Chapter 2
Organochlorinated Contaminants in General Population of Argentina and Other Latin American Countries

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Abstract Organochlorinated contaminants integrate the persistent organic pollutants (POPs) group, according to the Stockholm Convention. Persistent organic pollutants are synthetic chemicals highly lipophilic that cause harmful effects on human health. The extensive use of organochlorine pesticides (OCPs) in agriculture and polychlorinated biphenyls (PCBs) on industry, in confluence with its resistance to metabolic degradation, determined its persistence in the environment.

Studies on population of Argentina and other Latin American countries show exposure to POPs, whose levels in adipose tissue, serum, and breast milk mainly depend on the age, sex, and place of residence. Most studies were based on researches of OCP in exposed and unexposed populations. Blood was the biological matrix mostly used. The most frequently found pesticides were *pp′*-dichlorodiphenyldichloroethane (*pp′*-DDE), *pp′*-dichlorodiphenyltrichloroethane (*pp′*-DDT), hexachlorobenzene (HCB), and *β*-hexachlorocyclohexane (*β*-HCH) in all matrices investigated. Some studies examined the presence of PCBs in human tissues predominating the most persistent congeners 138, 153, 180, and 170.

While applications of POPs have decreased in recent years, human exposure in some Latin American countries continues because of their persistence and reuse of banned OCPs. Consequently, monitoring programs of the POP concentrations in general population are needed to provide data to government authorities in order to implement policies of bioremediation and to minimize the risk of human exposure to these persistent compounds.

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2.1 Introduction

Organochlorinated contaminants integrate the persistent organic pollutants (POPs) group. These compounds have been added to the POP Stockholm Convention list (Stockholm Convention 2001). POPs are synthetic chemicals highly lipophilic, resistant to degradation that accumulated in the environment with harmful effects on human health (Porta et al. 2008).

Organochlorinated compounds are stored in the fat of human tissues, particularly dichlorodiphenyldichloroethylene [DDE, the main product of degradation of dichlorodiphenyltrichloroethane (DDT), hexachlorobenzene (HCB), the hexachlorocyclohexanes (HCH), and polychlorinated biphenyls (PCBs)] (Porta et al. 2008). Humans can be exposed to these contaminants primarily through food intake by their working activities or environmental exposure. Many studies have shown that the main contributors to the total intake of POPs are meat, fish, and dairy (Gasull et al. 2011).

Although some POPs have been banned or restricted, others remain deeply rooted in our societies and human exposure continues even to compounds banned like DDT (Porta and Zumeta 2002).

Organochlorine environmental contaminants such as *pp′*-DDE, PCBs, and other related compounds may be causally related to decreased male reproductive capacity and infertility; also an increased incidence of breast cancer and neurodevelopmental deficits in children are based on environmental, laboratory, animal, and human data. Organochlorine pesticides (OCPs) and hydroxy-PCBs modulate endocrine responses in animal models and cell culture; these compounds are identified as potential endocrine disruptors (Safe 2001; López-Cervantes et al. 2004; McKinlay et al. 2008; Varayoud et al. 2008). Milesi et al. (2012) found that low doses of neonatal exposure to endosulfan affect uterine function in adulthood.

Although the human exposure to PCBs and persistent chlorinated pesticides has declined, there are evidences that the presence of organochlorines in food may be a risk factor for neurologic, hormonal, genotoxic, and immunologic effects in infants and children (ATSDR 2000; Glynn et al. 2003; Freire et al. 2012; Boccolini et al. 2013).

Organochlorine pesticides were widely used in Argentina from the 1940s to the 1970s, although different government resolutions were restricting and prohibiting their use (Villaamil Lepori et al. 2013). However, endosulfan is still the most widely used insecticide in agriculture both in Argentina and other Latin American countries (Souza Casadinho 2008). By their known toxic effects, the Argentine National Health Service and Food Quality (SENASA) enacted a law which banned the importation of endosulfan (from June 30, 2012, resolution 511/11) (Souza Casadinho 2008; SENASA 2013; Villaamil Lepori et al. 2013).

Polychlorinated biphenyls have been widely used as additives to oils in electrical equipment (transformers and capacitors), hydraulic machines, adhesives, textiles, printing, and sealants. Coplanar PCBs (non-*ortho* and mono-*ortho*) are the most toxic, producing effects like 2,3,7,8-tetrachlorodibenzodioxin (2,3,7,8-TCDD or dioxin) in experimental animals. Toxic equivalency factors (TEFs) have been established based on the aryl hydrocarbon receptor (AhR) affinity. Dioxin toxic equivalencies (TEQs) were used to evaluate the toxicological risk associated to the presence of PCBs in humans (Mariottini et al. 2000). Polychlorinated biphenyls were banned in Argentina from 2002 (law 25670/02), but even today continue the decontamination of transformers (Miglioranza et al. 2013).

The lipophilic properties of POPs promote transport via circulating lipids and lipoproteins and storage in tissues with high-fat content. Assessing levels of organic pollutants in these matrices as biomarkers provide invaluable information on the biological effects observed in individuals exposed to these substances (Díaz-Barriga et al. 2003; Waliszewski et al. 2013). Body concentrations of OCPs and PCBs in human biological samples have been analyzed in several studies and indicate that most humans store POPs, although interindividual differences in concentrations are substantial in most populations worldwide (Cerrillo et al. 2005; Porta et al. 2008; Gasull et al. 2011). Hereby, assessing human contamination requires monitoring POP levels in representative samples.

Argentina has reported several studies on the impact of organochlorinated contaminants in the environment, food, and wildlife. In the 1970s were investigated organochlorine pesticides in water of the Parana and Uruguay rivers. Hexachlorobenzene, α-, β-, and γ-HCH isomers, and *pp*′-DDT were detected more frequently. Higher concentrations corresponded to *pp*′-DDT (5.6 μg/L) (García Fernández et al. 1979).

Organochlorine pesticide residues were analyzed in 101 samples of infant formula and dairy products collected in the local market of Buenos Aires, Argentina. The groups of pesticides that were found mainly were heptachlor (57 %) and HCH (53 %). Heptachlor and its epoxide exceeded the acceptable daily intake (ADI) in dairy products which would imply an increased risk to health of infants and toddlers (Villaamil Lepori et al. 2003, 2006).

Colombo et al. (2011) evaluated the bioaccumulation and risk associated to consumption of lipid-rich detritivorous fish. Chlorinated pesticides and PCBs were analyzed in Sábalo fish (*Prochilodus lineatus*) collected in the polluted Metropolitan Buenos Aires coast and in migrating specimens. In this study, very high concentrations of organic pollutants were found in fatty fish muscles. These data correlate with that reported in fish from different river basins of Brazil by Torres et al. (2010).

In a recent study along the Río Negro basin (Argentinean Patagonia), Miglioranza et al. (2013) report the occurrence and distribution of OCPs and PCBs in soil, sediment, suspended particle matter (SPM), stream water, and macrophytes. They found a clear predominance of OCPs among all matrices indicating the impact of agriculture on the watershed. The highest levels were found for *pp*′-DDE in agricultural soils from the Upper Valley. The insecticide endosulfan was also found in all matrices. Levels of PCB (153, 138, 110, and 101 congeners) were directly related with the presence of hydroelectric power plant.

Researches on the impact of POPs in the environment, food, and nontarget species have also been conducted in other Latin American countries (Díaz-Barriga et al. 2003; Pérez-Maldonado et al. 2010; Torres et al. 2010; Avancini et al. 2013;

Benítez-Díaz and Miranda-Contreras 2013). Several studies suggested that banned OCPs were being used to protect illegal crops from pests. *pp*′-Dichlorodiphenyltrichloroethane is still used for vector control in several tropical and subtropical areas of South America and there is evidence of recent illegal use in agriculture (Varona et al. 2010; Mercado et al. 2013), hence the importance of monitoring these persist compounds in the environment and in humans.

In Argentina, Brazil, Mexico, and Bolivia, new studies have evaluated the genotoxic risk of occupational exposure to pesticides in agricultural workers that may be correlated with the exposure time (Simoniello et al. 2008; Poma et al. 2010; Da Silva et al. 2012; Benedetti et al. 2013; Gómez-Arroyo et al. 2013).

In many countries throughout Europe, Asia, and North America OCP and PCBs in humans (blood serum, breast milk, adipose tissue) have been reported, showing a reduction of human exposure in recent years due to the prohibitions and restrictions of these substances (Dua et al. 1996; James et al. 2002; Glynn et al. 2003; Jaraczewska et al. 2006; Petrik et al. 2006; Sudaryanto et al. 2006; Thomas et al. 2006; Lucena et al. 2007; Tanabe and Kunisue 2007; Kozul and Romanić 2010; Shen et al. 2010; Kalantzi et al. 2011; Mishra et al. 2011; Bräuner et al. 2012; Cok et al. 2012).

Although adipose tissue levels have been preferentially used as an indicator of chronic human exposure to organochlorine contaminants, serum levels have been adopted in epidemiological studies as a less invasive and more practical alternative (Díaz-Barriga et al. 2003).

In this document, we present a data collection of exposure to OCP and PCBs in adipose tissue, serum, and breast milk in Argentine general population and population from other Latin American countries, in order to evaluate the degree of exposure to POPs and compare the levels found in inhabitants of different regions.

2.2 Persistent Organic Pollutants and Polychlorinated Biphenyls: Human Exposure in Latin America

2.2.1 Adipose Tissue

Organochlorine pesticides and PCBs accumulate in fatty tissue due to its lipophilic properties. In human adipose tissue, the average life of most of these compounds is several years. Adipose tissue biopsy has been used in epidemiological studies to assess chronic exposure to organochlorine. This tissue is selected because it gives adequate results with regard to accumulation status (Botella et al. 2004; Waliszewski et al. 2011). Although the presence of organochlorine compounds in human adipose tissue has been reported throughout the world, in Argentina and Latin America few studies are found. Muñoz-de-Toro et al. (2006a) investigated the residues of OCPs and PCBs in mammary fat tissue from 76 women not occupationally exposed to organochlorines living in a littoral region in northeastern Argentina. Organochlorine compounds that appeared most frequently were *pp*′-DDE (100 %), HCB (87 %), and *β*-HCH (75 %). On the other hand, the incidence of PCBs congeners was very low. *pp*′-Dichlorodiphenyldichloroethane and *β*-HCH residues reached the highest levels (4,794 and 1,780 mg/g, respectively). Significant positive association was found between organochlorine levels, body mass index, and women age ($P<0.05$). The diet was a relevant source of exposure, particularly the consumption of animal fat and freshwater fish. Organochlorines accumulated in the mother could be transferred transplacentally to the developing fetus and from breast milk to the nursing infant. The frequencies and concentrations of organochlorine residues in adipose tissue reported in this study were higher than those reported in developed countries (Botella et al. 2004; Porta et al. 2008).

Another study by the same group of researchers in a subsample of 55 women diagnosed with invasive breast carcinoma from Santa Fe city area (a littoral region in Argentina) found a positive correlation between progesterone receptor (PR) expression (an estrogen-induced protein) in the neoplastic cells and organochlorine levels in adipose tissue. It was observed that when organochlorine levels in adipose tissue reached levels higher than 2,600 mg/g, the estrogen receptor alpha (ERα)-positive breast carcinomas from postmenopausal women exhibited high proliferation. The authors conclude that organochlorine residues in adipose tissue adjacent to breast carcinoma generate an estrogenic microenvironment that may influence the biological behavior of the tumor through ERα activation and ERα-dependent proliferation (Muñoz-de-Toro et al. 2006b).

Ridolfi et al. (2008) conducted a case-control study in order to assess the levels of 17 PCB congeners in breast adipose tissue from 88 women (aged between 30 and 85 years) with malignant and benign breast tumors from Buenos Aires city, Argentina. Concentrations of PCB congeners reported in this study were higher in malignant tumors compared to benign (Fig. 2.1). The sum of dioxin-like congeners adjusted for age, body mass index (BMI), residence, and occupation was higher in malignant tumors than in benign (200.1 vs. 61.1 ng/g lipid, respectively)

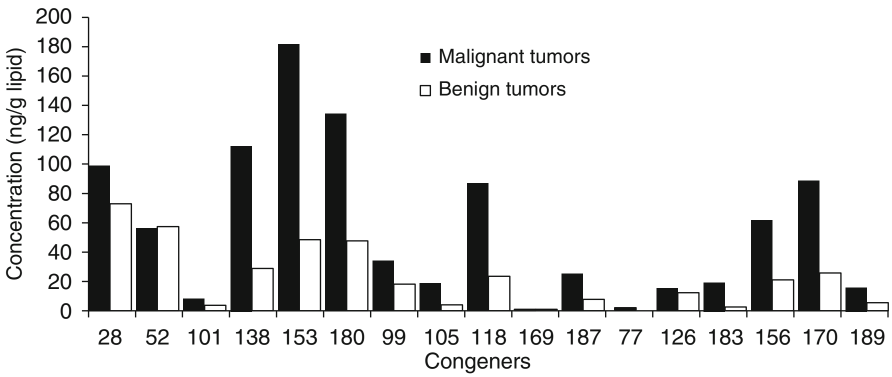

Fig. 2.1 Concentrations of congeners of polychlorinated biphenyls in malignant and benign breast tumors in Argentinean women

predominating the mono-*ortho* congeners 118 and 156. Association was found between toxicity equivalence to dioxins (TEQs) and breast cancer risk ($P < 0.05$). The results suggest that exposure to PCBs dioxin-like will increase the risk of breast cancer in the studied population.

In Latin America, Mexico had reported the highest number of studies in human adipose tissue. Levels of DDT and its metabolites in human biological media from different communities of Mexico had been assessed by Díaz-Barriga et al. (2003). The authors found higher values of DDE in adipose tissue of exposed population compared to an urban non-exposed community (21.5 and 8.3 μg/g lipid, respectively). In this study the authors also observed higher concentrations of DDT in adipose tissue from workers of the malaria program compared with adipose tissue from general people living in malarious areas. It was clear that children and elderly people were identified as the most exposed when the concentrations of DDTs in adipose tissue were correlated by age. The elderly findings can be explained by chronic exposure to DDT, whereas the children concentration may be the result of exposure to multiple pathways (soil, household dust, air, water, food). The authors conclude that a DDT monitoring program in adipose tissue is needed in order to assess the body burden in Mexican population.

Waliszewski et al. (2011) monitored HCB (α-β-γ-HCH), *pp′*-DDE, *op′*-DDT, and *pp′*-DDT in 150 adipose tissue samples from inhabitants of Veracruz (Mexico). The following pesticides were detected: *pp′*-DDE (100 % of the samples; 1.643 mg/kg); *pp′*-DDT (99.3 % of the samples, 0.227 mg/kg); β-HCH (97.3 % of the samples, 0.063 mg/kg); and *op′*-DDT (93.3 % of the samples, 0.022 mg/kg). The samples grouped according to age showed a positively associated factor with OCP levels in adipose tissue of Veracruz inhabitants. Comparing OCP levels between 2008 and 2010, a decreased tendency for β-HCH, *pp′*-DDE, Σ-DDT (DDT sum), and *pp′*-DDE/*pp′*-DDT ratio levels was observed.

At the same place (Veracruz), Herrero-Mercado et al. (2011) conducted another study to determine levels and calculate ratios of copartition coefficients among *β-HCH*, *pp′*-DDE, *op′*-DDT, and *pp′*-DDT in maternal adipose tissue, maternal blood serum, and umbilical blood serum of mother-infant pairs ($n = 70$). *pp′*-DDE was found as the major organochlorine component, detected in every maternal adipose tissue (0.770 mg/kg lipid). The mean concentrations of *pp′*-DDT and β-HCH were 0.101 mg/kg lipid and 0.027 mg/kg lipid, respectively. The authors postulated one mechanism to make out the differences between the copartition coefficients among samples. They identify significant increases in concentrations from adipose tissue to maternal blood serum and to umbilical blood serum. These increases indicated that maternal adipose tissue released OCP to blood serum and that they are carried over to umbilical cord blood.

Recently Waliszewski et al. (2013) conducted a review over human monitoring studies of adipose tissue in the inhabitants of the states of Veracruz, Puebla, and Tabasco (Mexico) to determine the degree of exposure to OCP. The results showed the presence of residues of organochlorine compound in general Mexican population; *pp′*-DDE was found in 100 % of samples and β-HCH, *pp′*-DDT, and *op′*-DDT predominated in Veracruz.

In Uruguay, the Poison Control Center (CIAT: Information and Toxicological Assessment Center), together with the Ministry of Agriculture, evaluated pesticide exposure through adipose tissue analysis of the Montevidean adult population. Both studies (1985 and 1996) on OCP in human fat show accumulation of these products in fat tissues (Mañay et al. 2004). Burger et al. (2000) evaluated a possible correlation between risk of breast cancer and residues of OCP in human body. Fifty-eight patients diagnosed with breast cancer and other 28 no-occupationally exposed with benign mammalian tumors were studied in Uruguay. There was a tendency toward higher levels of chlorinated pesticides in the problem cases than in the controls, but difference statistically significant was found only for *β*-HCH, being not possible to determine a consistent relationship between OCP residues and breast cancer. These results are consistent with studies in other regions (Safe 2001).

Another Latin American study compared human exposure to PCBs in adipose tissue samples collected from Concepción (Chile) and Siena (Italy). ΣPCBs were higher in Italian samples than those from Chile (493 and 53 ng/g, respectively). The congeners that prevailed in both groups were PCB 118, 138, 153, 170 180, and 187. Average concentrations of non-*ortho*-substituted coplanar congeners were below 1 pg/g. TEQ values were lower in Concepción, while in Sienese adipose tissue, toxic potential was much higher. According to the authors, the levels of PCBs found in Siena were similar to those reported by other industrialized countries of the northern hemisphere (Mariottini et al. 2000).

Table 2.1 presents the comparison of the mean concentrations of OCP most frequently found in human adipose tissue in different studies conducted in Argentina and other countries. Mexico had the highest level of DDT and metabolites, especially in malaria's areas. The concentrations found in Argentina and Uruguay were similar to those found in Spain by Botella et al. (2004), but higher than those reported in other European countries.

Table 2.1 Organochlorine pesticides (ng/g lipid) detected in human adipose tissue from Argentina and other countries (1998–2013)

Region	HCB	*β-HCH*	*pp′*-DDE	*pp′*-DDT	References
Argentina	68.3	367.4	918.8	7.9	Muñoz-de-Toro et al. (2006a)
Mexico (malarious zone)	ND	ND	28,900.0	8,100.0	Díaz-Barriga et al. (2003)
Mexico (urban zone)	ND	ND	6,000.0	1,100.0	Díaz-Barriga et al. (2003)
Mexico—Veracruz	ND	63.0	1,643.0	227.0	Waliszewski et al. (2011)
Mexico—Puebla	ND	73.0	916.0	83.0	Waliszewski et al. (2013)
Mexico—Tabasco	ND	49.0	1,034.0	116.0	Waliszewski et al. (2013)
Mexico—Veracruz	ND	27.0	770.0	101.0	Herrero-Mercado et al. (2011)
Uruguay (1996)	400.0	300.0	700.0	ND	Mañay et al. (2004)
USA—Long Island	19.7	22.2	546.7	17.0	Stellman et al. (1998)
Spain	ND	ND	508.0	61.0	Botella et al. (2004)

ND not determined

2.2.2 Human Blood

Persistent organic pollutant investigations in human blood from different regions of Argentina are scarce; however, its presence has been determined often in measurable concentrations (García Fernández et al. 1987; Lucero et al. 2008; Villaamil Lepori et al. 2013). Álvarez et al. (2006) investigated OCP in 100 samples of plasma from healthy volunteers not occupationally exposed of general population living in the metropolitan area of Buenos Aires (35 women and 65 men aged 18–82 were evaluated). Results showed that DDT group appeared most frequently (71 %) with a prevalence of *pp′*-DDE metabolite (57 %), followed by HCB (70 %), HCH group (57 %), and heptachlor and its epoxide (49 %). Maximum values of 9.87 and 8.05 μg/L were registered by *pp′*-DDD and *op′*-DDE, respectively (Table 2.2). OCP levels detected in this study were lower those reported by García Fernández et al. (1987) two decades earlier (Table 2.3). Results showed a significant decrease in the *β*- and *γ*-HCH isomers and *pp′*-DDT, banned in Argentina from the 1970s to 1980s. These results reinforce the concept that persistence of OCP metabolites is related with age and period of exposure (de Boer and Fiedler 2013; Waliszewski et al. 2013).

Another study conducted in Argentina evaluated the OCP levels in 649 blood samples analyzed during 2004–2012. People (aged between 1 and 85) came from different regions of the country. Results showed that the environment was the predominant etiology in children (97 %) and adults (85 %). Percentages of OCP were highest in children (0–15 years) [ΣDDT (53 %) and ΣHCH (41 %)] (Fig. 2.2). On the other hand, aldrin's group had a higher frequency in young adults [(16–30 and 31–50 years, 39 and 51 %, respectively)], followed by DDT (33 and 44 %) and HCH (31 and 47 %). Thirty percent of samples of older adults (51–85 years) presented HCH and others OCP. Concentration (μg/L) of pesticides (Fig. 2.3) showed different results with respect to the frequency of them (Fig. 2.2). Children had the lowest value of DDT (0.02 ± 0.63 μg/L), while the HCH group had the highest concentration (0.2012 ± 0.046 μg/L) exceeding the reference value for adults (0.09 ± 0.22 μg/L) reported by Álvarez et al. (2006). In young and adults DDT concentrations present low mean; the highest levels were found for HCHs and aldrin. In the older population (51–85 years) HCB had the highest mean concentration. The prevalence of OCP in this study was consistent with those reported in other countries (Fernandes Delgado et al. 2002; Petrik et al. 2006). However, endosulfan (a pesticide permitted until 2013 and used intensively in cultivating soybeans in Argentina) showed very low frequency of occurrence (4 %) and concentration (0.01–0.06 μg/L) which could be due to its shorter half-life compared to other chlorinated groups (Stockholm Convention 2001; Souza Casadinho 2008).

In the same study (between 2004 and 2012) the different chemical forms of DDT and its metabolites were compared between children and adults. Results revealed higher percentages of *pp′*-DDE and *op′*-DDD in children (Fig. 2.4a) and significantly lower concentrations in adults ($P < 0.05$) (Fig. 2.4b). On the other hand, the analysis of the HCH isomers showed a high percentage of adult samples containing *β*-HCH (Fig. 2.5a) while concentration of this isomer was low (Fig. 2.5b). In children,

Table 2.2 Concentration and frequency of organochlorine pesticides in plasma from people living in the metropolitan area of Buenos Aires, Argentina (2006)

Organochlorine pesticide	Mean concentration (μg/L)	Maximum concentration (μg/L)	Frequency (%)
HCB	0.19 ± 0.19	1.11	70
α-HCH	0.18 ± 0.32	1.78	38
β-HCH	0.08 ± 0.17	0.81	21
δ-HCH	0.01 ± 0.06	0.33	6
$\sum$ HCH	0.09 ± 0.22	1.78	57
Lindane	0.03 ± 0.09	0.59	10
Heptachlor	0.32 ± 0.86	6.09	32
Heptachlor epoxide	0.04 ± 0.09	0.50	15
$\sum$ Heptachlor	0.18 ± 0.63	6.09	49
Aldrin	0.20 ± 0.95	5.94	19
Dieldrin	0.02 ± 0.11	0.67	11
$\sum$ Aldrin	0.11 ± 0.68	0.36	19
Endrin	0.03 ± 0.17	1.08	3
α-Chlordane	0.00 ± 0.00	0.00	4
γ-Chlordane	0.03 ± 0.09	0.60	6
$\sum$ Chlordane	0.01 ± 0.06	0.67	8
α-Endosulfan	0.00 ± 0.00	0.00	4
β-Endosulfan	0.02 ± 0.08	0.67	6
$\sum$ Endosulfan	0.01 ± 0.06	0.67	8
op′-DDE	0.10 ± 0.81	8.05	5
pp′-DDE	0.30 ± 0.44	2.46	57
op′-DDD	0.04 ± 0.20	1.80	6
op′-DDT	0.04 ± 0.20	1.57	2
pp′-DDD	0.11 ± 0.99	9.87	3
pp′-DDT	0.07 ± 0.64	6.35	71
$\sum$ DDT	0.11 ± 0.63	10.28	71
Mirex	0.03 ± 0.16	1.29	6

Table 2.3 Comparison of organochlorine pesticides (μg/L) detected in 2 years (1987 vs. 2006) in blood samples from people living in the metropolitan area of Buenos Aires, Argentina

Organochlorine pesticides	1987	2006
α-HCH	3.30 ± 2.71	0.18 ± 0.32
β-HCH	9.41 ± 5.60	0.08 ± 0.17
δ-HCH (lindane)	2.81 ± 2.12	0.03 ± 0.09
Heptachlor	2.20 ± 1.60	0.32 ± 0.86
Heptachlor epoxide	0.81 ± 0.32	0.04 ± 0.09
pp′-DDE	8.10 ± 4.31	0.30 ± 0.44
pp′-DDT	7.70 ± 4.51	0.07 ± 0.64

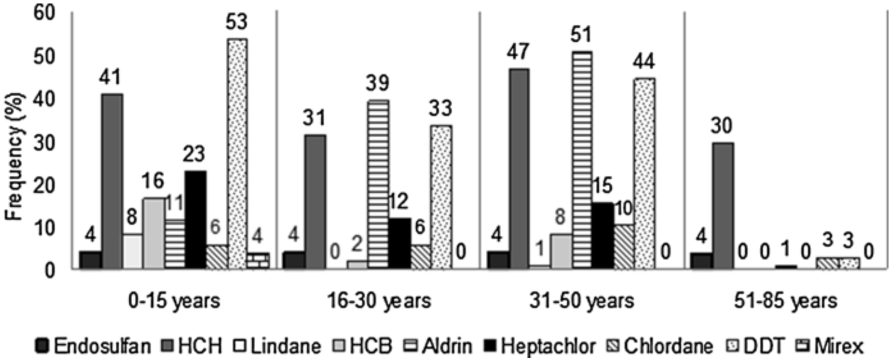

Fig. 2.2 Frequency of organochlorine pesticides by range of ages in Argentinean population

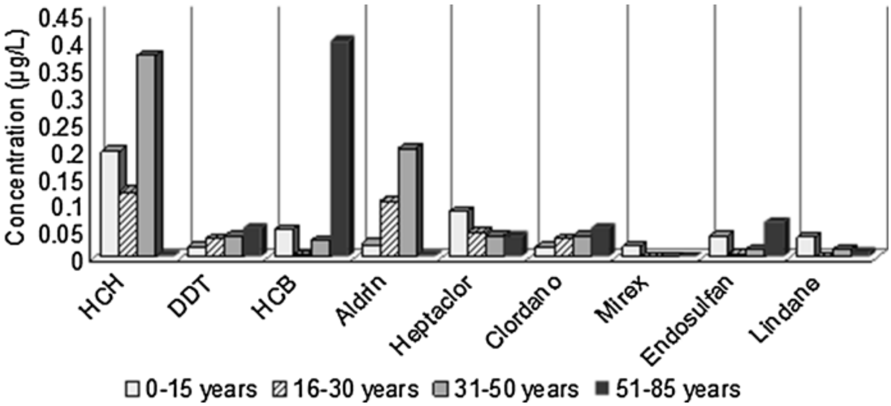

Fig. 2.3 Concentration of organochlorine pesticides by range of ages in Argentinean population

β-HCH showed higher concentration [(0.11 ± 0.43 µg/L), Fig. 2.5b]. It can be concluded that β-HCH isomer prevailed in the adults, which would result from their persistence, bioaccumulation, and prior use. In contrast, *pp′*-DDE and *op′*-DDD prevailed in children, probably because of its persistence in foods and in soils where DDT was applied. These compounds would also be transferred to the child through the placenta and breast milk. Concentrations of DDT were higher in adults, due to the property of biomagnifications (Lucena et al. 2007; Waliszewski et al. 2013).

During 2007, a descriptive and cross-sectional study of presence of OCPs in blood samples of healthy children was performed in Córdoba city (Argentina). Samples were collected in children (aged between 1 and 14 years) from two villages: Ituzaingó neighborhood [with high values and high frequency of OCPs registered in 2005 ($n = 142$)] and from children of the rest of the province [without exposure, reference population ($n = 62$)]. DDT group showed the highest frequency of occurrence in both groups (Ituzaingó: 65 %, reference: 27 %), followed by HCH isomers (Ituzaingó: 41 %, reference: 31 %) (Fig. 2.6a). Concentration of HCH was

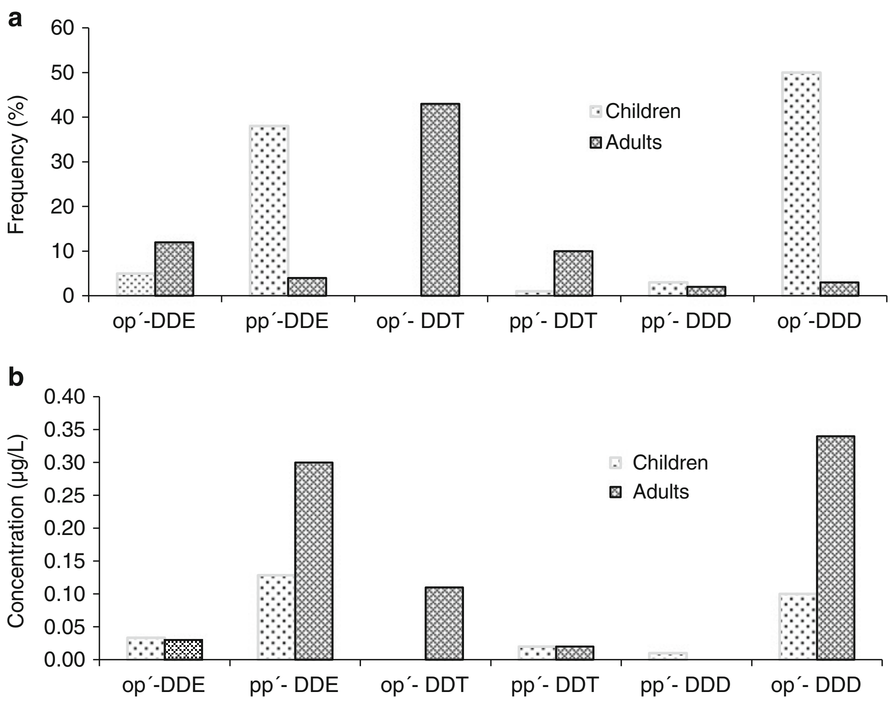

Fig. 2.4 Comparison of frequency (**a**) and concentration (**b**) of dichlorodiphenyltrichloroethane group in children and adults from Argentina

similar in both groups, but DDT was significantly lower in the reference, with respect to Ituzaingó population ($P<0.05$) (Fig. 2.6b) (Ridolfi et al. 2006, 2007; Rodríguez Girault et al. 2012).

In Argentina, as in many other countries, another side to the problematic POP pollution comes from waste management of both OCP and PCBs. It has been found that different provinces used pesticides expired or banned (Pechen de D´Angelo et al. 1998). In order to assess levels of OCP, Lucero et al. (2008) analyzed 167 blood samples from people living nearby undeclared deposits of pesticides (Cordoba province). The authors found mainly *pp′*-DDE, HCB, and *β*-HCH. The maximum concentration detected was 7.31 µg/L and corresponded to *pp′*-DDE. The *β*-HCH showed higher values than the other isomers of HCH. Good correlation between the concentration of pesticides *pp′*-DDE and age was observed. People of one of the studied sites showed *pp′*-DDT/*pp′*-DDE ratio of 0.76, according to DDT stored for over 20 years in their living place. This study provides information of the blood levels of OCP in a population with prolonged environmental exposure.

Occupational exposure is another relevant source of exposure to POPs where the income of these compounds occurs primarily through the skin and respiratory tract. Plasma levels of PCBs have been shown to be good biomarkers of recent exposure and have been proposed for monitoring exposed workers. In this context, analysis of plasma samples from 202 people working in the Electric Energy Argentina company

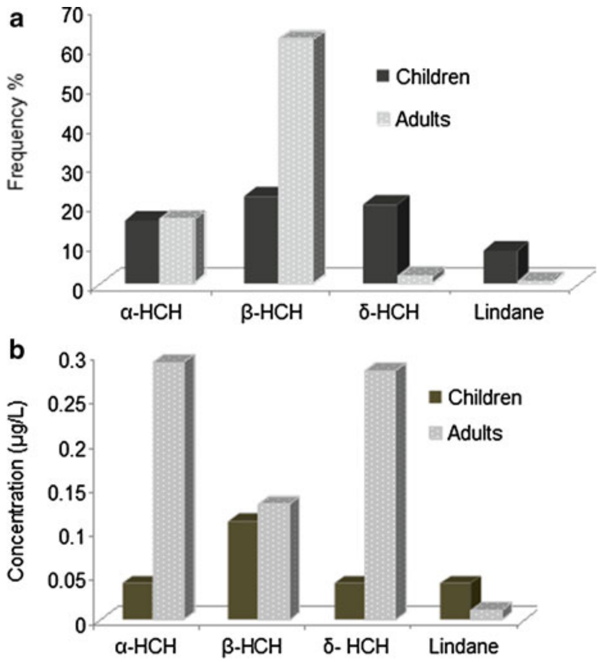

Fig. 2.5 Comparison of frequency (**a**) and concentration (**b**) of the hexachlorocyclohexane group in children and adults from Argentina

was performed (Rodríguez Girault et al. 2009). Samples were collected in 2007 after implementing preventive work practices. The values obtained were compared with values found in workers from the same company in 2005. At least one congener (among 28, 52, 101, 138, 153, and 180) was found in 92 % of the samples. The mean concentration of the sum of congeners was 0.86 ± 0.96 μg/L. Congener 52 had the highest concentration (range: ND–3.6 μg/L) (Table 2.4). In contrast, congeners that occurred more frequently were the most persistent, 138 (65 %) and 153 (63 %), followed by 52 (54 %), 180 (49 %), 101 (21 %), and 28 (20 %). Levels of total PCBs found in this study (2007) were significantly lower compared with those obtained in 2005 by the same working group ($P<0.05$) (Fig. 2.7). The average level of the sum of PCB congeners found in both studies was below that reported by Turci et al. (2006) and (Fitzgerald et al. 2007) for the general population.

Polychlorinated biphenyls "dioxin-like" present a risk of encouraging the development of cancer and may act as endocrine disruptors. Due to these characteristics the presence of PCBs in the environment brings concerns (Safe 2001; James et al. 2002). In 2011 17 PCB congeners were investigated in adults and children from different regions of Argentina. Toxic equivalencies (TEQs) of dioxin-like congeners were calculated. Two hundred and twenty plasma samples [73 from adults from the metropolitan area of Buenos Aires and Mar del Plata city and 147 from children from two Argentinean towns, Palpalá (Jujuy province) and neighborhood Ituzaingó (Cordoba province)] were analyzed. The most prevalent congeners in adult and child population were 180, 153, 138, and 170. Of the PCBs "dioxin-like," the congener 118 had the highest frequency (Table 2.5). Concentration of total PCBs and TEQ was significantly lower in children ($P<0.05$) probably because of long

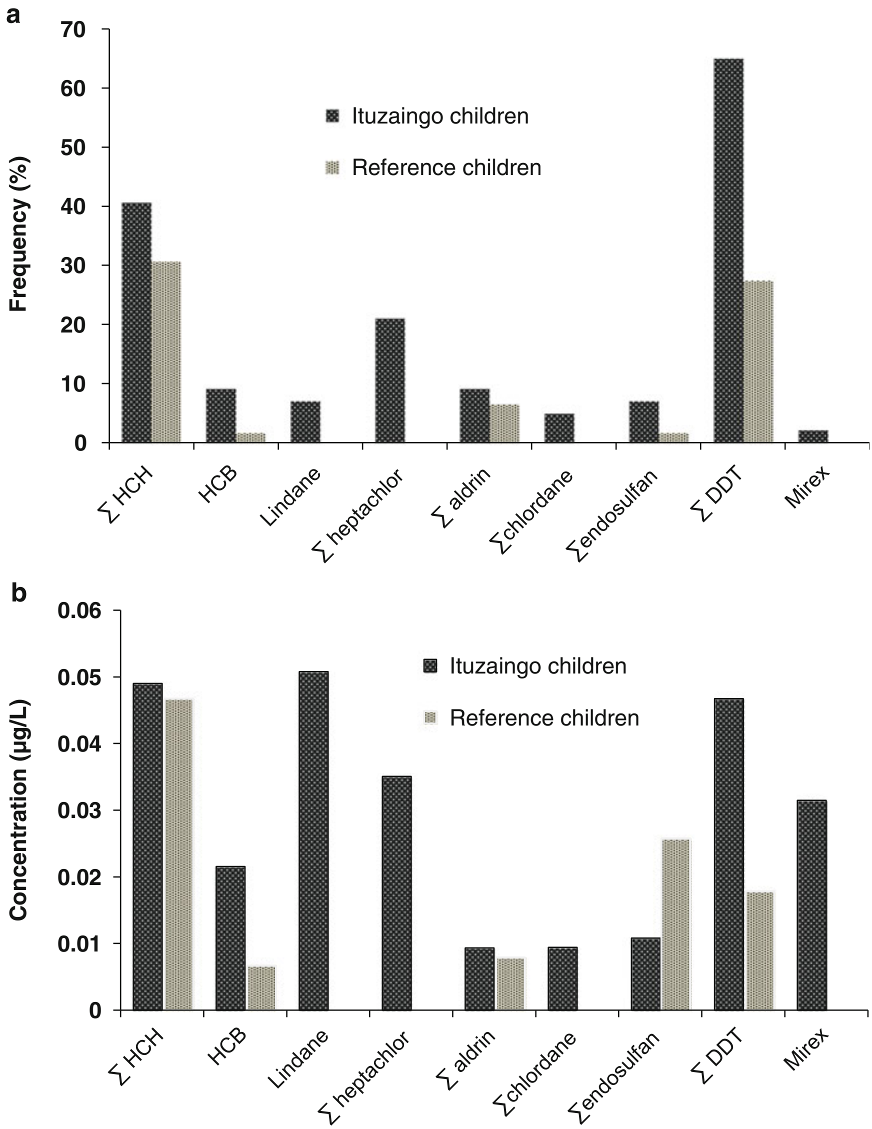

Fig. 2.6 Comparison of frequency (**a**) and concentration (**b**) of organochlorine pesticides from exposed children (Ituzaingó city) and not exposed (reference: Cordoba city)

Table 2.4 Polychlorinated biphenyl congeners (µg/L) determined in occupationally exposed population from Argentina

	Congeners						
	28	52	101	138	153	180	∑ Congeners
Concentration	0.1 ± 0.1	0.4 ± 0.7	0.1 ± 0.2	0.2 ± 0.2	0.1 ± 0.2	0.1 ± 0.1	1.0 ± 0.9
Range	ND–0.9	ND–3.6	ND–2.2	ND–2.1	ND–1.5	ND–1.0	ND–7.2

ND not detectable

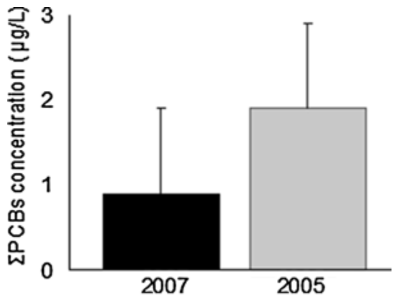

Fig. 2.7 Polychlorinated biphenyls in plasma obtained from occupational population exposed in 2 years (2005 and 2007) ($P<0.05$, Tukey test)

Table 2.5 Polychorinated biphenyl congeners (ng/g lipid) detected in adults and children from Argentina

	Adults			Children		
Congener	Mean	Maximum	Median	Mean	Maximum	Median
28	132.1 ± 142.3	795.1	92.0	28.1 ± 31.2	214.2	27.1
52	194.0 ± 306.2	1,410.0	84.1	87.0 ± 298.1	3,155.0	0.0
101	83.3 ± 110.0	720,2	58.2	73.4 ± 163.0	1,361.3	23.2
138	115.1 ± 57.4	391.1	109.4	46.1 ± 61.0	402.1	34.1
153	147.2 ± 174.0	835.0	75.1	70.2 ± 97.1	706.4	47.0
180	220.1 ± 154.1	876.2	200.0	42.1 ± 61.0	541.1	32.4
99	144.2 ± 261.0	1,212.0	33.1	54.2 ± 121.2	643.0	0.0
105	115.2 ± 89.0	3,359.2	114.3	8.0 ± 22.3	115.3	0.0
118	102.5 ± 84.2	615.1	101.1	32.4 ± 31.0	175.2	32.1
169	60.1 ± 82.3	302.0	24.0	40.1 ± 48.5	225.1	26.2
187	52.0 ± 58.1	270.3	44.2	12.1 ± 19.3	94.2	0.0
77	118.2 ± 186.2	1,044.2	18.1	30.0 ± 64.2	307.0	0.0
126	26.4 ± 26.1	110.0	22.0	19.4 ± 32.0	200.0	0.0
183	47.0 ± 53.3	194.1	30.3	11.2 ± 16.1	67.1	0.0
156	50.1 ± 45.0	172.3	50.1	12.3 ± 25.4	164.3	0.0
170	57.3 ± 33.2	198.2	55.2	30.1 ± 26.0	156.6	30.1
189	32.1 ± 53.0	195.1	0.0	1.2 ± 5.2	30.4	0.0
∑ PCB	1,696.0 ± 833.1	3,858.0	1,487.1	595.0 ± 465.1	3,997.0	475.4

exposure time in adults. These results were similar to those registered in other countries (Petrik et al. 2006; Shen et al. 2010).

Several studies have investigated levels of POPs in blood exposed occupationally and unexposed population in different countries of Latin America. In Mexico human exposure to DDT has been reported as a result of the presence of this insecticide in different environmental media. Women living in an area of malaria exposed to DDT (Veracruz city) had higher levels of DDT in blood than women who lived in San Luis Potosí, a control area not exposed to DDT (*pp′*-DDE: 14.5 μg/L and 1.8 μg/L, respectively). Correlation was also found between serum levels of DDE in mother's blood and umbilical cord blood, which shows the transplacental passage of this compound and the potential harmful effects on the fetus (Waliszewski et al. 1999).

Another study conducted in the Chiapas state (southwest of México) reported higher levels of DDTs in serum with respect to Veracruz. In Chiapas, children had higher levels of DDTs than adults and some senior workers occupationally exposed to DDT. Samples collected in San Luis Potosí and Chiapas showed high levels of DDE metilsulfonated ($MeSO_2$-DDE) that has been associated with toxic effects on the adrenal gland. The results obtained in this study identified three high-risk populations in Mexico: children, pregnant women, and workers (Díaz-Barriga et al. 2003).

Another study in Mexico assessed blood levels of PCBs and OCP in 229 children among 6–12 years who attended schools near contaminated sites of Mexico. Samples were collected in 2004 from nine sites with a history of contamination (agricultural areas with intensive use of pesticides, mining, and industrial) and urban or rural distribution. The results showed that all children had levels of *pp′*-DDE. The least persistent pesticide *pp′*-DDT was detected in 14 % of the study population. The community with higher concentrations of DDT and DDE was Puerto Madero, Chiapas, in southern Mexico, and this was due to the massive use of DDT in the malaria program. While the mean level of DDE for all the sites was approximately 2,000 ng/g lipid, the mean level was 11 times higher in Chiapas. In this study exposure to HCB was seen in 10 % of children. Three of the five sites with positive data of HCB were influenced from brick. Lindane was detected in a high percentage of children (85 %). This exposure could be due to the use of shampoos containing lindane that was still used to control scabies and lice. In this study PCB congener levels (52, 118, 138, 153, 170, and 180) were detected only in the community of Nicolas, and later it was identified that the source of PCB contamination was an oil used as fuel in brick kilns. As for health risks, the authors believe that while there is no health indicator clearly associated with DDT/DDE, previous studies have shown levels in exposed children correlated with apoptosis of immune cells, immunosuppression, and neurocognitive effects (Trejo-Acevedo et al. 2009).

In Uruguay, levels of OCP were compared in blood of exposed and not exposed population from 1979 to 1983; OCP residues were detected in all samples. The exposed population showed higher levels of *β*-HCH, *pp′*-DDE, *pp′*-DDT, dieldrin, and HCB compared to unexposed. Further studies (1985 and 1996) also assessed OCPs in the Montevidean adult population. A marked decrease in 1996 compared to 1979 was observed (Mañay et al. 2004).

In Rio de Janeiro city (Brazil), Fernandes Delgado et al. (2002) investigated levels of OCP and PCBs in blood samples from 33 volunteers (16 men, 17 women) aged between 19 and 63 years. Organochlorine residues of *op′*-DDT, *pp′*-DDT, *pp′*-DDD, *pp′*-DDE, aldrin, dieldrin, endrin, heptachlor, heptachlor-epoxide, *α-β-γ-HCH,* and HCB were detected in all samples. Polychlorinated biphenyl congeners (28, 52, 101, 138, 153, 180) were detected, as well. The *pp′*-DDE was found more frequently in a range of concentrations from 1.4 to 8.4 μg/L serum or from 0.200 to 3.452 μg/g lipids. Positive association between increased frequency and concentration of *pp′*-DDE with the age was found. This study suggests that the inhabitants of the urban area of Rio de Janeiro city would be exposed to relatively low levels of POPs.

In Colombia, Varona et al. (2010) explored exposure to OCP in blood of individuals living in a region of illegal crops. During 2005 and 2006, 99 serum samples were collected. Heptachlor (73 %), *pp′*-DDE (19 %), aldrin (15 %), *γ*-chlordane (12 %), dieldrin (11 %), α-chlordane (10 %), α-endosulfan (8 %), β-endosulfan (5 %), oxychlordane (3 %), *pp*′-DDT (3 %), and *op*′-DDT (2 %) were detected. Heptachlor presented a median of 8.7 ng/L and maximum of 43.8 ng/L. In this context, authors opined that prohibited OCPs were being used in some regions of Colombia.

Considering their persistence and toxicity, the Pan American Health Organization (PAHO) implemented a surveillance program in Mesoamerica in order to detect DDT residues in children's blood. This program was carried out in communities from Mexico, Guatemala, El Salvador, Honduras, Nicaragua, Costa Rica, and Panama. Results showed that in some areas the children had high concentrations of DDT, particularly in Mexico (mean level 50.2 ng/mL). The *pp*′-DDT/*pp*′-DDE quotient was higher than one in some communities reflecting recent exposure. Authors suggested that more efforts are needed to prevent the reintroduction of DDT into the region (Pérez-Maldonado et al. 2010).

In eastern Bolivia, human exposure to DDT in blood samples of farmers in three rural communities was investigated. *pp*′-DDE was found in 100 % of the samples, with a median concentration of 19.7 ng/mL (4,788.7 ng/g lipid), while *op*′-DDT was detected in three samples (4.3 %). The *pp*′-DDE serum concentrations were associated with length of residence in the study area, personal hygiene after work, and body mass index. The results revealed high levels of *pp*′-DDE in the population under study, which is due to high occupational exposure in the past and very polluted environment (Mercado et al. 2013). In Argentinean population, OCP levels are lower than other countries, with prevalence of HCH and DDT groups (Table 2.6).

Table 2.6 Organochlorine residues (µg/L) founded in human blood from different countries

Country/city	Period	*n*	∑ HCH	∑ DDT	References
Brazil	–	42	32.4	76.9	Minelli and Ribeiro (1996)
Germany	–	25	6.4	15.4	DeVoto et al. (1998)
Mexico	–	65	1.6	16.4	Waliszewski et al. (1999)
Sweden	–	790	51.6	836.1	Glynn et al. (2000)
Spain	1997–1999	141	1.1	2.2	Sala et al. (2001)
Japan	–	41	0.5	6.3	Hanaoka et al. (2002)
Belgium	–	251	–	8.2	Charlier and Plomteux (2002)
Portugal	2001–2002	203	13	93.5	Cruz et al. (2003)
Spain	1998–2000	102	13.8	49.4	Falcon et al. (2004)
Uruguay	1996	68	1.1	6.4	Mañay et al. (2004)
Spain	1997–1998	682	–	370.0	Zumbado et al. (2005)
Portugal	1997–2001	160	24.9	74.7	Lino and Silveira (2006)
Romania	2005	142	1,114.0	2,420.0	Dirtu et al. (2006)
Poland	2004	44	3.9	401.1	Jaraczewska et al. (2006)
UK	2003	154	15.0	100.2	Thomas et al. (2006)
Spain	1992–1995	405	1,291.4	4,895.8	Porta et al. (2008)
Delhi	–	112	23.5	1.7	Pathak et al. (2009)
Dibrugarh	2009–2010	169	348.1	417.0	Mishra et al. (2011)
Bolivia	2013	70	–	19.7	Mercado et al. (2013)
Argentina	2004–2012	649	0.3	0.4	Ridolfi (unpublished)

2.2.3 Breast Milk

Persistent organic pollutants are accumulated in the fat of the different tissues due to its lipophilic properties. These compounds are translocated and excreted through breast milk during lactation. Therefore it is an important route of exposure for infants during the first months of life. Early exposure to POPs during the neonatal period may affect child neurodevelopment and growth. The concentrations of these compounds in human milk are a nice indicator of environmental contamination (Díaz-Barriga et al. 2003; Der Parsehian 2008).

Der Parsehian (2008) studied in Argentina the presence of OCP in breast milk of 248 postpartum women from the Hospital Materno Infantil Ramon Sarda of Buenos Aires (from 2000 to 2001 and 2003 to 2004). Ninety-one percent of the samples showed OCP residues. Most frequently used pesticides were *pp′*-DDE (86.7 %), HCB (26.6 %), heptachlor epoxide (25.4 %), *β*-HCH (23.0 %), and chlordane (15.7 %). The highest concentration and the maximum value corresponded to *pp′*-DDE (8.98 ng/ml and 200.4 ng/ml, respectively). These results are consistent with other studies conducted in many countries.

Della Ceca et al. (2012) recently assessed exposure to OCP and PCBs in 59 breast milk samples from women living in Buenos Aires, Argentina, collected during 2010 and 2011. Concentrations of POPs found in breast milk decreased in the following order: DDTs ≈ PCBs > HCHs > heptachlor. PCB values ranged from 22 to 258 ng/g lipid, with a clear predominance of the most persistent congeners: 118 (13.0 ± 9.0 %), 138 (21.0 ± 9.0 %), 153 (27.0 ± 8.0 %), and 180 (20.0 ± 5.9 %). This reflects the persistence of higher molecular weight PCBs and their levels were comparable to previously reported in the literature for other areas (Kalantzi et al. 2004; Tanabe and Kunisue 2007; Polder et al. 2009; Kozul and Romanić 2010; Cok et al. 2012). Dichlorodiphenyltrichloroethane levels ranged from 7.7 to 500 ng/g lipid (76.0 ± 91.0), prevailing in all samples the main metabolite *pp′*-DDE (90.0 ± 17.0). This is consistent with increased lipophilicity and therefore higher bioaccumulation in fat tissue of DDE in comparison to DDT. Both compounds showed very low levels compared with values reported in other countries (Kalantzi et al. 2004; Jaraczewska et al. 2006; Azeredo et al. 2008; Polder et al. 2009). Hexachlorocyclohexane levels were higher (range: 5.8–197, mean: 33.0 ± 37.0 ng/g lipid) than those reported in literature (Sudaryanto et al. 2006; Polder et al. 2009). In both studies reported in Argentina, it was concluded that in maternal milk of the studied populations, the degradation products (DDE, heptachlor epoxide) and more persistent congeners and isomers (*β*-HCH, PCBs penta-hexa 180, 138, 118, and 153) predominated. This reflects the greater stability of these compounds and their high bioaccumulation potential.

In Latin America, data reported belong preferentially to levels of DDT and its metabolites, due to intensive use of this pesticide in areas of malaria.

Studies in Mexican population by Díaz-Barriga et al. (2003) reported that concentrations of DDT and metabolites (DDT, DDD, and DDE) in human milk samples collected between 1994 and 1995 were higher in agricultural communities exposed to this pesticide and in samples from areas of malaria with intensive use of DDT with respect to urban areas, where DDT was never applied (Mexico city). Following the banning of DDT since 2000, a decline in DDT levels in later years is expected. The estimated infant's daily intake in this study, considering the average concentrations of DDT and its body weight, was three times higher than that established by the World Health Organization's Acceptable Daily Intake (ADI) of 20 μg/kg/day.

In order to assess environmental exposure to OCP in Colombian population, Rojas-Squella et al. (2013) conducted a study in 32 milk samples from Colombian mothers. The results obtained corresponded to the most frequent *pp′*-DDE with a mean value of 203 ng/g lipid, and a range of <17 and 14,948 ng/g lipid. According to the authors, Colombia ranks fourth from bottom to top in terms of *pp′*-DDE average concentrations based on the results obtained from the POP Global Monitoring Plan report of 2009 of the Stockholm Convention.

Dichlorodiphenyltrichloroethane and its metabolites *pp′*-DDE and *pp′*-DDD were detected in 69 samples of maternal milk of inhabiting of Rio Madeira (Brazil). The range of concentrations found were *pp′*-DDE (10.7–7,271.5 ng/g lipid), *pp′*-DDD (ND–400.7 ng/g lipid), and *pp′*-DDT (3.0–2,534.1 ng/g lipid). Whereas the sum of all the average DDT concentration was 369.6 ng/g lipid and ranged from 25.4 to 9,361.9 and 8.7 % of the estimated daily intake (EDI), based on total DDT, it was higher than the acceptable daily intake proposed by the WHO. Although the studied population belonged to an area of numerous cases of malaria in the past, and intensive use of DDT until 1990, the concentrations found were lower than those reported in another study in Rio de Janeiro, in 2000, prior to the banned use of DDT for agriculture programs and malaria control (Azeredo et al. 2008).

Figure 2.8 shows the levels of DDTs and PCBs found in breast milk in Latin American countries. Argentina's population showed low levels compared with values reported in other areas.

The decreased concentrations of OCP and particularly DDT in different works reported in recent years are probably due to the prohibition of their use for agriculture in various Latin American countries, although their metabolites are still detected in the biological samples, due to their high persistence and bioaccumulation.

2.3 Concluding Remarks

The extensive use of OCP in agriculture and PCB on industry, in confluence with its lipophilicity and resistance to environmental and metabolic degradation, determined its persistence over the environment. These characteristics have defined the ability of POP bioaccumulation, increased by the generation of toxic metabolites of similar chemical characteristics which has developed current scenario of chronic exposure to residues of these compounds.

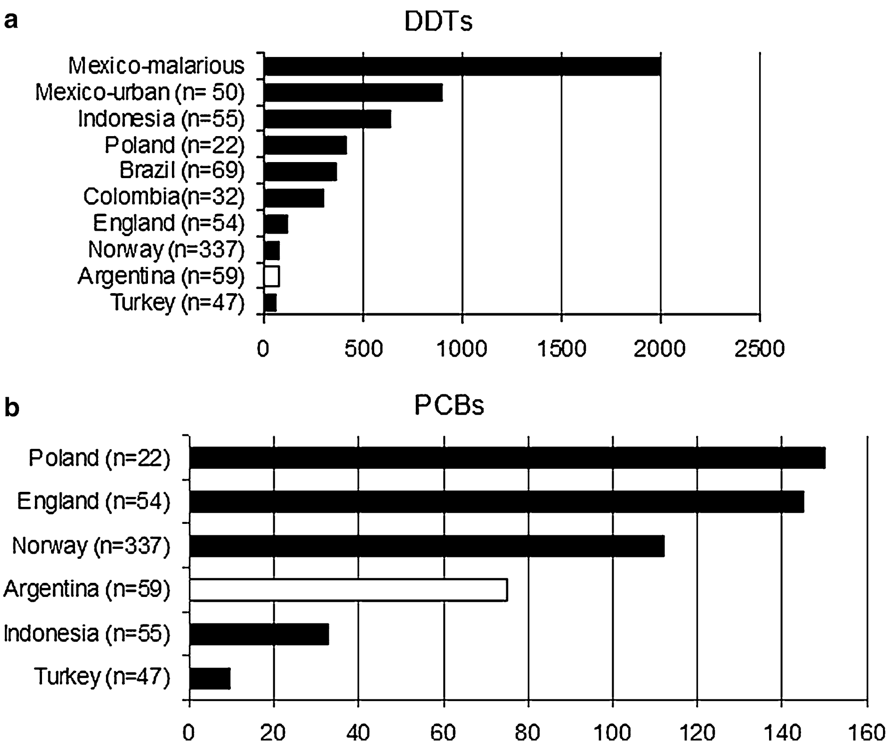

Fig. 2.8 Concentration (ng/g lipids) of dichlorodiphenyltrichloroethane (**a**) and polychlorinated biphenyls (**b**) in breast milk from different countries

In Argentina and Latin America there is little information on levels of POPs in human samples. Most studies are based on the investigation of OCP in exposed and unexposed populations. Blood was the most commonly used biological matrix. HCB, *pp*′-DDE, *pp*′-DDT, and *β*-HCH were the OCPs that were seen most frequently.

Some studies examined the presence of PCBs in serum, breast milk, and adipose tissue, predominantly the most persistent congeners 138, 153, 180, and 170.

The concept of toxicity due to POPs in child population is related to environmental long-term exposure to low concentrations, and increased susceptibility. Studies in Argentina show the presence of OCP in children even after decades of prohibition; for this reason children should be evaluated through epidemiological monitoring because they are not occupationally exposed, but reflect environmental exposure to a greater extent than adults.

The results of studies in Latin American populations demonstrate exposure to POPs, whose levels depend on age, sex, and place of residence.

Although the concentrations of these compounds in human biological samples have decreased in recent years, exposure continued due to their persistence and evidence of reuse of banned OCP in some Latin American countries.

Health consequences resulting from the use of OCP and PCBs are mainly due to the lack of information, failure to apply the existing laws, and inadequate supervision and awareness of the problem.

Population monitoring programs in conjunction with environmental evaluations will provide data that will allow government authorities to implement policies to bioremediation of contaminated sites and minimize the risk of human exposure to these persistent toxic compounds in the region.

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