Adetona et al.
397 (2013) reported that the concentration of PCBs in plasma was relatively stable across
398 trimesters, suggesting that pregnancy would have a limited influence on circulating

399 PCBs. The high frequency of PCB detection in the French women observed here is in
400 line with a previous study in which all the four PCBs were detected in 100% of breast
401 milk samples collected from French women between 2011 and 2014 (Antignac et al.,
402 2016). Of the four PCB congeners, PCB 180 was the predominant congener in both
403 Chinese women and French women (Fig. 1 and Table 1), even though PCB 180 was
404 much less consumed than the remaining three congeners in both countries (Breivik et
405 al., 2002). Likewise, PCB 180 was the most abundant congener of these four PCBs
406 (PCB 101, 138, 153, and 180) in the hair samples collected from Iranian pregnant
407 women (Behrooz et al., 2012) and French Children (Iglesias-González et al., 2020).
408 However, in hair samples from Romanian adolescents, PCB 153 was more frequently
409 detected and more abundant than PCB 180 (Covaci et al., 2008). These results likely
410 reflect the difference in historical use of commercial PCB products between countries.
411 Here the frequent detection of PCB 180 in the two female populations probably
412 suggests ongoing exposure to PCBs because of their lipophilic properties and
413 persistence in the environment, even though their production and usage have been
414 restricted or banned in China and France since the 1980s (Xing et al., 2005; Arnich et
415 al., 2009). The ongoing exposure of the French women is supported by the fact that
416 PCBs were still detected in hair samples collected in 2014-2015 from French children
417 aged from 1 to 12 years old, suggesting that exposure was still present in France
418 recently (Iglesias-González et al., 2020). However, the contribution of past exposure
419 to the body burden of PCBs revealed by hair analysis cannot be discounted, as PCBs
420 are persistent in the human body as well, with estimated intrinsic elimination
421 half-lives at background levels of 10.8, 14.4 and 11.5 years for PCBs 138, 153 and
422 180, respectively (Ritter et al., 2011). Additionally, because PCB 180 was much less
423 consumed than the remaining three congeners in both countries (Breivik et al., 2002),

424 the higher presence of PCB 180 could also suggest higher potential to transfer and/or
425 accumulate in human than lower chlorinated PCBs, as suggested by Norström et al.
426 (2010). Compared with previously reported data on general populations (Table S6),
427 median concentrations of all PCBs determined here were lower than those reported
428 for Philippine pregnant women (Malarvannan et al., 2013) and Romanian adolescents
429 (Covaci et al., 2008). Our populations also had lower levels of PCBs 138 and 180 than
430 Iranian women (Behrooz et al., 2012), and lower levels of PCBs 101, 138 and 153
431 than Chinese adults living in Zhejiang Province (Zhao et al., 2008). However, the
432 interpretation of these comparisons should be careful, as the sample size was very
433 small in the studies mentioned above (n = 4-42; Table S6) and different analytical
434 methods were applied between studies.

435

436 Similar to PCBs, higher frequencies of detection and higher levels of all PBDEs were
437 observed in the French women than in the Chinese women. Again, this may reflect
438 their greater consumption in France than in China in the past, since commercial
439 penta-BDE and octa-BDE have been phased out in the European Union since 2004
440 and in China since 2009 (La Guardia et al., 2006; Chen et al., 2018). This hypothesis
441 is further supported by the fact that deca-BDE was the predominant BDEs produced
442 and used in China (Mai et al., 2005). In both female populations, tetra- (BDE 47) and
443 penta-BDEs (BDEs 99 and 100) were more frequently detected than tri- (BDEs 28_33)
444 and hexa-BDEs (BDEs 153 and 154). This is most likely related to the penta-BDE
445 technical formulation, as BDEs 47, 99 and 100 were the major constituents of
446 commercial penta-BDE mixtures (La Guardia et al., 2006). Although the content of
447 BDE 99 was slightly higher than BDE 47 in penta-BDE products (DE-71 and
448 Bromkal 70-5DE; La Guardia et al., 2006), BDE 47 was the predominant congener in

449 both female populations (Table 1). This could be explained by differences in
450 assimilation efficiencies between BDE 47 and 99, by debromination of higher
451 brominated congeners (e.g., BDE 99 and 209), or by their combinations, as reported
452 by Stapleton et al. (2004). For instance, debromination of BDE 99 to BDE 47 has
453 been demonstrated in the gut of common carp (*Cyprinus carpio*) (Stapleton et al.,
454 2004). Similar results have been reported for hair samples from Canadian women in
455 Ontario (Carnevale et al., 2014) and Chinese adults in Pearl River Delta (Kang et al.,
456 2011) and serum samples from American pregnant women (Kalloo et al., 2018).
457 However, in hair samples from Spanish, Canadian and French children, BDE 99 was
458 more predominant than BDE 47 (Tadeo et al., 2009; Aleksa et al., 2012;
459 Iglesias-González et al., 2020). This difference may result from the difference in
460 exposure routes in children (diet versus dust), difference in use of specific BDE
461 commercial products, or differences in bioaccumulation depending on the congener.
462 In comparison with existing data on general populations in the literature (n = 4-65;
463 Table S6), concentrations of all the BDEs in our study populations were lower than
464 those in Canadian adults (Poon et al., 2014), Philippine pregnant women
465 (Malarvannan et al., 2013), and American adults (Liu et al., 2016). The median
466 concentration of BDE 47 in our French women was comparable to that reported for
467 reference adults in Zhejiang Province (1.17 pg/mg; Zhao et al., 2008), but was higher
468 than those reported for the Spanish (0.51 and 0.60 pg/mg; Tadeo et al., 2009), Polish
469 adults (0.40 pg/mg; Król et al., 2014), Canadian children (0.31 pg/mg; Aleksa et al.,
470 2012) and Chinese in Guangdong Province (0.33 and 0.42 pg/mg; Zheng et al., 2011)
471 and Shanghai (0.12 and 0.34 pg/mg; Tang et al., 2013). These differences suggest
472 differences in the body burden of BDEs, which is due to bioaccumulation over time

473 and/or to current exposure, although the influence of the analytic methods cannot be
474 excluded.

475

476 Numerous studies demonstrated the ubiquitous exposure of the general population to
477 BPA and BPS based on analyses on blood and urine (e.g., Larsson et al., 2017; Jin et
478 al., 2018; Lehmler et al., 2018; Owczarek et al., 2018). Likewise, these bisphenols
479 were detected in all the hair samples from both the Chinese and the French women
480 involved in the present study, indicating the widespread exposure to BPA and BPS in
481 these two populations. The higher exposure of the French women indicated by higher
482 hair levels of bisphenols than the Chinese women (Fig. 4) is in line with the greater
483 estimated BPA daily intake in French adults (~56.0 ng/kg bw/day) than in Chinese
484 adults (~33.3 ng/kg bw/day) (Huang et al., 2017). The significantly higher
485 concentration of BPA relative to BPS in both female populations suggests higher
486 exposure to BPA than BPS. Likewise, the BPA level was found to be higher than BPS
487 in plasma samples collected from Chinese adults (mean concentration: 0.40 versus
488 0.15 ng/mL; Jin et al., 2018) and in urine samples from American adults participating
489 in the National Health and Nutrition Examination Survey (NHANES) 2013-2014
490 (median concentration: 1.24 versus 0.37 ng/mL; Lehmler et al., 2018). Additionally,
491 BPA ($\text{Log } K_{ow}=3.32$; Chen et al., 2016) is more lipophilic than BPS ($\text{Log } K_{ow}=1.65$;
492 Michałowicz et al., 2014), which could increase BPA accumulation in human body
493 relative to BPS. Comparing BPA determined here with previously reported data on
494 general populations (Table S6), the median concentrations of BPA in our study
495 populations were higher than those reported in Greek adults (27.7 and 13.8 pg/mg;
496 Tzatzarakis et al., 2015; Karzi et al., 2018), Greek children (12.4 pg/mg; Karzi et al.,
497 2018), Spanish adults (4.60 pg/mg; Rodríguez-Gómez et al., 2017), and Korean adults

498 (< 22.9 pg/mg; Lee et al., 2017) but lower than that reported in Polish adults (337.5
499 pg/mg; Nehring et al., 2017). However, the small sample size ($n = 6-72$) in the studies
500 above may also explain the differences between them. Unlike BPA, there are no data
501 on BPS occurrence in human hair. Therefore, the present study is the first one to
502 report BPS concentrations in human hair.

503

504 We found stronger correlations between chemicals belonging to the same chemical
505 class than between chemicals from different classes, which is in line with previous
506 findings that chemicals with similar structures were more strongly correlated
507 compared to unrelated chemicals (Kalloo et al., 2018; Béranger et al. 2018; Peng et al.,
508 2020). Among the French women, we found strong correlations between PCBs 138
509 and 153 ($r = 0.80$), between BDEs 47, 99 and 100 ($r = 0.43-0.53$), between BDEs 153
510 and 154 ($r = 0.65$), as well as between BPA and BPS ($r = 0.67$) (Fig. 2). This is most
511 likely related to their combined use in industrial and commercial products and similar
512 exposure source (e.g., food products). For example, PCBs and PBDEs that have been
513 classified as POPs were sold as commercial mixtures, e.g. Aroclor 1254, Aroclor
514 1260, DE-71 and Bromkal 70-5DE (Breivik et al., 2002; La Guardia et al., 2006).
515 Likewise, in serum samples from American pregnant women, BDE 47 was strongly
516 correlated with BDE 99 ($r = 0.9$) and BDE 100 ($r = 0.9$) (Kalloo et al., 2018).
517 However, the correlation between BPA and BPS in Chinese women was much weaker
518 than that in the French women ($r = 0.31$ versus 0.67). This discrepancy may be related
519 to the difference in the use of specific products containing BPA and BPS, since BPA
520 and BPS have been found in foodstuffs (e.g., dairy products and meat) from the
521 United States (Liao and Kannan, 2013) and in personal care products from China and
522 the United States (Liao and Kannan, 2014). We did not find any hypothesis

523 explaining why BPA and BPS were weakly (although statistically significantly)
524 correlated with PCB 180, BDEs 47, 99 and 100 (Fig. 2).

525

526 The hair concentrations of the three dominant pollutants (PCB 180, BPA and BPS)
527 were found to be significantly associated with several population characteristics
528 (Table S5). PCB 180 concentration increased with the age of French women, despite
529 the narrow age range (18-44 years). A similar association has been reported in serum
530 samples from a Romanian population (age: 8-90 years; Dirtu et al., 2006). These
531 findings may indicate the lifetime cumulative exposure to PCBs among general
532 populations. The significant positive association between BPA concentrations in the
533 Chinese women's hair and frequency of consuming red meat may suggest that red
534 meat could be a source of exposure to BPA in China, as it has been reported that BPA
535 was detected in 76.5% of meat and meat products from the United States (Liao and
536 Kannan 2013). Although fish/seafood has been recognized as a dietary source of
537 exposure to BPA in addition to red meat (Liao and Kannan, 2013), there was no
538 significant association between frequency of consuming fish/seafood and hair BPA
539 concentration. However, we were not able to confirm such relations in the French
540 women, as there were no data on consumption of red meat or fish/ seafood for this
541 population. Unlike in Chinese women, BMI and education were found to be
542 significantly associated with hair BPA level in the French women. Likewise, previous
543 studies also reported discordant results concerning the relationship between BPA level
544 and BMI (Carwile and Michels, 2011; Arbuckle et al., 2014). Therefore, these results
545 should be interpreted cautiously. In agreement with our results, a negative relation
546 was observed between education and BPA level in urine samples collected from
547 women in a Canadian pregnancy cohort study (Arbuckle et al., 2014). Similar to BPA,

548 hair BPS concentration was associated with different characteristics between the
549 Chinese (living cities) and French women (BMI and smoking habit). The relation
550 between living cities and BPS level in the Chinese women may be caused by
551 difference in food contamination, as it has been suggested that diet is the most
552 important source of exposure to bisphenols in the general population (Wang et al.,
553 2015). The positive association between BMI and hair BPS concentration in the
554 French women is in line with the findings of Liu et al. (2017), who reported that
555 adults who were obese had higher urinary BPS level than adults who were not obese
556 in the National Health and Nutrition Examination Survey (NHANES) of the US
557 population.

558

559 Our study is subjected to a limitation. The analysis of PBDEs was restricted to
560 penta-BDEs in the present study, based on the results from most of previous
561 biomonitoring studies. However, deca-BDE (BDE-209) has also been detected in
562 non-occupationally exposed humans (e.g., breast milk and blood), although
563 bioaccumulation of BDE-209 was assumed to be low because of its large molecular
564 size, extreme hydrophobicity and low bioavailability (Frederiksen et al., 2009; Van
565 den Berg et al., 2017). Despite this limitation, our study aims, to show cumulative
566 exposure to EDCs in female adult populations and to identify potential regional
567 variability in exposure, are accomplished. Additionally, our study has several
568 strengths. The large sample size enables us to identify potential differences in
569 biomarkers concentrations between populations. Measurements were done in the same
570 laboratory and therefore our comparisons between the two cohorts are not impacted
571 by differences in analytical techniques. To our knowledge, this is the first study
572 reporting BPS concentrations in human hair.

573

574 **5. Conclusions**

575 The present study demonstrate that hair analysis is sufficiently sensitive to detect
576 exposure to endocrine disruptors such as PCBs, PBDEs and bisphenols, and highlight
577 differences in exposure between populations even at environmental levels. The body
578 burden of target PCBs and PBDEs determined by hair analysis was different between
579 the Chinese and the French women studied, indicating that the two cohorts had
580 different PCBs and PBDEs exposure profile. Although the French women were more
581 extensively exposed to BPA and BPS compared to the Chinese women, these two
582 contaminants were detected in all the hair samples from both cohorts, suggesting the
583 widespread exposure to BPA and BPS in general populations. Our results demonstrate
584 the multiple exposure of the general population and provide baseline data on women's
585 exposure to target EDCs.

586

587 **Acknowledgements**

588 ELFE is a study conducted conjointly by National Institute of Demographic Studies
589 (Ined), French National Institute for Health and Medical Research (Inserm), French
590 blood establishment (EFS), French Institute for Public Health Surveillance (InVS),
591 French National Institute for Statistics and Economic Studies (Insee), General
592 Directorate for Health (DGS, Ministry of Health), General Directorate for Risk
593 Prevention (DGPR, Ministry of Environment), Directorate for Research, Studies,
594 Evaluation and Statistics (Drees) and French National Family Allowance Fund (Cnaf).
595 It receives additional funding from the Ministry of Research, Committee on the
596 human and social sciences (CCDSHS), and the Ministry of Culture and

597 Communication (Deps). As part of the RECONAI platform, Elfe also receives
598 funding from the National research agency (ANR-11-EQPX-0038).

599 The author would like to acknowledge the financial support of L'Oréal for the hair
600 analysis performed on the Chinese women.

601

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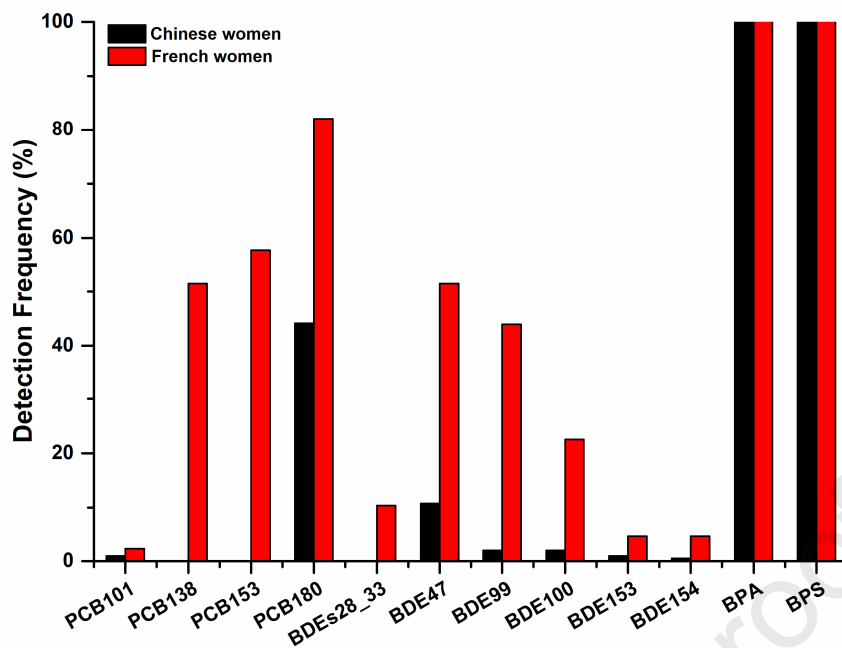
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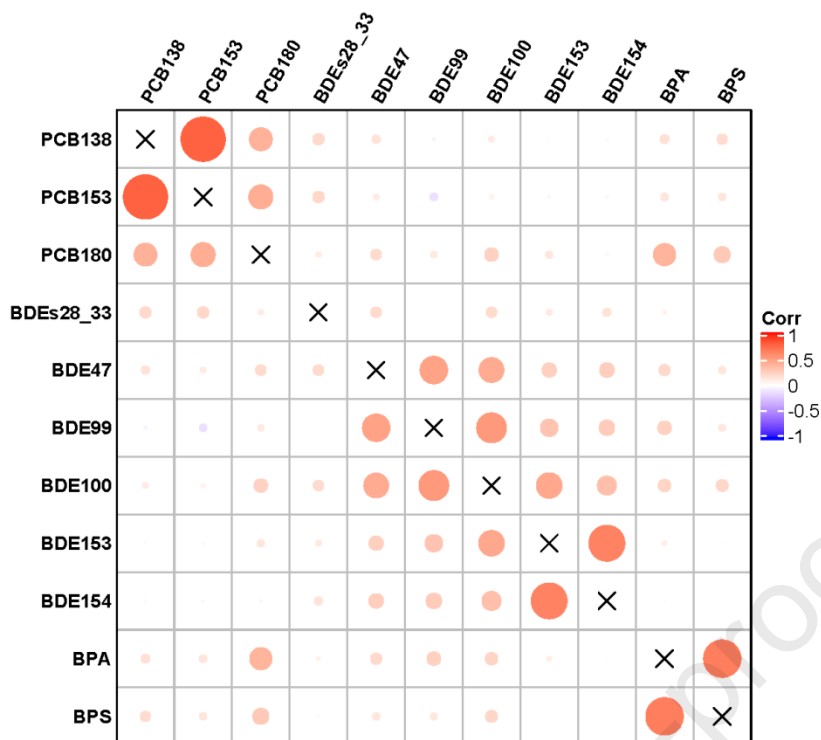
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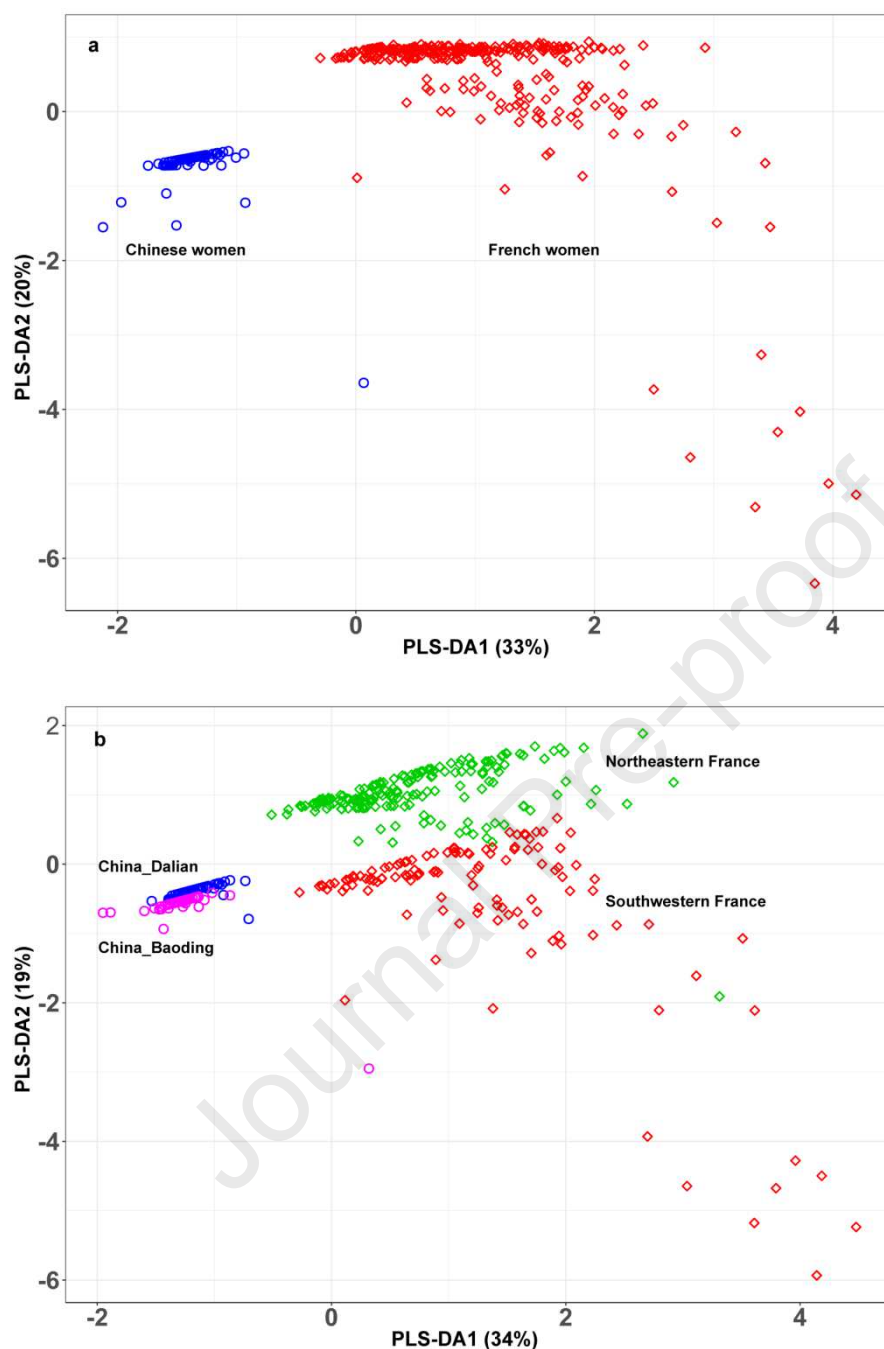
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873 **Figure 1.** Detection frequencies of target compounds in hair samples collected from
874 the Chinese (n=204; black bars) and the French women (n=311; red bars).



875

876 **Figure 2.** Heatmap showing pairwise correlations between analyte concentrations in
 877 hair samples collected from the French women in 2011. Blue and red colors indicate
 878 negative and positive correlations based on Spearman tests, respectively. The size of
 879 circle symbols represents the strength of correlations. A cross symbolizes that the
 880 correlation coefficient is 1. Only analytes detected in more than ten samples are
 881 presented in the graph. Abbreviations for all compounds are shown in the Supporting
 882 Information.



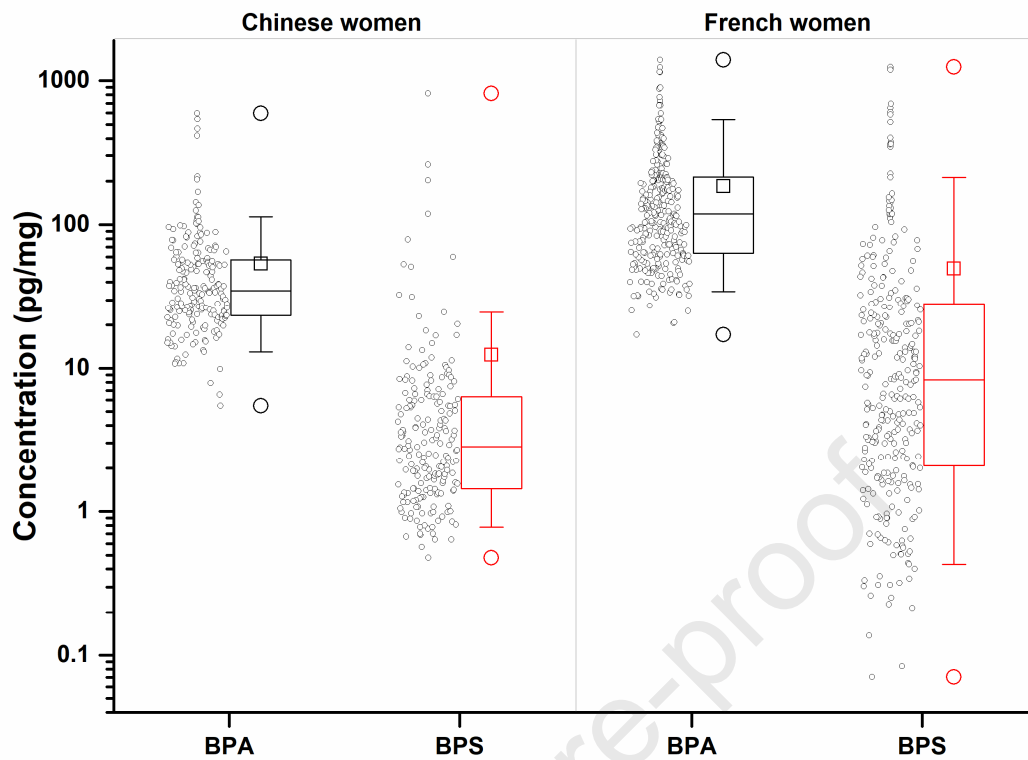
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886 **Figure 3.** Sample plots of partial least squared-discriminant analyses (PLS-DA)
887 performed on POPs (i.e., PCBs and PBDEs) determined in hair samples collected
888 from the Chinese (n=204) and the French women (n=311). In graph a, blue circle and
889 red diamond symbols indicate hair samples from the Chinese and the French women,
890 respectively; In graph b, blue and magenta circle symbols represent hair samples from the
891 Chinese women living in Dalian (n=102) and Baoding (n=102), respectively,
892 while green and red diamond symbols indicate hair samples from the French women
893 living in northeast (n=182) and southwest (n=129), respectively.

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895

896 **Figure 4.** Concentrations of BPA and BPS in hair samples collected from the Chinese
897 (n=204) and the French women (n=311). Black circles indicate hair samples. Boxes
898 represent 25-75 percentiles, with whiskers of 5th and 95th percentiles. The lowest and
899 highest circles indicate the minimum and maximum. Lines inside the boxes represent
900 medians while squares represent means.

901

902

903 **Table 1.** Concentrations (pg/mg) of PCBs, BDEs, BPA and BPS in human hair collected from the Chinese and the French women.

Analytes	China (n=204)								France (n=311)								p-value	LOD
	DF (%)	Arithmetic mean	Min	25% P	50% P	75% P	95% P	Max	DF (%)	Arithmetic mean	Min	25% P	50% P	75% P	95% P	Max		
PCB 101	1	4.63	< LOD	< LOD	< LOD	< LOD	< LOD	63.2	2	0.07	< LOD	< LOD	< LOD	< LOD	< LOD	1.12	0.558	0.26
PCB 138	0	-	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	45	0.37	< LOD	< LOD	< LOD	0.28	0.83	3.36	2.61E-28	0.03
PCB 153	0	-	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	50	0.63	< LOD	< LOD	< LOD	0.3	0.746	9.68	4.07E-32	0.08
PCB 180	44	0.24	< LOD	< LOD	< LOD	0.12	0.47	2.12	82	0.54	< LOD	0.1	0.25	0.45	1.12	6.79	6.80E-28	0.02
BDEs 28_33	0	-	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	7	0.26	< LOD	< LOD	< LOD	< LOD	0.13	3.37	7.23E-05	0.04
BDE 47	11	0.75	< LOD	< LOD	< LOD	< LOD	0.55	7.87	57	16	< LOD	< LOD	1.37	5.09	30.9	152	4.81E-29	0.15
BDE 99	2	1.02	< LOD	< LOD	< LOD	< LOD	< LOD	14.8	49	38.5	< LOD	< LOD	< LOD	4.02	26.6	396	2.94E-29	0.63
BDE 100	2	0.66	< LOD	< LOD	< LOD	< LOD	< LOD	9.49	20	5.5	< LOD	< LOD	< LOD	< LOD	4.23	73.7	2.01E-09	0.23
BDE 153	1	0.3	< LOD	< LOD	< LOD	< LOD	< LOD	4.75	5	14.7	< LOD	< LOD	< LOD	< LOD	0.98	234	0.011	0.91
BDE 154	0.5	0.09	< LOD	< LOD	< LOD	< LOD	< LOD	1.61	4	7.72	< LOD	< LOD	< LOD	< LOD	< LOD	112	0.017	0.67
Bisphenol A	100	72.6	5.47	23.4	34.9	57.2	122	596	100	199	17.1	63.4	118	213	541	1398	2.53E-43	5.47
Bisphenol S	100	62.1	0.48	1.43	2.84	6.34	29.9	817	100	149	0.07	2.11	8.32	27.9	217	1246	1.46E-10	0.07

904 DF: detection frequency; Min: minimum; LOD: limit of detection; Max: maximum; p-values were obtained from Mann-Whitney U-test.

Highlights

- Hair analysis was used to assess exposure of two female populations to multiclass EDCs
- All PCBs and PBDEs analyzed were more frequently detected in French women than in Chinese women
- PCB 180 was detected in 82% of the French women and 44% of the Chinese women
- BPA and BPS were found in 100% of the hair samples from both female populations
- French women had significantly higher levels of BPA and BPS than the Chinese women

Declaration of interests

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

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

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Human exposure to PCBs, PBDEs and bisphenols revealed by hair analysis: a comparison between Chinese and French women

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Fig. S1. Concentrations of BPA and BPS in hair samples from the Chinese women in Baoding (n=102), the Chinese women in Dalian (n=102), the French women in northeast (n=182), and the French women in southwest (n=129).

References

Table S1 Physicochemical properties of target compound in this study.

Analyte	Abbreviation	CAS	Formula	M.W. (mol/g)	Log Kow	References
2,2',4,5,5'-Pentachlorobiphenyl	PCB101	37680-73-2	C ₁₂ H ₅ Cl ₅	326.43	6.44	Veith et al. (1979)
2,2',3',4,4',5-Hexachlorobiphenyl	PCB138	35065-28-2	C ₁₂ H ₄ Cl ₆	360.88	6.83	Norström et al. (2010)
2,2',4,4',5,5'-Hexachlorobiphenyl	PCB153	35065-27-1	C ₁₂ H ₄ Cl ₆	360.88	6.92	Norström et al. (2010)
2,2',3,4,4',5,5'-Heptachlorobiphenyl	PCB180	35065-29-3	C ₁₂ H ₃ Cl ₇	395.32	7.62	Norström et al. (2010)
2,4,4'-Tribromodiphenyl ether	BDE28	41318-75-6	C ₁₂ H ₇ Br ₃ O	406.89	5.88	Llorca-Porcel et al. (2006)
2',3,4-Tribromodiphenyl ether	BDE33	147217-78-5	C ₁₂ H ₇ Br ₃ O	406.89	5.88	Llorca-Porcel et al. (2006)
2,2',4,4'-Tetrabromodiphenyl ether	BDE47	5436-43-1	C ₁₂ H ₆ Br ₄ O	485.79	6.77	Llorca-Porcel et al. (2006)
2,2',4,4',5-Pentabromodiphenyl	BDE99	60348-60-9	C ₁₂ H ₅ Br ₅ O	564.69	7.66	Llorca-Porcel et al. (2006)
2,2',4,4',6-Pentabromodiphenyl	BDE100	189084-64-8	C ₁₂ H ₅ Br ₅ O	564.69	7.66	Llorca-Porcel et al. (2006)
2,2',4,4',5,5'-Hexabromodiphenyl	BDE153	68631-49-2	C ₁₂ H ₄ Br ₆ O	643.58	8.55	Llorca-Porcel et al. (2006)
2,2',4,4',5,6'-Hexabromodiphenyl	BDE154	207122-15-4	C ₁₂ H ₄ Br ₆ O	643.58	8.55	Llorca-Porcel et al. (2006)
Bisphenol A	BPA	80-05-7	C ₁₅ H ₁₆ O ₂	228.29	3.32	Chen et al. (2016)
Bisphenol S	BPS	80-09-1	C ₁₂ H ₁₀ O ₄ S	250.27	1.65	Michałowicz et al. (2014)

Table S2 Characteristics of participants providing hair samples for analysis of target pollutants.

Characteristics	Chinese women (n=204)		French women (n=311)	
	N (%)	Mean \pm STD	N (%)	Mean \pm STD
Age ^a	204	34.2 \pm 5.7	310	30.1 \pm 5.0
BMI ^a	204	22.7 \pm 3.4	310	23.2 \pm 4.6
Education ^a				
High school or less	58 (28.4%)		140 (45.2%)	
University	146 (71.6%)		170 (54.8%)	
Smoking habit ^b				
Non-smoker	204 (100%)		230 (74.4%)	
Smoker	0		79 (25.6%)	
Consumption frequency of fish/seafood				
< 1 time/week	62 (30.4%)		NA	
1-3 times/week	126 (61.8%)		NA	
4-7 times/week	16 (7.8%)		NA	
Consumption frequency of fish ^c				
< 1 time/month	NA		93 (29.9%)	
\geq 1 time/month	NA		182 (58.5%)	

BMI, body mass index;

^a one missing value for the French women;

^b two missing values for the French women;

^c Thirty six missing values for the French women;

NA, not applicable.

Table S3 Method validation parameters of analysis of target PCBs, PBDEs and BPs in hair.

Compound	Analysis mode	ISTD	R ²	LOQ (pg/mg)	Accuracy (%; n = 5)					Variability (%; n = 5)					Recovery (%)		
					LOQ	0.1 pg/mg	1 pg/mg	10 pg/mg	100 pg/mg	LOQ	0.1 pg/mg	1 pg/mg	10 pg/mg	100 pg/mg	1 pg/mg	10 pg/mg	100 pg/mg
PCB 101	SPME-GC-NCI-MS/MS	γ -HCH-D ₆	0.963	20	112	N.D.	N.D.	N.D.	104	13	N.D.	N.D.	N.D.	11	N.D.	N.D.	132
PCB 138	SPME-GC-NCI-MS/MS	γ -HCH-D ₆	0.984	10	116	N.D.	N.D.	116	103	6	N.D.	N.D.	6	10	N.D.	116	131
PCB 153	SPME-GC-NCI-MS/MS	β -endosulfan-D ₄	0.955	20	100	N.D.	N.D.	157	99	22	N.D.	N.D.	18	12	N.D.	106	127
PCB 180	Liquid Injection-GC-NCI-MS/MS	Permethrin-D ₆	0.995	0.3*	N.A.	102**	88**	101**	101**	11	16	14	4	7	116	155	104
BDEs 28 + 33	SPME-GC-NCI-MS/MS	γ -HCH-D ₆	0.990	20	79	N.D.	N.D.	68	97	8	N.D.	N.D.	15	7	N.D.	109	120
BDE 47	Liquid Injection-GC-NCI-MS/MS	Permethrin-D ₆	0.990	5	90	N.D.	N.D.	79	101	20	N.D.	N.D.	5	9	N.D.	117	134
BDE 99	Liquid Injection-GC-NCI-MS/MS	Permethrin-D ₆	0.988	5	95	N.D.	N.D.	106	100	19	N.D.	N.D.	11	8	N.D.	141	92
BDE 100	Liquid Injection-GC-NCI-MS/MS	Permethrin-D ₆	0.991	2	96	N.D.	N.D.	105	99	22	N.D.	N.D.	15	6	N.D.	136	117
BDE 153	Liquid Injection-GC-NCI-MS/MS	Permethrin-D ₆	0.990	5	94	N.D.	N.D.	108	102	13	N.D.	N.D.	9	6	N.D.	134	87
BDE 154	Liquid Injection-GC-NCI-MS/MS	Permethrin-D ₆	0.986	5	114	N.D.	N.D.	96	101	12	N.D.	N.D.	16	6	N.D.	150	98
Bisphenol A	Liquid Injection-GC-NCI-MS/MS	BPA-d ₁₆	0.879	50	112	N.D.	N.D.	N.D.	106	16	N.D.	N.D.	N.D.	8	N.D.	N.D.	15
Bisphenol S	LC-MS/MS	Thiabendazole- ¹³ C ₆	0.964	9*	N.A.	1444	109	92	103	5	7	15	6	11	109	109	120

ISTD, internal standard; LOQ, limit of quantification.

* LOQ determined from concentrations in blank hair.

N.A., Accuracy Not Applicable.

** Accuracy of level supplemented (i.e. determined concentration minus background concentration in blank hair).

N.D., not determined.

Table S4 Concentrations (pg/mg) of PCBs, BDEs, BPA and BPS in human hair collected from the Chinese women in Baoding and Dalian and the French women in northeast and southwest.

Analytes	Baoding (n=102)								Dalian (n=102)								p-value	LOD
	DF (%)	Mean	Min	25% P	50% P	75% P	95% P	Max	DF (%)	Mean	Min	25% P	50% P	75% P	95% P	Max		
PCB101	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	2	0.95	< LOD	< LOD	< LOD	< LOD	< LOD	63.2	0.156	0.26
PCB138	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1	0.03
PCB153	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1	0.08
PCB180	49	0.16	< LOD	< LOD	< LOD	0.15	0.89	2.12	39	0.06	< LOD	< LOD	< LOD	0.07	0.26	1.03	0.021	0.02
BDEs28_33	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1	0.04
BDE47	12	0.2	< LOD	< LOD	< LOD	< LOD	0.55	4.99	10	0.23	< LOD	< LOD	< LOD	< LOD	0.9	7.87	0.684	0.15
BDE99	2	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.24	2	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	14.8	0.992	0.63
BDE100	2	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.83	2	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	9.49	1	0.23
BDE153	1	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.2	1	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	4.75	0.994	0.91
BDE154	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.61	0.317	0.67
BPA	100	60.6	7.85	25.6	38.1	64.8	160	544	100	47.1	5.47	21.8	30.8	53.2	123	596	0.029	5.47
BPS	100	5.84	0.48	1.33	2.05	5.85	14.7	202	100	18.9	0.64	1.84	3.51	8.22	58.8	817	0.006	0.07

Analytes	Northeast (n=182)								Southwest (n=129)								p-value
	DF (%)	Mean	Min	25% P	50% P	75% P	95% P	Max	DF (%)	Mean	Min	25% P	50% P	75% P	95% P	Max	
PCB101	3	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.12	0	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.058
PCB138	44	0.22	< LOD	< LOD	< LOD	0.3	0.94	2.44	47	0.21	< LOD	< LOD	< LOD	0.25	0.75	3.36	0.932
PCB153	48	0.27	< LOD	< LOD	< LOD	0.31	0.88	9.68	53	0.21	< LOD	< LOD	0.11	0.27	0.6	3.41	0.979
PCB180	80	0.33	< LOD	0.1	0.23	0.42	1.04	2	84	0.42	< LOD	0.11	0.27	0.49	1.16	6.79	0.226
BDEs28_33	2	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.13	15	0.12	< LOD	< LOD	< LOD	< LOD	0.66	3.37	2.30E-05
BDE47	56	4.34	< LOD	< LOD	1.1	4.36	19.3	82.1	59	9.15	< LOD	< LOD	1.73	8.09	49.4	152	0.081
BDE99	49	3.15	< LOD	< LOD	< LOD	3.07	12.7	64.8	49	17.2	< LOD	< LOD	< LOD	4.38	109	396	0.389
BDE100	15	0.53	< LOD	< LOD	< LOD	< LOD	2.37	24.2	26	2.13	< LOD	< LOD	< LOD	0.72	11.4	73.7	0.01
BDE153	1	0.49	< LOD	< LOD	< LOD	< LOD	< LOD	6.89	11	4.48	< LOD	< LOD	< LOD	< LOD	21.7	234	1.10E-04
BDE154	2	0.39	< LOD	< LOD	< LOD	< LOD	< LOD	8.17	7	2.55	< LOD	< LOD	< LOD	< LOD	9.49	112	0.015
BPA	100	184	20.6	63.5	117.5	223	540	1242	100	185	17.1	63.5	120	203	570	1398	0.901
BPS	100	64.5	0.08	2.03	7.44	29.5	401	1246	100	29.4	0.07	2.14	8.94	25.6	147	365	0.853

DF: detection frequency; LOD: limit of detection; Min: minimum; Max: maximum; p-values were obtained from Mann-Whitney U-test.

Table S5 Multiple linear regression models of dominant chemicals on population characteristics.

Country	Response	Explanatory variable	Coefficient	95% CI	<i>P</i>
China	Log10 BPA	-	-	-	-
	Log10 BPS	Living in Dalian (vs. Baoding)	0.209	(0.003, 0.345)	< 0.001
France	Log10 PCB180	Age (years)	0.015	(0.000, 0.030)	0.045
		University (vs. High school or less)	-0.207	(-0.355, -0.059)	0.006
	Log10 BPA	BMI (kg/m ²)	0.015	(0.007, 0.024)	< 0.001
		University (vs. High school or less)	-0.142	(-0.222, -0.061)	< 0.001
	Log10 BPS	BMI (kg/m ²)	0.021	(0.001, 0.040)	0.036
		Smoking (vs. not)	0.240	(0.037, 0.443)	0.021

BPA in the hair of Chinese women was not significantly associated with any of population characteristics evaluated; Consumption frequency of fish/seafood or fish was not retained in any final models and is thus not shown in the table.

Table S6 Comparison of concentrations of target compounds in human hair from various countries.

Country	Survey year	Age	N	PCB101	PCB 138	PCB 153	PCB 180	BDE 28	BDE 47	BDE 99	BDE 100	BDE 153	BDE 154	BPA	BPS	References	
Canada	NA	20-65	50					nd-22.6	4.6-815 (21)	5.8-1128 (20.3)	nd-207	nd-113.5	nd-85.4			Poon et al. (2014)	
Canada	NA	Female adults	65					nd-19.4 (nd)	nd-207.9	nd-139.4 (6.9)	nd-23.3 (nd)	nd-15.7 (nd)	nd-10.1 (nd)			Carnevale et al. (2014)	
Canada	NA	1-15	12					nd-0.09 (nd)	0.07-0.84	0.04-1.18	nd-0.45	nd-0.16 (0.03)	nd-0.12 (nd)			Aleksa et al. (2012)	
USA	2014	19-38	50					0.23-8.6 (1.0)	5.2-880 (33)	2.2-1020 (21)	0.48-176	1.4-78 (2.4)	0.16-54 (1.2)			Liu et al. (2016)	
Germany	NA	Adults	4											<41		Martín et al. (2015)	
Greece	2015-2016	2-12	50											nd-205.5 (12.4)		Karzi et al. (2018)	
Greece	2015-2016	25-91	72											nd-53.8 (13.8)		Karzi et al. (2018)	
Greece	2011-2014	Adults	8											27.7		Tzatzarakis et al. (2015)	
Greece	2011-2014	Children	61											45.6		Tzatzarakis et al. (2015)	
Poland	2012	25-30	12					nd-2.8 (0.4)	nd-0.65 (0.4)	nd-0.42 (0.21)	nd	nd	nd			Król et al. (2014)	
Poland	NA	16-55	42											26.1-1498.6		Nehring et al. (2017)	
Romania	2002-2003	Adolescents	42		0.2-7.3 (0.9)	0.2-20.5 (2.1)	0.2-10.7										Covaci et al. (2008)
Spain	NA	NA	6											nd-158 (44.5)		Martín et al. (2016)	
Spain	NA	24-44	9					nd	0.31-3.9 (0.6)	0.3-2.1 (0.85)	0.3-1.63 (0.5)	nd	nd			Tadeo et al. (2009)	
Spain	NA	1-11	7					nd	0.31-0.69	0.33-1.22 (0.8)	0.3-0.58	nd	nd			Tadeo et al. (2009)	
Spain	NA	> 18	6											nd-45 (4.6)		Rodríguez-Gómez et al. (2017)	
Iran	2007-2008	18-36	21	nd-2.5 (nd)	nd-4.5 (0.4)	nd-4 (nd)	nd-138 (6.5)										Behrooz et al. (2012)
Iran	2007-2008	16-43	16	nd-1.0 (nd)	nd-0.6 (0.3)	nd	3.5-13.5 (7)										Behrooz et al. (2012)
Iran	2007-2008	15-36	19	nd	nd-1.0 (0.4)	nd	nd-15 (nd)										Behrooz et al. (2012)
Korea	NA	25-28	10											17.0-22.9			Lee et al. (2017)
Philippines	2008	18-36	10	0.55-3.6 (1.7)	0.91-7.3 (3.7)	0.70-8.0 (3.8)	0.91-5.9	0.11-0.46	1.8-28 (3.4)	0.73-6.7 (1.0)	<0.02-0.86	0.25-4.5	0.10-0.71				Malarvannan et al. (2013)
China (Guangdong)	NA	22-42	18					nd	0.86-5.24	0.22-1.47	0.13-0.49	nd	nd				Kang et al. (2011)

China (Guangdong)	2014	21-25	43					(0.02)	(nd)	(nd)	(nd)	(0.06)	(nd)				Qiao et al. (2018)
China (Guangdong)	NA	NA	29						0.13-5.27	0.07-2.62	nd-0.76	nd-0.82 (0.08)	nd-0.22				Zheng et al. (2011)
China (Guangdong)	NA	NA	32						0.09-2.42	0.09-1.17	0.03-0.20	0.03-0.51	nd-0.11				Zheng et al. (2011)
China (Shanghai)	2010	Male adults	14					0.13-0.81	0.05-2.17	0.03-2.51	nd-0.45 (nd)	nd-0.27 (0.03)	nd-0.17 (nd)				Tang et al. (2013)
China (Shanghai)	2010	Female adults	11					0.26-3.43	0.14-1.68	nd-0.87 (0.26)	nd	nd-0.46 (0.2)	nd-0.23 (nd)				Tang et al. (2013)
China (Shanghai)	2007	18-50	11					0.42-3.66	2.70-32.6	0.3-13.1	0.12-2.62	nd-1.13	nd-0.71				Ma et al. (2011)
China (Zhejiang)	2015	23-62	65					nd-20.8	nd-1.9	nd-6.8	nd-0.5	nd-3.0	nd-0.4				Liang et al. (2016)
China (Zhejiang)	2015	24-57	22					nd-1.3	nd-9.5	nd-3.7	nd-1.8	nd-6.6	nd-3.9				Liang et al. (2016)
China (Zhejiang)	2007	Adults	4	0.47-1.79	0.87-3.26	0.13-0.44	0.10-0.47	0.53-1.14	1.07-1.57	0.25-0.45	0.25-1.81	0.30-1.39	nd				Zhao et al. (2008)
China (Zhejiang)	2005	Female adults	5					nd-0.42	nd-1.07 (0.34)	nd-0.44 (0.15)	nd-0.12	nd-0.41 (0.04)	nd-0.10				Leung et al. (2010)
China	2016	25-45	204	nd-20.4 (nd)	nd	nd	nd-2.12 (nd)	nd*	nd-7.87 (nd)	nd-14.8 (nd)	nd-9.49 (nd)	nd-4.75 (nd)	nd-1.61 (nd)	5.47-596 (34.9)	0.48-817		This study
France	2011	18-43	311	nd-1.12 (nd)	nd-3.36 (nd)	nd-9.68 (nd)	nd-6.79	nd-3.37 (nd)	nd-152 (1.37)	nd-396 (nd)	nd-73.7 (nd)	nd-234 (nd)	nd-112 (nd)	17.0-1398 (118)	0.10-1246		This study

NA: not available; N: number of samples; nd: not detected; Medians are presented in brackets. Asterisk: BDE 28 represents BDE 28 and 33 (BDE 28+33). We did not include data on occupational exposures or residence around e-waste recycling area or plants.

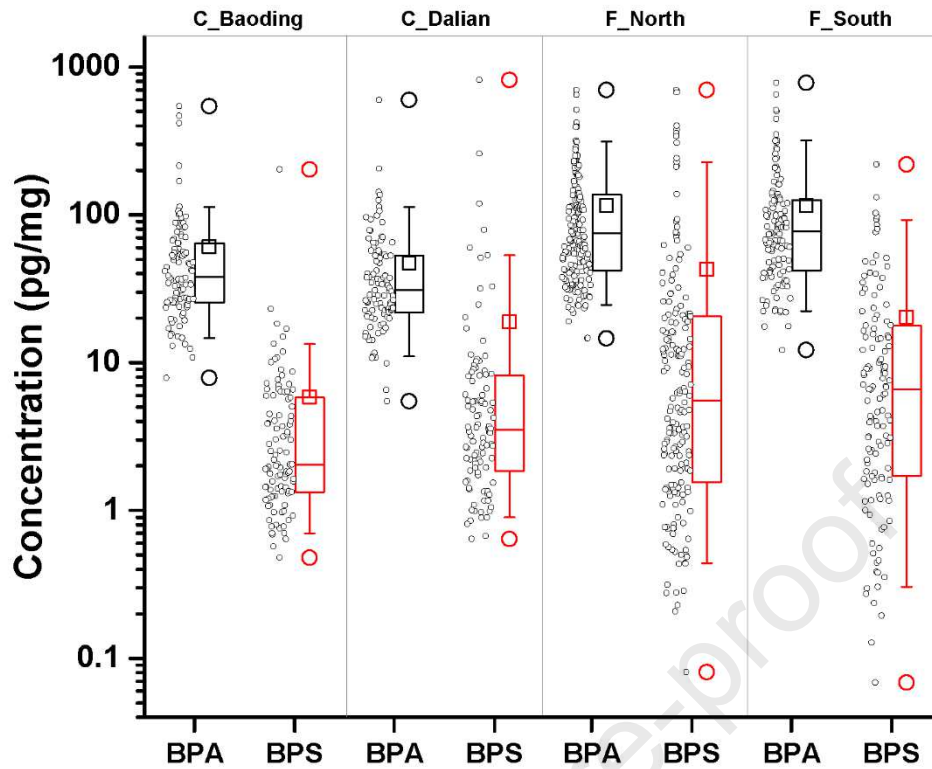


Fig. S1. Concentrations of BPA and BPS in hair samples collected from the Chinese women in Baoding (n=102), the Chinese women in Dalian (n=102), the French women in northeast (n=182), and the French women in southwest (n=129). Black circles indicate hair samples. Boxes represent 25-75 percentiles, with whiskers of 5th and 95th percentiles. The lowest and highest circles indicate the minimum and maximum. Lines inside the boxes represent medians while squares represent means.

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