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Serum concentrations of selected organochlorine pesticides in a Lebanese population and their associations to sociodemographic, anthropometric and dietary factors: ENASB study

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Abstract Organochlorine pesticides (OCPs) were banned by the Stockholm Convention many years ago; however, they are still detected in the environment due to their high persistence, their current illegal use, and their import from countries where they have not been banned. We evaluated the serum concentrations of selected OCPs (hexachlorobenzene (HCB), β-hexachlorocyclohexanes (β-HCH), p,p′-dichlorodiphenyltrichloroethane (DDT) and its metabolite p,p′-dichlorodiphenyldichloroethylene (DDE) in a sample of Lebanese adults using gas chromatography coupled to an ion trap mass spectrometer detector. The mean concentrations of HCB, β-HCH, DDT, and DDE were 7.1, 8.6, 2.1, and 18.9 ng/g of lipids, respectively, and the major contributor among the four OCPs was DDE. The OCP levels in the present study were in general lower than the values observed in several countries worldwide and their concentrations at the 95th percentile were lower than the biomonitoring equivalents (BEs) excluding any appreciable health risk. We observed an inverse association between HCB concentrations and body mass index (BMI) as well as HCB, β-HCH, and DDE levels, and smoking habits. Milk consumption however was positively associated with an increased serum level of β-HCH. This study, which was the first to investigate OCP serum levels in a Lebanese population, provides a baseline to which future measurements can be compared.

Keywords Human biomonitoring · Organochlorine pesticides · HCB · β-HCH · DDT · DDE · Lebanon

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Introduction

Organochlorine pesticides (OCPs) are chemical pesticides that were widely used in the past to control insects or pests on crops. Their agricultural use was first banned in the 1970s in developed countries and worldwide in 2001 and 2004, respectively (Stockholm Convention 2008a). Despite their ban by the Stockholm Convention, OCPs are still detected in the environment and remain a source of public concern. This is mainly due to their heavy use prior to their restrictions, their high persistence in the environment, their illegal use in some countries, and their import from countries where they were not banned (Choi et al. 2015). Long-term exposure to OCPs has been associated with deleterious health effects including neurologic and reproductive disorders, obesity, diabetes, and cancer (CDC 2009; Mrema et al. 2013). In Lebanon, several studies have detected OCPs in soils and sediments particularly hexachlorobenzene (HCB), β-hexachlorocyclohexanes (β-HCH), p,p′-dichlorodiphenyltrichloroethane (DDT), and its metabolite p,p′-dichlorodiphenyldichloroethylene (DDE) (MoE/ELARD 2005). Low levels of HCB, DDT, and DDE were detected in analyzed sediments from El Kabir Watershed in Akkar (the second most agricultural plain in the country following the Bekaa, located in the north of Lebanon) (MoE/ELARD 2005). Another study found DDT residues in soil samples from agricultural regions around Lebanon (coastal plains, Bekaa valley, and Mount Lebanon) (Bashour et al. 2004). A recent study by Chbib et al. (2017) showed that groundwater on the plain of Akkar was remarkably contaminated by DDT and HCH from recent use of these compounds. In view of the above, the aim of the present study was to evaluate for the first time the serum levels of four OCPs (DDT, DDE, HCB, and β-HCH) in a Lebanese sample and explore their correlations with sociodemographic and lifestyle habits.

Materials and methods

Study design and population

The study was a cross-sectional survey conducted between October 2013 and June 2015 in Saint Joseph University of Beirut (Université Saint Joseph (USJ)). The study sample consisted of randomly selected students and staff members of USJ aged between 18 and 65 years. A sample of 500 students and staff members was drawn using a stratified random cluster sampling by campus. Recruitment efforts targeted a sample with a status and sex distribution proportional to that of the university population per campus. In order to be included in the study, the participants had to be Lebanese and not taking any medication that could affect their blood tests. Three hundred fifty-eight subjects provided written consents. Sociodemographic, anthropometric, and clinical data were collected for 314 participants (133 men and 181 women), 44 individuals withdrew from the study, another 12 subjects were lost to follow up, and the dietary characteristics were evaluated for 302 individuals (see flow chart of study participants in Online Resource 1).

Data collection

Each participant completed an interviewer-administered questionnaire regarding age, gender, governorate of residence, crowding index, domestic use of pesticides, and smoking status. Crowding index was defined as the total number of co-residents per household, excluding newborn infants, divided by the total number of rooms, excluding the kitchen and the bathrooms. Anthropometric measurements including height, weight, and waist circumference (WC) were also measured. The body mass index (BMI) was calculated using the formula (weight in kg) / (height in meters)^2 and the participants were classified as overweight/obese if the BMI value ranged between 25 and 29.9 kg/m^2 and ≥30 kg/m^2, respectively, based on WHO cutoff points (World Health Organization (WHO) 2016). Values ≥94 cm for men and 80 cm for women were used as cutoff points for waist circumference according to the International Diabetes Federation (IDF) criteria (Alberti et al. 2009). Body composition analysis was performed using a bioelectrical impedance analyzer, "Inbody 720" (InBody, CA, USA), to estimate percent body fat. Dietary habits of the study sample were estimated using a quantitative, 164-items food-frequency questionnaire (FFQ). We assessed the intake of 14 major food categories, over the previous year, including culturally adapted food items. These categories included among others: fish and shellfish, red meat, poultry, eggs, dairy products, fruits, and vegetables. The frequency of consumption of each food item was noted per day, week, month, or year. The number of food portions consumed each time was also indicated by the participants, and the portion sizes were estimated using measuring cups and food models (Biró et al. 2002). The questionnaires were administered by the same interviewer, and food consumption frequencies were estimated in portions per day by multiplying the frequency of consumption (per week, per month, or per year) by the number of portions consumed each time.

Blood sampling and storage

Blood samples were collected from fasting subjects in 5-ml vacutainer tubes (BD vacutainer, Plymouth, UK) without anticoagulants and were centrifuged at 3500 rpm for 15 min. The serum was divided in two aliquots: the first one was analyzed for total lipids using an automatic biochemistry analyzer HumaStar 300 (Human GmbH, Wiesbaden, Germany). Total lipids were selectively extracted by a dioxanne-ethanol solving solution (1.2–15 M, respectively). Extracted lipids reacted with phenol (110 M) to produce turbidity. The absorbance of samples and standard was measured against the blank at 620 nm. The second aliquot was tested for OCPs using gas chromatography coupled to an ion trap mass spectrometer detector (Agilent Technologies, CA, USA). The human serum samples (10 ml) were homogenized manually by shaking for 1 min. A mixture of standard grade organochlorine references (Sigma-Aldrich, Darmstadt, Germany) were used for the validation and the standardization of the analysis method. The organic solvents used for extraction of OCPs were HPLC grade hexane, dichloromethane, acetone, and ethyl alcohol (Scharlau Chemicals, Scharlab, Barcelona, Spain). Pesticide residues were extracted (Trejo-Acevedo et al. 2009), and 2 ml of the sample was used for liquid-liquid extraction with a mixture of ethyl alcohol, ammonium sulfate, and hexane (1:1:3). The serum sample and the mixture were mixed for 3 min using a vortex and then centrifuged at 3000 rpm for 7 min. Extracts were then concentrated in a rotary evaporator. The concentrated extract was cleaned in a Florisil column (Alltech Associates Inc., IL, USA) and eluted with 6 ml of dichloromethane:hexane (30:70). Analysis of OCPs was performed using a gas chromatograph (Agilent Technologies, CA, USA) equipped with a split/splitless injector port and ion mass spectrometer detector. The injector was operated in the splitless mode (1.5 min). Hydrogen (H_2) served as a carrier gas with a flow rate of 1 ml/min. Chromatographic separation was accomplished with a CP-SIL 8 CB (Agilent Chrompack 35MS), (30 m × 0.25 mm, i.d.; 0.25-μm film thickness) capillary column. The temperature program used was 110 °C for 2 min, increased at 15 °C/min to 285 °C for 5 min and finally at 5 °C/min to 300 °C for 15 min. The detector was with a N_2 makeup gas. The corresponding limits of detection (LOD) were 0.012 μg/l for DDT (2.29 ng/g lipids), 0.027 μg/l for DDE (5.14 ng/g lipids), and 0.022 μg/l for HCB and β-HCH (4.19 ng/g lipids). The recovery was performed based on the fortification study and showed mean values from 88 to 94% depending on the pesticide.

Statistical analysis

Geometric means (log10 of quantitative variable) were used in case of non-normality of distribution. Given that more than 30% of the samples had non-detected concentrations of OCPs, a multiple imputation (MI) analysis was carried out (Lubin et al. 2004). All missing data type were created by imputing random values after generating multiple linear regression and creating standardized β which provided M estimators of the parameters of interest. The predictors in the imputation process were determined based on chi-square test. The upper bound was fixed and known in advance (LOD). No differences were observed in the linear regression analysis between the multiple imputation and half the limit of detection methods. Therefore, OCPs with concentrations below the LOD were reported as LOD/2. Resulting OCP concentrations were adjusted for total lipids. Sociodemographic, anthropometric, and dietary characteristics of the population were compared between men and women using chi-square test. Mean BMI, WC, and percentage of body fat were compared between the two groups using *t* test whereas BMI, WC, and percent body fat categories were compared using chi-square test. OCP mean concentrations were compared between categories of independent variables using *t* test and analysis of variance (ANOVA). Statistical analyses were performed using IBM SPSS (IBM SPSS Statistics for Windows, Version 20, IBM corp., Armonk, NY) and confidence interval of 95% was used for all tests with a p value <0.05.

Results and discussion

Characteristics of the population

Characteristics of the participants are summarized in Table 1. The study sample was composed of 314 students and staff members (42.4% men and 57.6% women) with an average age of 25.6 years. The overall mean BMI was in the normal range with 8.3% obese, 29% overweight, and 6.4% underweight. Obesity and overweight prevalence were significantly higher among men than women (12.8 vs 5%; 39.1 vs 21.5%, respectively). However, women had higher levels of abdominal obesity and percent body fat compared to men (44.8 vs 31.6% and 66.4 vs 51.1%, respectively; $p < 0.05$). In relation to diet, few differences were evident between genders: a higher intake of red meat, poultry, and eggs in men as compared to women.

OCP concentrations in serum samples

Since OCP concentrations depend on the amount of lipid in the sample, the results were expressed as nanograms per gram (ng/g) of lipids. Descriptive statistical data on the levels of lipid adjusted OCPs are given in Table 2. The four analyzed OCPs were detected in half of the serum samples and the mean concentrations of HCB, β-HCH, DDT, and DDE were 7.1, 8.6, 2.1, and 18.9 ng/g lipids, respectively.

HCB mean serum concentration was relatively low (7.1 ± 0.6 ng/g lipids) and the maximum value was 85.1 ng/g lipids. HCB was used as a fungicide and is released as an unintentional by-product during the production of chemicals and pesticides formulations. It has a half-life of 3 to 6 years in soil (ATSDR 2015). HCB was banned by the Lebanese government in 1998 and there is no evidence of its common use among the population especially that Lebanon is not an industrial country. However, low levels of HCB were detected in analyzed sediments from El Kabir Watershed in Akkar, north Lebanon (MoE/ELARD 2005).

The mean level of β-HCH in the present sample was 8.6 ± 0.6 ng/g lipids and the maximum concentration observed was 154.9 ng/g lipids. β-HCH is still present in the environment as an unintentional by-product during the production of lindane (γ-HCH), used historically as insecticide and a common treatment for scabies and lice. β-HCH is slowly metabolized, accumulates in fatty tissues, and has a half-life of 7 years. The general population is mainly exposed to β-HCH via dietary intake of animal food sources (Agency for Toxic Substances and Disease Registry (ATSDR) 2005; Stockholm Convention 2008b; Centers for Disease Control and Prevention 2009). Limited data on β-HCH exist in Lebanon except for one study, which detected levels of HCH in grape samples processed into a Lebanese alcoholic drink: "Arak." These levels however were below the maximum residue levels (MRL) (Kawar and Dagher 1976).

The geometric means of DDT and DDE were 2.1 ± 0.3 ng/g lipids and 18.9 ± 0.9 ng/g lipids, respectively, and their maximum concentrations were 15.8 and 630.9 ng/g lipids. DDT contributed to only 10% of the sum of DDT and DDE (∑DDT,DDE) (21 ng/g lipids) whereas DDE predominated and contributed to 51.64% of the sum of the four OCPs with no differences between men and women (Fig. 1). DDT was commonly used in Lebanon as an outdoor insecticide by

Table 1 Characteristics of the population (n = 314)

Variables	Total sample		Men (n = 133)		Women (n = 181)		p value
	N	%	*N*	%	*N*	%	
Age (mean in years ± SD)	25.6 ± 0.15		25.3 ± 0.14		25.9 ± 0.13		0.543
Governorate of residence							0.088
Beirut	104	34.4	49	36.8	59	32.6	
Mount Lebanon	192	63.6	77	57.9	121	66.9	
North	2	0.7	3	2.3	0	0.0	
Bekaa	1	0.3	1	0.8	0	0.0	
South	2	0.7	1	0.8	1	0.6	
Nabatieh	1	0.3	2	1.5	0	0.0	
Crowding index[a]							0.976
≤1	251	79.4	106	79.7	144	79.6	
>1	65	65	27	20.3	37	20.4	
Domestic use of pesticides							0.124
Yes	140	44.6	66	49.6	74	40.9	
No	174	55.4	67	50.4	107	61.5	
Smoking							0.275
Non-smoker	189	60.2	61	45.9	128	70.7	
Previous smoker	19	6.1	7	5.3	12	6.6	
Current smoker	106	33.8	65	48.9	41	22.7	
BMI (mean in kg/m^2 ± SD)	23.6 ± 0.07		25.1 ± 0.07		22.7 ± 0.07		0.000
BMI classification (kg/m^2)							0.000
Underweight (<18.5)	20	6.4	7	5.3	12	7.2	
Normal weight (18.5–24.9)	177	56.4	57	42.9	120	66.3	
Overweight (25–29.9)	91	29.0	52	39.1	39	21.5	
Obese (≥30)	26	8.3	17	12.8	9	5.0	
Waist circumference (mean in cm ± SD)[b]	84.1 ± 11.1		88.8 ± 11.6		80.6 ± 9.2		0.000
Waist circumference (%)[b]							0.018
<80 ♀ and 94 ♂	191	60.8	91	68.4	100	55.2	
≥80 ♀ and 94 ♂	123	39.2	42	31.6	81	44.8	
Percentage of body fat (mean ± SD)[c]	26.6 ± 9.7		21 ± 8.2		30.7 ± 8.7		0.000
Percentage of body fat (%)[d]							0.009
<30 ♀ and 25 ♂	171	56.6	83	48.5	88	33.6	
≥30 ♀ and 25 ♂	131	43.4	44	51.1	87	66.4	
Fish and shellfish consumption[c]							0.129
<2 portion per week	48	15.9	16	12.6	32	18.3	
2–4 portions per week	112	37.1	43	33.9	69	39.4	
>4 portions per week	142	47	68	53.5	74	42.3	

Table 1 (continued)

Variables	Total sample		Men (n = 133)		Women (n = 181)		p value
	N	%	*N*	%	*N*	%	
Red meat consumption (including organ meat and cured meat)[c]							0.000
<6 portions per week	103	33.9	26	20.5	77	43.5	
6–10 portions per week	104	34.2	32	25.2	72	40.7	
>10 portions per week	97	31.9	69	54.3	25	15.8	
Poultry consumption[c]							0.000
<3 portions per week	115	38.1	34	26.8	81	46.3	
3–4 portions per week	94	31.1	34	26.8	60	34.3	
>4 portions per week	93	30.8	59	46.5	34	19.4	
Eggs consumption[c]							0.002
<1 portion per week	87	28.8	22	17.3	65	37.1	
1-2portions per week	111	36.8	37	29.1	74	42.3	
>2 portions per week	104	34.4	68	53.5	36	20.6	
Dairy products consumption[c]							0.051
<2 portions per day	85	28.1	27	21.3	58	33.1	
2–3 portions per day	97	32.1	48	37.8	49	28.0	
>3 portions per day	120	39.7	52	40.9	68	38.9	
Milk consumption[c]							0.214
<1 portion per week	96	31.8	34	26.8	62	35.4	
1–4 portions per week	105	34.8	50	39.4	55	31.4	
>4 portions per week	101	33.4	43	33.9	58	33.1	
Fruits and vegetables consumption[c]							0.942
<5 portions per day	180	59.6	76	59.8	104	59.4	
≥5 portions per day	122	40.4	51	40.2	71	40.6	

All quantitative variables were normalized by calculating the geometric mean ± SD; $p < 0.05$ was considered as significant

SD standard deviation, *BMI* body mass index

[a] Crowding index was defined as the total number of co-residents per household, excluding newborn infants, divided by the total number of rooms, excluding the kitchen and the bathrooms

[b] Values ≥94 cm for men and 80 cm for women were used as cutoff points for waist circumference according to the International Diabetes Federation (IDF) criteria (Alberti et al. 2009)

[c] $n = 302$

[d] Values >25% for men and 30% for women were used to indicate high level of body fat (Lysen and Israel 2012)

Table 2 OCP concentrations (ng/g lipids) found in human serum samples ($n = 314$)

	% > LOD	% > LOD		GM	Min	50th percentile	95th percentile	Max
		Men	Women					
HCB	50	50.4	49.7	7.1 ± 0.6	<LOD	5.8	32.0	85.1
β-HCH	50	50.4	49.7	8.6 ± 0.6	<LOD	7.6	45.0	154.9
DDT	49.7	50.4	49.2	2.1 ± 0.3	<LOD	2.0	5.0	15.8
DDE	50	50.4	49.7	18.9 ± 0.9	<LOD	17.3	180	630.9

Geometric means were used in order to normalize the distribution. No significant differences observed between men and women ($p > 0.05$)

OCPs organochlorine pesticides, *LOD* limits of detection, *GM* geometric mean, *HCB* hexachlorobenzene, *β-HCH* β-hexachlorocyclohexanes, *DDT* p,p′-dichlorodiphenyltrichloroethane, *DDE* p,p′-dichlorodiphenyldichloroethylene

municipalities as well as in households and was largely used to treat hair and body lice (MoE/ELARD 2005; CDC 2009). It is metabolized in the human body into DDE, which persists for several years longer than DDT. In fact, DDE has a half-life reaching 16 years in temperate regions and is an indicator of the past exposure to DDT (ATSDR 2002; CDC 2009). Even though the Lebanese government banned its usage in 1982, the civil war between 1975 and 1990 made the enforcement of this law difficult and the smuggling of these chemicals possible. In fact, in a study measuring 113 soil samples from agricultural regions around Lebanon for total DDT residues, 40 samples were tested positive; 50% of them were located in the coastal plain, 44% in the Bekaa valley, and 38% in Mount Lebanon (Bashour et al. 2004). Most of the detected levels were below the "total threshold limit concentration" of 1000 ng/g (permissible levels for agricultural use), which identifies whether waste is hazardous or not (Martz 1992). Only two samples had levels exceeding 1000 ng/g and these samples were originally from the South Caza, in Saida (Sidon), located in the South Governorate of Lebanon. Three sources of contamination were postulated: an old DDT storage site, a small-scale dumping site, and an accidental disposal of DDT (MoE/ELARD 2005). In another study, sediments from El Kabir Watershed in Akkar, north Lebanon, were tested for the presence of selected pesticides. Small levels of DDT and DDE were detected in all analyzed samples (MoE/ELARD 2005). Moreover, Chbib et al. (2017) found that groundwater on the plain of Akkar was remarkably contaminated by DDT and HCH from recent use of these molecules.

Predictors of OCP concentrations

Sociodemographic, anthropometric, and dietary predictors of OCP concentrations are shown in Table 3. We did not observe significant differences in OCP levels between men and women. Similar results have been observed in Greece and Turkey with regard to HCB and β-HCH (Kalantzi et al. 2011; Ulutaş et al. 2015). On the contrary, women had higher levels of the four OCPs compared to men in France and Spain (Fréry et al.

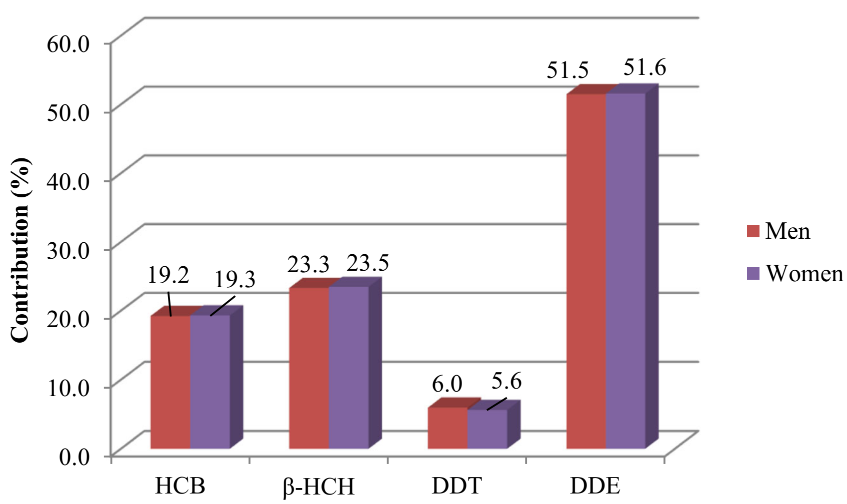

Fig. 1 Contribution of HCB, β-HCH, DDT, and DDE to the sum of the four OCP concentrations in men and women ($n = 314$). Statistical test used: chi-square test; $p > 0.05$. *HCB* hexachlorobenzene, *β-HCH* β-hexachlorocyclohexanes, *DDT* p,p′-dichlorodiphenyltrichloroethane, *DDE* p,p′-dichlorodiphenyldichloroethylene, *OCPs* organochlorine pesticides

Table 3 Geometric means (±SD) of OCP concentrations (ng/g lipids) according to categories of sociodemographic, anthropometric, and dietary factors

	HCB	β-HCH	DDT	DDE
Gender				
Men	7.1 ± 0.6	8.6 ± 0.6	2.2 ± 0.3	19.0 ± 0.9
Women	7.1 ± 0.6	8.6 ± 0.6	2.1 ± 0.3	18.9 ± 0.9
p for trend	0.979	0.995	0.52	0.982
Age				
17–24	7.5 ± 0.5	9.1 ± 0.6	2.2 ± 0.3	20.2 ± 0.9
25–39	8.0 ± 0.5	10.1 ± 0.6	2.3 ± 0.3	23.9 ± 0.9
40–65	4.9 ± 0.5	5.5 ± 0.6	1.7 ± 0.3	11.5 ± 0.9
p for trend	0.062	0.055	0.066	0.135
Pesticides usage				
Yes	7.2 ± 0.6	8.7 ± 0.6	2.1 ± 0.3	19.3 ± 0.9
No	7.0 ± 0.5	8.5 ± 0.6	2.1 ± 0.3	18.7 ± 0.9
p for trend	0.957	0.966	0.956	0.957
Smoking habits				
Non-smoker/previous smoker	7.9 ± 0.5[b]	9.7 ± 0.6[b]	2.2 ± 0.3	22.5 ± 0.9[b]
Current smoker	5.7 ± 0.7	6.7 ± 0.6	1.9 ± 0.3	13.5 ± 0.9
p for trend	0.025	0.034	0.109	0.033
BMI				
<18.5	11.2 ± 0.5[c]	13.9 ± 0.6	2.6 ± 0.3	36.4 ± 0.9
18.5–24.9	7.1 ± 0.5	8.6 ± 0.6	2.1 ± 0.3	18.8 ± 0.9
25–29.9	7.4 ± 0.5	9.1 ± 0.6	2.2 ± 0.3	20.9 ± 0.9
≥30	4.2 ± 0.5	4.8 ± 0.6	2.1 ± 0.3	8.7 ± 0.9
p for trend	0.066	0.083	0.129	0.098
Waist circumference				
<80 ♀ and 94 ♂	7.5 ± 0.5	9.2 ± 0.6	2.2 ± 0.3	20.8 ± 0.9
≥80 ♀ and 94 ♂	6.5 ± 0.6	7.7 ± 0.6	2.0 ± 0.3	16.4 ± 0.9
p for trend	0.312	0.282	0.273	0.316
Percentage of body fat[a]				
<30 ♀ and 25 ♂	7.9 ± 0.6	9.6 ± 0.6	2.3 ± 0.3	22.3 ± 0.9
≥30 ♀ and 25 ♂	6.3 ± 0.5	7.5 ± 0.6	1.9 ± 0.3	15.7 ± 0.9
p for trend	0.167	0.175	0.105	0.18
Fish and shellfish consumption[a]				
<2 portion per week	6.5 ± 0.5	7.8 ± 0.6	1.9 ± 0.3	16.5 ± 0.9
2–4 portions per week	7.4 ± 0.5	9.1 ± 0.6	2.2 ± 0.3	20.9 ± 0.9
>4 portions per week	7.3 ± 0.5	8.9 ± 0.6	2.2 ± 0.5	19.6 ± 0.9
p for trend	0.769	0.775	0.694	0.78
Red meat consumption (including organ meat and cured meat)[a]				
<6 portions per week	6.3 ± 0.5	7.3 ± 0.6	1.9 ± 0.3	15.2 ± 0.9
6–10 portions per week	7.5 ± 0.6	9.3 ± 0.6	2.2 ± 0.3	22.1 ± 0.9
>10 portions per week	8.1 ± 0.5	10.0 ± 0.6	2.3 ± 0.3	22.4 ± 0.9
p for trend	0.341	0.279	0.219	0.293
Poultry consumption[a]				
≤2 portions per week	7.1 ± 0.5	8.5 ± 0.6	2.1 ± 0.3	19.1 ± 0.9
3–4 portions per week	7.3 ± 0.5	8.9 ± 0.6	2.1 ± 0.3	19.6 ± 0.9
>4 portions per week	7.2 ± 0.5	8.9 ± 0.6	2.2 ± 0.3	19.7 ± 0.9
p for trend	0.977	0.958	0.797	0.993
Eggs consumption[a]				

Table 3 (continued)

	HCB	β-HCH	DDT	DDE
<1 portion per week	6.0 ± 0.5	7.2 ± 0.6	1.9 ± 0.3	15.2 ± 0.9
1-2portions per week	8.2 ± 0.5	10.2 ± 0.6	2.3 ± 0.3	23.5 ± 0.9
>2 portions per week	7.8 ± 0.5	9.2 ± 0.6	2.2 ± 0.3	21.5 ± 0.9
p for trend	0.163	0.181	0.174	0.257
Dairy products consumption[a]				
<2 portions per day	6.6 ± 0.5	8.0 ± 0.6	2.0 ± 0.3	17.5 ± 0.9
2–3 portions per day	8.7 ± 0.5	10.8 ± 0.6	2.4 ± 0.3	25.2 ± 0.9
>3 portions per day	6.6 ± 0.5	7.8 ± 0.6	2.0 ± 0.3	17.2 ± 0.9
p for trend	0.261	0.288	0.127	0.388
Milk consumption[a]				
<1 portion per week	5.6 ± 0.6	6.9 ± 0.6[d]	1.9 ± 0.3	14.2 ± 0.9
1–4 portions per week	7.0 ± 0.6	8.7 ± 0.6	2.1 ± 0.3	1.3 ± 0.9
>4 portions per week	8.9 ± 0.6	11.2 ± 0.6	2.4 ± 0.3	27.4 ± 0.9
p for trend	0.062	0.045	0.101	0.059
Fruits and vegetables consumption[a]				
<5 portions per day	6.9 ± 0.5	8.3 ± 0.6	2.1 ± 0.3	18.1 ± 0.9
≥5 portions per day	7.7 ± 0.5	9.5 ± 0.6	2.2 ± 0.3	21.9 ± 0.9
p for trend	0.472	0.453	0.642	0.429

Geometric mean was used for organochlorine pesticides (OCPs) concentrations in order to normalize the distribution; $p < 0.05$ was considered as significant

SD standard deviation, *OCPs* organochlorine pesticides, *HCB* hexachlorobenzene, *β-HCH* β-hexachlorocyclohexanes, *DDT* p,p′-dichlorodiphenyltrichloroethane, *DDE* p,p′-dichlorodiphenyldichloroethylene, *BMI* body mass index

[a] $n = 302$

[b] Significant difference between first/second categories ($p < 0.05$)

[c] Significant difference between first/fourth categories ($p < 0.05$)

[d] Significant difference between first/third categories ($p < 0.05$)

2013; Porta et al. 2010). Determining OCP levels among women is of particular importance because they have been positively associated with breast cancer (Charlier et al. 2003). Moreover, evaluating the exposure among women of childbearing age is important because OCPs can be transmitted via the breast milk to their descendants (CDC 2009). Age is known to be an important predictor of serum OCP levels (Becker et al. 2002; Porta et al. 2010; Fréry et al. 2013; Ben Hassine et al. 2014; Chovancová et al. 2014); however, this association was not reported in the present study (Table 3) probably due the low average age of the participants. The positive association between OCPs and age might be explained by age itself and a previous exposure effect. Older participants have had a longer time to accumulate the metabolites of these compounds in their body. In addition, they had higher level exposures to these compounds because they were lived during the period prior to the ban of production and use of certain OCPs such as DDT.

We found no association with domestic use of pesticides, while this factor has been proven to influence OCP levels in the French Nutrition and Health Survey (Fréry et al. 2013). This could be due to the ban of pesticides or to the limited presence of illegal pesticides in certain spots on the Lebanese territory. In relation to smoking, non-smokers or previous smokers had higher levels of HCB, β-HCH, and DDE than current smokers. This has been observed in prior studies as well (Fréry et al. 2013; Chovancová et al. 2014). This association might be explained by the fact that smoking can induce the enzymatic activity responsible of the OCP metabolism (Bachelet et al. 2011; Fréry et al. 2013).

In relation to BMI, there are conflicting results in the literature on the association between the obesity rate and OCP burden, with studies reporting negative, positive, or no association (Laden et al. 1999; Glynn et al. 2003; Wolff et al. 2005; Llop et al. 2010; Porta et al. 2010; Bachelet et al. 2011; Fréry et al. 2013). The results of this study showed no associations between OCP serum levels and BMI except for HCB levels. In fact, we observed a negative association between the lowest and the highest BMI categories and HCB concentrations. These findings might be due to the dilution effect of the OCP accumulation in the adipose tissue, which induces a decrease in the serum OCP levels (Lee et al. 2014). In fact, BMI reflects the body's adipose reservoir. Unmetabolized OCPs are stored in the adipose tissue, and for people with similar overall intake, those with more adipose tissue have lower serum concentrations of OCPs (Wolff et al. 2005). It is estimated that the highest concentrations of DDT and DDE in the human body are present in the adipose tissue (CDC 2009) and the prevalence of overweight and obesity in our sample was 29 and 8.3%, respectively. We did not observe any statistically significant associations with the four OCP serum concentrations and waist circumference or percentage of body fat.

The primary source of exposure of the general population is through the food chain. OCPs are lipophilic substances that tend to accumulate in fatty tissues, which lead to their biomagnification in the food chain, mainly fish, dairy, and meat products (CDC 2009). We did not observe a significant association between the four OCP levels and fish and shellfish consumption. This result could be due to the low level of fish and shellfish consumption by the study participants. Moreover, data from 1988 showed that DDT and DDE levels in Sultan Ibrahim (*Mullus barbatus*) and Ghobos (*Boops boops*), reaching a maximum of 0.1 mg/kg, are below the limit of 5 mg/kg set by the FDA (Abu Jawdeh 2006; FDA 2016). Bachelet et al. also found no association between DDE levels and fish consumption, but they observed a positive association with shellfish consumption (Bachelet et al. 2011) and so did the French Nutrition and Health Survey (Fréry et al. 2013). Mean OCP levels increased slightly with increasing red meat, eggs, and fruits and vegetables consumption, but the test for trend was not statistically significant. However, we found a positive association between the lowest and highest milk consumption category and β-HCH. A positive association was observed in the French survey with consumption of dairy products (Fréry et al. 2013).

OCP serum concentrations and comparison to international studies

The comparison with worldwide studies shows that the mean serum concentration of HCB was lower than the values reported in Tunisia, Greece, Italy, Turkey, Spain, France, Germany, Belgium, Canada, and USA. However, β-HCH mean concentration was higher than the values observed in Canada and USA (8.6 ± 0.6 ng/g lipids vs 6.4 and 7.9 ng/g lipids, respectively) and DDT and DDE mean concentrations were also higher than the values found in Turkey, Canada, and USA (Health Canada 2010; CDC 2015; Ulutaş et al. 2015) (see Online Resource 2 for more details).

OCP serum concentrations and health-based guidance values

Human biomonitoring survey data are used to evaluate human exposure to chemical contaminants. They can be interpreted using HBM-related guidance values currently developed in Germany and USA. In Germany, these guidance values are referred to as HBM-I and HBM-II and they are based on exposure data of the general population whereas biomonitoring equivalents were developed in the USA based on toxicokinetics extrapolated from animal studies. Biomonitoring equivalents reference dose (BE_{RfD}) and biomonitoring equivalents point of departure (BE_{POD}) rather than HBM-I and HBM-II values were established for OCPs (Choi et al. 2015).

The interpretation of the present study in combination with BE values will allow an initial screening of the Lebanese sample exposure to the selected OCPs. In fact, the concentrations of HCB, DDT, and DDE on the 95th percentile were 32, 5, and 180 ng/g lipids, respectively, much lower than the BE_{RfD} and BE_{POD} values (340 and 3384 ng/g lipids for HCB and 5 and 16 μg/g lipids for the sum of DDT and DDE (∑DDT,DDE) (Choi et al. 2015). These findings exclude any appreciable health risk related to the selected OCP exposure in the present population. Moreover, these concentrations at the 95th percentile were also much lower than the values associated with diabetes risk by Pal et al. (36.4 ng/l for HCB, 44.12 ng/l for β-HCH, 11.15 ng/l for DDT, and 97.2 ng/l for DDE vs 8.3 μg/l for HCB, 2 μg/l for β-HCH, and 396.9 μg/l for DDE) (Pal et al. 2013).

The risk of breast cancer associated with OCP exposure was examined by Charlier et al. (2003) who evaluated the concentrations of HCB and the ∑DDT,DDE in a sample of breast cancer patients and healthy women. The mean levels of OCPs among breast cancer patients were 90 ng/g lipids for HCB and 1830 ng/g lipids for ∑DDT,DDE much higher than the values reported in this study for women of childbearing age (7.6 ng/g lipids for HCB and 23.8 ng/g lipids for ∑DDT,DDE).

Strengths and limitations of the study

This research aimed to assess, for the first time, serum concentrations of HCB, β-HCH, DDT, and DDE in a sample of Lebanese adults and to evaluate sociodemographic, anthropometric, and dietary predictors of the exposure. A limitation of this study is that the majority of the sample resided in Mount Lebanon and Beirut because of the conflicts and the endangering situation during the study period. Nevertheless, it gives a preliminary idea on the current exposure of the Lebanese population to the four selected OCPs. Finally, since this is a cross-sectional human biomonitoring study, the findings cannot be used to evaluate causal correlations but rather test associations.

Conclusion

In conclusion, our study revealed relatively low levels of OCPs when compared to worldwide concentrations. The levels were also lower than the biomonitoring equivalents (BE_{RfD} and BE_{POD}) which exclude the potential health risks related to OCP exposure in the study sample. Further research is required to evaluate serum concentrations of the measured four OCPs on a larger sample more representative of the Lebanese population and distributed on a wider range of areas located outside Beirut and Mount Lebanon. It will also be important for future investigations to include measures of other OCPs in the human serum.

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Compliance with ethical standards

Ethics approval This study was approved by the Ethics Committee of USJ (Reference number: USJ-2012-19) and informed consent was obtained from all participants prior to their recruitment into the study.

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

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