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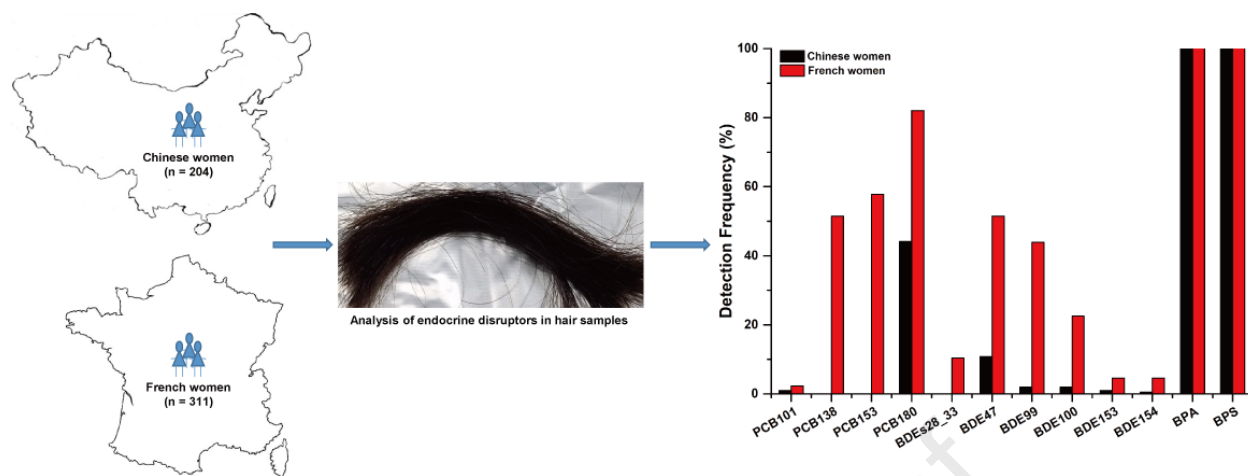
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**Human exposure to PCBs, PBDEs and bisphenols revealed by hair
analysis: a comparison between two adult female populations in
China and France**

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

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

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

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1. Introduction

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

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

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

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

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

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

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

2. Materials and methods

2.1. Chemicals and reagents

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

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

2.2. Sample collection

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

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

2.3. Sample preparation and analysis

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

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

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

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

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

2.4. Statistical analysis

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

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

3. Results

3.1. Hair concentrations of EDCs

3.1.1. Chinese women

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

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

3.1.2. French women

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

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

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

3.2. Comparing EDCs in Chinese women and French women

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

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

4. Discussion

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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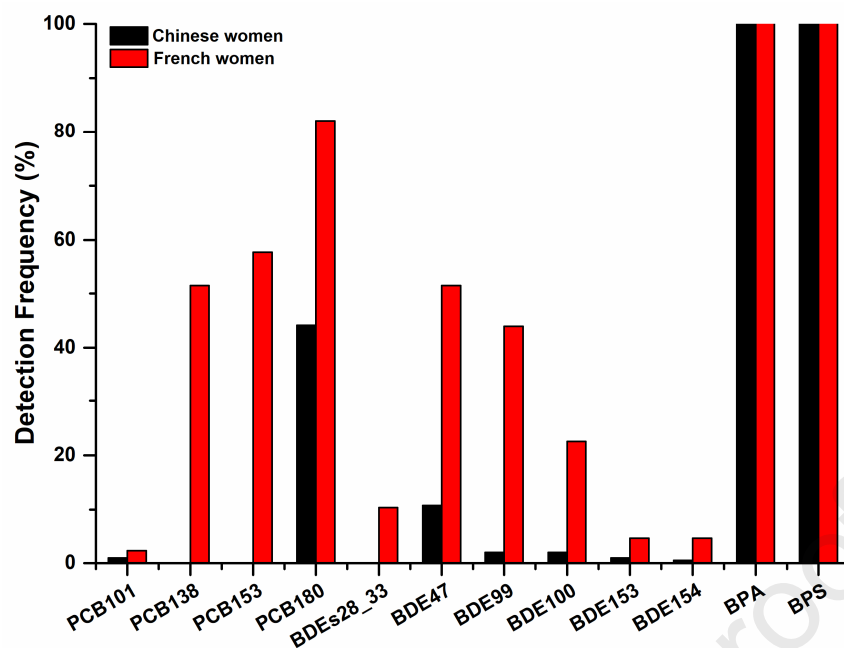


Figure 1. Detection frequencies of target compounds in hair samples collected from the Chinese (n=204; black bars) and the French women (n=311; red bars).

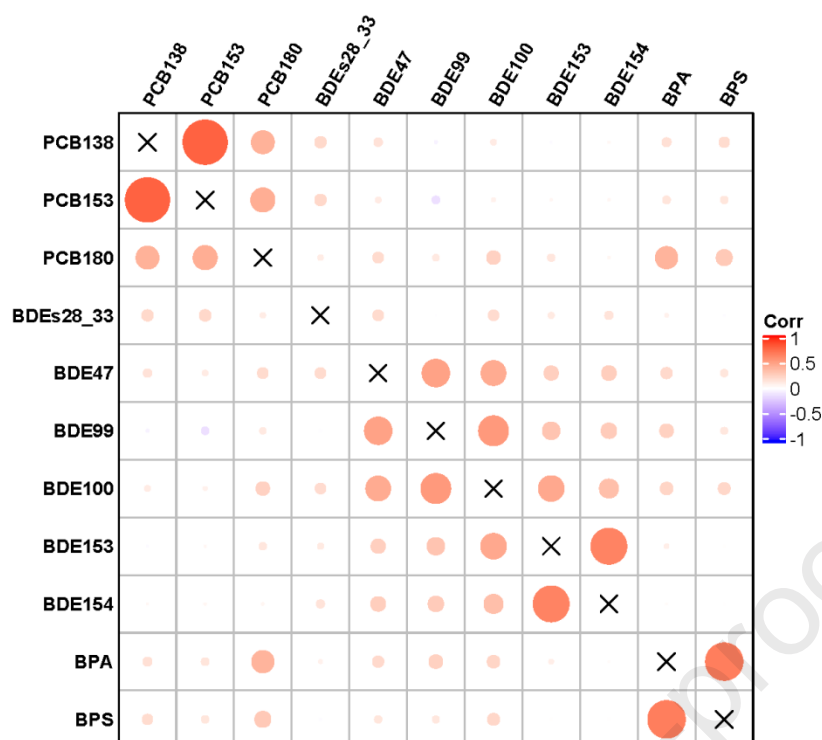


Figure 2. Heatmap showing pairwise correlations between analyte concentrations in hair samples collected from the French women in 2011. Blue and red colors indicate negative and positive correlations based on Spearman tests, respectively. The size of circle symbols represents the strength of correlations.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

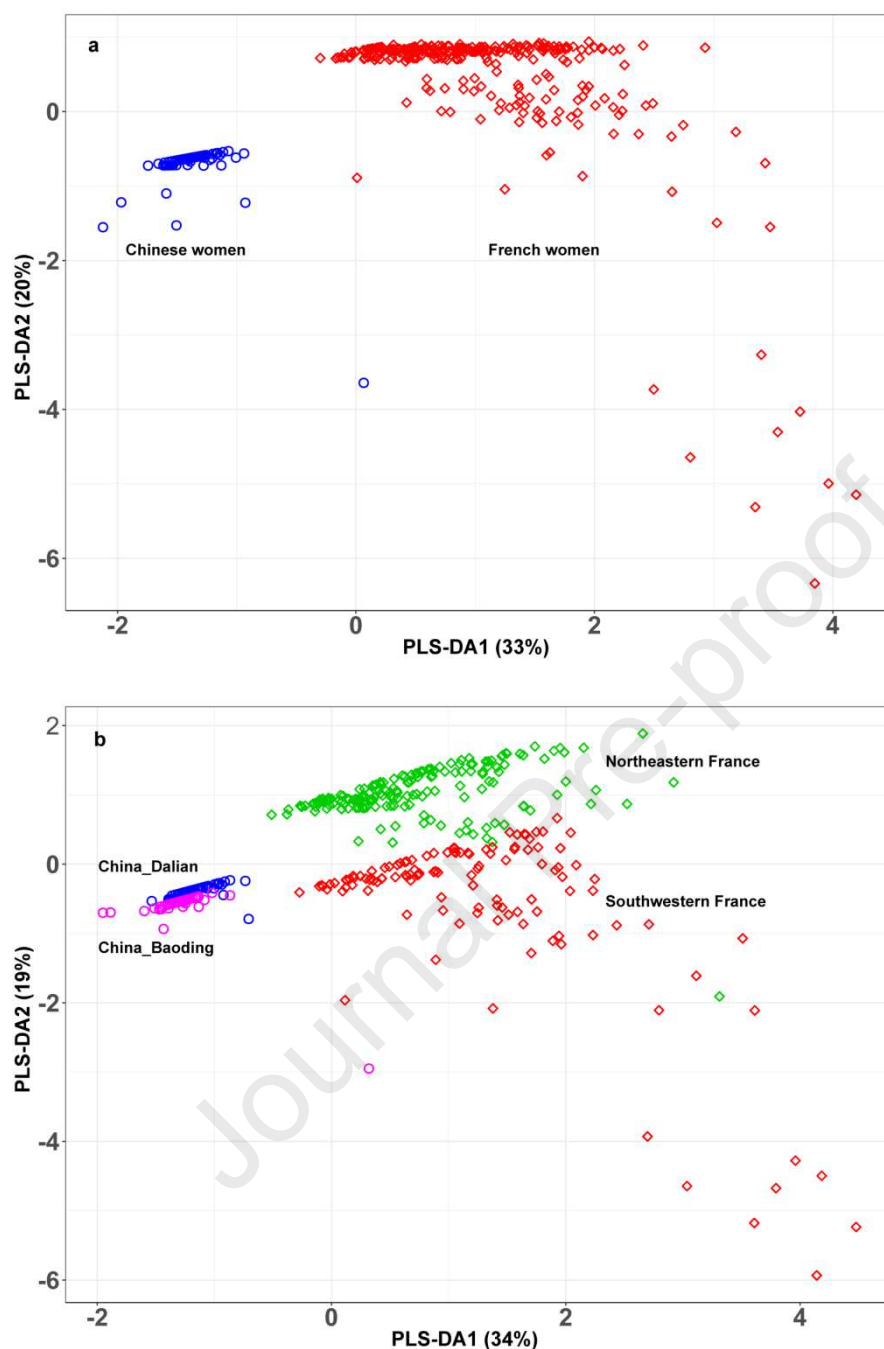
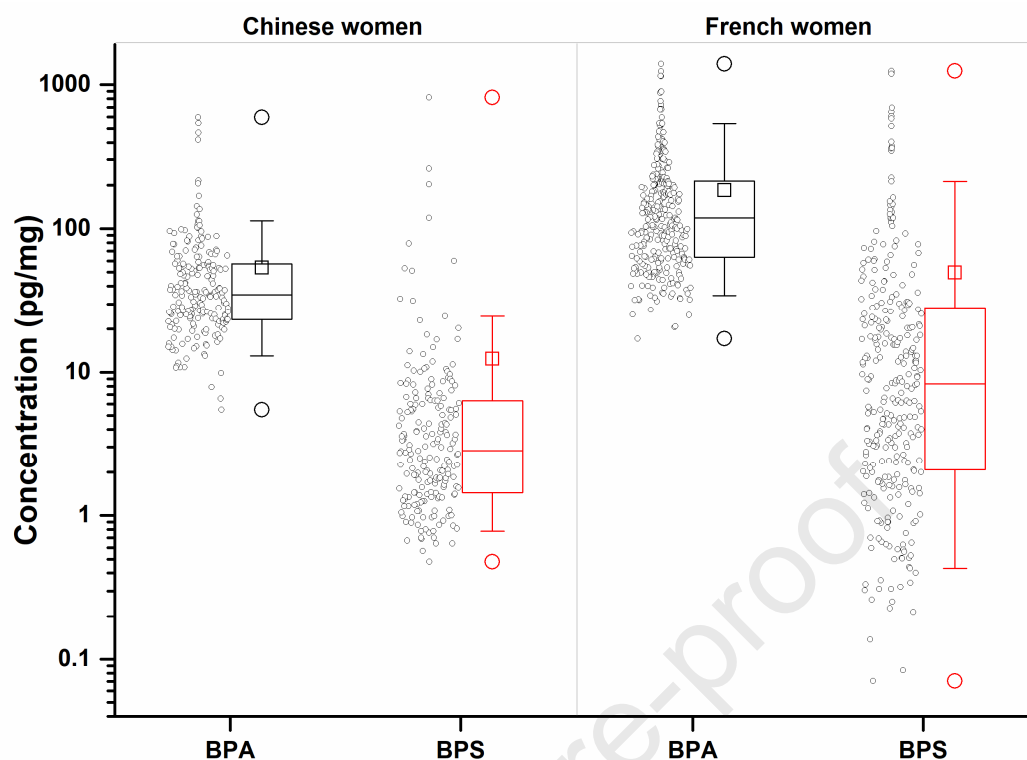


Figure 3. Sample plots of partial least squared-discriminant analyses (PLS-DA) performed on POPs (i.e., PCBs and PBDEs) determined in hair samples collected from the Chinese (n=204) and the French women (n=311). In graph a, blue circle and red diamond symbols indicate hair samples from the Chinese and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

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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.

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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)								<i>p</i> -value	LOD
	DF	Arithmetic	Min	25% P	50% P	75% P	95% P	Max	DF (%)	Arithmetic	Min	25% P	50% P	75% P	95% P	Max		
	(%)	mean								mean								
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:

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

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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)								<i>p</i> -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)								<i>p</i> -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.

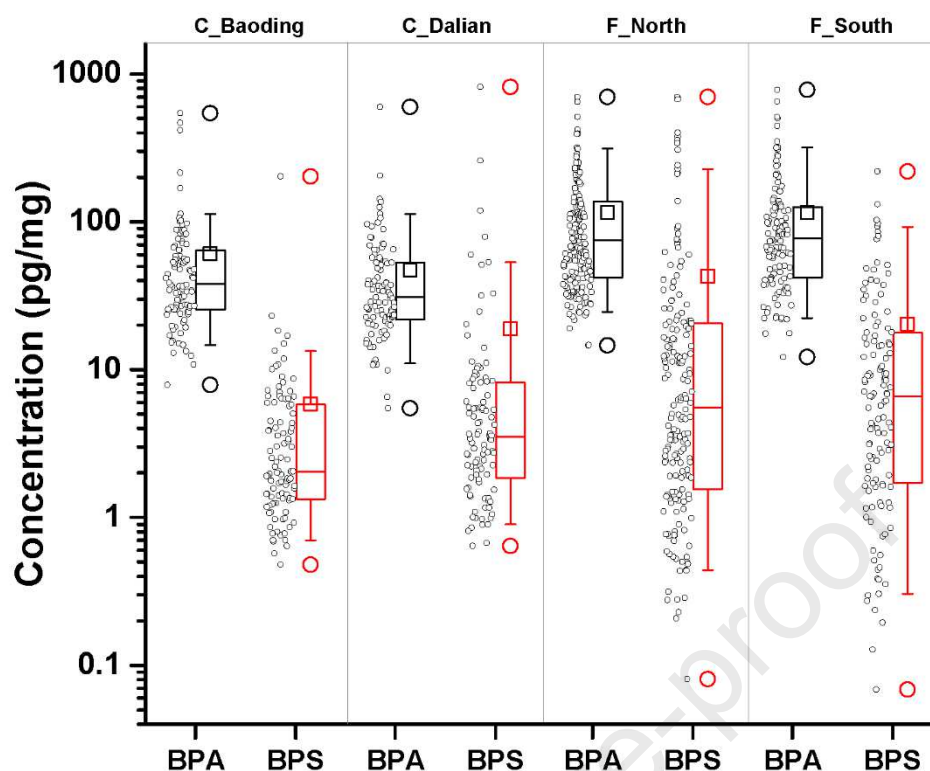


Fig. S1. Concentrations of BPA and BPS in hair samples collected from the Chinese women in Baoding (n=102), the Chinese women in Dalian (n=102), the French women in northeast (n=182), and the French women in southwest (n=129). Black circles indicate hair samples. Boxes represent 25-75 percentiles, with 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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