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Review

A review of organochlorine pesticides and polychlorinated biphenyls in Lebanon: Environmental and human contaminants

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HIGHLIGHTS

- We collected data from 12 studies of PCBs and OCPs levels in Lebanon.
- Akkar groundwater and Tripoli sediments have recorded high OCPs and PCBs levels.
- Lebanese soil studied at national level showed acceptable levels of DDT residues.
- A biomonitoring study in Beirut reported normal levels of PCBs and OCPs.

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ABSTRACT

The country of Lebanon banned organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in 1982 and 1997, respectively, and adopted the Stockholm Convention on persistent organic pollutants (POPs) in 2003. Compliance with the Stockholm Convention began immediately, and research related to POPs in Lebanon had already been completed. A National Implementation Plan for POPs was formulated and updated several times, and includes a national inventory of PCBs that were mainly detected in insulating oils and equipment in power stations. High levels of PCBs have also been detected in sediments from the Port of Tripoli, the second major sea port in Lebanon. High levels of OCPs, which are illegally smuggled into Lebanon and improperly handled and used by farmers, have been detected in underground and surface waters for many years. There have also been human biomonitoring studies of PCBs and OCPs; for example, in 1999, measurable amounts of DDE were found in breast milk, and a 2018 study reported measurable amounts of PCBs and OCPs in human serum. While these levels were well below concentrations observed in other countries, they were slightly higher than the levels observed by the National Health and Nutrition Examination Survey (NHANES). This review provides an overview of the available PCB and OCP data from Lebanon between 1999 and 2017. In total, 12 studies of PCBs and OCPs in environmental samples, human serum samples, and human milk samples are included in this review, and the results of these studies are compared in terms of geography and chronology.

Abbreviations: Arithm., arithmetical; av, average; BE, biomonitoring equivalent; CCME, Canadian Council of Ministers of the Environment; Cl, Chlorine; DDD, dichlorodiphenyldichloroethane; DDE, dichlorodiphenyldichloroethylene; DDT, dichlorodiphenyltrichloroethane; dw, dry weight; EDL, Électricité du Liban; EPA, Environmental Protection Agency; EC, European Commission; EU, European Union; GC-MS, gas chromatography-mass spectrometry; Geom., geometrical; HBM, human biomonitoring; HCB, hexachlorobenzene; HCH, hexachlorocyclohexane; γ-HCH, lindane; HBM, human biomonitoring; LSB, Lebanese Serum Biomonitoring; LOD, limit of detection; LOQ, limit of quantification; LP-GC-MS, low-pressure gas chromatography-mass spectrometry; Max, maximum; Min, minimum; MRL, maximum residue level; nd, non-detectable; nq, non-quantifiable; NHANES, National Health and Nutrition Examination Survey; OCPs, organochlorine pesticides; PEL, probable effect level; POPs, persistent organic pollutants; POCIS, polar organic chemical integrative sampler; PCBs, polychlorinated biphenyls; PCDD, polychlorinated dibenzo-p-dioxins; PCDF, dibenzofurans; rang, range; SC, Stockholm Convention; samp, sampling; SPE, solid phase extraction; TEL, threshold effect level; USA, United States of America; WHO, World Health Organization.

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Contents

1. Introduction

1.1. Human exposure to persistent organic pollutants (POPs)

Persistent organic pollutants (POPs) are liposoluble organic compounds that are resistant to environmental degradation; therefore, POPs persist in soil, water, sediments, and air (Mcginn, 2000; Marvin et al., 2002; Zhang et al., 2002). Persistent organic pollutants are volatile in hot climates, such that they can travel in the atmosphere and be deposited thousands of kilometers away, and they are harmful to land and aquatic animals that accumulate POPs in their adipose tissue before eliminating them very slowly (Ren et al., 2017; Braune et al., 2005; Barni et al., 2016; Tekin and Pazi, 2017). Humans are mainly exposed to POPs through food, water, and air, and they also accumulate POPs in their adipose tissue. Under some circumstances, human exposure to POPs, even at low levels, can result in increased cancer risk, reproductive disorders, alterations of the immune system, neurotoxicity, endocrine disruption, genotoxicity, and birth defects (WHO, 2018; Salihovic et al., 2012). These issues were discussed at the Stockholm Convention (SC) on POPs in 2001, which ended with an agreement to eliminate or severely restrict the production of POPs. Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) are two common types of POPs that are intentionally manufactured and are listed in the SC (Stockholm Convention, 2008).

1.2. Organochlorine pesticides (OCPs)

The use of OCPs has been severely restricted in accordance with the SC because of their persistence in the environment and their potential harmful effects on wildlife and humans (Zhou et al., 2008). Organochlorine pesticides are volatile and can enter the environment through agricultural applications, domestic use, disposal of contaminated wastes into legal or anarchic landfills, and emissions from stockpiles or manufacturing plants. These compounds can adhere to soil (Harner et al., 1999) and particles in the atmosphere (Bozlaker et al., 2009; Cindoruk, 2011), and in aquatic systems, OCPs adsorb to sediments and bioaccumulate in fish and aquatic animals. Because of their liposolubility, OCPs can concentrate in foods rich in fat (Bulut et al., 2011; Ahmad et al., 2010; Fenik et al., 2011), such as dairy products and fish; these are the main sources of human exposure (CDC, 2017; Al-Shamary et al., 2016).

1.3. Polychlorinated biphenyls (PCBs)

Polychlorinated biphenyls comprise 209 individual chlorinated biphenyls or “congeners,” which can contain one to eight chlorine atoms. These compounds have been used since the 1930s as insulating fluids in electric equipment and as additives in sealants. Worldwide, emissions of PCBs to the environment come from industries (Cardellicchio et al., 2007), improper disposal, leakage from landfills, improper incineration, volatilization (Cardellicchio et al., 2007; Smedes and de Boer, 1997), and port activities (Hong et al., 2005, 2006). Low-chlorinated PCBs are often metabolized and eliminated by humans, whereas highly chlorinated PCBs tend to bioaccumulate (Beyer and Biziuk, 2009; Borja et al., 2005). The Environmental Protection Agency (EPA) classifies PCBs as probable human carcinogens (US EPA, 2015) whereas the International Agency for Research on Cancer (IARC) classifies them as Group I human carcinogens (IARC; Lauby-Secretan et al., 2013); their numerous negative health effects (US EPA, 2015; Arfaeinia et al., 2017) make them a major toxicological concern worldwide (Cetin et al., 2017).

1.4. Aim of this review

This review provides an overview of the available PCB and OCP data from Lebanon between 1999 and 2017, since there has yet to be a comprehensive study of this issue. Twelve academic studies of PCBs and OCPs in environmental samples, human serum samples, and human milk samples are included in this review, and their results have been compared in terms of geography and chronology.

2. Materials and methods

The articles included in this review were selected using research engines such as PubMed, Ovid, Cochrane, Elsevier/ScienceDirect, Research Gate, and Google Scholar. Keywords such as POPs, PCBs, OCPs, dichlorodiphenyltrichloroethane (DDT), and hexachlorobenzene (HCB) were used with the word Lebanon, as well as names of matrices and other words of interest: environment, water, groundwater, sediments, abiotic, soil, biomonitoring, animal, marine, serum, and breast milk, among others. Google Scholar was used to find geographical information about Lebanon.

Data related to OCPs in rainwater and drinking water networks were excluded since they did not identify contamination or unsafe levels. Articles published prior to 1999 and those related to the validation of analytical techniques for measuring OCPs were also excluded. Diet is the major route of human exposure to OCPs (Wang et al., 2009, 2011; Luo and Zhang, 2010; Panuwet et al., 2009), but only one article related to food was found – an article investigating pesticide residues in low-fat foods of plant origin – and it was excluded because OCPs are more likely to accumulate in animal fat than foods of plant origin. Ultimately, 12 academic studies performed in Lebanon were included in this review (see Table S1 of the Supplementary Information for a summary of the available data on OCP and PCB research in Lebanon) and information on legal authorities' actions related to OCPs and PCBs were gathered from reports and interviews of the Lebanese Ministry of Environment.

3. Lebanon

Lebanon is located on the eastern Mediterranean coast between 33°03′–34°45′N latitude and 35°05′–36°30′9″E longitude. From west to east, Lebanon can be geographically divided into four areas: a coastal plain along the west, the Mount Lebanon mountain chain, the fertile Bekaa Plain, and the Anti-Lebanon mountain chain that runs along the eastern border with Syria; the fertile Bekaa Plain lies between the two mountain chains (Ministry of Environment, 2006). The area of Lebanon is 10,452 km^2 and its population, including foreigners, is 6.3 million people (CIA, 2018); hence, Lebanon has a high population density of 600 persons/km^2. In 2017, industry contributed 13.1% to the Gross Domestic Product, and agriculture, which employed more than 39% of the population in 2009 (Chbib et al., 2017), contributed 3.9% (CIA, 2018). Both of these sectors are considered to be major contributors to OCP and PCB contamination in Lebanon (Chen, 2007; Wilson and Tisdel, 2001; Nisbet and Sarofim, 1972).

4. Legal actions regarding OCPs and PCBs

Lebanon became a member of the SC in 2003 (Stockholm Convention, 2008), created national inventories of OCPs and PCBs, and formulated a National Implementation Plan in 2006 (Ministry of Environment, 2006) in order to develop efficient legal frameworks for eliminating or reducing the release of POPs to the environment. Updated inventories and reports identified OCP and PCB hotspots in the country (Chbib et al., 2017) and revealed serious gaps in law enforcement on this issue. Legal authorities' actions that aim to implement the SC are supposed to be complemented by research, as stated by the SC itself, and academic institutions have played a major role in detecting, measuring, and monitoring the listed hazardous chemicals (Chbib et al., 2017). Unfortunately, the lack of funds and other obstacles have limited their efforts to advance this issue, nevertheless they continue to carry out environmental monitoring and biomonitoring studies and enforce bridging with legal authorities in order to improve information and expertise exchange.

5. Organochlorine pesticides

5.1. OCP hotspots in Lebanon

Of the land area in Lebanon, 63.3% is arable but only 22–24% has been cultivated (Fig. S1 of the Supplementary Information) (CIA, 2018); cultivated land corresponds to major OCP hotspots, such as the Bekaa Plain, Akkar (in the northern coastal plain), and Tyre (in the southern coastal plain). During the past two decades, agricultural growth has been the result of expanded water availability, especially groundwater, and the excessive use of pesticides, up to 5.5 kg/ha. In 2007, the United Nations Economic and Social Commission for Western Asia reported that these pesticides included banned OCPs. Another estimation of pesticide use reported that more than 10.7 kg/ha of pesticides were used in Akkar in 2000 (Chbib et al., 2017).

Lebanon does not produce pesticides and relies on legal imports to meet its needs (Stockholm Convention, 2008; E//State an, 2011; Youssef et al., 2015); however, banned OCPs continue to be smuggled through legal airport and ports, as well as through uncontrolled parts of the northern and eastern borders with Syria. Other gaps in the regulation of OCPs are that the ministries of agriculture and environment are understaffed with respect to inspection personnel, municipalities and farmers do not hold proper records of pesticide use, and a huge number of farmers have poor knowledge about the harmful effects of OCPs, their persistence in the environment, and their potential to accumulate in humans and animals (ATSDR, 2018a). Farmers misuse and mishandle OCPs, and improperly discard their containers, all of which result in their release into the environment (Stockholm Convention, 2008). Therefore, Lebanon is currently incapable of controlling the smuggling of OCPs and the misuse of OCPs by farmers (Ministry of Environment, 2006). Furthermore, less than 0.1% of applied pesticides are estimated to reach their target crops, while the rest contaminate the air, soil, and water (Arias-Estévez et al., 2008). Hence, agricultural fields are potentially contaminated sites, as are the streams and rivers flowing into them. Gravity irrigation is the predominant method of irrigation in Lebanon; this method washes OCPs out of agricultural fields and introduces them into surface water and groundwater (Arias-Estévez et al., 2008). Despite agricultural growth, local agricultural production does not meet the needs of the country and the majority of the country's food needs are imported (Investment Development Authority of Lebanon, 2018); imported foods may also contribute to the ingestion of OCPs in Lebanon.

Accredited and renowned laboratories have not reported detecting OCPs in fruits, cereals, or vegetables since 2011; however, this does not mean they are no longer present, as only a portion of locally produced or imported crops are sampled and tested for OCP residues (Ministry of Environment, 2018). Academic research on OCPs in Lebanon is limited, but existing studies have been included in this review.

5.2. OCPs identified in Lebanon

The OCPs, impurities, and degradation products that have been identified and have measurements included in this review are: DDT and its residues, which include dichlorodiphenyldichloroethane (DDD); dichlorodiphenyldichloroethylene (DDE); aldrin; dieldrin; chlorobenzilate; β-endosulfan; endosulfan sulfate; endrin; endrin ketone; HCB; and hexachlorocyclohexane (HCH); isomers such as lindane (γ-HCH); heptachlor; heptachlor epoxides A and B; methoxychlor; and tetradifon. These compounds are all listed in the SC, except for chlorobenzilate and tetradifon (Stockholm Convention, 2008).

6. PCB hotspots in Lebanon

Polychlorinated biphenyls are present in 100% of adult Americans (Wahlang et al., 2014); hence, they are a public health concern in the United States of America (USA) and worldwide (US EPA, 2015). The Lebanese national inventory of PCBs revealed that insulating oils formerly used by Lebanon's electricity company “Électricité du Liban” (EDL), which accounts for 90% of the power sector, are the primary known source of PCBs in the country. While Lebanon never produced PCBs, local industries could still be an unintentional source of PCBs. Virtually all power plants, distribution stations, and workshops operated by EDL are hotspots for PCBs; these include 7 thermal power plants, 12 hydropower plants, and 58 distribution stations comprising 22,551 transformers that contain insulating oils across the country (Ministry of Environment, 2018 – updated data was not reported in the most recent, 2017 inventory).

Before PCBs were banned, insulating oils contained Aroclor 1260 and Clophen A60, which are both composed of highly chlorinated PCBs. Aroclor 1260 is 99% PCBs with five to eight chlorine atoms, and 21% of those are PCB180 (7 chlorines) and PCB153 (6 chlorines) (WHO, 2017), which likely bioaccumulate in humans (Yang et al., 2002; Hsu et al., 2014). Despite the use of PCB-free oils since 1997, 1130 transformers continue to reveal contamination, with PCB residues at levels exceeding 50 ppm (Ministry of Environment, 2018 – updated data was not reported in the most recent, 2017 inventory). This contamination is the result of cross contamination between old oil residues and PCB-free oils in transformers and EDL filtration units. The EDL repair shop in the suburbs of Beirut is the country's sole storage area for the disposal of damaged transformers (The World Bank, 2011), and is the primary PCB hotspot in Lebanon since it includes a water well where 55 tons of PCB-contaminated oil were dumped recklessly because Lebanon has no PCB incineration kilns (The World Bank, 2011). Other sources of PCBs could be the volatilization of old contaminated oil that EDL sold as fuel prior to the ban of PCBs, and the bombing and burning of seven EDL distribution stations during wars in the past four decades (The World Bank, 2011). In 2015, the Lebanese Ministry of the Environment began assembling contaminated EDL equipment and volumes of oil greater than 0.05 L containing more than 50 ppm PCBs. These items and oils were shipped to France for proper incineration in compliance with the SC, and this work is still on-going (The World Bank, 2011).

7. Previous studies on OCPs and PCBs

Research on OCPs and PCBs in Lebanon published between 1999 and 2017 covered abiotic matrices such as surface water, groundwater, river and sea sediments, soil, and biological matrices, namely human serum and breast milk.

7.1. Surface water

Lebanon has 16 non-navigable perennial rivers and 23 seasonal rivers or streams (MOE/UNDP/ECODIT, 2011). These rivers and streams all have sources within the country, except the Kebir River (77 km), and they all discharge in estuaries in Lebanon, except for two perennial rivers that flow abroad: the Orontes and the Hasbani-Wazzani. Four of them are longer than 50 km: the Orontes, the Hasbani-Wazzani, the Litani, and the Kebir River (Fig. 1). Each of these rivers and streams provide drinking and irrigation water and are vulnerable to contamination by OCPs and PCBs because of agricultural and industrial activities around their basins. However, only four rivers in Lebanon have been studied, namely the Orontes, the Hasbani-Wazzani, the Litani, and the Ibrahim.

The Orontes (572 km) flows north for 47 km in the Bekaa Plain, then crosses Syria and Turkey where it discharges into the Mediterranean Sea. The Hasbani-Wazzani crosses 25 km of agricultural lands in South Lebanon before flowing into the Jordan River. The Litani is 170 km long and flows exclusively in Lebanese territory. It is the longest river in the country and the lifeline of agriculture in central and southern Bekaa and South Lebanon, as the artificial

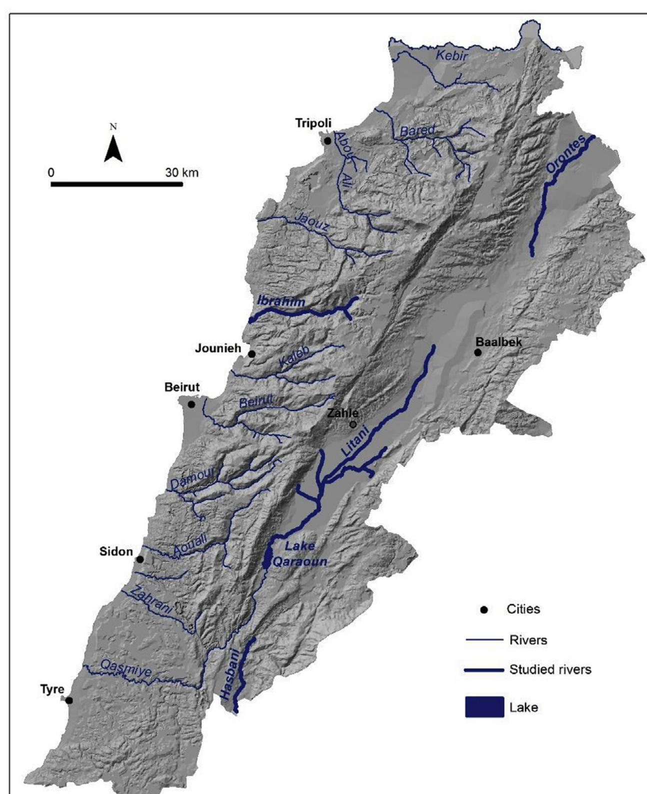

Fig. 1. Surface waters in Lebanon that have been sampled and analyzed for POPs (in bold).

Lake Qaraoun is located on the Litani and feeds three irrigation networks. Finally, the Ibrahim River is a 30 km long perennial river, that feeds irrigation projects in Mount Lebanon. Unauthorized discharge continues to occur in the Litani and Ibrahim rivers, and unlicensed projects continue to develop on their riverbanks (MOE/UNDP/ECODIT, 2011).

Four previous studies included the above-mentioned four rivers (Youssef et al., 2015; Kouzayha et al., 2013; Badr et al., 2014; Hneine et al., 2017) and each aimed to analyze OCPs, PCBs, non-OCPs, heavy metals, and bacteria in their surface waters. The SC lists 14 OCPs under Annex A (Prohibition & Elimination) and one under Annex B (Restriction); the majority of these OCPs were included in the four studies. Their metabolites were also included – such as DDE and DDD (the metabolites of DDT in Annex B of SC), endosulfan sulfate (the metabolite of endosulfan α and β), heptachlor epoxide (the metabolite of heptachlor) – since they are as persistent as their mother products, if not more so. The four studies also measured some OCPs that are not listed by the SC but are banned in the USA or the European Union (EU).

Sampling for three of the four studies was completed between 2012 and 2013 (Youssef et al., 2015; Kouzayha et al., 2013; Badr et al., 2014); duplicate or triplicate water samples were collected in opaque or amber glass bottles (1.5–2.5 L) from different sites on the targeted rivers at different periods of the year. The fourth study (Hneine et al., 2017), completed in 2015, used the Polar Organic Chemical Integrative Sampler (POCIS) to collect samples from three of the four rivers. The POCIS is a passive sampler equipped with a specific adsorbent (Oasis® hydrophilic-lipophilic balance [HLB] sorbent) that retains OCPs and other pesticides and is mainly used to monitor hydrophilic compounds with an octanol-water partition coefficient ($\log k_{ow}$) < 4 (Stockholm Convention, 2008; Alvarez et al., 2004; Herrero-Hernández et al., 2013). The POCIS can detect low concentrations of contaminants that cannot usually be detected through grab sampling by continuously collecting them onto the adsorbent phase (Stockholm Convention, 2008). In the fourth study, POCISs were deployed in triplicate every 14 days in three rivers, one deployment site in each: deployed in Ibrahim River for 12 months, on Lake Qaraoun for six months, and on the Hasbani for six months. In this study, DDE was the only OCP that could be calculated by the POCIS method, and whose concentration reached 23.16 ng/L in the Ibrahim River, 1.73 ng/L in the Lake Qaraoun and 15.43 ng/L in the Hasbani River. Table 1.

It was impossible to calculate the concentrations of β-endosulfan, heptachlor, and HCB in ng/L by the same method because the parameters needed for the calculation were not available. Therefore, the concentrations of β-endosulfan, heptachlor, and HCB have been expressed in ng/(g sorbent phase of POCIS) (Table 2) (Stockholm Convention, 2008). The four studies used either solid phase extraction (SPE) with low-pressure gas chromatography-mass spectrometry (LP-GC-MS) or gas chromatography-mass spectrometry (GC-MS) to measure OCPs and PCBs and overall, the four studies of the four rivers detected and/or quantified 13 OCPs and one PCB (Tables 1 and 2).

7.1.1. *Hasbani River*

Three of the four studies discussed above included the Hasbani River (Youssef et al., 2015; Badr et al., 2014; Hneine et al., 2017) and they sampled the river between 2011 and 2014. Four banned OCPs and one banned PCB congener were detected at measurable levels: DDE and HCB by all three studies, β-endosulfan by two studies, heptachlor by one study, and PCB52 by one study. The most abundant contaminant detected in three studies was DDE. In two

Table 1
Surface water results.

	Sampling in 2011–2012(Published 2013)ng/L			Sampling in 2013(Published 2014) ng/L		Sampling in 2013(Published 2015)ng/L		Sampling in 2014 (Published 2017)ng/L					
Authors	Kouzayha, A. et al.			Badr, R. et al.		Youssef, L. et al.		Ashi, A. et al.					
LOD & LOQ	LOQ ranged from 0.1 ng/L (Aldrin) to 2 ng/L (Endosulfan sulfate)			Not mentioned		LOQ ranged from 0.5 ng/L (HCB) to 2 ng/L (Endosulfan sulfate)		LOD and LOQ for DDE were 0.29 & 0.5 ng/L, respectively					
Method	SPE + GC-MS			SPE + GC-MS		SPE + GC-MS		SPE + LP-GC-MS/ MSTriplicate samples					
Location	Litani		Orontes	Hasbani – Wazzani[a]		Hasbani		Ibrahim		Qaraoun[b]		Hasbani	
Num. of sites	2		3	20		3		1		1		1	
Num. of samples	4		6	60		15		72		36		36	
Samp. Method	Grab sample Duplicates			Grab sample Triplicates		Grab sample		POCIS[c]		POCIS[d]		POCIS[d]	
OCPs/PCBs	av	Rang	Rang	av	Rang	av	Rang	av	Rang	av	Rang	av	Rang
PCB52[e]				64.75	35.43–121.61								
Aldrin						nq	nd–nq						
DDD	1	nd–3.4				1.29	nq–5.6						
DDE[f]	1.1	nd–2.7	< LOQ	2.03	1.12–4.99	23.25	nq–135.6	23.16	nd–137.66	1.73	nd–8.63	15.43	nd–31.79
DDT						nq	nd–nq						
Β-Endosulfan[g]				24.46	3.26–107.39								
Endosulfan sulfate	5.3	nd–9.2											
HCB			< LOQ	1.02	0.17–2.70	0.6	nq–0.7						
Heptachlor Epox. A						nq	nd–nq						
Heptachlor Epox. B						nq	nd–nq						
Lindane	7.8	nd–9.5											
Methoxychlor	1.8	nd–4.7	< LOQ										
Tetradifon	6.6	nd–8.6	< LOQ										

[a]The Hasbani River flows into the Wazzani River.
[b]Lake Qaraoun is an artificial lake on the Litani River.
[c]POCIS passive samplers deployed in triplicate every 14 days for 12 months.
[d]POCIS passive samplers deployed in triplicate every 14 days for 6 months.
[e]EPA Max for PCB52 = 1.7 ng/L.
[f]EPA Max for DDE = 8.3 ng/L.
[g]CCME PEL (the level above which more than 50% adverse effects occur in marine environments) for β-Endosulfan = 3 ng/L.

Table 2
Results of surface waters study sampled by the POCIS method.

Detected OCPs	2014 ng/g *(nanogram per gram of sorbent phase of POCIS)*					
Authors	Ashi A et al.					
Method	SPE + LP-GC-MS/MS					
	Triplicate samples					
	Since it was impossible to calculate the concentrations of β-endosulfan, Heptachlor, and HCB in ng/L using POCIS passive samplers because of lack of parameters, these compounds were discussed as mass (ng) accumulated in each g of POCIS sorbent phase.					
	Ibrahim		Litani[a] (Lake Qaraoun)		Hasbani	
	1 site (n = 72)(POCIS method)[b]		1 site (n = 36)(POCIS method)[c]		1 site (n = 36)(POCIS method)[c]	
	av	rang	Av	Rang	av	Rang
B-Endosulfan	26.67	nd−143.38	5504.90	nd−27201.42	50728.13	nd−142672.08
HCB	3.22	0.71−10.46	13.14	2.18−51.38	7.44	3.02−18.01
Heptachlor	2.72	nd−8.13	2.42	1−5.63	8.16	4−17.44

[a]Lake Qaraoun is an artificial lake on the Litani River.
[b]POCIS passive samplers deployed in triplicate every 14 days for 12 months.
[c]POCIS passive samplers deployed in triplicate every 14 days for 6 months.

studies, its levels far exceeded the maximum admissible level set by the EPA (8.3 ng/L) and it exhibited a positive trend over the years (Youssef et al., 2015; Badr et al., 2014; Hneine et al., 2017) that could indicate historical and/or recent use of DDT despite its ban. The second most abundant contaminant detected in the Hasbani was HCB, which indicates recent use in the Hasbani basin; recent use may have been intentional or unintentional since HCB can be used directly or exist as an impurity in other banned OCPs. The detected HCB levels were all below the upper limit that affects aquatic animals. B-endosulfan was the third most detected OCP in the Hasbani River and its levels in one study far exceeded 3 ng/L, the probable effect level (PEL) set by the Canadian Council of Ministers of the Environment (CCME) (Badr et al., 2014).

The PCB52 congener was detected in the Hasbani River (Table 1) at concentrations exceeding the safe limit fixed by the EPA for PCBs: 0.0017 μg/L (1.7 ng/L) (Badr et al., 2014; ATSDR, 2018b). In addition to its probable sources from the power sector, this PCB could have multiple sources that have not been accurately determined, as industrial sector and atmospheric PCBs have yet to be studied in Lebanon. The study by Badr et al. (2014) is the only study to measure a PCB congener in surface water in Lebanon.

7.1.2. Litani River and Lake Qaraoun

The Litani River has been covered by two studies: a study in 2012 grab sampled riverbed water (Kouzayha et al., 2013) and a study in 2015 sampled Lake Qaraoun using the POCIS method (Hneine et al., 2017). Nine OCPs were detected in measurable amounts by at least one of the two studies: DDD, DDE, endosulfan sulfate, β-endosulfan, HCB, heptachlor, lindane, methoxychlor, and tetradifon. The DDE levels were below the maximum limit set by the EPA in the 2012 riverbed water study, but were higher than this limit in the 2014 Lake Qaraoun study, where POPs could accumulate. B-endosulfan in Lake Qaraoun was measured in the adsorbent phase of the POCIS. There is no PEL for it in sediments, but its level there is of concern. In the Litani riverbed, the concentration of endosulfan sulfate was also considered to be high at 5.3 ng/L (Kouzayha et al., 2013), even though no upper limit is available. The concentration of endosulfan sulfate was evaluated relative to the PEL of β-endosulfan in surface waters, 3 ng/L, which was set by the CCME (PEL is the level above which more than 50% adverse effects occur in marine environments). The detection of endosulfan sulfate could indicate historical or recent use of endosulfan sulfate as well as β-endosulfan. Measurable levels of HCB, lindane, methoxychlor, and tetradifon were also detected in the Litani riverbed; HCB and lindane were within the acceptable levels set by the EPA, and reference values have yet to be set for methoxychlor and tetradifon (Tables 1 and 2). Measurable levels of methoxychlor and tetradifon in the Litani riverbed indicate that they have recently been used in the area.

7.1.3. Orontes River

Only one study included the Orontes River, wherein DDE, HCB, methoxychlor, and tetradifon were detected below the limit of quantification (LOQ) and the other OCP analytes were not detected (Kouzayha et al., 2013). No other studies have included the Orontes, which is probably due to its proximity to the Syrian border and proximal risks due to the war in Syria between 2012 and 2017 (Table 1).

7.1.4. Ibrahim River

The waters of the Ibrahim River have been included in one study, which revealed the presence of β-endosulfan, HCB, heptachlor, and high levels of DDE relative to the upper limit set by the EPA, 8 ng/L (Hneine et al., 2017) (Tables 1 and 2).

7.2. Groundwater

Groundwater in Lebanon provides 50% of irrigation water, 80% of potable water, and water for domestic use through private and public wells (Youssef et al., 2015). According to the Lebanese Ministry of Water and Energy Resources, there were 42,824 wells in Lebanon in 2011 and 52% of them were illegal (Youssef et al., 2015). Numerous wells are located in agricultural areas; these wells are at risk of contamination because rain and irrigation waters flush OCPs that have been applied to the soil and PCBs from discharge areas into aquifers.

Four studies have investigated groundwater wells in Lebanon: three wells in a nationwide study in 2011 (Kouzayha et al., 2013), 10 wells in Akkar in 2012 (El-Osmani et al., 2014), three wells in the Hasbani basin, South Lebanon in 2012 (Youssef et al., 2015), and 15 wells in Akkar in 2015 (Chbib et al., 2017). The analytical laboratory techniques used in the four studies included SPE with LP-GC-MS or GC-MS, and a total of 21 OCPs were detected in groundwater samples from the four studies (Table 3).

7.2.1. Nationwide study

Three wells were assessed by grab sampling (Kouzayha et al., 2013); however, three wells is a very small sample size that is not representative of the entire country. The OCPs detected in groundwater samples from these wells were below the maximum admissible concentrations for drinking water set by the European Commission (EC) for each individual pesticide.

7.2.2. Groundwater in Akkar

Akkar province (798 km^2) in North Lebanon is the second largest agricultural plain in Lebanon (Chbib et al., 2017). Two studies

Table 3
Groundwater results.

OCPs/PCBs	Sampling in 2011 −2012(Published 2013)ng/L		Sampling in 2012(Published 2014)ng/L		Sampling in 2012(Published 2015)ng/L		Sampling in 2015(Published 2017)ng/L		EU Max. ng/LDrinking Water
Authors	Kouzhaya, A. et al.		El-Osmani, R. et al.		Youssef, L. et al.		Chbib, C. et al.		
LOD & LOQ	LOQ ranged from 0.1 ng/L (Aldrin) to 2 ng/L (Endosulfan sulfate)		LOD ranged from 5 ng/L (Aldrin and Heptachlor) to 60 ng/L (DDT)		LOQ ranged from 0.5 ng/L (HCB) to 5 ng/L (DDT)		LOD and LOQ for DDE were 0.29 & 0.5 ng/L, respectively		
Method	SPE + GC-MS		SPE + GC-MS		SPE + GC-MS		SPE + LP-GC-MS/MS		
Location	Dennyeh (North) Terbol (Bekaa) Abbassieh (South)		Akkar		Hasbani		Akkar		
Num. of sites	3		10, considered most vulnerable		3		15		
Num. of samples	15		30		11		30		
Samp. Method	Grab sample		Grab sample\Triplicates		Grab sample		Grab sampleDuplicates		
	av	rang	av	rang	av	Rang	Av	Rang	
Aldrin	0.4	nd−0.7	280	nd−720	Nq	nd−nq	2300	40−9330	30
DDD	1.6	0.9−2.2			2.80	nq−9,6	11142	90−19170	100
DDE	1.1	0.7−1.3			28.5	nq−144.8	30	nd−77	100
DDT			260	nd−690	nq	nd−nq	340	330−1290	100
ΣDDT							11840	610−19980	–
Chlorobenzilate			1111	140−6090					100
Dieldrin	0.6	nd−1.5					2721	50−9560	30
β-Endosulfan							280	20−640	100
Endosulfan sulfate			791	nd−1570			Nd	Nd	100
Endrin			1290	nd−2470			48	nd−80	100
Endrin Aldehyde							83.05	nd−0.19	100
Endrin Ketone			240	nd−2150			592	150−1650	100
HCB	1.5	1.2−1.6			0.52	nq−1.1			100
α-HCH			1460	nd−6420			1787	20−5760	100
β-HCH			126	nd−720			160	30−360	100
γ-HCH			311	nd−770			1304	100−3160	100
δ-HCH			423	nd−1350			17	nd−70	100
ΣHCH			2420	nd−7860			3268	150−9350	–
Heptachlor Epox. A			64	nd−120					30
Heptachlor Epox. B			679	nd−1610					30
Σ Heptachlor Epox.A + B			740	nd−2050	nq	nd−nq	9005	1390−25120	30
Heptachlor							3375	nd−6130	30
Methoxychlor					0.88	nd−3.4	57	nd−250	100
Tetradifon	0.2	nd−0.5							NA

assessed groundwater in Akkar: one in 2012 (El-Osmani et al., 2014) and one in 2015 (Chbib et al., 2017). Groundwater in Akkar is considered to be highly vulnerable to contamination because of heavy and uncontrolled use of pesticides in this region (Chbib et al., 2017), and the composition and nature of the agricultural land leads to poor filtration and rapid infiltration of pesticide residues leached by rainwater and irrigation water into aquifers. Therefore, inhabitants are at high risk of contamination because they drink groundwater. In the 2012 study, 28 OCPs were analyzed and 16 were detected (El-Osmani et al., 2014), while in the 2015 study, 20 OCPs were analyzed and all were detected (Chbib et al., 2017). In both studies, the detected OCPs implied recent, illegal use with an increasing trend, and the measured concentration of each OCP exceeded the maximum limit of 100 ng/L set by the EC. Moreover, the total concentration of detected OCPs also exceeded the limit set by the EC, 500 ng/L (Table 3). Most of the contaminated sites were inland and sites near the coast reported significantly lower concentrations (Youssef et al., 2015). Both studies demonstrated extensive recent use of low purity lindane and technical grade HCH, both of which are illegal, as the α, β, δ, and γ isomers that are present in technical grade HCH were detected at high levels in nearly all sampled sites in both studies (Chbib et al., 2017; El-Osmani et al., 2014; Zhao et al., 2009).

7.2.3. Groundwater in the Hasbani basin

In the Hasbani basin, 11 samples of groundwater were taken from three wells during 2012 (Youssef et al., 2015). Seven OCPs were detected at levels <100 ng/L (EC standard); however, DDE exceeded 100 ng/L in October after the first rainfalls.

7.3. DDT residues in lebanese soil

Only one nationwide study, conducted in 2004, has measured OCPs in Lebanese soil (Bachour et al., 2004). In major agricultural regions, 113 surface soil samples (0–20 cm depth) were collected from cultivated fields: 29 samples from Mount–Lebanon, 48 samples from the Bekaa Plain, and 36 samples from the coastal plain. The total residual DDT (ΣDDT, sum of p,p-DDT, -DDE, and -DDD) in soil samples, collected in triplicate, was extracted using methanol and measured by one of two methods: immunoassay was used to measure 82 samples and GC with electron capture detector (GC-ECD) was used to measure 33 samples. The results were as follows: 64 samples (57%) had concentrations < 2.5 ng/g, 44 samples (39%) had concentrations between 2.5 and 200 ng/g, three samples (3%) between 200 and 500 ng/g, and two samples were above 1000 ng/g; the maximum detected concentration was 1190 ng/g. Therefore, the residual DDT levels were less than 1000 ng/g (permissible levels for agricultural use) or the "total threshold limit concentration" for 98% of the samples; only two samples exceeded these limits and they were collected near a stockpile of DDT. The frequency of detection was higher in samples collected from sites near hubs of human activity (villages, gas stations, roads, etc.) (Bachour et al., 2004).

In order to assess the historic usage of DDT, DDE was quantified in 11 samples. In nine samples, the residue was entirely DDE, but DDT was present in two samples. In these samples, DDT was present in lower concentrations than DDE, which means that the application of DDT occurred long before sampling in 2004 (see Table S2 for details on DDT residues in Lebanese soils) (Bachour et al., 2004). The total residual DDT in soil in Lebanon has not been measured since the 2004 study, but if we extrapolate recent results from groundwater and surface water studies to soil, we would expect the levels of residual DDT in soil to have increased. This extrapolation should be confirmed by further studies.

7.4. Sediments

Two studies of sediments were published between 1999 and 2017: one on OCPs in sediments of the Kebir River (Thomas and Khawlie, 2003) and one on PCBs in sea sediments in Tripoli Harbor (Merhaby et al., 2015).

7.4.1. Sediments in the Kebir River

The Kebir River (77 km) is the natural northern border between Lebanon and Syria; the river has sources and tributaries on both sides of the border. Six sediment samples were taken from the river in 2002: two samples from Syria and four samples from Lebanon near junctions between tributaries and the main riverbed (Bachour et al., 2004). Sediments were analyzed for OCPs using either GC-MS or GC-ECD, following standard extraction and clean-up procedures (Bachour et al., 2004). The sediment samples contained varying amounts of HCB, DDT, and DDE: two samples contained HCB, one from Syria and one from Lebanon, and each had 80 ng/g; DDE was present in all samples and the average concentrations was 2020 ng/g (range: 170–8400 ng/g); and DDT was detected in five samples at an average concentration of 2210 ng/g (range: nd to 9960 ng/g). The average sum of DDT residues was 3840 ng/g (range: 170–12170 ng/g).

The results from this study (Bachour et al., 2004) indicate that DDT was used in the Kebir River watershed on both sides of the border despite its ban at the time of sampling (2002). The two samples taken in Syria had higher DDE concentrations than those collected in Lebanon. The mean values of DDE (1420 ng/g) and DDT (1190 ng/g) exceeded the Threshold Effect Level (TEL) and the Interim Standard Quality Guidelines (ISQG) set by the CCME (TEL is the ceiling below which fewer than 25% adverse effects occur in marine environments), but were within the PEL set by the CCME.

7.4.2. Sediments in Tripoli Harbor

The PCB pollution in sediments of Tripoli Harbor, the second largest port in the country (3,000,000 m^2), was assessed in December 2013 (Merhaby et al., 2015). Tripoli Harbor is located on the Mediterranean Sea, 80 km north of Beirut and 30 km south of the Syrian border. The harbor's basins are eight to 10 m deep and receive 450 ships per year (Office d'exploitation du port de Tripoli, 2015). Shipping and transit activities, storm water runoff, dredged materials, sewage outfall, landfill leachate, and commercial fishing put the harbor at risk of contamination (see Fig. S2 for Map of Tripoli Harbor) (Merhaby et al., 2015). Fifteen superficial sediments samples (0–5 cm) were collected from the harbor's basins in glass tubes by a diver.

Twenty-eight PCBs were analyzed: the 12 dioxin-like PCBs (PCB-DL) – PCB77, PCB81, PCB105, PCB114, PCB118, PCB123, PCB126, PCB156, PCB157, PCB167, PCB169, and PCB189; the seven indicators PCB (PCBi) – PCB28, PCB52, PCB101, PCB118 (DL), PCB138, PCB153, and PCB180; and nine other PCBs – PCB8, PCB18, PCB44, PCB66, PCB128, PCB170, PCB187, PCB195, PCB206, and PCB209. The analysis method included accelerated solvent extraction (ASE) followed by GC-MS-MS. Of the 28 congener analytes, 24 were detected and quantified; PCB101, PCB138, and PCB153 were present in all samples and DL-PCB156, DL-PCB167, DL-PCB189, and PCB195 were below the LOQ. The summary of the results was as follows: the ΣPCBs concentration at 11 sites exceeded 21.5 μg/kg, the TEL set by the CCME (2001) including one site where the ΣPCBs concentration exceeded 189 μg/kg, the PEL set by the CCME (2001); at the other four sites the ΣPCBs concentration was below the TEL. According to the ISQG (Long et al., 1995) that use the TEL/PEL approach (Macdonald et al., 1996), most of the samples exceeded the TEL, and one exceeded the PEL (see Table S3 for PCB concentrations in Tripoli Harbor sediments). Another approach to classifying PCB concentrations in sediments is the sediment quality guidelines for dredged materials in ports sediments set by the "Groupe d'Etude et d'Observation sur le Dragage et l'Environnement" of the maritime authorities of France (1990). According to this classification, all sediment samples from Tripoli Harbor's basin were below the N1 level: 500 μg/kg (dw). Moreover, 60% of the detected PCBs were tetra-, penta-, and hexa-chlorinated PCBs. Their abundance may be the result of atmospheric emissions from port equipment, generators, ships, vehicles, and truck exhaust (Merhaby et al., 2015).

7.5. Human biomonitoring (HBM)

One study of OCPs in human milk and one study of OCPs and PCBs in human serum are included in this review.

7.5.1. Human milk

In the study of OCP residues in human milk in Lebanon (Dagher et al., 1999), 32 milk samples (10 mL each) were taken from 12 primiparous and 15 multiparous nursing mothers between the ages of 17 and 35. The samples were taken after delivery and while the mothers were hospitalized at the American University of Beirut Hospital in 1995 (Bordet et al., 1993). The current World Health Organization (WHO) sampling recommendations for human milk biomonitoring state that mothers should be primiparous, aged ≤ 30 years, and 50 mL of milk should be collected three to eight weeks after delivery (WHO - UNEP, 2007). Despite the deviations from these recommendations, the study has been included in this review because it is the only one to date and its results offer interesting information about the history of OCP use in Lebanon.

The dietary intakes of the recruited mothers were assessed by 24 h dietary recalls and food frequency questionnaires, and milk samples underwent fat extraction before analysis by SPE and GC-ECD. The most common OCPs found in the milk samples were DDT residues (DDT, DDD, and DDE) – DDE was present in 97% of samples – and β-endosulfan was present in 19% of samples. The DDE concentrations in milk ranged from 47 μg/L to 1563 μg/L, and the mean DDE value was 362 ± 34 μg/L. At the time of the study, the authors considered these DDE levels to be comparable to those of studies from other countries during the same period, and thus they were not considered to be unusually high. However, the WHO's current maximum residue level (MRL) for ΣDDT in human milk is 20 μg/L, therefore, the concentrations of ΣDDT measured in 1995 were very high. Finally, positive correlation of DDE levels with high fat meat consumption was observed as well as positive correlation with tuna, weak correlation with poultry, and negative correlation with vegetable oils.

7.5.2. Human serum

In Lebanon, the first human serum biomonitoring of PCBs and OCPs was reported in 2017 (Harmouche-Karaki et al., 2017a, 2017b). From 2013 to 2015, 133 men and 181 women between the ages of 18 and 65 were recruited from students and employees of the Saint-Joseph University of Beirut; 97.5% of the subjects were from Beirut and the Mount Lebanon Governorate. Data collection included samples of blood, anthropometric measures, blood pressure measurements, 24 h dietary recall, and food frequency and socio-demographic questionnaires. Four OCPs were screened – HCB, β-HCH, DDT, and DDE –and six of the seven PCBs were screened – PCB28, PCB52, PCB101, PCB138, PCB153, and PCB180. These analytes were measured using targeted analytical methods: SPE followed by GC-MS/MS.

7.6. OCPs results

Biomonitoring equivalent (BE) limits were developed in the USA in 2008 to indicate concentrations above which health risks are high (Choi et al., 2015). The mean concentrations of HCB, β-HCH, DDT, and DDE were below the BE limits (Harmouche-Karaki et al., 2017a), except for one DDE value that was above the BE limit for cancer risk. Furthermore, concentrations at the 95th percentile were much lower than the values associated with diabetes risk by Pal et al. (2013).

7.7. PCBs results

The PCB concentrations in human serum samples from Lebanon were less than those of many European countries (Table 4) (Harmouche-Karaki et al., 2017b). Highly chlorinated PCB congeners containing more than five chlorine atoms tend to bioaccumulate in human serum lipids and adipose tissue (Grimm et al., 2015) and can persist for 10–47 years according to the Agency for Toxic Substances and Disease Registry (ATSDR). Low-chlorinated congeners containing fewer than six carbon atoms, such as PCB101 (five Cl), PCB52 (four Cl), and PCB28 (three Cl), are

Table 4
OCPs and PCBs in serum (Harmouche-Karaki et al., 2017a) (Harmouche-Karaki et al., 2017b).

PCB congener	% > LOD (n = 316)	Geom. Mean ng/g lipids	Arithm. Mean ng/g lipids	Min.ng/g lipids	50^{th} percentile ng/g lipids	95th percentile ng/g lipids	Max. ng/g lipids
HCB[a]	50.0	7.1 ± 0.6	–	< LOD	5.8	32.0	85.1
βHCH	50.0	8.6 ± 0.6	–	< LOD	7.6	45.0	154.9
DDT[b]	49.7	2.1 ± 0.3	–	< LOD	2.0	5.0	15.8
DDE[c]	50.0	18.9 ± 0.9	–	< LOD	17.3	180.0	630.9
ΣDDE, DDT[d]	–	21	–	< LOD	–	–	646.7
PCB28	56.3	0.9	2.2	< LOD	2.2	5.6	18.2
PCB52	58.2	0.3	0.6	< LOD	0.3	1.5	17.4
PCB101	57.6	0.6	1.3	< LOD	0.6	3.6	17.4
PCB138	59.2	1.9	7.1	< LOD	8.2	16.7	50.1
PCB153	58.2	3.0	16.5	< LOD	16.4	45.9	87.1
PCB180	58.2	3.7	24.7	< LOD	24.1	66.8	170.0
Σ PCB138,153,180	–	9.0	48.3	< LOD	52.6	128.0	302.0
ΣPCBi	–	10.3	52.4	< LOD	57.9	135.0	339.0

[a] Biomonitoring equivalents reference dose of non-cancer risk $BE_{RfD} = 340$ ng/g lipids; Biomonitoring equivalents point of departure of non-cancer risk $BE_{POD} = 3384$ ng/g lipids (Choi et al., 2015).
[b] Biomonitoring equivalents BE of 1E-05 cancer risk = 300 ng/g lipids (EPA).
[c] Biomonitoring equivalents BE of 1E-05 cancer risk = 500 ng/g lipids (EPA).
[d] Biomonitoring equivalents reference dose of non-cancer risk $BE_{RfD} = 5000$ ng/g lipids; Biomonitoring equivalents point of departure of non-cancer risk $BE_{POD} = 16000$ ng/g lipids (Choi et al., 2015).

relatively rapidly metabolized by the human body and their half-lives can reach 5 years. In this study, PCB138 (six Cl), PCB153 (six Cl), and PCB180 (seven Cl) were the most frequently detected and accounted for 75% of the analyzed PCBs. PCB180, the most chlorinated congener, was the most prevalent and accounted for 34.1% of the PCBs. The results of this study are in agreement with studies from around the world and may be explained by EDL's long-time use of Aroclor 1260, which is 99% highly chlorinated PCBs (five to eight Cl) and predominantly PCB180 (Harmouche-Karaki et al., 2017b). This would explain the high fraction of PCB180 in the human serum study in Lebanon, as PCB153 is typically the primary PCB in biomonitoring studies from other countries (Table 4). There were no significant differences between the serum samples from men and women.

The results were compared to available standards, HBM-I and HBM-II, which are limit values established by the German Commission. The HBM-I standard represents concentrations in the human body below which there is no risk of adverse health effects; conversely, HBM-II represents concentrations in the human body above which there is an increased risk of adverse health effects (Choi et al., 2015). For women of childbearing age and children, the concentration limits of ΣPCB138,153,180 set by HBM I and HBM II are 3.5 and 7 μg/L, respectively (Lehmphul, 2015). A study on the association between PCB153 concentration and breast cancer risk set the critical value of PCB153 as 1.63 μg/L (Charlier et al., 2003). Pal. et al. (2013) also evaluated relationships between PCB concentrations and diabetes, and they found that the mean ΣPCB138,153,180 concentration in diabetics is 395.3 μg/L (Pal et al., 2013).

The results of this human serum study demonstrated that the PCB levels in Lebanese women of childbearing age were 11 times less than the HBM II values, 7 times less than the critical value for breast cancer, and 600 times less than the PCB levels reported for diabetics. Finally, the French Agency for Food, Environmental and Occupational Health & Safety (ANSES, 2018) established critical limits for total PCB concentration below which there is a negligible risk of health effects for women of childbearing age and children (700 ng/g lipids) and the general population (1800 ng/g lipids) (AFSSA, 2018). In this human serum study, the PCB concentrations at the 95th percentile for women of childbearing age and the total population are 5 times lower than 700 ng/g lipids (138.38 ng/g lipids) and 13 times lower than 1800 ng/g lipids (134.58 ng/g lipids), respectively (Harmouche-Karaki et al., 2017b).

8. Discussion & conclusions

This review summarizes official authorities' actions regarding PCBs and OCPs in Lebanon; these actions include creating and updating the national inventories, and shipping PCB-containing equipment and oils from the power sector abroad to ensure proper disposal. However, more action needs to be taken to reduce and eliminate PCBs and OCPs in Lebanon. Reports issued by the Ministry of the Environment indicate that supplementary resources are needed in order for law enforcement to stop the smuggling of OCPs into the country and for farmers to receive education about the dangers of OCPs.

Studies investigating OCPs over the past decade include four surface water studies of four rivers in Lebanon. The levels of banned OCPs measured in the Litani, Ibrahim, and Hasbani rivers exceeded the MRL and the Hasbani River was highly polluted with PCB52 (Badr et al., 2014). Compared to studies in other countries during the past decade, the DDE levels in the surface waters of the Litani, Ibrahim, and Hasbani rivers were much higher than those detected in Turkey's Ankara River (Sevin et al., 2018), Kenya's Nairobi River (Ndunda et al., 2018), and Nigeria's Niger River (Unyimadu et al., 2017). However, DDT was used intensively (553.8 tons in 1978) on agricultural fields and swamp areas in Turkey until the 1980s (Ayas, 2007). Furthermore, DDT continued to be used intensively in Kenya until 2007 and is still used indoors to fight malaria (Ministry of Environment, 2014). In Nigeria over 130,000 tons of pesticides, including DDT, are applied every year to protect agriculture and to stop vector-borne diseases (Mansouri et al., 2017). The β-endosulfan levels found in the surface water of the Hasbani and Litani rivers were also much higher than those found in the Niger River and Nairobi River in 2009 (Unyimadu et al., 2017; Ayas, 2007). The sum of OCPs found in surface water of each river included in this review, excluding the Orontes, far exceed that of 19 OCPs found in Ñuble River in central Chile in 2014 (Montory et al., 2017); the main OCPs in the Ñuble River were endosulfan and lindane, whereas DDE, endosulfan, and HCB were the main OCPs in the three Lebanese rivers (Montory et al., 2017). In the surface water of the Hasbani-Wazzani River, the level of PCB52 alone was much higher than the sum of 32 PCBs found in the Tiber River in Italy (Montuori et al., 2016). These figures are of concern regionally, since the Litani and Ibrahim rivers flow into the Mediterranean Sea, and the Hasbani-Wazzani flows into the sea of Galilee, contributing thus to the bioaccumulation of OCPs and PCBs in the marine and aquatic biota.

In the last ten years, the groundwater from 25 wells used for drinking and irrigation in Akkar has been highly contaminated with OCPs, whereas groundwater in the Hasbani basin is less contaminated, except for some OCPs that exceed the MRLs in some samples. There has yet to be a national study that covers enough wells to develop a more accurate assessment of groundwater, as there are more than 42,000 wells in Lebanon. Akkar's groundwater is much more polluted with aldrin, dieldrin, endosulfan, ΣHCH, and DDT residues than the groundwater of the vegetable planting area in Tianjin City, Northern China in 2011 (Pan et al., 2017). The contaminated underground water of Akkar likely finds its way to the eastern Mediterranean Sea due to its proximity, contributing thus to the pollution of neighboring countries' shores.

There has only been one national study of Lebanese soil, conducted in 2004. While the study revealed acceptable levels of DDT residues, it is insufficient for assessing the current contamination of soil in Lebanon because it is outdated and other studies are needed to observe variations in OCP concentrations with time. Since the OCPs in soil have not been measured by any other studies (Bachour et al., 2004), it is not relevant to compare the small set of results to recent studies in other countries.

The study of river sediments from the Kebir River, located between Lebanon and Syria, in 2002 revealed the levels of DDE, DDT, and HCB that exceeded the TEL but were within the PEL; these DDT and DDE levels were much higher than those of surface sediments in the Hooghly estuary in India (Mitra et al., 2019) and the Nairobi River in Kenya (Ndunda et al., 2018). Similarly, the study of PCBs in marine sediments from Tripoli Harbor revealed PCB levels that were higher than those of sediments collected along the north-south axis of the western Adriatic Sea (Combi et al., 2016). Furthermore, the sum of PCBi concentrations in the sediments from Tripoli Harbor was much higher than that of the sediments from Venice Lagoon in Italy (Cassin et al., 2018). Knowing that this harbor has one-tenth of the shipping activities than occur in Beirut harbor, it would be interesting to carry out a study on Beirut port sediments to have a better estimation about the status of PCBs and OCPs on the eastern Mediterranean shore.

Finally, the biomonitoring study of human breast milk in 1995 reported DDT residue concentrations that exceed the current limit set by the WHO. Furthermore, the biomonitoring study of human serum carried out during 2013–2015 revealed levels of PCBs and OCPs below reference values but greater than the levels set by the

Table 5
Comparison of PCB and OCP results between the Lebanese Serum Biomonitoring (LSB) study (Harmouche-Karaki et al., 2017a, 2017b) and the NHANES (CDC, 2019).

PCBs & OCPs	LSB concentrations in serum (ng/g lipids)	LSB years	NHANES concentrations in serum (ng/g lipids)	NHANES years
PCB28	0.9	(2013–2015)	0.27	(2013–2014)
PCB52	0.3	(2013–2015)	0.3	(2007–2008)
PCB101	0.6	(2013–2015)	0.3	(2007–2008)
PCB153	3.0	(2013–2015)	0.26	(2013–2014)
PCB180	3.6	(2013–2015)	0.26	(2013–2014)
HCB	7.1 ± 0.6	(2013–2015)	1.3	(2013–2014)
βHCH	8.6 ± 0.6	(2013–2015)	1.3	(2013–2014)
DDT	2.1 ± 0.3	(2013–2015)	1.3	(2013–2014)
DDE	18.9 ± 0.9	(2013–2015)	1.8	(2013–2014)

National Health and Nutrition Examination Survey (NHANES) in the USA (CDC, 2019) (Table 5).

The 12 studies included in this review are largely insufficient to establish national statistics or correlations between the level of exposure and OCP and PCB concentrations in human serum or breast milk. Further studies are needed on a national scale since Lebanon is still a hotspot for PCBs, OCPs, and other POPs.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2019.05.109.

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