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Reductions in blood concentrations of persistent organic pollutants in the general population of Barcelona from 2006 to 2016

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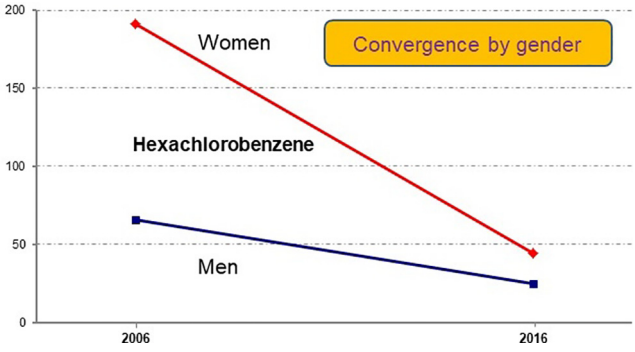
HIGHLIGHTS

- Concentrations of the nine most prevalent POPs decreased markedly during the 10-years in almost all sex, age and BMI subgroups.
- A relevant component of the success is a reduction of differences (convergence) by gender.
- For some POPs the decrease was larger in the younger groups.
- Reductions in PCBs and DDE were observed in individuals with normal weight but not in individuals with obesity.
- Barcelona is one of few cities that biomonitor POP levels through periodic health surveys representative of the population.

GRAPHICAL ABSTRACT



Abbreviations: BHS, Barcelona Health Survey; BMI, body mass index; CI, confidence interval; DDD, dichlorodiphenyldichloroethane; DDE, dichlorodiphenyldichloroethene; DDT, dichlorodiphenyltrichloroethane; GLM, General Linear Model; HCB, hexachlorobenzene; HCH, hexachlorocyclohexane; LOD, limit of detection; LOQ, limit of quantification; OCs, organochlorine compounds; OCPs, organochlorine pesticides; PAHs, polycyclic aromatic hydrocarbons; PBDEs, polybrominated diphenyl ethers; PCBs, polychlorinated biphenyls; POPs, persistent organic pollutants; TL, total serum lipids.

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ABSTRACT

Background: Few cities in the world biomonitor changes in human levels of persistent organic pollutants (POPs) through periodic health surveys representative of the general population.
Objectives: To analyze changes in serum concentrations of POPs in Barcelona from 2006 to 2016, and to analyze socio-demographic correlates of concentrations of 62 POPs in 2016.
Methods: Participants in the Barcelona Health Surveys of 2006 and 2016 ($N = 231$ and 240, respectively) were interviewed face-to-face, gave blood, and underwent a physical exam. POPs were analyzed by gas chromatography - mass spectrometry.
Results: Concentrations of all nine most prevalent compounds decreased markedly during the 10-years . Reductions occurred in almost all sex, age and BMI subgroups. For most organochlorine compounds the reduction was larger in women than men (for HCB, −77% and − 62%, respectively). For β-HCH, PCBs, naphthalene and phenanthrene the decrease was larger in the younger groups. Large reductions in concentrations of PCBs and DDE were observed in individuals with normal weight but not in individuals with obesity.
Conclusions: While concentrations of most POPs are decreasing in Barcelona, significant sociodemographic differences in such reductions warrant strengthening public and private policies towards groups making slower progress. A relevant component of the success in the current decreasing is a reduction of differences (convergence) by gender.

1. Introduction

A solid body of evidence shows that persistent organic pollutants (POPs) are highly lipophilic and degradation-resistant synthetic chemicals that bioaccumulate in the environment, food webs and living organisms. Exposed to such compounds throughout life, mostly from the ingestion of animal fats, virtually all humans store POP mixtures in adipose tissues –at highly different concentrations. Production and use of numerous POPs have been periodically banned or restricted during past decades. However, large differences in the decline or stabilization of environmental and human levels of the pollutants have been observed across populations, largely due to their physico-chemical and economic persistence. Today, human contamination and health effects from POPs remain significant (Gore et al., 2015; Henkler and Luch, 2011; Kassotis et al., 2020; LaMerrill et al., 2020; Lee et al., 2014; Marraudino et al., 2019; Peinado et al., 2020).

Biomonitoring POPs and other environmental chemical agents is thus relevant because throughout the lifecourse and history individuals and societies experience regular changes in their contamination (Demeneix, 2017; Trasande, 2019; Porta, 2018; Henríquez-Hernández et al., 2020; Porta et al., 2008). Documenting such changes by sociodemographic factors, for instance, can identify groups where regulation is less effective to prevent exposure and, thus, in whom private and public policies need strengthening (Porta, 2004). When biomonitoring shows that POP body concentrations decrease, we can see the beneficial effects of such policies (Henríquez-Hernández et al., 2020; Porta et al., 2008; Porta, 2004; Angerer et al., 2007; Schulz et al., 2011; Bjerregaard-Olesen et al., 2017; Buekers et al., 2018; Calafat, 2016; Joas et al., 2017; Dennis et al., 2017; Haines et al., 2017; Li et al., 2020; Nøst et al., 2019; Schoeters et al., 2017; Schröter-Kermani et al., 2013; Stubleski et al., 2018).

Often guided by global initiatives as the Stockholm convention and similar frameworks, an increasing number of countries and some cities in the world produce valid analyses of trends in POP levels based on studies representative of the general population (UNEP - United Nations Environment Program, 2020; Porta and Zumeta, 2002). In some instances, analyses are integrated within health surveys, which is a strength from scientific and policy standpoints (Porta et al., 2008). Barcelona is one of the few such cities. The Barcelona Health Survey (BHS) conducted by the Barcelona Agency for Public Health is the oldest of such surveys in Spain, with more editions and scientific productivity (ASPB, 2020). It is also the only study in the country based on representative samples of the general population in which blood has been collected on two time points to analyze changes in biomarkers of environmental contaminants as POPs.

In addition to the organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) analyzed in the 2006 BHS study on serum concentrations of POPs, the BHS of 2016 also included polybrominated diphenyl ethers (PBDEs) and polycyclic aromatic hydrocarbons (PAHs). PBDEs are POPs used as flame retardants, and some of them are banned (UNEP - United Nations Environment Program, 2020). PAHs are not banned; they are produced by natural and anthropogenic pyrogenic processes. The continuous presence of PAHs in the environment supports their consideration as POPs (EEA - European Environment Agency, 2019).

Therefore, the aim of the present study was two-fold: first, to analyze changes in serum concentrations of POPs in the non-institutionalized adult population of Barcelona city from 2006 to 2016; and second, to analyze socio-demographic correlates of concentrations of 62 POPs in 2016.

2. Methods

2.1. Study population and health interview survey

Methods of the Barcelona Health Survey (BHS) have been described in detail (Bartoll et al., 2018; Gasull, 2019; Porta et al., 2012a). Briefly, the BHS is a health survey that is representative of the adult non-institutionalized population of the city of Barcelona. At the end of the 2006 and 2016 BHS interviews, participants ≥15 years old were offered to take part in the study on POPs (Porta et al., 2009); the youngest persons of the 2006 and 2016 BHS who actually participated in the POP study were 18 and 19 years old, respectively. Subsequently, a nurse personally interviewed again each person who accepted to participate in the POP study, measured the weight, height, and the hip and waist circumference, and collected blood and urine samples. Participants had been asked to fast for at least eight hours before blood extraction. As in 2006, in 2016 blood was collected in a vacuum system tube and centrifuged for 15 min × 3000 rpm at 4 °C to obtain serum. Right after centrifugation, serum was divided in 1–3 mL aliquots and stored at −80° until 2018–2019, when POP concentrations were analyzed. The additional interview for the POPs study included structured questions about recent and past changes in body weight (last 6 months), and on whether the person had been breastfed. Women were also asked about parity, breastfeeding their children, and abortion histories.

The average age of the 240 participants of the 2016 BHS was 50 years, and 53% were women (Supplemental Table 1). Body mass index (BMI) was computed by measured weight [kg] divided by measured height squared [m^2]. 16% of participants were obese (BMI ≥ 30 kg/m^2), 85% were breastfed in childhood, 23% were born

outside Spain, 90% had completed at least primary schooling, and 31% were from occupational social classes IV or V (Supplemental Table 1). While occupational and educational indicators were slightly more favorable for women, there were no significant differences in the distribution of age, having been breastfed in childhood, birth place, educational level, social class and employment status by sex. However, a slightly greater proportion of women than men had a normal weight (47% vs. 41%, respectively, $p < 0.09$). The most relevant differences between participants in the 2016 BHS and the 231 participants in the 2006 BHS (Porta et al., 2012a; Porta et al., 2009) were birth place, social class, educational level, and employment status (Supplemental Table 1). Therefore, such differences will subsequently be controlled through multivariate adjustment (Section 2.3. Statistical analyses). The Ethics Committee of the Parc de Salut Mar approved the study protocols, and all participants signed an informed consent before completing questionnaires at baseline (Porta et al., 2009).

2.2. Analytical chemical methods

Analyses of serum samples of the 2016 BHS were carried out in the Research Institute of Biomedical and Health Sciences (IUIBS) in the University of Las Palmas de Gran Canaria, Spain. Serum concentrations of 62 compounds were measured (Supplemental Table 2 and Supplemental Fig. 1). Of these, 38 were organochlorine compounds (OCs): 20 were organochlorine pesticides (OCPs): *o,p'*-dichlorodiphenyltrichloroethane (DDT), *p,p'*-DDT, *o,p'*-dichlorodiphenyldichloroethene (DDE), *p,p'*-DDE, *o,p'*-dichlorodiphenyldichloroethane (DDD), *p,p'*-DDD, hexachlorobenzene (HCB), hexachlorocyclohexane (isomers α-, β-, γ-, and δ-HCH), aldrin, endrin, dieldrin, endosulfan (α-, and β-isomers), endosulfan sulphate, mirex, methoxychlor, and heptachlor; the OCs analyzed included as well 18 polychlorinated biphenyl (PCB) congeners: marker-PCBs (IUPAC numbers 28, 52, 101, 138, 153, and 180), and dioxin-like PCBs (congeners 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189). Analyses included also 8 polybrominated diphenyl ethers (PBDEs): the tribrominated congener BDE-28, the tetrabrominated BDE-47, the pentabrominated BDE-85, -99 and -100, the hexabrominated BDE-153 and -154, and the heptabrominated BDE-183; and the 16 most environmentally relevant polycyclic aromatic hydrocarbons (PAHs) listed by the United States Environmental Protection Agency (Keith and Telliard, 1979; Loibner et al., 2004) (Supplemental Table 2 and Supplemental Fig. 1).

In serum samples of the 231 participants of the 2006 BHS we analyzed the 8 PBDEs –congeners #28, 47, 85, 99, 100, 153, 154, and 183– and the 16 PAHs not analyzed in the previous study (Porta et al., 2012a) (Supplemental Table 3 and Supplemental Fig. 1): naphthalene, acenaphthene, acenaphthylene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, dibenzo[*a,h*]anthracene, benzo[*ghi*]perylene and indeno[1,2,3,-*cd*]pyrene (Luzardo et al., 2019). Serum samples from the 2006 BHS had been stored at IMIM frozen at −80 °C, with no incidents.

In addition, in 2018–2019 the laboratory at IUIBS analyzed the concentrations of the 18 POPs previously analyzed by CSIC in the 231 subjects from the 2006 BHS (Porta et al., 2012a) (Supplemental Fig. 1). IUIBS did so as a methodological control in samples of 30 (13%) such individuals, since it was not justified to repeat the analyses of these 18 POPs for all 231 subjects (biological samples from a population sample that is representative of the general population are particularly precious). To compare the concentrations of the compounds analyzed in 2016 and 2006 we computed by linear regression a conversion factor from values obtained in the above-mentioned 30 individuals of the 2006 BHS. The conversion factor was applied to concentrations of the 18 POPs previously analyzed in the 231 participants of 2006 BHS (Supplemental Fig. 1 and Supplemental Methods). After applying the conversion factor to the concentrations of 2006, we applied to these concentrations the same LODs and LOQs used by IUIBIS to the samples of 2016.

2.2.1. Sample preparation and instrumentation

Half-milliliter aliquots of serum samples were mixed with 0.4 mL of water/n-propanol (85:15) and subsequently centrifuged at 3000 rpm for 5 min. Then, 0.1 mL of acetic acid was added to each sample and loaded to 200 mg (3 mL) Chromabond® C18ec columns (Macherey-Nagel, Germany) mounted in a vacuum manifold (Waters Corporation, USA). The columns were previously conditioned with 2 × 1 mL methanol followed by 2 × 1 mL isopropanol:water (15:85). After passing the samples the columns were washed with 1 mL of isopropanol:water (15:85), and to a drying thereof under vacuum for 30 min. Finally, the analytes were eluted with 1 mL of dichloromethane. Briefly, we employed a Gas Chromatography (GC) System 7890B equipped with a 7693 Autosampler (Agilent Technologies, Palo Alto, CA, USA) for gas chromatographic separations. Two fused silica ultra-inert capillary columns Agilent J&WHP-5MS (Crosslinked 5% phenylmethylpolysiloxane, Agilent Technologies) each with a length of 15 m, 0.25 mm i.d., and a film thickness of 0.25 μm were connected in series and used as the stationary phase. Both columns were connected by a Purged Ultimate Union (PUU; Agilent Technologies). Helium (99.999%) at a constant flow rate of 1.0 mL/min for column 1 was used as the carrier gas. A back-flushing technique was incorporated to the GC. The oven temperature program was programmed as follows: a) an initial temperature of 60 °C held for 1 min; b) increase to 170 °C at a rate of 40 °C/min; c) increase to 310 °C at a rate of 10 °C/min with 3 min hold time; and d) cool down to 60 °C. Injector and transfer line were set at 280 °C. Standards and samples were injected (1 μL) in the splitless mode using a 4-mm ultrainert liner with glass wool (Agilent Technologies). The detection of the analytes was performed using a Triple Quad 7010 mass spectrometer (Agilent Technologies, Palo Alto, CA, USA). The quantification was done using point calibration curves, which were constructed using a least-squares linear regression from the injection of standard solutions ranging from 0.025 to 25 ng/mL. The limits of quantification (LOQ) were 0.03 to 0.1 ng/mL (Supplemental Table 2). The details of validated chromatographic method and quality control have been previously reported (Luzardo et al., 2019; Henríquez-Hernández et al., 2017).

To be conservative, the main statistical analyses were limited to compounds that were detected above the detection limit in >75% of participants of BHS 2016. The limits of detection (LODs) for the 11 compounds included in the main analyses ranged from 0.009 to 0.021 ng/mL, and the LOQs from 0.030 to 0.070 ng/mL (Supplemental Table 2). When the assigned value of an analyte was below the LOD of IUIBS, it was assigned the mid-value of this limit; and when a POP concentration was between the LOD and the LOQ, the mid-value between LOD and LOQ was used (Porta et al., 2012a; Porta et al., 2009; Porta et al., 2010).

Concentrations of total cholesterol and triglycerides were determined enzymatically, using serum obtained at the same time as the serum used for POP analyses. Total serum lipids (TL) were calculated by the standard formula 2 of Phillips et al. (Gasull, 2019; Porta et al., 2012a). POP concentrations were individually corrected for TL and are expressed in nanograms of analyte per gram lipid (ng/g of lipid). Among the 240 participants in the 2016 study, mean (standard deviation) serum concentrations of total cholesterol, triglycerides and TL were 193.4 (40.3), 95.9 (65.5) and 597.3 (132.8) mg/dL, respectively.

2.2.2. Quality of analyses and quality control (QA/QC)

All measurements were performed in triplicate, and the geometric mean was used for the calculations. In each batch of samples, three controls were included for every 18 vials: a reagent blank consisting of a vial containing only cyclohexane; a vial containing 2 ng/mL of each of the pollutants in cyclohexane; and an internal laboratory quality control sample (QC) consisting of melted butter spiked at 10 ng/mL of each of the analytes, which was processed using the same method of extraction as the serum samples. The results were considered to be acceptable when the concentration of the analytes determined in the QC sample was within 15% of the

deviation of the theoretical value. Further details on quality of analyses and quality control (QA/QC) were provided previously (Cabrera-Rodríguez et al., 2019).

2.3. Statistical analyses

Univariate statistics were computed as customary (Kleinbaum et al., 2007). Kruskal-Wallis' test and Mann-Whitney's *U* test were used to assess differences in concentrations of POPs by sociodemographic characteristics of the participants. The Kolmogorov-Smirnov test for normality was used to check the distributions of POPs; as none was normal, log-transformed values were used in regression analyses. The selection of a representative sample of the non-institutionalized population in each BHS followed a complex design (Bartoll et al., 2018; Gasull, 2019; Porta et al., 2012a). In order to guarantee that the demographic structure of the population is preserved with respect to age and gender, and thus the representativeness of the study population with respect the general population of Barcelona, sampling weights were used in the statistical analyses of the present study.

We calculated the number of POPs detected in each person at high concentrations (nPhc) in the two BHS studies as follows: for each subject we added the number of POPs whose respective serum concentrations were equal to or greater than the percentile 75 of each POP distribution in the 2006 BHS (Porta et al., 2012a; Porta et al., 2012b; Pumarega et al., 2016). In the analyses we included the 9 POPs that had been detected (each) in >70% of the study subjects of 2006 BHS (Table 1).

Generalized Linear Models (GLM) were used to analyze variations in lipid-corrected and log-transformed concentrations of POPs (Kleinbaum et al., 2007). The main effects of all predictors were independently explored in base models (Lash et al., 2021). Sociodemographic differences between participants in the 2016 and 2006 BHS were also controlled with GLM to compute geometric means adjusted for the relevant confounders. The level of statistical significance was set at 0.05 and all tests were two-tailed. The statistical significance, the precision of the estimates, and the magnitude of the effects were all taken into consideration to assess significance (Lash et al., 2021; Amrhein et al., 2019). Statistical analyses were conducted using SPSS version 22.0.0.0 (IBM SPSS Statistics, Armonk, NY, USA, 2013), R version 3.5.2 (2018), and Stata 15.0.

3. Results

Concentrations of all nine most prevalent compounds decreased markedly during the 10-year study period. This was so for all four main measures: lipid-adjusted (ng/g lipid) and crude (ng/ml) concentrations assessed by medians and by adjusted geometric means, with most changes from about −30% to −80% or more (all $p < 0.05$) (Table 1 and Fig. 1).

Table 1
Change in serum concentrations of persistent organic pollutants in the general population of Barcelona, 2006 to 2016.

	Barcelona Health Survey of 2006 ($N = 231$)			Barcelona Health Survey of 2016 ($N = 240$)			Change 2006–2016		
	% > LOD	Median (P25-P75)	aGM (CI 95%)	% > LOD	Median (P25-P75)	aGM (CI 95%)	% > LOD (*p*-value)[a]	Medians (%)[b]	aGM (%)[c]
p,p'-DDE									
ng/g lipid		323.1 (142.0–977.5)	379.7 (330.3–436.5)		177.9 (98.9–549.3)	226.8 (197.7–260.2)	−0.4	−45	−40
ng/ml	100.0	2.13 (0.86–6.53)	2.37 (2.06–2.73)	99.6	1.02 (0.55–3.48)	1.33 (1.16–1.52)	(>0.999)	−52	−44
HCB									
ng/g lipid		111.0 (41.5–313.8)	119.6 (108.0–132.4)		25.5 (15.6–70.7)	34.9 (31.6–38.6)	+1.2	−77	−71
ng/ml	98.4	0.75 (0.24–2.08)	0.75 (0.68–0.83)	99.6	0.16 (0.09–0.43)	0.20 (0.18–0.23)	(0.208)	−79	−73
β-HCH									
ng/g lipid		69.6 (35.9–220.4)	89.8 (80.0–100.8)		17.3 (7.08–52.7)	20.1 (18.0–22.5)	−5.2	−75	−78
ng/ml	96.9	0.48 (0.20–1.51)	0.56 (0.50–0.63)	91.6	0.11 (0.03–0.32)	0.12 (0.10–0.13)	(0.016)	−78	−79
PCB 118									
ng/g lipid		12.5 (9.30–27.4)	15.5 (13.7–17.6)		1.71 (1.28–15.5)	3.96 (3.50–4.47)	−41.6	−86	−74
ng/ml	82.5	0.06 (0.06–0.17)	0.09 (0.09–0.11)	40.9	0.01 (0.01–0.09)	0.02 (0.02–0.03)	(<0.001)	−87	−76
PCB 138									
ng/g lipid		59.6 (35.0–107.6)	56.9 (51.8–62.6)		40.1 (21.4–74.8)	36.9 (33.7–40.6)	−0.3	−33	−35
ng/ml	97.8	0.38 (0.19–0.68)	0.36 (0.32–0.39)	97.5	0.24 (0.12–0.44)	0.22 (0.20–0.24)	(>0.999)	−38	−39
PCB 153									
ng/g lipid		81.0 (48.4–144.2)	77.8 (70.7–85.6)		64.6 (31.6–119.6)	59.0 (53.7–64.9)	+1.4	−20	−24
ng/ml	97.8	0.52 (0.27–0.97)	0.49 (0.44–0.54)	99.2	0.38 (0.18–0.70)	0.35 (0.31–0.38)	(0.277)	−27	−29
PCB 180									
ng/g lipid		80.7 (45.3–124.9)	72.1 (64.9–80.1)		64.6 (24.2–121.1)	46.4 (41.8–51.4)	−3.0	−20	−36
ng/ml	98.1	0.53 (0.25–0.82)	0.45 (0.41–0.50)	95.1	0.37 (0.13–0.73)	0.27 (0.24–0.30)	(0.073)	−31	−40
Naphthalene									
ng/g lipid		294.9 (27.5–444.9)	80.5 (60.5–107.2)		1.71 (1.26–81.3)	8.16 (6.16–10.8)	−37.1	−99	−90
ng/ml	82.5	2.03 (0.20–2.61)	0.50 (0.38–0.67)	45.4	0.01 (0.01–0.47)	0.05 (0.04–0.06)	(<0.001)	−100	−91
Phenanthrene									
ng/g lipid		115.4 (2.34–206.5)	39.3 (30.7–50.5)		51.4 (6.61–101.4)	26.2 (20.5–33.4)	+2.9	−55	−33
ng/ml	73.2	0.70 (0.01–1.26)	0.25 (0.19–0.31)	76.2	0.30 (0.05–0.57)	0.15 (0.12–0.20)	(0.459)	−57	−38

The Table shows results for the 9 POPs that had been detected (each) in >70% of the study subjects of the 2006 BHS.
LOD: limit of detection. % > LOD: percent of subjects with concentrations above the LOD. P25, P75: 25th, and 75th percentile, respectively. aGM: Geometric mean adjusted for age, sex and body mass index. CI: confidence interval. Concentrations of the seven first compounds in the 2006 Barcelona Health Survey (BHS) reflect the conversion factor explained in the Methods section.

[a] Difference in percentage points between the 2006 and 2016 BHS in the percent of subjects with concentrations above the limit of detection (Fisher's exact test, two-tail test).

[b] Percent change in the median of the 2006 and 2016 BHS; the differences between such pairs of medians were statistically significant (*p*-value <0.05, Mann-Whitney's *U* test, two-tail) for all 9 compounds: for HCB, β-HCH, PCBs 118, and 138, naphthalene and phenanthrene the *p*-values were ≤ 0.001.

[c] Percent change in the aGM of the 2006 and 2016 BHS; the differences between such pairs of aGM were statistically significant (*p*-value ≤0.001, Wald test) for all 9 compounds.

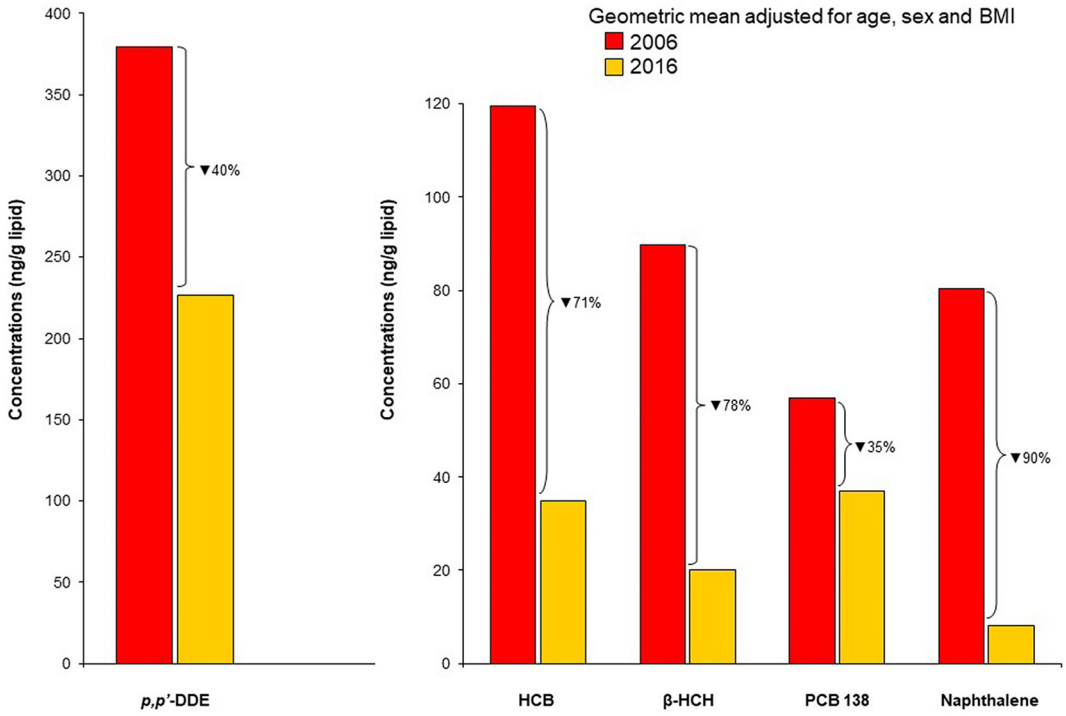

Fig. 1. Change from 2006 to 2016 in the geometric mean (adjusted for age, sex and BMI) of the serum concentration of *p,p'*-DDE, HCB, β-HCH, PCB 138, and naphthlene in the corresponding representative samples of inhabitants of the city of Barcelona. *Footonote:* See figures in the last right column of Table 1.

Many POPs were still detected in 2016, and the change in the % > LOD was only significant (e.g., larger than −35% and $p < 0.001$) for two of the nine compounds (PCB 118 and naphthalene) (Table 1). Nevertheless, in 2016 *p,p'*-DDE, HCB, β-HCH, PCBs 138, 153, 156 and 180, fluorene, phenanthrene, pyrene and fluoranthene were detected in >75% of the population of Barcelona, whereas in 2006 PCB 118 and naphthalene were also detected in >75% of participants; by contrast, in 2006 fluorene, pyrene and fluoranthene were not detected ($p < 0.001$ in the % > LOD for all five compounds) (Table 1 and Supplemental Tables 2 and 3).

Endrin, methoxychlor, *o,p'*-DDE and γ-HCH were not detected in any participant in 2016; *o,p'*-DDT, *p,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, α- and β-endosulfan, heptachlor, α- and δ-HCH, and seven of the 18 PCBs were detected in <6% of participants (Supplemental Tables 2 and 3).

Seven of the 8 PBDEs and 8 of the 16 PAHs were detected in ≤7.5% of individuals in both 2006 and 2016. BDE-153 and acenaphthylene were detected >20 percentage points more in 2016 than in 2006 (both $p < 0.001$) (Supplemental Tables 2 and 3). Mixtures found in human bodies change incessantly.

Happily, in the 10-year study period, substantial reductions in concentrations (most, greater than 30%, and up to 80% and 90%) occurred in almost all sex, age and BMI subgroups. For instance, from 2006 to 2016 there was a 77% decrease in the concentrations of HCB among women, or a reduction of 91% for naphthalene among men (Table 2). For most OCs the reduction was larger in women than in men; this was so, specifically, for β-HCH (−80% vs. –74% in women and men, respectively), *p,p'*-DDE, HCB, and PCBs 138, 153 and 180 (Table 2). However, in 2016 women still had significantly higher median

Table 2
Percent change from 2006 to 2016 in the adjusted geometric mean of serum concentrations of persistent organic pollutants (ng/g lipid) in the population of the city of Barcelona, by sex, age, and body mass index.

	Sex[a]		Age[b]			Body Mass Index[c]		
	Men	Women	18–44 years	45–64 years	≥65 years	Normal weight	Over-weight	Obese
p,p'-DDE	−38*	−42*	−38*	−55*	−18	−54*	−25	−34
HCB	−62*	−77*	−63*	−77*	−72*	−69*	−75*	−73*
β-HCH	−74*	−80*	−80*	−77*	−72*	−81*	−79*	−67*
PCB 118	−78*	−71*	−80*	−73*	−61*	−82*	−76*	−44*
PCB 138	−30*	−39*	−44*	−30*	−20	−49*	−33*	5
PCB 153	−16	−31*	−31*	−20*	−14	−38*	−25*	26
PCB 180	−32*	−39*	−56*	−12	−10	−47*	−40*	21
Naphthalene	−91*	−88*	−94*	−89*	−74*	−92*	−84*	−91*
Phenanthrene	−33	−34	−57*	−18	27	−45*	15	−58*

The Table shows results for the 9 POPs that had been detected (each) in >70% of the study subjects of the 2006 Barcelona Health Survey study on serum concentrations of POPs.
[a] Models adjusted for age and body mass index.
[b] Models adjusted for sex and body mass index.
[c] Models adjusted for age and sex.
* *p*-value <0.05 (Wald test) for the percent change from 2006 to 2016 in each stratum; e.g., the 42% decrease from 2006 to 2016 in the concentrations of *p,p'*-DDE among women was statistically significant (*p*-value <0.05).

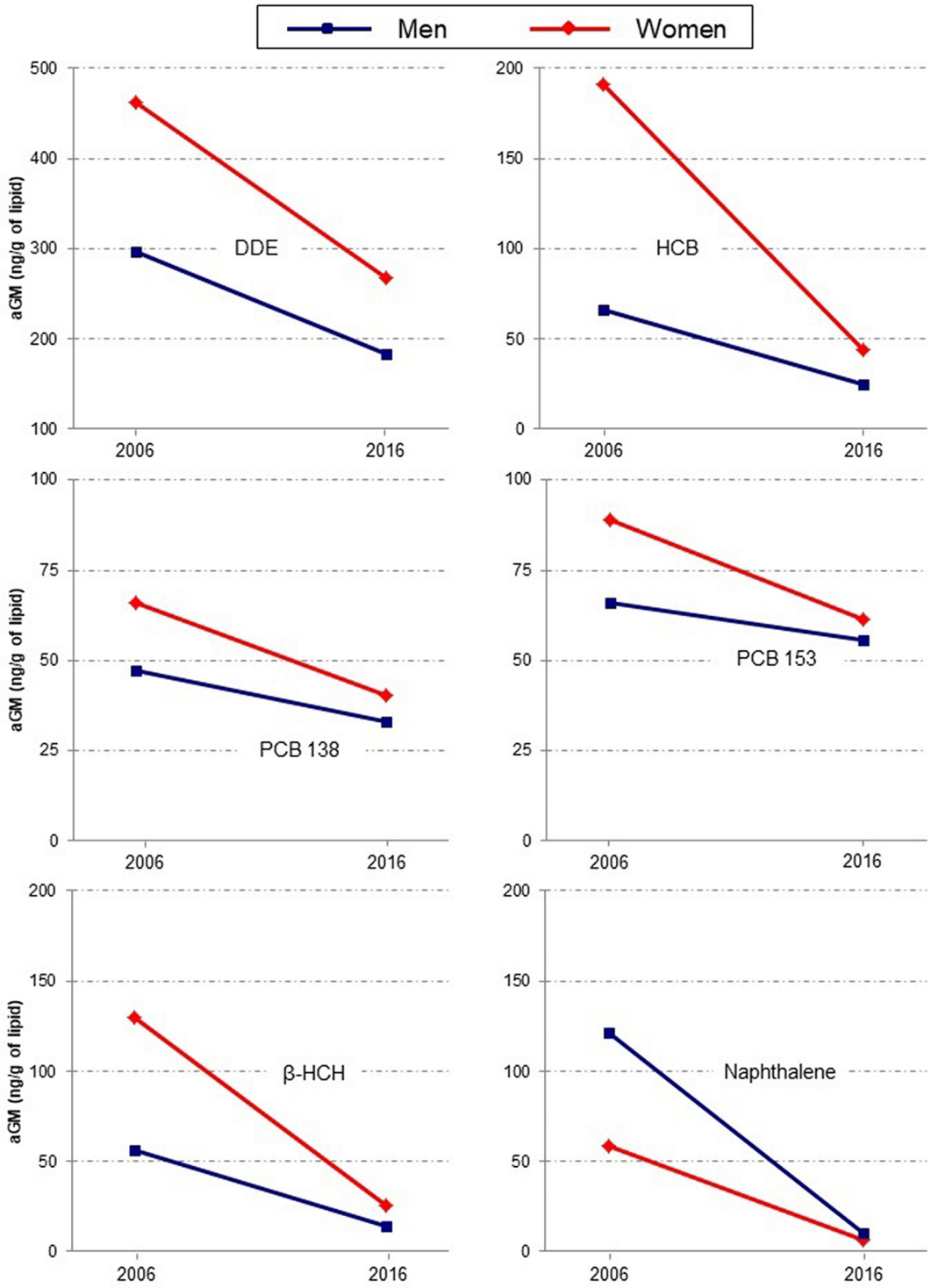

Fig. 2. Convergence by gender: Change from 2006 to 2016 in the geometric mean (adjusted for age and BMI) of the serum concentrations (in ng/g lipid) of six persistent organic pollutants in the population of the city of Barcelona by gender. *Footnote:* The figure shows a selection of the results included in the first two columns of Table 2. The geometric means of each Barcelona Health Survey were adjusted for age and body mass index of the two studies simultaneously.

concentrations than men of DDE, HCB and β-HCH. Table 2 and Fig. 2 show the reduction of differences (convergence) by gender.

No compound showed a larger decrease in the 10 years in the oldest than in the younger groups, whereas for β-HCH, PCBs, naphthalene and phenanthrene the decrease was larger in the younger groups, often with monotonic trends (Table 2).

Larger reductions in concentrations of PCBs and DDE were observed in individuals with normal weight and overweight than in the obese. Among the latter, concentrations of PCBs 138, 153 and 180 did not change or even increased non-significantly from 2006 to 2016 (Table 2).

In 2006, 33.4% of participants had concentrations of 3 or more POPs at high concentrations; this figure was 15.9% in 2016, a remarkable absolute reduction of 17.4 percentage points, and a 52% relative reduction. The number of participants with 6 or more POPs at high concentrations was 13.7% in 2006 and 4.0% in 2016, a difference of 9.6 percentage points, and a 71% relative reduction (Table 3).

Table 3
Absolute and relative change from 2006 to 2016 in the prevalence of subjects with POPs at high concentrations.

Number of POPs at high concentrations	2006 (N = 231)			2016 (*N* = 240)			Cumulative number of POPs	2006 (%)	2016 (%)	2016 vs. 2006	
	N	(%)	Cumulative percent	N	(%)	Cumulative percent				Absolute difference[a]	Relative difference[b]
0	74	(31.8)	31.8	136	(56.6)	56.6					
1	42	(18.1)	50.0	41	(17.1)	73.7	≥1	68.2	43.4	−24.7	−36.4%
2	39	(16.8)	66.8	25	(10.5)	84.1	≥2	50.0	26.3	−23.7	−47.4%
3	17	(7.5)	74.2	8	(3.5)	87.7	≥3	33.2	15.9	−17.4	−52.1%
4	10	(4.3)	78.6	10	(4.3)	92.0	≥4	25.8	12.3	−13.4	−52.3%
5	18	(7.8)	86.3	10	(4.0)	96.0	≥5	21.4	8.0	−13.4	−62.6%
6	10	(4.3)	90.6	2	(0.9)	96.9	≥6	13.7	4.0	−9.6	−70.8%
7	18	(7.9)	98.5	5	(1.9)	98.8	≥7	9.4	3.1	−6.3	−67.0%
8	3	(1.5)	100.0	0	(0.0)	98.8	≥8	1.5	1.2	−0.3	−20.0%
9	0	(0.0)	100.0	3	(1.2)	100.0	9	0	1.2	+1.2	––

Based on concentrations above percentile 75 of the nine most frequently detected POPs in the 2006 Barcelona Health Survey study of POPs (see Table 1, Table 2, and Section 2.3. Statistical analyses).
N, number of subjects.
[a] Absolute difference from 2006 to 2016 (in percentage points) of the percent of subjects with POPs at high concentrations (>P75).
[b] Relative difference or change from 2006 to 2016 (in %) of the percent of subjects with POPs at high concentrations (>P75).

We next focus exclusively on findings in 2016. Concentrations of OCPs and PCBs increased with increasing age (Tables 4 and 5). This was not so for PAHs, neither in women nor in men. In men, most POP concentrations tended to increase with increasing BMI, with the exception of PAHs, which were lower in obese men (Table 4). In women, adjusting for age, concentrations of all OCPs increased with increasing BMI, whereas this pattern was not apparent for PCBs, which showed lower levels in women who were overweight (Table 5).

Concentrations of OCPs and PCBs were slightly higher in men who lost 4–5 kg in the previous 6 months. In men, concentrations of all compounds were non-significantly lower in participants who reported being breastfed as a child (Table 4); in women this was also so for most POPs (Table 5). Concentrations of all compounds were non-significantly lower in women who breastfed their children longer.

In men, we mutually adjusted BMI and weight-change (during previous 6 months) to assess whether concentrations were highest in subjects with high BMI and higher weight-loss. This was so for DDE, HCB, β-HCH, and the four PCBs, with figures similar to those already shown in Table 4. These associations were not apparent for PAHs. In women, we further adjusted for duration of lactation. Only concentrations of DDE were highest in women with high BMI and higher weight-loss. Figures for the other POPs were similar to those already shown in Table 5.

Men and women born outside Spain showed higher concentrations of *p,p'*-DDE and lower concentrations of the rest of OCs. Men born abroad also had lower concentrations of fluorene.

No simple patterns were apparent for occupational social class. In men (Table 4), no compounds showed statistically significant differences in the geometric means adjusted for age (aGM). Concentrations of PCBs were often higher in men and women of the more affluent social classes I-II but, again, none was statistically significant. Higher concentrations in classes IV-V (less affluent) were only apparent for *p,p'*-DDE, fluorene, phenanthrene and pyrene in men, but none was statistically significant. In men the aGM was never higher in class III than in the other class categories. In women (Table 5), concentrations of most POPs were similar across classes; they were almost never higher in classes IV-V than in classes I-II; and concentrations of *p,p'*-DDE, HCB, β-HCH, and PAHs were highest in class III (statistically significant for HCB and pyrene).

4. Discussion

During the 10-year period from 2006 to 2016, there were marked reductions in blood concentrations of most POPs across most population subgroups defined by gender, age, and BMI. The largest decreases were seen for OCPs (>70% for concentrations of HCB and β-HCH), followed by PCBs (>70% for PCB 118 and >20% for the other three most detected PCBs). Reductions were more marked among women, younger individuals, and people who were not obese. We found remarkable equality of POP levels in the different social classes.

Slightly larger reductions of concentrations in women than men have been previously reported (Fernyhough et al., 1999; Partearroyo et al., 2019). Perhaps women reduced more the intake of dietary animal fats. Also, concentrations of all compounds were lower in women having breastfed their children longer (Song et al., 2018). Nevertheless, in 2016 women still had significantly higher concentrations than men of DDE, HCB and β-HCH, as also observed before (Zubero et al., 2017; Carreño et al., 2007). Hence, a relevant component of the success seen in the current decreasing human concentrations of POPs in Barcelona appears to be a convergence of such concentrations between women and men (i.e., a reduction in gender differences). Available studies could be reanalyzed to formally assess this possibility while accounting for confounders; hence, to identify common and separate causes of the decrease –by gender.

Both in women and men concentrations of OCs (OCPs and PCBs) increased with age, as often reported. The observation probably results from age-cohort-period effects (Porta, 2018; Porta et al., 2008; Porta, 2004; Quinn and Wania, 2012; Gasull et al., 2013). In fact, no compound showed a larger decrease from 2006 to 2016 in the oldest than in the younger groups, whereas for β-HCH, PCBs, naphthalene and phenanthrene the decrease was larger in the younger groups, mostly with monotonic trends. This is also excellent news. Younger cohorts have probably been less exposed to OCs because of policies operating during recent periods (Porta et al., 2008; Porta, 2004; Porta, 2012). By contrast, the decrease in the concentrations of these highly persistent pollutants was less marked among older individuals, whom obviously had more years of exposure and (influenced as well by the higher OC contamination in their specific historical period), higher body concentrations. But the observation is also likely due to age-related physiological factors and behaviors, such as decreased detoxification activity of liver enzymes (Sotaniemi et al., 1997), and higher consumption of POP-rich foods by older individuals (Porta, 2018; Henríquez-Hernández et al., 2020; Porta et al., 2008; Partearroyo et al., 2019; Gasull et al., 2013).

Larger reductions in concentrations of PCBs and DDE were observed in individuals with normal weight and overweight than in the obese; among the latter, concentrations of some PCBs did not change or even increased from 2006 to 2016. These findings reinforce reasons to fight obesity through individual behavior and policies, and suggest that reductions in internal POP concentrations are less likely among the obese even when levels decrease in society. In agreement, it has been

Table 4
Serum concentrations of POPs (ng/g lipid) (compounds detected in over 75% of the population of Barcelona city) in 2016 by sociodemographic characteristics, men.

	p,p'-DDE		HCB		β-HCH	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
All men	145.4 (87.6–355.0)	189.5 (154.6–232.2)	23.2 (13.9–44.6)	25.4 (21.5–29.9)	10.8 (6.0–35.9)	15.9 (13.4–18.9)
Age (years)						
18–44	103.0 (73.2–151.6)*	112.4 (83.8–150.7)	15.4 (10.5–24.2)*	15.8 (12.6–20.0)	6.6 (5.3–8.3)*	6.6 (5.1–8.4)
45–64	176.8 (112.0–377.6)	176.8 (124.1–251.9)	23.9 (13.1–43.0)	22.7 (17.2–30.1)	17.6 (8.6–36.5)	16.2 (12.0–21.8)**
≥65	543.3 (183.6–910.2)	456.0 (292.2–711.8)**	55.0 (30.3–84.3)	61.0 (42.9–86.8)**	52.8 (36.0–123.0)	59.5 (40.9–86.6)**
Body Mass Index						
Normal	94.4 (49.4–145.9)*	122.6 (87.4–172.1)	15.2 (10.2–30.8)*	26.5 (20.0–35.2)	7.8 (5.6–11.3)*	17.0 (12.7–22.7)
Overweight	268.8 (130.1–815.2)	249.7 (185.6–336.1)**	23.2 (15.3–49.7)	21.4 (16.7–27.3)	17.7 (5.9–39.0)	13.1 (10.1–16.9)
Obese	275.5 (128.2–491.5)	231.1 (142.4–375.0)**	39.3 (26.1–68.8)	36.5 (24.4–54.5)	26.5 (18.3–55.1)	23.2 (15.3–35.2)
Weight change in last 6 months						
Lost 4–5 Kg	200.3 (91.4–1417)	386.9 (192.6–777.2)**	18.0 (13.6–54.4)	30.5 (17.1–54.3)	8.9 (5.1–38.9)	18.5 (10.2–33.8)
Little or no change (± 3 Kg)[a]	151.6 (89.2–455.3)	183.0 (147.3–227.4)	23.5 (13.9–45.8)	25.3 (21.1–30.3)	10.8 (6.1–35.9)	15.8 (13.1–19.1)
Gained 4–5 Kg	129.0 (72.0–138.6)	134.9 (67.1–271.2)	18.7 (9.3–26.4)	22.0 (12.3–39.3)	12.4 (4.7–18.3)	14.6 (8.0–26.7)
Was breastfed						
No	139.9 (117.4–555.8)	228.8 (138.5–377.8)	24.4 (18.5–53.5)	35.5 (23.4–53.7)	8.1 (6.8–35.4)	19.2 (12.4–29.7)
Yes	151.6 (87.4–355.0)	185.0 (146.4–233.7)	22.2 (13.3–46.3)	24.8 (20.4–30.1)	12.1 (6.1–36.2)	15.7 (12.9–19.3)
Birth place						
Catalonia	146.6 (86.5–347.8)	169.6 (133.4–215.5)	27.6 (15.3–49.0)*	35.5 (23.4–53.7)	17.1 (7.4–36.2)*	17.3 (14.1–21.3)
Rest of Spain	138.3 (74.2–306.3)	152.7 (87.1–267.7)	24.7 (10.7–67.8)	27.9 (17.9–43.5)	16.6 (5.6–54.5)	14.8 (9.1–24.0)
Abroad	148.6 (92.7–531.2)	324.5 (206.9–508.8)**	11.9 (7.0–18.1)	14.2 (9.9–20.3)**	6.5 (4.3–9.3)	12.2 (8.3–18.0)
Occupational social class						
I-II (more affluent)	132.0 (74.3–473.6)	169.9 (127.9–225.8)	132.0 (74.3–473.6)	28.1 (22.3–35.4)	9.1 (5.6–46.5)	16.0 (12.6–20.4)
III	136.7 (82.5–206.8)	179.5 (115.7–278.3)	136.7 (82.5–206.8)	24.7 (17.3–35.3)	10.6 (6.6–22.9)	16.2 (11.2–23.5)
IV-V (less affluent)	183.8 (121.5–531.2)	238.5 (163.5–347.8)	183.8 (121.5–531.2)	21.7 (15.9–29.4)	15.1 (6.7–36.2)	15.5 (11.2–21.3)

	PCB 138		PCB 153		PCB 156		PCB 180	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Men	39.2 (21.3–75.3)	36.4 (30.9–42.8)	64.9 (30.3–122.9)	60.9 (52.3–70.9)	7.4 (4.9–14.3)	7.2 (6.1–8.4)	54.7 (24.7–134.7)	53.2 (44.1–64.2)
Age (years)								
18–44	21.4 (8.1–28.6)*	16.0 (12.6–20.4)	32.4 (18.4–47.9)*	29.1 (23.2–36.5)	5.2 (1.5–7.0)*	3.9 (3.1–4.8)	25.3 (8.4–32.6)*	18.3 (13.9–24.0)
45–64	56.6 (34.4–76.3)	47.8 (35.7–64.0)**	91.7 (60.0–137.4)	77.2 (58.8–101.4)**	10.6 (5.2–16.4)	8.4 (6.5–10.9)**	112.1 (71.4–145.1)	85.5 (61.4–118.9)**
≥65	82.7 (74.9–129.2)	86.4 (59.9–124.7)**	131.1 (113.8–202.8)	134.4 (95.4–189.3)**	16.0 (11.0–26.2)	15.9 (11.5–21.9)**	152.4 (122.9–184.2)	141.5 (93.5–214.4)**
Body Mass Index								
Normal	22.3 (13.5–47.6)*	34.9 (26.3–46.3)	37.7 (21.3–75.3)*	58.2 (44.7–75.8)	5.7 (3.3–8.1)*	7.5 (5.7–9.9)	30.2 (13.3–102.6)*	57.1 (41.3–79.0)
Overweight	50.1 (28.5–78.3)	34.3 (26.8–43.9)	81.2 (33.3–132.7)	58.3 (46.2–73.5)	8.1 (4.1–16.8)	6.6 (5.2–8.4)	82.4 (27.8–152.0)	47.2 (35.5–62.8)
Obese	56.9 (29.7–95.0)	46.7 (31.2–69.9)	87.2 (46.0–162.8)	75.4 (51.7–109.9)	11.9 (5.3–17.2)	8.2 (5.6–12.1)	86.1 (43.1–145.5)	62.8 (39.5–99.9)
Weight change in last 6 months								
Lost 4–5 Kg	45.4 (20.1–69.8)	45.3 (25.5–80.2)	78.4 (35.1–118.9)	85.9 (50.5–146.3)	6.0 (5.2–13.6)	8.4 (4.8–14.6)	67.8 (25.5–150.5)	68.4 (35.5–132.0)
Little or no change (± 3 Kg)[a]	40.1 (20.8–77.4)	35.2 (29.5–42.1)	67.0 (30.3–127.9)	58.5 (49.6–69.1)	7.8 (4.8–15.5)	7.3 (6.1–8.6)	55.1 (24.7–139.5)	51.3 (41.9–62.9)
Gained 4–5 Kg	27.7 (23.1–40.2)	41.3 (23.3–73.3)	50.2 (30.5–68.5)	66.5 (39.0–113.2)	5.1 (3.2–6.6)	5.6 (3.2–9.7)	42.7 (20.8–80.0)	61.7 (31.9–119.0)
Was breastfed								
No	50.0 (8.1–77.0)	40.2 (26.2–61.8)	79.8 (18.4–127.1)	71.7 (48.2–106.6)	7.9 (5.3–12.3)	8.6 (5.7–12.9)	56.3 (8.1–123.8)	54.3 (33.5–88.2)
Yes	40.1 (21.5–75.7)	35.7 (29.2–43.6)	65.2 (30.8–113.9)	59.4 (49.3–71.4)	7.5 (4.3–15.2)	7.1 (5.9–8.6)	58.3 (25.5–138.8)	54.0 (43.1–67.6)
Birth place								
Catalonia	47.8 (22.1–78.3)*	41.5 (34.6–49.9)	79.5 (41.2–127.9)*	68.9 (57.9–81.9)	7.8 (5.2–15.6)*	8.4 (7.0–10.0)	83.3 (30.3–147.2)*	66.6 (54.7–81.1)
Rest of Spain	54.2 (23.2–76.5)	49.6 (32.3–76.3)	84.2 (33.8–155.0)	78.8 (52.6–117.9)	6.9 (5.2–13.4)	7.4 (4.8–11.3)	72.7 (31.5–153.5)	74.2 (46.8–117.9)
Abroad	23.5 (3.9–32.3)	18.5 (13.1–26.1)**	29.4 (10.1–57.9)	33.1 (23.9–45.8)**	3.2 (1.4–7.8)	4.2 (3.0–5.9)**	21.7 (3.2–33.5)	19.2 (13.2–27.8)**

Table 4 (*continued*)

	PCB 138		PCB 153		PCB 156		PCB 180	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Occupational social class								
I-II (more affluent)	42.9 (21.4–77.4)	39.2 (31.1–49.3)	72.2 (33.8–127.0)	64.4 (51.9–79.9)	7.8 (5.2–14.5)	7.9 (6.3–9.8)	54.5 (25.3–144.1)	59.7 (45.8–77.6)
III	27.3 (8.9–70.1)	30.9 (21.7–44.0)	41.5 (16.9–118.6)	55.9 (40.1–77.9)	5.7 (1.5–13.1)	6.4 (4.5–9.0)	53.3 (8.8–107.3)	46.3 (30.9–69.6)
IV-V (less affluent)	39.2 (24.0–65.1)	35.9 (26.5–48.8)	63.0 (30.2–123.3)	58.7 (44.1–78.1)	6.2 (4.0–14.6)	6.7 (5.0–9.0)	62.5 (23.9–129.4)	48.1 (33.9–68.2)

	Fluorene		Phenanthrene		Pyrene		Fluoranthene	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Men	18.3 (11.0–35.7)	15.4 (12.5–18.9)	46.8 (7.1–104.0)	28.1 (20.2–39.0)	23.6 (16.9–32.1)	22.3 (19.6–25.4)	21.3 (13.4–37.2)	17.2 (14.0–21.2)
Age (years)								
18–44	22.3 (13.6–39.0)*	19.5 (14.5–26.2)	71.8 (7.0–123.0)*	31.3 (19.8–49.6)	24.2 (15.6–33.0)	24.7 (205–29.6)	21.3 (13.6–36.0)	18.5 (13.8–24.8)
45–64	14.0 (8.9–24.1)	11.7 (8.2–16.7)**	29.3 (1.8–65.9)	15.9 (9.1–27.6)	20.5 (15.0–29.8)	18.1 (14.5–22.6)	18.7 (6.8–30.2)	12.5 (8.8–17.9)
≥65	23.4 (10.0–39.5)	16.6 (10.6–26.0)	76.5 (36.6–137.9)	55.3 (27.6–110.9)	27.5 (20.6–44.6)	26.5 (20.0–35.0)	33.2 (18.7–44.5)	25.1 (16.0–39.2)
Body Mass Index								
Normal	22.4 (14.1–39.5)*	19.7 (13.9–28.1)	69.3 (7.1–137.6)	33.5 (18.9–59.4)	25.6 (17.0–33.3)	25.4 (20.3–31.7)	21.8 (15.2–41.1)	18.8 (13.0–27.0)
Overweight	17.4 (8.8–29.0)	14.3 (10.5–19.5)	36.7 (7.5–103.5)	26.0 (15.7–42.9)	22.2 (17.0–34.0)	21.3 (17.5–25.9)	20.3 (12.5–38.4)	16.9 (12.3–23.2)
Obese	15.1 (5.8–25.1)	11.0 (6.6–18.1)**	43.6 (7.0–72.0)	23.6 (10.4–53.4)	19.8 (13.4–30.2)	19.2 (14.0–26.4)	18.9 (10.0–30.3)	15.1 (9.0–25.4)
Weight change in last 6 months								
Lost 4–5 Kg	16.7 (11.9–25.5)	11.4 (5.6–23.2)	35.5 (1.9–86.6)	16.7 (5.3–52.4)	25.0 (16.5–29.2)	21.3 (13.6–33.5)	19.4 (16.3–29.0)	20.3 (9.8–42.3)
Little or no change (± 3 Kg)[a]	21.2 (11.6–37.3)	16.5 (13.2–20.6)	55.6 (7.8–107.1)	31.6 (22.1–45.1)	23.8 (17.1–34.5)	22.8 (19.8–26.3)	21.8 (12.4–41.4)	17.4 (13.9–21.9)
Gained 4–5 Kg	14.7 (7.2–18.7)	9.8 (4.8–20.0)	20.0 (1.4–77.4)	13.1 (4.2–41.0)	16.9 (14.2–23.4)	18.3 (11.7–28.9)	16.9 (14.2–23.8)	12.7 (6.1–26.5)
Was breastfed								
No	23.4 (14.2–39.6)	21.6 (12.6–37.3)	89.6 (23.7–120.2)	46.4 (19.8–108.6)	31.9 (14.7–57.4)	28.8 (20.6–40.4)	41.1 (14.4–42.0)	25.9 (15.4–43.7)
Yes	17.5 (10.3–36.8)	14.6 (11.3–18.7)	41.0 (6.1–114.3)	25.6 (17.2–38.1)	23.0 (16.8–31.4)	21.2 (18.1–24.8)	20.1 (12.9–32.9)	16.1 (12.7–20.6)
Birth place								
Catalonia	21.7 (12.8–38.8)	17.5 (13.7–22.4)	69.1 (7.2–119.0)*	33.9 (23.0–50.2)	25.3 (17.1–35.9)	24.1 (20.7–28.2)	22.1 (16.3–41.4)	19.7 (15.4–25.4)
Rest of Spain	15.3 (10.0–35.7)	14.7 (8.3–26.0)	11.8 (1.8–33.3)	10.1 (4.0–25.2)**	22.2 (17.0–26.6)	20.5 (14.3–29.5)	15.2 (9.8–32.8)	12.4 (6.9–22.3)
Abroad	15.4 (5.6–27.4)	10.1 (6.4–16.0)**	36.7 (15.9–99.8)	27.7 (13.3–57.7)	22.0 (14.0–30.2)	17.8 (13.3–23.8)	18.1 (10.6–28.4)	13.1 (8.2–20.9)
Occupational social class								
I-II (more affluent)	17.0 (5.4–31.3)	12.6 (9.5–16.7)	51.2 (4.2–117.5)	27.5 (17.3–43.8)	23.6 (16.8–31.4)	22.1 (18.4–26.5)	21.3 (15.2–41.4)	20.4 (15.2–27.3)
III	20.7 (12.0–37.4)	18.1 (11.7–28.1)	45.4 (6.0–82.7)	24.1 (11.8–49.4)	24.0 (17.9–32.0)	21.6 (16.3–28.6)	21.7 (8.6–35.0)	13.8 (8.8–21.7)
IV-V (less affluent)	17.6 (12.5–40.2)	19.5 (13.4–28.5)	47.3 (26.0–104.5)	32.4 (17.5–60.1)	22.9 (15.6–39.3)	23.2 (18.2–29.5)	20.2 (13.1–33.4)	15.1 (10.2–22.2)

aGM: Geometric mean adjusted for age. $N = 112$.

[a] Reference category (except where otherwise noted, the reference category is the first category mentioned above).

* *p*-value <0.05 (median, Kruskal-Wallis test).

** *p*-value <0.05 (aGM, Wald test; compared against the reference category).

suggested that bioaccumulated OCPs are frequently at higher concentrations and tend to persist longer in the adipose tissue of obese than in non-obese individuals (Arrebola et al., 2009; Wolff et al., 2005; Wolff et al., 2007).

Men with obesity and overweight had the highest levels of *p,p'*-DDE. Women with obesity and overweight had the highest levels of HCB and β-HCH. Women with overweight had the lowest levels of PCBs, as also observed in other populations (Ibarluzea et al., 2011).

In innovative studies, concentrations of DDE and PCBs were both positively associated with BMI and recent weight-loss; they were highest with higher BMI, higher weight-loss, and (in women) shorter lactation (Wolff et al., 2005; Wolff et al., 2007; Wolff and Teitelbaum, 2020). In our study, this was so in men for DDE, HCB, β-HCH, and the four PCBs; in women, for DDE only. However, numbers were small, and more measures per person would be needed to capture influences during the life course, especially in women. Further progress to elucidate these issues

Table 5
Serum concentrations of POPs (ng/g lipid) (compounds detected in over 75% of the population of Barcelona city) in 2016 by sociodemographic characteristics, women.

	p,p'-DDE		HCB		β-HCH	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Women	221.1 (114.6–968.3)	273.4 (230.4–324.4)	36.4 (17.5–104.7)	45.5 (39.2–52.7)	26.7 (10.7–81.1)	26.5 (21.7–32.4)
Age (years)						
18–44	135 (71.1–195.4)*	131.4 (99.8–172.9)	17.5 (13.6–25.4)*	19.2 (15.2–24.3)	10.5 (3.6–17.6)*	9.3 (6.8–12.8)
45–64	317.8 (125.2–640.3)	280.9 (207.4–380.5)**	57.8 (24.3–95.2)	51.2 (39.5–66.3)**	36.4 (16.2–60.0)	33.8 (23.7–48.2)**
≥65	1204 (871.6–1796)	936.4 (661.1–1326)**	139.4 (96.7–327.1)	170.7 (126.9–229.7)**	109.9 (68.4–208.5)	115.8 (77.1–173.8)**
Body Mass Index						
Normal	146.0 (70.3–293.9)*	228.7 (173.8–301.0)	21.4 (15.5–35.0)*	34.2 (27.4–42.7)	14.0 (8.0–30.6)*	21.1 (15.4–28.8)
Overweight	404.7 (149.0–1202)	300.7 (220.2–410.7)	84.4 (21.5–137.3)	50.2 (39.0–64.6)**	44.0 (16.2–106.3)	27.5 (19.3–39.2)
Obese	898.0 (260.3–1989)	360.7 (229.2–567.5)	132.1 (70.9–220.0)	72.2 (50.1–104.2)**	85.5 (38.5–151.8)	44.1 (26.3–73.6)**
Weight change in last 6 months						
Lost 4–5 Kg	329.5 (69.6–733.1)	322.4 (155.6–668.2)	24.8 (16.4–64.0)	39.1 (20.9–73.1)	32.8 (1.4–76.5)	22.7 (9.8–52.8)
Little or no change (± 3 Kg)[a]	223.8 (118.9–969.7)	269.7 (224.1–324.7)	38.8 (17.5–110.5)	44.1 (37.7–51.7)	24.8 (10.8–75.8)	25.2 (20.3–31.2)
Gained 4–5 Kg	142.1 (91.0–1074)	282.0 (150.6–528.2)	47.8 (15.6–251.9)	71.1 (41.5–121.8)	71.9 (16.5–176.2)	54.0 (26.2–111.6)
Was breastfed						
No	180 (118.2–1208)	310.1 (188.7–509.6)	48.4 (21.4–91.9)	48.9 (32.6–73.6)	26.0 (11.7–66.2)	28.2 (15.9–49.8)
Yes	222.7 (120.1–757.1)	278.5 (230.7–336.1)	37.9 (16.7–126.0)	48.5 (41.6–56.6)	32.2 (10.8–106.3)	29.0 (23.4–36.0)
Birth place						
Catalonia	206.3 (98.8–683.7)	250.5 (198.7–315.7)	52.8 (22.4–118.8)*	55.4 (45.9–66.9)	29.8 (10.7–92.7)*	28.4 (21.6–37.5)
Rest of Spain	349.9 (179.9–1192)	211.7 (143.9–311.5)	108.1 (56.7–177.7)	57.9 (42.2–79.4)	67.9 (26.6–117.5)	29.1 (18.3–46.1)
Abroad	193.4 (101.9–471.7)	403.7 (283.9–573.9)**	15.6 (12.1–21.4)	24.8 (18.6–33.1)**	14.6 (4.3–19.0)	21.3 (13.9–32.5)
Occupational social class						
I-II (more affluent)	186.1 (101.9–376.5)*	256.4 (195.2–336.7)	25.7 (17.3–59.5)	45.1 (35.9–56.6)	15.0 (8.9–39.8)*	23.5 (17.6–31.4)
III	256.7 (121.8–779.6)	320.5 (225.8–454.8)	72.0 (17.3–139.4)	65.7 (49.0–88.0)**	42.9 (12.2–109.0)	36.8 (25.4–53.4)
IV-V (less affluent)	614.1 (143.4–1208)	279.2 (203.7–382.7)	50.7 (18.6–137.3)	35.4 (27.2–46.1)	35.5 (15.4–109.9)	23.4 (16.8–32.8)
Breastfed her children						
Never	739.5 (154.2–1421)	468.0 (232.7–941.1)	78.8 (24.2–181.1)	63.8 (35.6–114.5)	60.6 (14.9–126.2)	44.9 (21.9–92.0)
≤6 months	374.8 (172.6–1021)	342.2 (232.4–503.9)	54.8 (22.6–115.2)	50.3 (36.4–69.5)	32.8 (16.2–80.8)	32.1 (21.6–47.8)
>6 months	367.3 (105.3–1192)	277.9 (208.9–369.7)	53.3 (18.8–139.1)	46.7 (36.8–59.5)	35.7 (10.4–109.9)	27.2 (20.3–36.5)

	PCB 138		PCB 153		PCB 156		PCB 180	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Women	42.2 (22.7–73.6)	42.1 (37.1–47.7)	63.9 (34.2–117.2)	64.6 (57.3–72.8)	8.7 (4.0–16.6)	7.1 (6.1–8.2)	67.8 (24.1–110.4)	48.7 (41.5–57.0)
Age (years)								
18–44	20.3 (14.4–31.0)*	20.6 (16.9–25.2)	31.4 (21.9–48.8)*	31.4 (25.8–38.1)	5.6 (1.5–7.4)*	3.9 (3.1–5.0)	20.7 (11.4–43.9)*	19.3 (14.9–25.0)
45–64	57.7 (39.1–74.7)	49.4 (39.6–61.5)**	86.7 (59.4–118.0)	79.3 (63.9–98.3)**	9.1 (5.2–15.5)	7.5 (5.7–9.8)**	80.3 (59.5–112.6)	71.3 (53.5–95.0)**
≥65	112.6 (63.7–252.4)	115.6 (89.8–148.8)**	168.8 (103.5–365.0)	170.5 (133.1–218.4)**	18.4 (12.7–34.6)	18.7 (13.7–25.4)**	127.9 (98.7–344.1)	143.7 (103.3–199.8)**
Body Mass Index								
Normal	29.0 (18.5–50.4)*	43.1 (35.3–52.5)	48.0 (27.0–83.9)*	71.6 (59.4–86.4)	6.3 (1.7–10.7)*	7.8 (6.1–9.8)	43.2 (18.7–80.9)*	62.4 (48.8–79.6)
Overweight	59.7 (31.1–80.2)	37.6 (30.0–47.1)	94.0 (39.2–122.7)	53.0 (42.9–65.6)**	12.6 (6.0–15.3)	6.1 (4.6–8.0)	84.0 (24.1–112.4)	33.9 (25.8–44.7)**
Obese	77.8 (40.3–148.7)	42.9 (30.9–59.6)	107.2 (73.1–198.4)	63.4 (46.5–86.3)	13.3 (6.4–22.2)	6.3 (4.2–9.3)	89.7 (50.6–144.0)	43.3 (28.9–64.6)
Weight change in last 6 months								
Lost 4–5 Kg	41.4 (12.6–59.6)	37.0 (21.8–63.1)	45.7 (26.5–88.1)	56.6 (34.1–94.0)	6.6 (2.8–9.0)	6.2 (3.2–11.7)	31.4 (14.3–80.8)	31.6 (16.3–61.2)
Little or no change (± 3 Kg)[a]	43.0 (25.3–75.6)	42.1 (36.8–48.2)	70.9 (38.4–118.9)	65.3 (57.4–74.3)	9.1 (4.4–17.3)	7.2 (6.1–8.5)	68.8 (25.1–112.2)	52.1 (44.0–61.6)
Gained 4–5 Kg	25.2 (17.3–107.4)	45.8 (28.9–72.4)	40.9 (23.9–150.5)	63.2 (40.8–97.8)	7.8 (1.6–17.9)	6.8 (3.9–11.8)	43.3 (4.8–102.3)	30.9 (17.5–54.6)**
Was breastfed								
No	45.0 (31.0–121.2)	55.8 (39.4–78.9)	63.9 (49.3–201.3)	95.0 (68.8–131.2)	15.8 (6.3–28.0)	11.1 (7.1–17.2)	72.6 (34.7–146.7)	80.4 (52.1–123.9)
Yes	43.1 (20.0–75.4)	42.7 (37.4–48.7)	72.5 (31.1–117.6)	64.1 (56.7–72.4)**	8.7 (1.7–14.5)	6.8 (5.7–8.0)**	69.2 (20.5–112.2)	47.2 (40.1–55.7)**
Birth place								
Catalonia	50.4 (29.0–77.7)*	51.5 (43.9–60.4)	80.8 (48.4–134.8)*	83.8 (72.7–96.5)	9.0 (5.8–17.9)*	8.3 (6.8–10.2)	80.9 (44.3–113.0)*	71.3 (59.6–85.3)
Rest of Spain	71.8 (42.6–132.8)	46.5 (35.6–60.6)	104.0 (75.1–199.8)	70.9 (55.9–89.8)	12.8 (6.5–26.9)	7.4 (5.3–10.4)	98.3 (67.9–178.2)	57.3 (42.5–77.3)

Table 5 (*continued*)

	PCB 138		PCB 153		PCB 156		PCB 180	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Abroad	18.6 (12.8–31.1)	25.6 (20.1–32.6)**	23.9 (14.8–45.9)	35.2 (28.4–43.7)**	1.7 (1.4–8.7)	4.9 (3.6–6.7)**	15.7 (5.6–33.3)	19.5 (14.9–25.6)**
Occupational social class								
I-II (more affluent)	35.9 (23.9–71.9)	44.9 (36.8–54.7)	60.0 (37.2–114.8)	73.2 (60.6–88.3)	6.8 (5.1–13.8)	8.0 (6.3–10.1)	65.0 (23.9–102.8)	60.6 (47.3–77.6)
III	60.8 (21.3–101.0)	48.0 (37.3–61.9)	89.9 (31.8–143.4)	67.8 (53.3–86.4)	12.1 (1.7–19.0)	7.4 (5.5–10.0)	83.2 (18.0–112.3)	44.8 (32.6–61.6)
IV-V (less affluent)	49.8 (24.5–101.9)	36.5 (29.0–45.9)	75.5 (36.0–144.5)	54.7 (44.0–68.0)	12.7 (1.7–19.3)	6.0 (4.6–7.8)	69.2 (24.5–114.0)	42.4 (31.8–56.5)
Breastfed her children								
Never	71.9 (41.3–115.5)	58.0 (34.9–97.9)	114.4 (69.4–180.0)	91.3 (55.2–150.9)	11.7 (6.4–19.0)	9.1 (5.3–15.8)	101.8 (82.1–147.1)	84.2 (43.9–161.4)
≤6 months	59.1 (33.1–85.0)	50.8 (38.1–67.9)	83.2 (51.2–139.8)	75.4 (57.1–99.6)	11.8 (6.8–16.6)	8.4 (6.2–11.4)	77.2 (44.3–121.2)	54.7 (38.1–78.4)
>6 months	54.0 (23.2–107.0)	41.5 (33.5–51.4)	88.8 (42.2–150.0)	68.1 (55.4–83.6)	9.6 (5.1–22.0)	7.4 (5.9–9.3)	83.2 (36.8–147.8)	52.6 (40.3–68.6)

	Fluorene		Phenanthrene		Pyrene		Fluoranthene	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Women	21.0 (5.7–40.2)	13.4 (10.5–17.0)	53.6 (2.9–95.0)	25.2 (18.4–34.6)	23.3 (13.6–33.6)	22.0 (19.3–25.0)	20.0 (11.4–31.3)	15.5 (12.6–19.2)
Age (years)								
18–44	25.4 (6.7–44.4)	15.4 (10.6–22.3)	50.8 (2.4–107.7)	22.2 (13.5–36.4)	23.8 (14.5–39.0)	24.4 (19.9–29.8)	22.9 (11.9–34.8)	17.9 (12.9–25.0)
45–64	18.1 (6.2–39.9)	14.2 (9.4–21.5)	51.7 (9.4–91.8)	29.9 (17.3–51.7)	23.6 (14.0–28.2)	19.9 (15.9–24.8)	19.7 (10.1–29.2)	13.7 (9.5–19.7)
≥65	13.3 (4.8–24.6)	9.7 (6.0–15.5)	64.7 (1.7–88.2)	25.0 (13.3–46.8)	18.6 (13.1–35.5)	21.1 (16.4–27.3)	16.2 (6.9–27.8)	14.3 (9.4–21.8)
Body Mass Index								
Normal	20.3 (6.3–36.8)	12.1 (8.3–17.8)	49.9 (2.9–99.0)*	25.0 (15.3–40.8)	23.0 (12.9–29.6)	20.4 (16.7–24.9)	19.5 (11.9–28.4)	14.0 (10.0–19.6)
Overweight	24.5 (11.9–46.9)	18.1 (11.7–28.0)	78.4 (18.9–116.1)	41.2 (23.6–72.0)	25.5 (15.2–38.8)	26.8 (21.4–33.6)	26.6 (13.4–45.6)	19.6 (13.4–28.7)
Obese	16.6 (1.5–32.9)	10.5 (5.6–19.7)	8.8 (1.6–78.0)	11.1 (4.9–24.9)	21.9 (13.2–38.5)	20.9 (15.1–29.1)	17.5 (6.9–29.0)	15.0 (8.6–25.9)
Weight change in last 6 months								
Lost 4–5 Kg	9.6 (1.2–33.4)	5.1 (1.9–13.8)	55.0 (7.1–111.1)	23.2 (6.0–89.7)	26.8 (13.3–62.9)	28.7 (16.7–49.2)	19.2 (15.0–34.7)	17.8 (7.2–43.7)
Little or no change (± 3 Kg)[a]	21.0 (6.2–40.2)	13.7 (10.7–17.7)	51.4 (2.6–92.2)	24.9 (17.7–35.1)	21.8 (13.1–29.1)	21.0 (18.3–24.1)	19.7 (11.2–29.2)	15.4 (12.3–19.4)
Gained 4–5 Kg	28.4 (18.6–51.1)	20.0 (8.5–47.3)	71.2 (6.5–141.5)	31.1 (9.7–99.4)	29.3 (27.3–64.7)	31.1 (19.6–49.6)	30.4 (5.4–46.8)	15.2 (7.0–33.2)
Was breastfed								
No	19.6 (9.2–54.5)	20.2 (9.9–40.9)	9.6 (1.6–137.6)	15.3 (6.0–39.4)	23.5 (13.1–42.3)	26.3 (17.9–38.6)	16.2 (1.4–29.2)	9.9 (5.4–18.2)
Yes	21.0 (5.2–39.4)	12.5 (9.6–16.4)	51.4 (5.9–91.4)	25.5 (17.8–36.4)	21.8 (13.3–29.2)	20.9 (18.1–24.2)	19.7 (12.3–29.2)	16.0 (12.7–20.2)
Birth place								
Catalonia	16.7 (3.5–44.6)	11.9 (8.5–16.5)	52.8 (2.9–112.7)*	27.2 (17.5–42.1)	21.2 (12.4–33.6)	22.1 (18.6–26.4)	20.2 (8.9–36.2)	16.9 (12.6–22.6)
Rest of Spain	15.3 (5.5–24.6)	13.8 (8.0–23.9)	45.0 (1.9–75.3)	16.7 (8.1–34.7)	21.8 (12.9–26.1)	18.2 (13.6–24.4)	18.6 (14.4–25.6)	14.5 (8.9–23.5)
Abroad	25.4 (16.9–43.0)	16.6 (10.0–27.3)	76.9 (5.9–103.3)	30.5 (15.7–59.2)	27.5 (17.4–39.5)	25.4 (19.4–33.2)	23.9 (8.6–43.0)	13.9 (8.9–21.6)
Occupational social class								
I-II (more affluent)	21.2 (6.4–40.3)	13.9 (9.5–20.4)	46.4 (2.5–93.5)*	22.7 (13.9–37.2)	21.8 (10.4–30.4)	19.4 (15.9–23.7)	17.8 (9.9–29.1)	15.1 (10.8–21.1)
III	25.2 (2.9–51.1)	14.8 (9.1–24.2)	88.4 (25.0–137.7)	48.2 (25.6–90.9)	27.3 (18.0–38.6)	28.1 (21.7–36.4)**	27.1 (13.7–38.5)	20.4 (13.3–31.4)
IV-V (less affluent)	17.0 (5.5–28.4)	11.5 (7.4–17.9)	41.0 (1.7–78.4)	16.4 (9.3–29.1)	20.9 (13.4–35.1)	21.6 (17.1–27.3)	18.9 (6.9–29.0)	13.1 (8.9–19.3)

(*continued on next page*)

Table 5 (*continued*)

	Fluorene		Phenanthrene		Pyrene		Fluoranthene	
	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)	Median (P25-P75)	aGM (CI 95%)
Breastfed her children								
Never	44.7 (11.0–52.7)	29.6 (11.8–74.0)	72.7 (14.6–211.7)	44.9 (13.6–148.6)	26.4 (15.2–48.2)	25.9 (16.3–41.3)	24.0 (5.7–36.5)	13.9 (6.0–32.1)
≤6 months	20.6 (8.4–25.9)	13.1 (7.9–21.8)	58.3 (9.4–86.0)	27.5 (14.2–53.3)	26.2 (17.9–39.5)	23. (18.3–30.7)	24.1 (14.2–38.6)	16.5 (10.4–26.2)
>6 months	19.2 (5.4–31.8)	11.4 (7.8–16.6)	53.4 (2.5–93.4)	21.5 (13.2–35.0)	19.4 (13.1–27.4)	19.9 (16.4–24.0)	16.1 (7.3–26.7)	13.5 (9.6–19.1)

aGM: Geometric mean adjusted for age. $N = 128$.
[a] Reference category (except where otherwise noted, the reference category is the first category mentioned above).
* p-value <0.05 (median, Kruskal-Wallis test).
** p-value <0.05 (aGM, Wald test; compared against the reference category).

could be made integrating empirical information on actual levels and trends of POP exposure during the specific historical periods preceding each analysis (Porta, 2018; Henríquez-Hernández et al., 2020; Porta et al., 2008; Li et al., 2020; Wolff et al., 2005; Wolff et al., 2007; Wolff and Teitelbaum, 2020).

The reduction in mean concentrations of most POPs was not inevitably to be accompanied by a substantial reduction in the number of participants with a high number of POPs at high concentrations (Porta, 2004; Porta et al., 2012b; Pumarega et al., 2016), but in this study it was: in the 10 years, the percent of participants with concentrations of 3 or more and 6 or more POPs each at high concentrations decreased by 52% and 70%, respectively. Again, good news.

There are local studies, like those carried out in areas where people are exposed to polluted environments or the emissions of industrial facilities, that have monitored different kinds of pollutants, including POPs, and which also support the main conclusions of the present study. When in 2005 a municipal solid waste plant started to operate in Bilbao (Basque Country, Spain), a biomonitoring project identified and quantified POPs from 2006 to 2013 in three cross-sectional studies, including 127 adults prospectively monitored, living in areas near to and further away from the plant. OCs decreased significantly during the study period: 80% (PCDD/Fs), 80% (dl-PCBs), and 37% (marker-PCBs) (Zubero et al., 2017).

The two PAHs most frequently detected in 2006 samples, naphthalene and phenanthrene, showed decreases in concentrations greater than 90% and 30%, respectively. They were detected in 83% and 73% of participants, respectively, in 2006, and in 45% and 76%, respectively, in 2016. While in 2006 fluorene, pyrene and fluoranthene were not detected in anybody, in 2016 they were detected in 85%–99% of citizens. These changes were significant for fluorene, pyrene, fluoranthene, and naphthalene, but not for phenanthrene. Uses and physicochemical characteristics of PAHs may explain these variations. Although PAHs are rightly considered as POPs (EEA - European Environment Agency, 2019), PAHs have a lower liposolubility coefficient than other OCs, they are partially metabolized in the organism (which limits their accumulation), and have less resistance to degradation in the environment (Mumtaz et al., 1996). Despite this, their constant presence in the environment and the continuous exposure of humans to them (mainly through diet) (Yebra-Pimentel et al., 2015), make PAHs relevant for biomonitoring studies, where they are frequently detected. Thus, phenanthrene and pyrene were the most frequently detected PAHs (both in more than 55% of 447 newborns) in the island of La Palma (Spain); naphthalene was the PAH detected at highest concentration (Cabrera-Rodríguez et al., 2019). A similar profile was reported in 121 Romanian subjects (Luzardo et al., 2019). In a series of 121 women from China, fluorene, phenanthrene, pyrene and fluoranthene showed frequencies of detection greater than 95% (Wang et al., 2015).

In the present study the percentages of detection of PBDEs were lower than in other studies in Europe; e.g. the HELIX study detected BDE-153 in 73% of participating mothers (Montazeri et al., 2019), and the same compound was detected in all 170 participants in a population-based study in Sweden (Bjermo et al., 2017), while in the BHS of 2016, BDE-153 was detected in 24% of participants, being the most detected PBDE compound.

p,p'-DDT was detected (by IUIBS) in only 3% of the 240 samples from 2016, whereas in 2006 it was detected (by IQAB-CSIC) in 97% of the 231 samples (median, 0.14 ng/mL) (Porta et al., 2012a). The nature of this difference is uncertain because *p,p'*-DDT was not detected in the 30 samples from 2006 re-analyzed by IUIBS in 2016 (see 2.2. Analytical chemical methods). The long period of storage could explain the difference. DDT can undergo dechlorination in frozen blood, with the subsequent increase in the concentrations of its metabolites (DDE and DDD) (Ecobichon and Saschenbrecker, 1967). DDT was detected in 4% of 447 newborns from the island of La Palma (Spain) (Cabrera-Rodríguez et al., 2019), in 23% of 121 adults from Romania, a country that only recently banned DDT (Luzardo et al., 2019), or in 13% of 1135 adolescents from Germany (Bandow et al., 2020). Such currently observed percentages contrast with those observed more than 10 years ago, with DDT detection frequencies close to 100% in some cases (Porta et al., 2008; Schoeters et al., 2017; Saoudi et al., 2014). While there have been significant improvements in the analytical techniques over the last decades, there remains a need for re-assessment of historical samples.

Compared to citizens born in Spain, citizens born abroad had higher concentrations of *p,p'*-DDE and lower of HCB and PCBs. These facts have also been observed in different cohorts in Spain (Ibarluzea et al., 2011). Citizens from Latin America represent a high percentage of citizens born abroad residing in Barcelona. The specific country of origin of citizens born abroad and the duration of their stay in the city are key to understanding differences in concentrations with the rest of the population of Barcelona. In comparison to Europe, where OCPs were banned in the mid- to late 1970s, in Latin American countries they were banned later, and in many their use is still allowed, albeit with restrictions (UNEP, 2002). Conversely, although HCB was formerly used as fungicide, non-dietary sources of HCB and PCBs are more common in industrialized areas (ATSDR, 2020). Also, concentrations of OCPs were higher in a group of 131 subjects from Morocco than in 100 subjects from Canary Islands, a region of Spain 100 km off the coast of Morocco (Henríquez-Hernández et al., 2016), demonstrating the influence of social and life conditions in the concentrations of OCPs in two populations that are geographically close.

Studies biomonitoring time trends in human contamination from environmental chemical agents may or not be representative of the target population; be more or less limited to a range of ages, socioeconomic positions and geographies; include larger or smaller numbers of individuals, chemicals, and periods of sample collection; integrate or not personal and social predictors of the concentrations; and be linked or not to institutions and policies to decrease exposure. They must, in any case, use meaningful biomarkers and, critically, apply accurate procedures for sample collection, processing, analysis, and quality assurance (Henríquez-Hernández et al., 2020; Porta

et al., 2008; Porta, 2004; Angerer et al., 2007; Schulz et al., 2011; Bjerregaard-Olesen et al., 2017; Buekers et al., 2018; Calafat, 2016; Joas et al., 2017; Haines et al., 2017; Li et al., 2020). We think our study fulfils the latter set of requirements (Luzardo et al., 2019; Henríquez-Hernández et al., 2017). We also think we were reasonably rigorous and pragmatic to maximize the comparability of chemical analyses performed in 2006 and 2016 in the two laboratories (e.g., re-analyzing at IUIBS in 2016, 30 samples from the 2006 BHS). It is often not possible to use the same laboratory in different studies; even when it is, protocols change (Porta et al., 2008; Porta, 2004; Angerer et al., 2007; Schulz et al., 2011; Bjerregaard-Olesen et al., 2017; Buekers et al., 2018; Calafat, 2016; Joas et al., 2017; Dennis et al., 2017; Haines et al., 2017; Li et al., 2020; Nøst et al., 2019; Schoeters et al., 2017; Schröter-Kermani et al., 2013; Stubleski et al., 2018). Therefore, we think the reported decreases in POP concentrations have a high degree of validity.

Our study used a cross-sectional design to study a representative sample of the target population, it included a small number of individuals (471 in total) from 18 to 80 years of age and of all socioeconomic positions, it measured a fair number of chemicals (62) in two periods, it integrated personal and social predictors of the concentrations, and it is linked to policies developed by the Barcelona Agency for Public Health. The common presence of results on POP studies in social networks and media has likely influenced citizens' behaviors, and policies of public and private organizations.

Finally, we can look further backwards to concentrations we measured in 2002 in the first representative sample of citizens of Barcelona:(Porta et al., 2012a) concentrations decreased markedly between 2002 and 2016, with changes of −72% for *p,p*'-DDE, −86% for HCB, −89% for β-HCH, −92% for PCB 118, and of more than −60% for the other three main PCBs (138, 153 and 180).

5. Conclusions

While concentrations of most POPs are decreasing in Barcelona, significant sociodemographic differences in such reductions warrant strengthening public and private policies towards groups making slower progress. A relevant component of the success in the current decreasing is a reduction of differences (convergence) by gender. For several pollutants the decrease was larger in the younger groups, mostly with monotonic trends, a finding coherent with the possibility that younger cohorts are being less exposed to POPs. Even in a society with decreasing levels, reductions were less frequent in individuals with obesity than with normal weight.

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Declaration of competing interest

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

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