RESEARCH ARTICLE



Serum concentrations of polychlorinated biphenyls (PCBs) in a Lebanese population: ENASB study

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Abstract Polychlorinated biphenyls (PCBs) are persistent organic pollutants that are still routinely detected 30 years after their restriction in many countries. PCBs have been associated with several non-communicable diseases. They are best measured via human biomonitoring (HBM). The concentrations of six indicator PCBs (PCBs 28, 52, 101, 138, 153, and 180) were measured in the serum samples of 316 Lebanese students and employees from Saint Joseph University of Beirut, Lebanon, using gas chromatography coupled to an iron trap mass spectrometer detector. PCBs were detected in 56.3 to 59.2% of the serum samples. The sum of PCB (Σ PCBs) levels ranged from <LOD to 338.84 ng/g lipids, with a geometric mean level of 10.34 ± 0.98 ng/g lipids. The major contributor to the Σ PCBs was PCB 180. In the present study, the levels were, in general, lower than the values observed in several Western and European countries. No association was found between age and concentration of any of the PCBs. In terms of risk for health, the highest levels were lower than critical limits such as HBM I and II values. We observed an inverted Ushaped association between levels of serum PCBs and the risk of overweight/obesity (OR = 2.140; CI = 1.095-4.185;

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p = 0.026). Regarding potential food contributors, we found no relation between PCB levels and fish consumption and a moderate relation with dairy product consumption (moderate consumers of dairy products had higher PCB levels compared to lower consumers) ($16.92 \pm 0.1/6.92 \pm 0.12$; p = 0.025). The present study is the first to provide information regarding PCB levels in a Lebanese population. Larger studies are required in order to estimate the PCB exposure parameters of the Lebanese population.

Keywords Human biomonitoring · Polychlorinated biphenyls · Persistent organic pollutants · Lebanon

Introduction

Human exposure to persistent environmental chemicals is best determined via human biomonitoring (HBM) which detects levels of pollutants in human matrices such as blood, serum, urine, and others (World Health Organization (WHO) 2015). Combining biomonitoring to health risk assessment is of particular importance when it comes to pollutants that accumulate in the body for long periods of time (World Health Organization (WHO) 2015).

Several HBM programs have been launched in many western countries worldwide, in order to assess the nationwide concentration of specific persistent pollutants, notably the *German Environment Surveys*, the *French Nutrition and Health Survey* (ENNS), the *United States National Health and Nutrition Examination Survey* (NHANES), the *Canadian Health Measures Survey* (CHMS), and the *BIOAMBIENT.ES* in Spain, among others (Choi et al. 2015).

Polychlorinated biphenyls (PCBs) were identified among the initial 12 persistent organic pollutants (POPs) by the 2001 Stockholm Convention. Since then, they have been largely

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investigated due to their toxicity, resistance to degradation, long-distance transport, and accumulation in the ecosystem (2008 Stockholm Convention). Exposure to PCBs has been linked to serious health effects, such as certain types of cancer, and not only reproductive, immunological, and neurological effects but also obesity and type 2 diabetes mellitus (Centers for Disease Control and Prevention 2009; Crinnion 2011). PCBs can leak from old contaminated electrical devices in the power sector, buildings constructed prior to 1979, as well as landfills and incineration of refuse (Agency for Toxic Substances and Disease Registry (ATSDR), Public Health Service 2000; Ministry of Environment (MOE), United Nations Development Programme (UNDP), ECODIT 2011).

These pollutants can accumulate in the fatty human tissues and can be subsequently monitored in the human matrices (Tehrani and Van Aken 2014). The Community Bureau of Reference of the European Commission selected seven congeners among the 209, to be primarily monitored in environmental and biological matrices, known as "indicator PCBs" (INERIS 2011). These PCBs are 28, 52, 101, 118, 138, 153, and 180. In HBM studies, these POPs are most commonly monitored in blood, due to the blood's standardized sampling procedures, the familiarity of this intervention, as well as the pollutants' equilibrium with the human organs (World Health Organization (WHO) 2015). However, among the seven indicator PCBs, the dioxin-like PCB (118) is generally not included in the PCB monitored; thus, six non-dioxin-like indicator congeners are monitored in main HBM studies.

Given the highly toxic nature of the POPs and knowing that they have not been previously monitored in Lebanon, the aim of the present study was to determine, for the first time, the serum levels of six indicator PCBs (28, 52, 101, 138, 153, and 180) in a sample of students and employees of *Saint Joseph University*, Lebanon, and to consider these levels as first values of reference. Furthermore, possible difference in PCB levels by gender, age, and body mass index (BMI) was also evaluated. The results obtained were compared with data from other countries.

Materials and methods

Study design and population

The study design was a cross-sectional survey conducted between October 2013 and June 2015. For technical reasons, the study was done in Saint Joseph University campuses that are located in many areas across Lebanon.

A list of currently enrolled students and employees was obtained from the university's records and registration department. Three hundred sixteen students and employees were randomly selected for recruitment using an automated random selection procedure. To be eligible for the study, the participants had to be Lebanese, aged between 17 and 65 years, and should have spent the last 10 years in Lebanon. Each participant completed a "face-to-face" questionnaire comprising sociodemographic information, details about their housing locations, smoking, and dietary habits. In order to determine the geographical vicinity to the sources of pollution, the distance in kilometers (km) from each participant's housing location to the nearest source of pollution was determined. Participants were therefore categorized by living within or out of 5 km range from known sources of exposure.

A previously validated food frequency questionnaire (FFQ) was administered to the participants in order to assess the intake of 14 major food categories, over the last year, including culturally adapted food items. These categories comprised cereals and grains, dairy products, fruits, vegetables, legumes, meats including fish and shellfish, fast food, nuts and seeds, oils and fats, salty snacks, sweets, and beverages. The frequency of consumption of each food item was noted per day, week, month, or year, in addition to the number of portions consumed each time. The portion sizes of food were estimated using measuring cups and food models (Biró et al. 2002).

Height and weight were taken by previously trained dietitians, using a scale-mounted stadiometer. The weight was measured twice, and the mean value was calculated. The percentage of body fat was measured using the bioelectrical impedance analyzer *InBody 720*.

Ethical considerations

This study was approved by the Ethics Committee of *Saint Joseph University of Beirut* (USJ-2012-19). The participants were fully informed about the purpose and procedures of the study before reading and signing the informed consent form. The confidentiality of the results was maintained.

Blood sampling and storage

Blood samples were collected during the period of October 2013–June 2015 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 into two aliquots: the first one was analyzed for total lipids using an automatic biochemistry analyzer (HumaStar) and the second one was stored in *Eppendorf* tubes at –80 °C to be tested for PCBs at the Lebanese laboratories of *Industrial Research Institute* (IRI), using gas chromatography coupled to an ion trap mass spectrometer detector.

Sample preparation

The human serum samples (10 ml) were homogenized manually by shaking for 1 min; 5 ml of the human serum was centrifuged. One hundred microliters of internal standard (103 PCB + PCB 193) (125 ng/ml in isooctane) was added, and the samples were sonicated for 20 min. Then, the samples were stored overnight at 4 °C before adding 1.5 ml formic acid and 2 ml demineralized water.

Sample extraction

The solid-phase extraction (SPE) cartridge was washed and conditioned with 5 ml dichloromethane (DCM) and 5 ml (5% methanol-water) with a flow of 1.5 ml/min; the sample extract was added to the SPE cartridge with a flow of 0.4 ml/min. Then, the cartridge was dried under a nitrogen stream at 20 psi for 10 min, followed by centrifugation (15 min, 4000 rpm); the PCBs were eluted with 5 ml hexane followed by 3 ml DCM, and the extracts were concentrated to approximately 1 ml in a water bath (\leq 40 °C) under a nitrogen flow.

Sample purification

The samples were cleaned up over an Al_2O_3 column, fractioned over a 1.5% (*w/w*) deactivated silica column, and then purified over an acidic silica column (40% H₂SO₄-silica).

Instrumental analysis

Analysis of PCBs was performed using a gas chromatograph (Agilent Technologies) equipped with a split/splitless injector port and an electron capture detector. The injector was operated in the splitless mode (1.5 min). Helium (He) 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 CP8753) (60 m \times 0.25 mm, i.d.; 0.25 μ m film thickness) capillary column. The temperature program used was set at 90 °C initially for 3 min and increased at 30 to 200 °C/min for 15 min, then at 5 to 265 °C/min for 5 min, and finally, at 3 to 27 °C/min for 15 min. The detector was with an N2 makeup gas. A CP-Sil 19 CB (Agilent Chrompack CP8722) (60 m \times 0.25 mm, i.d.; 0.25 μ m film thickness) capillary column was used as a second column to confirm the identifications of PCBs in the human serum. The corresponding limit of detection (LOD) was 1.4 ng/l of serum (0.27 ng/g lipids).

Statistical analysis

Given that more than 30% of the values were not detected, a multiple imputation (MI) analysis was carried out. All missing data were created by imputing random values after generating multiple linear regression and creating standardized β which provides M estimators of the parameters of interest. The independent predictors used were age, BMI, fish, shellfish, and dairy product consumption. The upper bound was fixed and

known in advance (1.4 ng/l). PCBs with concentrations below the LOD were also reported as LOD/2, and the results were compared to those obtained from the MI method. Resulting PCB concentrations were adjusted for total lipids. Continuous variables were determined as means and standard deviations while categorical variables were determined as frequencies and percentages. Geometric means (GM) (\log_{10} and square root of quantitative variable) was used in case of nonnormality of distribution. Bivariate analysis (Spearman test) was carried out to determine the linear correlation between continuous variables.

Results and discussion

Characteristics of the study population

Demographic and anthropometric characteristics of the participants are summarized in Table 1.

All quantitative variables were normalized by calculating the geometric mean \pm SD.

Elevated percentage of body fat is defined by a percentage of fat $\geq 25\%$ for men and 30% for women (Lysen and Israel 2012).

The study sample was 42.7% men and 57.3% women. The mean age of the participants was 25.6 years. Of our participants, 97.5% reside in Beirut and Mount Lebanon. It was very difficult to include participants from Bekaa and North and South Lebanon because of the conflicts and the endangering situation during the study period. Fifty percent live within 5 km from an ongoing or accidental source of exposure. The BMI of the participants varied between 16.9 and 45.9 kg/m² with a mean value of 25.4 kg/m² for men and 23.0 kg/m² for women. The mean percentage of body fat was 20.9% in men and 30.7% in women. 60.1% were non-smokers, while 39.9% had ever smoked.

Polychlorinated biphenyl concentrations in the serum samples

In order to determine which value to attribute to the PCB concentrations below the LOD, the results obtained from MI and LOD/2 methods were compared (Tables 2 and 3). Given that no differences were found, LOD/2 was used as a replacement for all missing values obtained while doing our analytical analysis for PCBs.

Table 4 shows the concentrations of PCB in the human serum as well as the percentage of measurements above the LOD for each congener. Since PCB concentrations depend on the amount of lipid in the sample, the results are expressed as nanograms per gram serum lipid. The mean level of lipid in the serum was $523.6 \pm 120.5 \text{ mg/dl}$. The concentrations of individual serum PCB congeners were above the LOD in

Table 1Characteristics of the study population (n = 316)

Variables	Number	Percent
Gender		
Male	135	42.7
Female	181	57.3
Age (mean in years \pm SD)	25.6 ± 0.2	
Men	25.2 ± 0.1	
Women	25.9 ± 0.2	
Governorate of residence		
Beirut	110	34.8
Mount Lebanon	198	62.7
North Lebanon	3	0.9
Bekaa	1	0.3
South Lebanon	2	0.6
Nabatieh	2	0.6
Geographical vicinity to source of	exposure	
≤5 km	160	50.6
>5 km	156	49.4
BMI (mean in $kg/m^2 \pm SD$)	24 ± 4.3	
Men	25.4 ± 4.3	
Women	23 ± 4.1	
BMI classification (kg/m ²)		
Underweight (<18.5)	21	6.6
Normal weight (18.5–25)	177	56.0
Overweight (25–30)	92	29.1
Obese (>30)	26	8.2
Percentage of body fat (mean \pm SI))	
Men	20.9 ± 8.2	
Women	30.7 ± 8.7	
Percentage of body fat (%)		
$<30 \stackrel{\bigcirc}{_{-}}$ and 25 $\stackrel{\nearrow}{_{-}}$	173	57.1
\geq 30 \bigcirc and 25 \Diamond	130	42.9
Smoking		
Non-smoker	190	60.1
Previous smoker	19	6.0
Current smoker	107	33.9
Fish and shellfish consumption		
≤ 1 portion per week	49	16.1
2–4 portions per week	112	36.7
≥5 portions per week	144	47.2
Dairy product consumption		
<2 portions per day	85	28.0
2-3 portions per day	99	32.6
>3 portions per day	120	39.5

56.3 to 59.2% of the cases. PCB congeners 138, 153, and 180 predominated and accounted for about 15.7, 25.2, and 34.1% of the amount of the indicator congeners analyzed. This is in agreement with previous findings (Hirai et al. 2005; Černá et al. 2008; Henríquez-Hernández et al. 2011; Kalantzi et al. 2011; Zani et al. 2013; Pavuk et al. 2014; Ben Hassine et al. 2014; Huetos et al. 2014; Ulutas et al. 2015). The main contributor was PCB 180, similar to the BIOAMBIENT study (Huetos et al. 2014), whereas the main contributor found in the other studies was PCB 153 (Kalantzi et al. 2011; Pavuk et al. 2014; Ben Hassine et al. 2014; Ulutas et al. 2015). In fact, the higher the chlorination, the more a congener is persistent and resistant to biodegradation. PCB 180 has the highest number of chlorine atoms with seven atoms, while PCBs 138 and 153 have six atoms, PCB 101 has five, PCB 52 has four, and PCB 28 has three (Agence de l'eau Seine-Normandie et al. 2009). The lowly chlorinated congeners are more rapidly metabolized by the human body, while the highly chlorinated congeners tend to bioaccumulate in serum lipids and in the adipose tissue (Grimm et al. 2015). PCBs 25, 52, and 101 can reach half-lives of 5 years in the human body. In contrast, the highly chlorinated PCBs can persist for 10 to 47 years (Agency for Toxic Substances and Disease Registry (ATSDR), U.S. Department of Health and Human Services, Public Health Service 2000; European Food Safety Authority (EFSA) 2005; Ritter et al. 2010). This explains the high contribution of PCBs 138, 153, and 180 (75%) to the total PCB level. Another contributor is the source of contamination. Historically, technical mixtures of PCBs were manufactured under several trademarks, among which the most widely used was Aroclor (Monsanto Company, USA). Several types of Aroclor mixtures exist depending on the composition of chlorine in PCBs (Agency for Toxic Substances and Disease Registry (ATSDR), U.S. Department of Health and Human Services, Public Health Service 2000; Agence de l'eau Seine-Normandie et al. 2009). In particular, Aroclor 1260 is richer in PCB 180 than in PCB 153, whereas Aroclor 1254a contains more PCB 153 than PCB 180 (U.S. Environmental Protection Agency 2006). Clophen A-60 (Germany) and Phenoclor DP-6 (France) are similar in composition to Aroclor 1260 whereas Kanechlor 500 (Japan) is similar to Aroclor 1254 PCBs. When used as coolants in the electrical devices, Aroclor 1254 was used to fill transformers and capacitors while Aroclor 1260 was used to fill only transformers (Agency for Toxic Substances and Disease Registry (ATSDR), U.S. Department of Health and Human Services, Public Health Service 2000; Agence de l'eau Seine-

Table 2	Geometric means of the
PCBs ob	tained with MI and
LOD/2 r	nethods, respectively

	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	∑PCBs
Multiple imputation	0.96	0.28	0.55	1.92	3.01	3.49	10.22
ND = LOD/2	0.86	0.34	0.58	1.88	2.95	3.72	10.34

Table 3 Correlates of the \sum PCBs for both methods of calculation using linear regression

	∑PCBs (multiple in	nputation	ı)		\sum PCBs (ND = LOI	D/2)		
	Standardized beta	р	95% CI		Standardized beta	р	95% CI	
			Lower bound	Upper bound			Lower bound	Upper bound
Age (years)	-0.05	0.43	-0.02	0.01	-0.06	0.36	-0.01	0.00
Dairy products	0.04	0.50	-0.03	0.06	0.04	0.51	-0.03	0.06
Fish and shellfish	0.02	0.69	-0.05	0.08	0.02	0.70	-0.06	0.08
BMI	0.03	0.57	-0.02	0.03	0.04	0.54	-0.02	0.03

Normandie et al. 2009; INERIS 2011). Therefore, the higher contribution of PCB 180 in the present study to the detriment of PCB 153 could be due to the contamination in PCBs by transformers in the power sector.

The sum of PCB concentrations defined as the sum of all detected and quantified congeners ranged from <LOD to 338.8 ng/g lipids with geometric mean (\pm SD), arithmetic mean (\pm standard error), and median values of 10.3 \pm 0.98, 52.38 \pm 2.88, and 57.9 ng/g lipids, respectively. Additional data are given in Online Resource 1. The figure shows the geometric means, 95th percentile, and maximum values for the six PCB congeners as well as $\sum PCB_{138,153,180}$ and $\sum PCB_{138,153,180}$

Comparison of polychlorinated biphenyl levels with international values

Table 4PCB concentrations (in
nanograms per gram lipid) found
in human serum samples

(n = 316)

The comparison with worldwide studies shows that the mean concentration of \sum PCBs in the human blood from Lebanon was lower than the value found in Tunisia (Ben Hassine et al. 2014), Greece (Costopoulou et al. 2006; Kalantzi et al. 2011), Italy (Zani et al. 2013; Esposito et al. 2014), Spain (Henríquez-Hernández et al. 2011; Huetos et al. 2014), France (Fréry et al. 2013), Germany (Becker et al. 2002;

Becker et al. 2008), Czech (Černá et al. 2008), Canada (Health Canada 2010), and the USA (Centers for Disease Control and Prevention (CDC) 2015) (Table 5). In particular, the values of the lowly chlorinated congeners (PCBs 28, 52, and 101) were higher than the Turkish, Tunisian, and Canadian concentrations (Health Canada 2010; Ben Hassine et al. 2014; Ulutaş et al. 2015). In addition, the concentration of the lowly chlorinated PCB 101 was higher than the value observed in Greece (Kalantzi et al. 2011). Additional data are given in Online Resource 2. The graph compares the serum PCB concentrations (ng/g lipids) in the present study to those of various countries in the world.

The relatively low levels in the present study could be due to the fact that Lebanon had never been a PCB-producing country, even though there were several uses of PCBs in the past that are still considered an important source of PCBs, namely the transformers and capacitors in the power sector, as well as the uncontrolled dumping (Ministry of Environment (MOE) et al. 2011). In Lebanon, ongoing exposure sources of PCBs included PCB-contaminated electrical devices, transformers, and capacitors that were identified in two major thermal power plants, Zouk (north of Beirut) and Jiyeh, (south of Beirut) as well as in a specialized repair workshop of electrical hardware in Bauchrieh (all three regions located in Mount

	%	% >L0	OD	GM^{a}	AM	Min	50th	95th p	Max
	>LOD	Men	Women				percentile	percentile	
PCB 28	56.3	55.6	56.9	0.9	2.2	<lod< td=""><td>2.2</td><td>5.6</td><td>18.2</td></lod<>	2.2	5.6	18.2
PCB 52	58.2	58.5	58	0.3	0.6	<lod< td=""><td>0.3</td><td>1.5</td><td>17.4</td></lod<>	0.3	1.5	17.4
PCB 101	57.6	57.8	57.5	0.6	1.3	<lod< td=""><td>0.6</td><td>3.6</td><td>17.4</td></lod<>	0.6	3.6	17.4
PCB 138	59.2	58.5	59.7	1.9	7.1	<lod< td=""><td>8.2</td><td>16.7</td><td>50.1</td></lod<>	8.2	16.7	50.1
PCB 153	58.2	58.5	58	3	16.5	<lod< td=""><td>16.4</td><td>45.9</td><td>87.1</td></lod<>	16.4	45.9	87.1
PCB 180	58.2	58.5	58	3.7	24.7	<lod< td=""><td>24.1</td><td>66.8</td><td>170</td></lod<>	24.1	66.8	170
∑PCB _{138,153,180}	-	-	-	9	48.3	<lod< td=""><td>52.6</td><td>128</td><td>302</td></lod<>	52.6	128	302
∑PCBs	_	-	_	10.3	52.4	<lod< td=""><td>57.9</td><td>135</td><td>339</td></lod<>	57.9	135	339

No significant difference has been found between men and women (p > 0.05)

AM arithmetic mean

^a Geometric mean (GM) had been used in order to normalize the distribution

Table 5 PCB mean concentration	is in the present :	study and in vari	ous countries	s worldwide (expressed in r	ig/g lipids and	in ng/l serum)			
Country	Sample	Year	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	ΣPCBs	Reference
Lebanon	316	2013-2015	0.9	0.3	0.6	1.9	3	3.7	10.3	Present study
Lebanon ^a	316	2013-2015	4.4	1.7	2.9	9.5	14.9	18.8	63.8	Present study
Tunisia	113	2011-2012	<lod< td=""><td><tod< td=""><td>0.4</td><td>26.1</td><td>52</td><td>34.6</td><td>113</td><td>Ben Hassine et al. (2014)</td></tod<></td></lod<>	<tod< td=""><td>0.4</td><td>26.1</td><td>52</td><td>34.6</td><td>113</td><td>Ben Hassine et al. (2014)</td></tod<>	0.4	26.1	52	34.6	113	Ben Hassine et al. (2014)
Turkey	57	2010-2012	0.2	0.1	0.1	0.6	0.7	0.4	2	Ulutaș et al. (2015)
Greece										
Athens	105	2002 - 2004	1.4	0.04	0.3	33.9	59.8	61.3	157	Costopoulou et al. (2006)
Kozani	22		0.4	0.3	1.5	11.8	22.6	20.4	57	
Greece	61	2007	I	Ι	0.1	24.9	43.2	31.4	Ι	Kalantzi et al. (2011)
Italy	58	Ι	4.4	1	3.2	36.9	56.3	72.8	155	Esposito et al. (2014)
Italy	527	2003–2004	I	Ι	Ι	188	332	424	I	Zani et al. (2013)
France (ENNS)	386	2010	2.2	1	1.1	70	110	90	274	Fréry et al. (2013)
Canada (1st cycle) USA	1661–1668	2007–2009	<lod< td=""><td><pre><tod< pre=""></tod<></pre></td><td><tod< td=""><td>10.1</td><td>18.3</td><td>15.2</td><td>43.7</td><td>Health Canada (2010)</td></tod<></td></lod<>	<pre><tod< pre=""></tod<></pre>	<tod< td=""><td>10.1</td><td>18.3</td><td>15.2</td><td>43.7</td><td>Health Canada (2010)</td></tod<>	10.1	18.3	15.2	43.7	Health Canada (2010)
NHANES	1866–1897	2003–2004	4.9	2.7	1.7	17.7 ^b	19.8	15.1	44.1	Centers for Disease Control and Prevention (CDC) (2015)
Anniston, AL	353	2005-2007	2	I	I	I	176	106	Í	Pavuk et al. (2014)
African-American White	412		1.7	I	I	I	58.5	45.1	I	
Germany										
GerES III ^a	2818-2823	1998	I	I	I	420	680	440		Becker et al. (2002)
GerES IV ^a	1079	2003-2006	\sim	$\overline{\vee}$	$\overline{\nabla}$	89	129	65	283	Becker et al. (2008)
Spain (BIOAMBIENT)	1880	2009–2010	Ι	Ι	Ι	31.9	43.6	56	Ι	Huetos et al. (2014)
Canary Islands	100	2011	10.9	5.2	9.2	6.8	12.1	8.1	54.4	Henríquez-Hernández et al. (2016)
Permanent Moroccan residents	131		21	5.5	5.9	11.4	25.5	13.2	83.3	
Czech	202	2006	14	5	I	186	423	374	I	Černá et al. (2008)
Japan	24	2004	6.3°	2.8 ^{d, c}	5.7°	64.7°	171 ^c	89.7 ^c	140	Hirai et al. (2005)

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^a Expressed in nanograms per liter of serum ^b PCB 138 + 158

 $^{\rm c}$ Expressed in picograms per gram of whole blood $^{\rm d}$ PCB 52 + 69

Lebanon Governorate). The latter is the most critical hotspot. where damaged, reparable, and out-of-service transformers and capacitors are scattered on the site, as well as barrels and wells where the transformers' dielectric oil was emptied (Ministry of Environment (MOE) et al. 2005a, b). It was estimated that 1250 ton of PCB-contaminated oil is present in the Electricité du Liban network and in the Bauchrieh workshop (El Jisr et al. 2011). Despite these levels, Lebanon has been using PCB-free oil for several years (Ministry of Environment (MOE) et al. 2006) and ratified the 2002 Stockholm Convention on the phaseout of PCBs by 2025. Uncontrolled landfilling and waste incineration also constitute an important issue in Lebanon, primarily in three major locations, Na'ameh, Bsalim (both in Mount Lebanon), and Zahlé, a region in Bekaa (Ministry of Environment (MOE) et al. 2011). Accidental exposure sources included three power stations (Deir Ammar and Deir Nbouh in North Lebanon, and Baalbeck in Bekaa) and three substations in Mount Lebanon (Bsalim, Jamhour, and Hazmieh) that were hit and damaged during the war (Ministry of Environment (MOE) et al. 2005a, b). Other sources included electronic waste and burning tires (Ministry of Environment (MOE) et al. 2011), even though the main source is considered to be the energy sector (United Nations Environment Programme (UNEP) et al.).

PCB levels in the present study were higher than the observed values in Turkey. Even though Istanbul is an industrialized area of Turkey (Ulutaş et al. 2015), the detected PCB values were relatively low. The authors explained this difference by the low consumption of fish of their population and by the fact that previously analyzed environmental and food sample contained PCBs at levels below the safety limits. Turkey banned the use of PCBs between 1993 and 1996. In contrast, in Lebanon, a national implementation plan was prepared in 2006 for the phaseout of PCBs by 2025. The abovementioned sources of PCB in Lebanon as well as the relatively delayed phase-out actions might explain the higher values observed in our sample when compared to Turkey.

PCB and critical limits

PCB are now considered to be "group 1 human carcinogens" by the International Agency for Research on Cancer (IARC) (2016, vol. 107) and "reasonably anticipated to be carcinogens" by the National Toxicology Program (NTP) (Centers for Disease Control and Prevention (CDC) 2009). In fact, PCB levels have been associated, in occupationally exposed individuals, with elevated risks of several cancers, such as cancers of the digestive system and the liver, malignant melanoma, and others (Centers for Disease Control and Prevention (CDC) 2009; World Health Organization (WHO) 2003). Moreover, PCBs are endocrine disruptors that, at low

doses, imitate or antagonize hormones (Gore et al. 2015). They have been associated with reproductive effects in males and females by anti-estrogenic and anti-androgenic effects, with alteration of the thyroid function, as well as obesogenic and diabetogenic effects (Agence de l'eau Seine-Normandie 2008; Centers for Disease Control and Prevention 2009; Crinnion 2011; Hamers et al. 2011; Gore et al. 2015). PCB levels were also associated with a higher risk of hypertension (Donat-Vargas et al. 2015; Van Larebeke et al. 2015; Perkins et al. 2016; Bergkvist et al. 2016), with a lower level of HDL cholesterol (Perkins et al. 2016) and a higher risk of myocardial infarction (Bergkvist et al. 2016). Among the abovementioned non-dioxin-like markers (NDL-ms) PCB, associations were most frequently detected with PCB 138, PCB 153, and PCB 180 (Van Larebeke et al. 2015).

HBM I and HBM II values (3.5 and 7 µg/l) were assigned for the sum of the PCBs 138, 153, and 180 for women of childbearing age and for children (Umweltbundesamt 2015). In a study evaluating the association between levels of PCB and breast cancer risk, a serum PCB 153 concentration of $1.63 \mu g/l$ was considered critical (Charlier et al. 2003). In a study by Pal et al. (2013) that evaluated the association of PCB concentrations with diabetes, diabetics had a Σ PCB_{138,153,180} concentration of 395.3 µg/l. When comparing the values in the present study at the 95th percentile (0.6 $\mu g/l$ for $\Sigma PCB_{138,153,180}$ and 0.23 $\mu g/l$ for PCB 153) to the critical levels, we notice that the Lebanese values are 11 times lower for the HBM values and 7 times lower for the breast cancer critical level and 600 times lower than the PCB levels in diabetics. The $\sum PCB_{138,153,180}$ levels in our study among women at childbearing age (0.6 µg/l) are also 11 times lower than the relative HBM values. The French Agency for Food, Environmental and Occupational Health & Safety (ANSES) also published critical limits of total PCBs associated to a negligible risk of health effects for women of childbearing age and children (700 ng/g lipids) as well as for the general population (1800 ng/g lipids) (Agence Française de Sécurité Sanitaire des Aliments (AFSSA) 2010). The values at the 95th percentile in the present study (138.38 ng/g lipids for women of childbearing age and 134.58 ng/g lipids for the total sample) are 5 times lower than 700 ng/g lipids and 13 times lower than 1800 ng/g lipids.

Correlates of PCB levels

Table 6 shows the comparison of the mean concentration of PCBs with sociodemographic and lifestyle factors. We found no significant difference of PCB concentrations between genders; in the literature, PCB concentrations were found to be higher in men than in women (Costopoulou et al. 2006; Becker et al. 2008; Porta et al. 2010; Esposito et al. 2014; Ben Hassine et al. 2014; Huetos et al. 2014; Helmfrid et al. 2015). However, three studies found no gender differences, in

 Table 6
 Mean (± standard error)

 levels of PCBs (ng/g lipids)
 according to different

 sociodemographic and lifestyle
 groups

	∑PCB _{138,153,180}	p value	∑PCBs	p value
Gender		0.504		0.462
Male	9 ± 0.1		12.6 ± 0.1	
Female	9.1 ± 0.1		12.6 ± 0.1	
Age		0.504		0.473
17–24	9.1 ± 0.1		12.7 ± 0.1	
25–39	11.4 ± 0.1		15.5 ± 0.2	
40–65	7 ± 0.1		9.9 ± 0.2	
BMI		0.655		0.672
<18.5	8.5 ± 0.2		12.3 ± 0.3	
18.5–24.9	7.1 ± 0.1		10.3 ± 0.1	
25–29.9	14 ± 0.1		18.4 ± 0.1	
≥30	10.1 ± 0.2		13.5 ± 0.2	
Percentage of body fat (%)		0.434		0.391
$<30 \stackrel{\bigcirc}{_{-}}$ and 25 $\stackrel{\bigcirc}{_{-}}$	9.6 ± 1.1		13.4 ± 0.99	
\geq 30 \bigcirc and 25 \bigcirc	7.6 ± 1.1		10.7 ± 0.99	
Geographical vicinity of source of PCB		0.504		0.462
≤5 km	8.2 ± 0.1		11.5 ± 0.1	
>5 km	10 ± 0.1		13.9 ± 0.1	
Smoking habits		0.860		0.869
Non-smoker	8.2 ± 0.1		11.5 ± 0.1	
Previous smoker	10.9 ± 0.2		14.7 ± 0.3	
Current smoker	10.5 ± 0.1		14.5 ± 0.1	
Frequency of fish and shellfish consumption		0.657		0.642
≤ 1 portion per week	6.7 ± 0.2		9.7 ± 0.2	
2–4 portions per week	8.1 ± 0.1		11.4 ± 0.1	
≥5 portions per week	11.5 ± 0.1		15.7 ± 0.1	
Frequency of dairy product consumption		1.000		0.053
<2 portions per day	$9.9 \pm 1.1^\dagger$		$6.9\pm0.1^\dagger$	
2–3 portions per day	$22.2\pm0.9*$		$16.9\pm0.1*$	
>3 portions per day	10.7 ± 0.9		7.5 ± 0.1	

Geometric mean had been used for PCBs in order to normalize the distribution. Elevated percentage of body fat is defined by a percentage of fat $\geq 25\%$ for men and 30% for women (Lysen and Israel 2012)

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

*Significant difference between second/third categories (p < 0.05)

accordance with the results in the present study (Kalantzi et al. 2011; Fréry et al. 2013; Zani et al. 2013).

No association with age was observed in the present study. PCB concentrations had been positively related to age and were found to be higher in men than in women (Costopoulou et al. 2006; Becker et al. 2008; Ben Hassine et al. 2014; Huetos et al. 2014). However, one study found an inverse association between PCB concentrations and age among German children (Becker et al. 2008). This trend is probably due to the presence of PCBs coming from breastfeeding among these children that decrease as they become older. The average age in our sample is 25.58 ± 0.15 years, which is relatively low. Knowing that this mean age is lower than all previous studies except the GerES IV, this might have affected the association of serum PCBs with age. Additional data are given in Online Resource 3. It shows the variation of PCB concentration between genders across different age groups. Males have lower concentrations than females in all three age groups, the biggest difference being in the last group (age 40 to 65 years). However, the differences were not significant between the two genders (p > 0.05).

Regarding smoking status, we found no significant association, in concordance with two previous studies (Bachelet et al. 2011; Chovancová et al. 2014).

As for BMI, we found no differences in serum PCB levels among BMI categories, similar to previous studies (Ben Hassine et al. 2014; Huetos et al. 2014). We also found no significant differences in PCB levels among percent body fat categories. Nevertheless, when we compared BMI levels



Fig. 1 Mean BMI levels (kg/m^2) according to tertiles of serum $\sum PCB$ levels (ng/g lipids)

among tertiles of PCB concentrations, we found a significant inverted U-shaped association (Fig. 1), in concordance with a study by Lee et al. (2011). In fact, two factors influence PCB levels: body fat and consumption of fatty products. In our study, the second tertile of PCB levels was associated with a twice higher risk of overweight/obesity (OR = 2.140; CI = 1.095-4.185; p = 0.026), after adjustment for age, gender, smoking status, dairy products, and fish and shellfish consumption. This was also determined by Lee et al. (2011) who concluded that low serum PCB levels are associated with an increased BMI. This could be due to higher consumption of fatty food, accompanied with an increase in body fat, which leads to an accumulation of PCBs in the adipose tissue. The highest tertile was not significantly associated with overweight/obesity (OR = 1.274; CI = 0.635-2.556; p = 0.496). This could be due to the dilution effect of the PCB accumulation in the body fat, which induces low serum levels among obese people and, therefore, inverse correlations between BMI and PCB levels in cross-sectional studies. Weight fluctuation can also play a major role in determining serum PCB levels, via release of PCBs into the blood after weight loss due to the redistribution of PCBs from fatty tissues after weight gain (Lee et al. 2014). In addition, the average BMI is 24.01 ± 4.32 kg/m² which is relatively low; this could explain the absence of significant difference in serum PCB levels between BMI categories. As a matter of fact, previous findings regarding the association of the serum PCBs with obesity, levels of triglycerides, hypertension, risk of diabetes,

glycemia, and risk of metabolic syndrome followed an inverted U-shaped pattern (Lee et al. 2007, 2011, 2012). It was hypothesized that low doses of PCB might be more harmful than higher ones (Lee et al. 2007).

We found no significant association with the geographical vicinity of the source of pollution. In fact, the main source of PCB exposure for the general population is dietary, particularly fatty foods, which could explain the absence of association with this parameter (Centers for Disease Control and Prevention (CDC) 2009).

Fish and shellfish consumption was not significantly associated with PCB concentrations, as reported in only one previous study (Helmfrid et al. 2015). Kalantzi et al. (2011) previously demonstrated that fish consumption is a predictor for PCB levels, and Bachelet et al. (2011) reported a positive association only among older participants (>50 years). In comparison to the one in Bachelet et al.'s study, the mean age of the present study is much younger. The absence of association in the current study could be due to the low consumption levels of seafood. In fact, only 47.2% of our participants consume the minimal recommended seafood intake of 240 g per week (U.S. Department of Health and Human Services, U.S. Department of Agriculture 2015). Moreover, in the year 1988, analysis of PCB levels in two of the most frequently consumed species of sea fish (Mullus barbatus commonly known as Sultan Ibrahim and Boops boops commonly known as Ghobos) showed that the detected concentrations (62 and 19.5 ng/g) were below the former maximum limits of 2 mg/kg in the USA and in Europe (Food and Drug Administration (FDA) 1996; Ministry of Environment (MOE) et al. 2005a; Observatoire Régional de la Santé 2008). Nowadays, we suppose that the fish content in PCB is also expected to be lower than the European maximum limits of 75 ng/g of fresh weight (Commission Européenne 2011).

Further analysis was carried out to evaluate the association of serum PCB levels with salmon and tuna consumption, but the results were not significant.

Regarding dairy product consumption, results showed a higher mean of \sum PCBs and \sum PCB_{138,153,180} among participants who consumed two to three portions of dairy products per day, when compared to those who consumed less than or

Table 7	Spearman correlation
coefficien	nt (r) between PCB
concentra	ations

	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	∑PCBs
PCB 28	_	0.880^{a}	0.876 ^a	0.872 ^a	0.856 ^a	0.860 ^a	0.880^{a}
PCB 52	$0.880^{\rm a}$	_	0.920 ^a	0.893 ^a	$0.885^{\rm a}$	$0.877^{\rm a}$	0.903 ^a
PCB 101	0.876 ^a	0.920 ^a	-	0.919 ^a	$0.937^{\rm a}$	0.918 ^a	0.941 ^a
PCB 138	0.872 ^a	0.893 ^a	0.919 ^a	_	0.922 ^a	0.921 ^a	0.946 ^a
PCB 153	0.856 ^a	0.885^{a}	$0.937^{\rm a}$	0.922 ^a	_	0.952 ^a	0.974^{a}
PCB 180	0.860^{a}	$0.877^{\rm a}$	0.918 ^a	0.921 ^a	0.952 ^a	_	0.982 ^a
∑PCBs	0.880 ^a	0.903 ^a	0.941 ^a	0.946 ^a	0.974 ^a	0.982 ^a	—

^a Correlation is significant at the 0.01 level (two-tailed)

equal to two portions $(16.92 \pm 0.1/6.92 \pm 0.12 \text{ ng/g lipids},$ p = 0.025, and 22.15 $\pm 0.91/9.92 \pm 1.08$ ng/g lipids, p = 0.029). Fréry et al. (2013) previously reported a positive association between dairy product consumption and PCB levels. In fact, PCBs, particularly highly chlorinated congeners, accumulate in the fatty compartments of food, such as dairy products (Centers for Disease Control and Prevention (CDC) 2009), which would explain the positive association of serum PCBs with dairy product consumption. However, eating more than three portions of dairy products per day was associated with a lower concentration of serum PCBs when compared to two to three portions $(7.5 \pm 0.1/16.92 \pm 0.1 \text{ ng/g lipids},$ p = 0.038, and $10.69 \pm 0.92/22.15 \pm 0.91$ ng/g lipids, p = 0.042). This inverse association could be due to the relatively higher proportion of participants who frequently consume dairy products, with 72.1% of the participants consuming more than two portions per day. In Lebanon, no previous studies have been carried out in order to measure PCBs in dairy products.

Correlation between pairs of PCBs

A very high correlation was found among the six PCBs (Table 7). The highly chlorinated PCB congeners correlated substantially with one another with the highest correlation found between PCBs 180 and 153. However, the correlations between PCB 28 and other highly chlorinated PCBs were lower. The difference could be due to the rapid metabolism of the lowly chlorinated PCBs (Grimm et al. 2015) and to the consequently shorter half-life, as opposed to the highly chlorinated PCBs go through cycles of volatilization and atmospheric deposition, while the highly chlorinated PCBs tend to remain closer to the source of contamination (Agency for Toxic Substances and Disease Registry (ATSDR), U.S. Department of Health and Human Services, Public Health Service 2000; European Food Safety Authority (EFSA) 2005; Ritter et al. 2010).

Strengths and limitations of the study

The limitations of our study are the relatively small size, as well as the restricted governorates. It was very difficult to include participants from Bekaa and North and South Lebanon because of the conflicts and the endangering situation during the study period. However, this study gives an idea of the exposure to these pollutants in the student and employee population of the Saint Joseph University in Beirut.

Conclusion

The present study is the first study to monitor levels of serum PCBs in a Lebanese population sample, which allows us to draw a first conclusion on the exposure to PCB, as well as the

influence of age, diet, and geographical vicinity of the sources of pollution.

Our study revealed relatively low levels of PCBs when compared to concentrations in western countries. Levels were lower than HBM I and HBM II values for PCBs, as well as the reported critical limits for health effects. This excludes the potential risks related to PCBs. Nevertheless, low levels of PCBs were associated with an increased risk of obesity while higher levels of PCBs were not.

Moderate consumption of dairy products was associated with a higher level of serum PCB, whereas fish and shellfish consumption did not show significant associations.

Future research should focus on a larger sample more representative of the Lebanese population and distributed on a wider range of areas, especially the ones located outside Beirut and Mount Lebanon, for a better identification of the dietary and environmental sources of exposure. Further analysis should also be held on food matrices in order to determine the estimated daily intake of PCBs in our population.

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Compliance with ethical standards This study was approved by the Ethics Committee of Saint Joseph University of Beirut (USJ-2012-19). The participants were fully informed about the purpose and procedures of the study before reading and signing the informed consent form.

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

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