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Levels of organochlorine contaminants in organic and conventional cheeses and their impact on the health of consumers: An independent study in the Canary Islands (Spain)

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ABSTRACT

In the present work we have evaluated the levels of polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) in 61 commercially available brands of cheese (54 conventional and 7 organic) to estimate their relevance as a source of organochlorines. Our results showed that hexachlorobenzene, α-HCH, dieldrin, *p,p′*-DDE, and PCBs 153 and 180 were present in most of the samples independent of the cheese type. The concentration of OCPs was low for both types of cheese, although organic had lower concentrations than conventional. The estimated daily intake (EDI) of OCPs was lower than the tolerable daily intake (TDI). The levels of PCBs in cheese were also low; however, there were higher levels of PCBs in organic than in conventional brands. Levels of dioxin- like PCBs (DL-PCBs) in both types of cheese reached concentrations in the 75th percentile higher than 3 pg WHO–TEQ/g fat, and above 100% of the levels established by the EU. People consuming the most contaminated brands could have an EDI well above the recommended TDI (2 pg WHO–TEQ/kg bw/day). These results are of concern as the adverse health effects exerted by dioxin-like compounds are well known.

1. Introduction

Organochlorine contaminants include a large variety of toxic substances, such as polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), and dioxins (PCDDs), and are characterized by the presence of halogenated atoms in their structure and their high lipophilicity which results in its ability to persist in the environment, ultimately accumulating and biomagnifying up the food chains.

Different sources of human exposure have been identified (Linares et al., 2000); however more than 90% of the human exposures to these environmental contaminants can be attributed to the consumption of contaminated food, where animal and fish products are the main sources of exposure (Liem et al., 2000). In fact, it has been reported that residues of these chemicals are found mainly in meat, fish, dairy products, and eggs (Domingo and Bocio, 2007; Llobet et al., 2003). Dairy products, because of the high fat content, are a dietary route of exposure for organochlorine compounds (Focant et al., 2002) and they supply approximately 30% of the total dietary intake with chlorinated contaminants in Western populations (Bordajandi et al., 2004; Focant et al., 2002). To monitor the presence of these substances in food, international agencies have developed food contaminant monitoring programs and total diet studies. The control of contaminants in food identified for human consumption and in animal feed are regulated through the EU Council Regulation 315/93/EEC, 1993 and Maximum Residue Levels (MRLs) have been established for the regulation of pesticides and other toxic contaminants in food (Regulation 396/2005/EC). Additionally, a tolerable daily intake (TDI) has been established by international agencies for these compounds.

The population of the Canary Islands has been studied in depth regarding its levels of contamination by OCPs and PCBs; the results show that this population has been exposed to a relatively high levels as a result of the chronic exposure to OCPs contamination by OCPs indicating the existence of a that persisted throughout the 1990's (Luzardo et al., 2006, 2009; Zumbado et al., 2005); and has been exposed to PCBs at low concentrations (Henriquez-Hernandez et al., 2011), in spite of the fact that such substances were banned in Spain in the late 1970's. As mentioned previously, the main route of exposure to organochlorines appears to be the intake of fish and dairy products (Agudo et al., 2009), and the population of the Canary Islands has been known to consume high quantities of dairy products (Serra Majem et al., 2000a,b). Additionally, we have

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recently reported that, among the consumers of dairy products from these islands, only milk consumption could account for exposures greater than the TDI for dioxin-like compounds established by the World Health Organization (WHO) (Luzardo et al., 2012). Under this scenario, and to complete the evaluation of the significance of dairy products as a source of organohalogenated contaminants exposure, the objectives of this study were to investigate the concentration levels and the patterns of organochlorine contaminants (OCPs and PCBs) in 54 of the top-selling, commercially available brands of cheese conventionally produced and 7 commercially available brands of cheese organically produced that are sold from various supermarkets in the Canary Islands (Spain), and to estimate the dietary exposure in humans of this archipelago.

2. Materials and methods

2.1. Study area

The Canary Islands are located in the Atlantic Ocean, near the African coast (southwest of Morocco). Geographically, the Islands are part of the African continent; however, from an historical, economic, political and socio-cultural point of view, the Canaries are entirely European. In the Canary Islands, as in the rest of Spain, there is a long tradition of production and consumption of the local cheeses (made from milk produced on the islands), but there is also wide distribution and consumption of cheeses from mainland Spain and other European countries.

The Canary Islands' economy relies primarily on a few economic sectors: tourism and, to a lesser extent, fishing and agriculture (where, in recent decades, agriculture has become more aggressive with an increased use in greenhouses). Other economic sectors, such as industries who contributed to pollutant emissions as traditional polluting have relatively limited presence in the Islands.

2.2. Sampling and collection

In this study, 54 commercial brands of conventionally produced cheeses were randomly selected from high delivery rate supermarkets of the Canary Islands. To investigate other possibilities, we also included 7 brands of organic cheese in the study. Samples were collected between December 2006 and April 2008. Whereas all the organic cheeses collected were produced in mainland Spain or in north European countries, 38 (70%) of the 54 conventionally produced cheeses, were locally produced.

Each of the 61 selected brands (six samples for any brand) was sampled monthly during this period of time to obtain a representative estimation for each brand and to study potential fluctuations in concentrations between different batches. The lipid content of the samples was determined in triplicate by the Gerber method with a butyrometer (with a graduation scale of 0 to 40%) to obtain the final lipid-corrected values. All samples were collected and frozen at −80 °C until analysis. The average fat content the in conventional cheese was 21%, whereas the average fat content in organic cheese was 29%.

2.3. Sample preparation and analytical procedure

The cheese stored at −80 °C was acclimated to room temperature and homogenized with 5 ml of water (previously cleaned with hexane) per gram of cheese. A total of 20 g of this homogenate was lyophilized for 72 h before removing 2 g of the lyophilized cheese for the extraction of fat, OCPs, and PCBs according to a Soxlet extraction method (FOSS Soxtec Avanti 2055) and EU recommendations (EN 1528-2, 1996). The extracts (extracted in 3 ml of dichloromethane; DCM) were purified using gel permeation chromatography (GPC), as recommended by the European Standard EN 1528-3:1996 for the determination of pesticides and polychlorinated biphenyls in fatty foods. To achieve maximum sensitivity in our analysis, two sequential cleanup steps were performed; the 3 ml fatty extract in DCM obtained by Soxlet was divided into three 1 ml aliquots that were then individually purified using GPC with a 100% Fluorinated divinylbenzene GPC column (50 cm × 10 mm i.d. EPA 3640a Pesticide Cleanup GPC Jordi column, Sorbtech Technologies, Atlanta, USA), and using DCM as the eluting solvent at a flow rate of 1.6 ml/min. This GPC system was operated using an automated apparatus (GPC-CL1, Cromlab S.L., Barcelona, Spain). The first fraction (22 ml) eluted, contained the lipids and was discarded. The second fraction (14 ml), contained the organochlorine compounds and was collected. The three organochlorines-containing fractions per sample were combined and evaporated to near dryness leaving a small amount of oily residue. The oily residue was dissolved in 1 ml DCM and subjected to GPC purification, thereby obtaining a new 14 ml organochlorine-containing fraction that was dried and diluted with cyclohexane up to a volume of 200 μl. This diluted sample was used for GC–MS analysis using the appropriate internal standards.

2.4. Analytes of interest

The following were the analytes of interest in this study: the diphenyl-aliphatic pesticides and metabolites (methoxychlor, *p*,*p*′-DDT, *o*,*p*′-DDT *p*,*p*′-DDE, *o*,*p*′-DDE, *p*,*p*′-DDD, and *o*,*p*′-DDD); the persistent and bioaccumulative contaminant hexachlorobenzene (HCB); the four isomers of hexachlorocyclohexane (α-, β-, γ-, and δ-HCH); the cyclodienes dieldrin, aldrin, endrin, heptachlor epoxide (cis- and trans-isomers), chlordane (cis- and trans-isomers) and mirex; and endosulfan (α- and β-isomers). In this study, we also included the measurement of the most relevant PCB congeners (IUPAC numbers# 28, 52, 77, 81, 101, 105, 114, 118, 123, 126, 138, 153, 156, 157, 167, 169, 180, and 189).

2.5. Procedure of chemical analyses

Chromatographic analysis was performed using 4a Thermo-Finnigan TRACE DSQ GC/MS instrument as previously reported (Dmitrovic and Chan, 2002; Luzardo et al., 2009). A fused silica capillary column BPX5 (Crosslinked 5% phenyl methylpolysiloxane, SGE Inc., USA) with a length of 30 m, an i.d. of 0.25 mm and a film thickness of 0.25 μm was used as the stationary phase. Helium, at a flow rate of 1 ml/min, was used as the carrier gas. The oven temperature was programmed as follows: an initial oven temperature of 80 °C held constant for 1 min, increased to 300 °C at 10 °C/min increments and then held at 300 °C for 9 min. The Injector and transfer line temperatures were set at 200 °C and 310 °C, respectively. Standards and samples were injected (2 μl) in splitless mode. Two chromatographic analyses were performed for each sample to obtain mass spectra in two different ionization modes. For DDT and metabolites, methoxychlor, endrin, and PCB congeners 28, 52, 101, and 118, mass spectra were obtained in electronic impact mode (GC/EIMS) at 70 eV, with an ion source temperature of 200 °C. For the remaining analytes included in this study, mass spectra were obtained in negative chemical ionization mode (GC/NCIMS) using methane as the buffer gas at a flow rate of 2.5 ml/min. Both GC/EIMS and GC/NCIMS analyses were conducted using selected ion monitoring (SIM). Tetrachloro-*m*-xylene was used as the nternal standard (IS) in the GC/EIMS mode, and PCB 202 as the IS in the GC/NCIMS mode. We determined the limit of quantification (LOQ) to be 10-fold the standard deviation of the blank and the limit of detection (LOD) as half of the LOQ. Nevertheless, only LOQ has been employed throughout this study (values below the LOQ have not been considered).

The MS system was routinely programmed in SIM using one target and two qualifier ions. Confirmation of the organochlorine pollutants was determined by the retention time of the target ion and the two qualifier-to-target ion ratios. The abundance of the target and qualifier ions were determined by injecting individual pollutant standards and using full scan mode (50–500 *m*/*z*) under the same chromatographic conditions. The qualifier-to-target ion ratio was then determined by dividing the abundance of the selected qualifier ion by that of the target ion (almost consistently the base peak) and multiplying by 100. Determination of the relative percent of the theoretical relative abundance uncertainty of the qualifier ions was conducted using the criteria described in the EU recommendations (SANCO/2007/3131). Quantification was based on the pollutant target ion/IS peak area ratio and was achieved by using linear regression: a six-point calibration curve was generated from the standard solutions ranging from LOQ of each pollutant to 10 ng/ml and by using the GC–MS Xcalibur 2.0.7 software. The standard analytes were purchased from Dr. Ehrenstorfer (Riedel-de Haën, Sigma–Aldrich Laborchemikalien GmbH, Germany).

The daily variation in the method was evaluated over 5 days using duplicates of these two different pools of spiked samples. The coefficient of variation was <20% for every case and was therefore considered acceptable.

In this study, we expressed the total value of OCP residues as the sum of the quantified 22 OCPs and its metabolites; the total value of HCH residues ($\sum$HCHs) as the sum of the 4 HCH isomers quantified (α-, β-, γ-, and δ-HCH); and the total value of cyclodienes quantified ($\sum$cyclodienes) as the sum of aldrin, dieldrin, endrin, cis-chlordane, trans-chlordane, and heptachlor epoxide (cis- and trans-isomers). Because the cyclodiene endosulfan was banned recently (December 2005, 2005/864/EC), we have considered this pesticide separately, expressing the total value of endosulfan residues ($\sum$endosulfan) as the sum of the 2 quantified endosulfan isomers (α-, and β-endosulfan).

Similarly, we expressed the total value of PCB residues ($\sum$PCBs) as the sum of the 18 PCBs quantified. Additionally, the congeners considered as markers of environmental contamination for PCBs (IUPAC congeners #28, 52, 101, 118, 138, 153, and 180) were considered as a group ($\sum$Marker-PCBs; $\sum$M-PCBs), and total value of dioxin-like-PCB residues (DL-PCBs) were also expressed as the sum of the 12 DL-PCBs quantified ($\sum$DL-PCBs; IUPAC congeners #77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189).

We also estimated the potential toxicity (in terms of toxic equivalence to dioxins; TEQs) of the DL-PCBs quantified using the toxicity equivalent factors (TEF), as revised by the World Health Organization (WHO) in 2005 (Van den Berg et al., 2006). We also expressed the total TEQ ($\sum$TEQs) as the sum of the TEQs obtained from the quantified DL-PCBs.

2.6. Exposure assessment

Dietary intake was calculated by multiplying the respective concentration (median) by the amount of cheese consumed by an average adult from Canary

Islands (18 years and above, average weight 70.1 kg) per day. Exposures for small children (6–10 years; average weight, 30.4 kg) were also estimated.

Exposures were assessed for both OCPs and DL-PCBs. For calculations, when a congener concentration was under the limit of quantification (LOQ), the value was assumed to be 0 (lower bound approach).

Consumption data of cheese by the Islands population were obtained from the Canary Islands Nutritional Survey (Serra Majem et al., 2000a,b).

The daily intake of dioxins (pg WHO–TEQ kg^{-1} b.w d^{-1}) is equal to the occurrence (pg WHO–TEQ g^{-1} w.w.) multiplied by the consumption (g kg^{-1} b.w. d^{-1}).

2.7. Statistical analyses

Database management and statistical analyses were performed using PASW Statistics v 17.0 (SPSS Inc., Chicago, IL, USA). The OCP and PCB concentrations did not follow a normal distribution; therefore, the results are expressed in terms of the median, the 25th and 75th percentiles (p25 and p75, respectively), and range (values maximum and minimum). Differences in the OCP and PCB levels between three groups were tested with the non-parametric Mann–Whitney U-test and Kruskal Wallis test. The categorical variables are presented as percentages and were compared between variables with the Chi-square test. A P value of less than 0.05 (two-tail) was considered to be statistically significant.

3. Results and discussion

The concentrations of OCPs and PCBs did not show significant differences among the six collected samples from each cheese brand during the period of collection (data not shown). As a consequence, we have used the median, maximum and minimum values (range), and 25th–75th percentiles of the distribution obtained for each chemical in all samples analyzed (Tables 1a, 1b, 2a, and 2b).

3.1. Occurrence of selected OCP residues in cheese samples

The results showed that all analyzed samples (100% of the cheeses) had a quantifiable amount of OCP residues, HCB and α-HCH being the most frequently observed residues (Tables 1a and 1b). The number of OCP residues was similar in both types of cheeses; therefore, an average of 10 OCP residues per sample was measured in non-organic cheese samples (within a range of 3–14), whereas organic cheese samples had an average of 8 OCP residues per sample (within a range of 6–11).

Only the cyclodiene trans-chlordane was observed more frequently in conventional cheese brands than in the organically produced cheese brands (89% vs. 43%, respectively; $p = 0.002$). Furthermore, all the analyzed samples (both, from conventional and organic cheeses) showed, to some extent, the presence of HCH-isomer residues, although the total HCH residue level (∑HCHs) was higher in conventional than in organic cheese samples (6.64 *vs.* 1.35 ng/g fat, respectively; $p = 0.007$) (Table 1a). This result may be attributed to lindane's (γ-HCH, currently banned in Spain) recent use as an ectoparasitic agent in livestock by non-organic farmers (Botella et al., 2004) and, was, in fact found at higher levels in conventional cheeses brands than in organic cheese brands (median values of 3.24 *vs.* 0.51 ng/g fat, respectively; $p = 0.005$). Similarly, the levels of α-HCH were also higher in conventional cheeses than in samples from organically produced brands of cheese (median values of 1 *vs.* 0.22 ng/g fat, respectively; $p = 0.008$) and higher levels of cyclodiene pesticides (Σcyclodienes) in conventional cheese brands than in organic cheese brands (median value of 10.52 and 2.73 ng/g fat, respectively; $p = 0.014$). Specifically, the cyclodienes dieldrin and trans-chlordane were found at higher levels in conventionally produced cheeses than in organic cheeses (median values 6.61 *vs.* 2.64 ng/g fat, respectively; $p = 0.031$ and 2.08 *vs.* 0.00 ng/g; $p = 0.002$, respectively); however, the pesticide dieldrin was found in all the organic brands cheese samples, and in 87% of the samples from conventionally produced cheeses.

There were no differences in the levels of endosulfan residues (Σendosulfan) between both types of cheeses. Similarly, the residue levels of DDT-derivatives (ΣDDTs) in the organic cheese brands were similar compared with the levels in the conventional brands of cheese. Nevertheless, the most ubiquitous DDT-derivative, *p*,*p*′-DDE, was quantified in most cheese samples (83% in conventional and 100% in organic brands) and was the pesticide found in the highest concentration in both types of cheese.

As a consequence of the aforementioned results, the total burden of OCPs (∑OCPs), was higher in conventional than in organic brands of cheese (median values of 42.73 *vs.* 14.44 ng/g fat, respectively; $p = 0,001$).

Nevertheless, as shown in Tables 1a, 1b, and 3, the median levels of OCPs in both types of cheeses were found to be relatively low and consistently below the maximum residue limit (MRL) established by the European Legislation (OJEC, 1993 and 1994).

The results obtained for OCP residues in cheese are consistent with those found for milk samples (Luzardo et al., 2012), and

Table 1a

Frequency of detection (%), and average concentrations (ng/g fat) of organochlorine pesticides found in conventional and organic cheese samples from the Canary Islands market (Spain).

OC–compound	Conventional cheese ($n = 54$)				Organic cheese ($n = 7$)				p^a	p^b
	Mean ± SD	Median (p25–p75)	Range	%	Mean ± SD	Median (p25–p75)	Range	%		
HCB	6.95 ± 7.77	4.62 (1.39–10.01)	n.d.–34.63	98.0	2.27 ± 1.46	1.77 (1.38–2.45)	1.13–5.38	100.0		
HCH										
α-HCH	1.65 ± 2.64	1.00 (0.32–1.80)	n.d.–17.35	98.0	0.23 ± 0.10	0.22 (0.19–0.23)	0.11–0.45	100.0	0.008	
β-HCH	21.07 ± 62.54	1.69 (0.43–5.82)	n.d.–335.98	85.7	0.65 ± 0.58	0.35 (0.23–1.16)	n.d.–1.60	85.7	0.054	
δ-HCH	0.03 ± 0.23	0.00 (0.00–0.00)	n.d.–1.73	1.9	n.d.	n.d.	n.d.	0.0		
Lindane (γ-HCH)	8.46 ± 21.01	3.24 (1.01–6.26)	n.d.–115.25	87.0	0.53 ± 0.51	0.51 (0.00–0.89)	n.d.–1.39	71.4	0.005	
∑HCH	31.22 ± 78.99	6.64 (3.09–18.20)	0.25–349.79	100.0	1.41 ± 0.50	1.35 (1.08–1.78)	0.62–2.18	100.0	0.007	
Cyclodienes										
Aldrin	1.19 ± 3.32	0.00 (0.00–0.97)	n.d.–22.06	38.9	0.08 ± 0.14	0.00 (0.00–0.25)	n.d.–0.31	28.6		
Dieldrin	10.47 ± 13.23	6.61 (2.83–11.96)	n.d.–68.24	87.0	3.01 ± 1.32	2.64 (2.06–4.02)	1.67–5.52	100	0.031	
Endrin	n.d.	n.d.	n.d.	0.0	n.d.	n.d.	n.d.	0.0		
cis-Chlordane	2.44 ± 5.26	0.99 (0.00–2.13)	n.d.–32.41	70.4	0.62 ± 0.61	0.76 (0.00–1.12)	n.d.–1.42	57.1		
trans-Chlordane	2.39 ± 2.02	2.08 (0.46–3.27)	n.d.–7.47	88.9	0.16 ± 0.21	0.00 (0.00–0.38)	n.d.–0.47	42.9	0.002	0.002
∑Chlordanes	4.83 ± 6.72	2.89 (0.97–5.42)	n.d.–39.88	88.9	0.77 ± 0.70	1.01 (0.00–1.24)	n.d.–1.80	71.4	0.012	
Heptachlor	2.09 ± 2.97	0.18 (0.00–4.09)	n.d.–12.31	57.4	0.18 ± 0.23	0.00 (0.00–0.43)	n.d.–0.53	42.9		
∑Cyclodienes	13.75 ± 15.69	10.52 (3.35–16.61)	n.d.–81.40	88.9	3.27 ± 1.37	2.73 (2.26–4.56)	2.06–5.77	100.0	0.014	

SD: standard deviation; p25: 25th percentile; p75: 75th percentile; %: percentage of detectable samples; HCB: hexachlorobenzene; HCH: hexachlorocyclohexanes; n.d.: non-detectable.

[a] values result from the comparison between the medians (Mann–Whitney test).

[b] values result from the comparison between the frequencies of detection (Chi-square test).

Table 1b
Frequency of detection (%), and average concentrations (ng/g fat) of organochlorine pesticides found in conventional and organic cheese samples from the Canary Islands market (Spain).

OC-compound	Conventional cheese (*n* = 54)				Organic cheese (*n* = 7)				p^a	p^b
	Mean ± SD	Median (p25–p75)	Range	%	Mean ± SD	Median (p25–p75)	Range	%		
Endosulfans										
α-Endosulfan	1.18 ± 2.30	0.00 (0.00–1.94)	n.d.–14.49	46.6	0.10 ± 0.16	0.00 (0.00–0.30)	n.d.–0.37	28.6		
β-Endosulfan	0.55 ± 1.15	0.00 (0.00–0.61)	n.d.–5.19	33.3	n.d.	n.d.	n.d.	0.0		
∑Endosulfans	1.73 ± 2.68	0.22 (0.00–3.00)	n.d.–14.49	51.9	0.10 ± 0.16	0.00 (0.00–0.30)	n.d.–0.37	28.6		
Diphenyl-alyphatics										
p,p′-DDE	26.21 ± 50.65	9.99 (2.28–21.38)	n.d.–303.15	83.3	4.81 ± 3.59	5.80 (1.31–8.44)	0.81–9.05	100.0		
p,p′-DDT	0.66 ± 1.26	0.04 (0.00–0.48)	n.d.–4.80	51.9	0.10 ± 0.17	0.00 (0.00–0.33)	n.d.–0.37	28.6		
p,p′-DDD	0.04 ± 0.10	0.00 (0.00–0.00)	n.d.–0.52	18.5	0.02 ± 0.04	0.00 (0.00–0.08)	n.d.–0.09	28.6		
∑DDTs	26.91 ± 51.35	10.05 (2.36–22.03)	n.d.–306.48	83.3	4.93 ± 3.72	5.80 (1.31–8.44)	0.81–9.51	100.0		
Methoxychlor	0.32 ± 1.20	0.00 (0.00–0.00)	n.d.–7.66	11.1	0.13 ± 0.25	0.00 (0.00–0.27)	n.d.–0.65	28.6		
∑OCP residues	85.16 ± 111.7	42.73 (25.91–96.88)	3.31–502.30	100.0	12.88 ± 4.44	14.44 (6.89–15.81)	6.49–17.99	100.0	0.001	

OCP: organochlorine pesticide; SD: standard deviation; p25: 25th percentile; p75: 75th percentile; %: percentage of detectable samples; HCB: hexachlorobenzene; HCH: hexachlorocyclohexanes; n.d.: non-determined.
[a] values result from the comparison between the medians (Mann–Whitney test.
[b] values result from the comparison between the frequencies of detection (Chi-square test).

Table 2a
Frequency of detection (%) and average concentrations (ng/g fat) of PCBs residues found in conventional and organic cheese samples from the Canary Islands market (Spain).

Congeners	Conventional cheese (*n* = 54)				Organic cheese (*n* = 7)				p^a	p^b
	Mean ± SD	Median (p25–p75)	Range	%	Mean ± SD	Median (p25–p75)	Range	%		
Marker PCBs										
PCB 28	0.36 ± 1.42	0.00 (0.00–0.00)	n.d.–8.74	11.1	n.d.	n.d.	n.d.	0.0		
PCB 52	0.46 ± 1.96	0.00 (0.00–0.00)	n.d.–11.69	18.5	n.d.	n.d.	n.d.	0.0		
PCB 101	0.11 ± 0.43	0.00 (0.00–0.00)	n.d.–2.53	9.3	n.d.	n.d.	n.d.	0.0		
PCB 118	0.34 ± 1.23	0.00 (0.00–0.00)	n.d.–6.73	9.3	n.d.	n.d.	n.d.	0.0		
PCB 138	5.07 ± 8.23	0.00 (0.00–7.39)	n.d.–33.24	46.3	4.94 ± 6.26	0.00 (0.00–11.21)	n.d.–13.55	42.9		
PCB 153	5.08 ± 6.89	1.98 (0.00–7.22)	n.d.–27.16	66.7	11.15 ± 4.53	10.46 (8.81–13.01)	5.61–20.10	100.0	0.004	
PCB 180	1.40 ± 1.53	1.00 (0.00–1.95)	n.d.–7.39	74.1	2.25 ± 0.93	2.19 (1.68–2.58)	0.87–3.89	100.0	0.032	
DL-PCB (Non-ortho)										
PCB 77	0.03 ± 0.06	0.00 (0.00–0.00)	n.d.–0.22	22.2	n.d.	n.d.	n.d.	0.0		
PCB 81	0.71 ± 2.11	0.00 (0.00–0.00)	n.d.–8.27	11.1	n.d.	n.d.	n.d.	0.0		
PCB 126	0.16 ± 0.30	0.00 (0.00–0.24)	n.d.–1.38	33.3	0.39 ± 0.68	0.00 (0.00–1.16)	n.d.–1.57	28.6		
PCB 169	0.02 ± 0.05	0.00 (0.00–0.00)	n.d.–0.27	9.3	n.d.	n.d.	n.d.	0.0		
DL-PCB (Mono-ortho)										
PCB 105	n.d.	n.d.	n.d.	0.0	n.d.	n.d.	n.d.	0.0		
PCB 114	n.d.	n.d.	n.d.	0.0	n.d.	n.d.	n.d.	0.0		
PCB 118	0.34 ± 1.23	0.00 (0.00–0.00)	n.d.–6.73	9.3	n.d.	n.d.	n.d.	0.0		
PCB 123	n.d.	n.d.	n.d.	0.0	n.d.	n.d.	n.d.	0.0		
PCB 156	0.44 ± 0.53	0.24 (0.00–0.82)	n.d.–1.91	51.9	0.12 ± 0.13	0.11 (0.00–0.19)	n.d.–0.37	57.1		
PCB 157	n.d.	n.d.	n.d.	0.0	n.d.	n.d.	n.d.	0.0		
PCB 167	0.24 ± 0.39	0.00 (0.00–0.34)	n.d.–1.28	40.7	0.03 ± 0.07	0.00 (0.00–0.00)	n.d.–0.18	14.3		
PCB 189	n.d.	n.d.	n.d.	0.0	n.d.	n.d.	n.d.	0.0		

SD: standard deviation; p25: represents the 25th percentile; p75: represents the 75th percentile; %: percentage of detectable samples; n.d.: non-detectable.
[a] values result from the comparison between the medians (Mann–Whitney test).
[b] values result from the comparison between frequencies of detection (Chi-square test).

Table 2b
Frequency of detection (%) and distribution of concentrations (ng/g fat) of PCBs residues, and TEQs (pg WHO–TEQ/g fat) found in conventional and organic cheese samples from the Canary Islands market (Spain).

Congeners	Conventional cheese (*n* = 54)				Organic cheese (*n* = 7)				p^a	p^b
	Mean ± SD	Median (p25–p75)	Range	%	Mean ± SD	Median (p25–p75)	Range	%		
∑PCBs	14.42 ± 16.90	9.57 (2.38–19.33)	n.d.–77.70	98.1	18.87 ± 7.25	22.55 (13.23–24.42)	6.48–25.89	100.0	0.074	
∑M-PCBs	12.81 ± 17.27	5.83 (0.64–19.11)	n.d.–77.40	81.5	18.34 ± 7.33	20.84 (11.51–24.30)	6.48–25.70	100.0	0.049	
∑DL-PCBs	1.94 ± 2.84	1.02 (0.18–2.26)	n.d.–11.07	77.8	0.53 ± 0.81	0.11 (0.00–1.71)	0.00–1.72	57.1		
∑TEQs	10.37 ± 28.45	0.91 (0.01–2.94)	n.d.–137.68		23.33 ± 39.96	0.00 (0.00–76.19)	0.00–87.09			

SD: standard deviation; p25: represents the 25th percentile; p75: represents the 75th percentile; %: percentage of detectable samples; ΣPCBs: Sum of all PCB congeners; ΣM-PCB: Sum of Marker PCBs (IUPAC numbers 28, 52, 101, 118, 138, 153 and 180); ΣDL-PCB: Sum of Dioxin Like PCBs in pg/g fat (IUPAC numbers 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189); ΣTEQs: Sum of TEQs for DL-PCBs in pg/g fat; n.d.: non-detectable.
[a] values result from the comparison between the medians (Mann–Whitney test).
[b] values result from the comparison between frequencies of detection (Chi-square test).

indicate that although in recent years there has been an evident decline in residue levels of these pollutants world-wide, these persistent pesticides remain present in the environment and are therefore present in the food chain; several decades may pass

Table 3
Tolerable daily intake (TDI) and maximum residue levels (MRL) of organochlorine pesticides (OCPs) and dioxin-like PCBs (DL-PCBs) in dairy products established by the European Authorities.

Contaminants	TDI (mg/kg b.w.)	MRL (mg/kg fat)
Aldrin and Dieldrin	0.0001	0.006
$\sum$Chlordanes	0.0005	0.002
$\sum$DDT	0.01	0.04
$\sum$Endosulfans	0.006	0.05
Endrina	0.0002	0.0008
Heptachlor	0.0001	0.004
HCB	NA	0.01
α-HCH	NA	0.004
β-HCH	NA	0.003
γ-HCH (lindane)	0.005	0.001
Methoxychlor	0.1	0.01
$\sum$DL-PCBs	2×10^{-9}	3×10^{-6}

TDI: Tolerable daily intake; MRL: maximum residue level; NA: not available; DL-PCBs: dioxin-like PCBs; Data related to OCPs were obtained from Regulation (EC) No 299/2008 (EU), and data related to DL-PCBs were obtained from Regulation (EC) No 466/2001 (EU).

before the residue levels are undetectable in food items. Nonetheless, geographical and regulatory variations in the use and restriction of OCPs around the world may explain the differences in residual levels of these chemicals, based on of different patterns of use and exposure routes.

Although OCPs are currently banned in Spain for use on agricultural practices, they have been used in vast quantities on vegetable crops in this archipelago over the last decades (as the result of the introduction of intensive agriculture in the Canary Islands) and even they have been used as ectoparasitic agents on domestic animals and livestock (Botella et al., 2004). This misuse of OCPs and the geological features of islands, may have caused heavy contamination of the soil and water (Allen et al., 1997; Díaz-Díaz et al., 1999). Additionally, a number of OCPs are currently used in the neighboring State of Morocco; therefore, some deposition of OCPs onto Archipelago soils via volatilization and atmospheric transport from Morocco may occur (Rapaport et al., 1985; Fries, 1995). In this context, milk producing livestock may be environmentally exposed to OCPs. Because 70% of the conventionally produced cheeses analyzed throughout this study are locally produced, the existence of active environmental sources of OCPs may explain the higher levels of OCPs in conventional cheese compared with organic brands. Further studies are required to evaluate this possibility.

3.2. Occurrence of selected PCB residues in cheese samples

We found that 100% of the cheese samples (both conventionally and organically produced brands) showed quantifiable levels of PCBs (Tables 2a and 2b). The number of PCB residues in both types of cheese was similar thus, an average of 4 PCB congeners per sample (range 1–8) were found in conventional brands, while an average of 3 PCB congeners (range 2–6) were detected in organic brands. Five congeners (105, 114, 123, 157 and 189) were not detected in any brand of cheese, and the most frequently detected congeners in our samples were PCBs 153 and 180. PCBs 28, 52, 101, and 118 were detected only in samples from conventional brands of cheese. All analyzed cheeses showed residue concentrations above the LOQ of some of the congeners which are considered to be indicators of environmental contamination by PCBs, (marker PCBs; M-PCBs) (Table 2a).

The total concentration of PCB residues (Σ-PCBs; Table 2b) was higher in organic than in conventional cheeses (although such differences were not statistically significant; 22.55 *vs.* 9.57 ng/g fat, respectively; $p = 0.07$). This result appear to be a consequence of M-PCB levels (ΣM-PCBs) being higher in organic than in conventional cheeses (median values of 20.84 *vs.* 5.83 ng/g fat, respectively; $p = 0.049$; Table 2b). The M-PCBs most frequently detected were congeners 153 and 180, (detected in 100% of the analyzed organic cheese samples, and 67% in the case of PCB 153 and 74% in the case of PCB 180 in conventional cheeses). Additionally, concentrations of congeners 153 and 180 were higher in organic than in conventional brands of cheese (p values of 0.004 and 0.032 respectively).

There were not significant differences between the both types of cheeses in the DL-PCB levels (Table 2a). Among the 12 DL-PCBs analyzed for in the samples, only congeners 105, 114, 123, 157 and 189 were not detected in any sample. In contrast, the most frequently DL-PCBs detected were the congeners 156 (52 and 57% in conventional and organic cheese samples, respectively), 167 (14 and 41% in organic and conventional cheese samples, respectively), and 126 (29 and 33% in organic and conventional cheese samples, respectively). Nevertheless, as shown in Tables 2a, 2b, and 3, the median levels of DL-PCBs in both types of cheeses were observed to be relatively low and were consistently below the maximum residue limit (MRL) established by the European Legislation (OJEC, 1993 and 1994).

Our results demonstrate that commercially available brands of cheese present in the Canary Islands market showed measurable levels of various PCBs and the results are consistent with the presence of these organochlorine contaminants found in milk brands available in the Canary Islands market (Luzardo et al., 2012) and with those data published by other authors (Durand et al., 2008; Focant et al., 2002; Marin et al., 2011; Windal et al., 2010) who detected measurable levels of PCBs in milk and dairy samples (including cheese) from France, Belgium, and mainland Spain. We, however, found a different profile of contamination by PCBs as compared with data describing dairy products from mainland Spain (Marin et al., 2011). For example, Marin et al. (2011) reported that PCB 118 was the most abundant congener followed by PCBs 105 and 156; however, in our study we found that PCBs 180 and 153 were the congeners most frequently measured (present in 100% of the organic brands and 67 and 74% of the conventional brands), whereas that PCB 105 was not measured in any sample, and PCB 118 was measured only in 9% of the conventional brands. The congener 156, however, was measured in more than 50% of both types of cheese (conventional and organic). Approximately 63% of the analyzed samples were from cheeses locally produced, which may explain such differences in levels of PCB residues among both studies. The environmental PCB contamination level in these islands (where the presence of traditional pollution via industries is scarce) is assumed to be relatively low.

As shown in Table 2b, median values of TEQ–PCBs in conventionally and organically-produced cheeses were very low (0.9 and 0.0 pg WHO–TEQ/g fat, for conventional and organic cheeses, respectively), and well below the levels established by International Agencies; therefore, a maximum concentration of 6 pg WHO–TEQ/g fat has been defined for the sum of PCDDs, PCDFs and DL-PCBs in food, and the action level for DL-PCBs is 3 pg WHO–TEQ/g fat for milk and dairy products (Fattore et al., 2006; Recommendation 2006/88/EC). Most contaminated cheeses (those included in the 75th percentile), however, were observed to have concentrations as high as 76 pg WHO–TEQ/g fat in organically produced cheese and 3 pg WHO–TEQ/g fat in conventionally produced cheese.

Our data suggesting that cheese could be a significant source of dioxin-like toxicants for the population under study (and to a greater extent with those cheeses organically produced), agree with the results described in commercially available brands milk from the Canary Islands (Luzardo et al., 2012). The origin of the organic cheese may explain these results: most brands of organic cheese available in the Canary Islands market are produced mainly in industrialized European countries (Holland, Belgium, Germany), where the level of environmental contamination by PCBs is potentially high (Covaci et al., 2002a,b); however, as cited previously, most of the conventional brands of cheese analyzed throughout this study were locally produced.

3.3. Assessment of cheese-related dietary exposure of the population of the Canary Islands to OCP residues and dioxin-like PCBs

As previously published (Luzardo et al., 2012) the population of the Canary Islands may have milk-related estimated daily intakes (EDI) of certain organochlorine contaminants (specifically, DL-PCBs) well above the recommended Tolerable Daily Intake (TDI) established by European Union Authorities (2 pg WHO–TEQ kg^{-1} -b.w. d^{-1}). Additionally, this population shows the highest consumption rates of milk and dairy products in Europe (ENCA, 1998; Serra Majem et al., 2000a,b) thus, a mean value of 26.1 g and 20.6 g of cheese are consumed daily by the adult population and children of the archipelago (respectively). Considering the mean body weight of adults (18–75 years) as 70.1 kg, and that of children (6–10 years) as 30.4 kg (ENCA, 1998), we have calculated the cheese-related EDI for the organohalogenated contaminants measured throughout this study using the deterministic method for chronic exposure as previously published (Dorne, 2010).

As expected, the cheese-related EDI of OCPs for adults and children living in the Canary Islands are low in comparison with the TDI established by the European Food Safety Authority (EFSA) (2002) (Tables 3, 4a, and 4b). Nevertheless we have to consider that the population of the Canary Islands has been subjected to a chronic exposure to OCPs that persisted in the late 1990's (Henriquez-Hernandez et al., 2011; Luzardo et al., 2006; Zumbado et al., 2005) up to the early 2000's current century (Luzardo et al., 2009). It is well known that the main route of exposure to organochorinated contaminants is through the dietary intake diet (Bordajandi et al., 2004; Darnerud et al., 2006; Hanaoka et al., 2002), and that milk and dairy products could be a relevant source of these environmental contaminants in the general population (Focant et al., 2002). Under this premise and considering the concentrations (and the intake) of OCPs present in cheese in addition to those concentrations reported previously from milk (Luzardo et al., 2012), the possibility exists that this population may be subject to a high dietary exposure of OCPs. Because many OCPs have endocrine- and metabolic-disrupting properties (mainly, DDT, aldrin or dieldrin, and its metabolites), they have been linked to environmentally induced diseases, such as obesity, diabetes and cancer (Everett et al., 2007; Soto et al., 1995; Snedeker, 2001; Wolff et al., 2000; Holtcamp, 2012). Consequently, the existence of active sources of OCPs for this population, especially children and pregnant women, should not be overlooked.

As shown in Table 2b, TEQ levels were similar in organic and in conventional cheese samples. In any case it should be highlighted the existence of huge differences in the Total TEQ levels among the cheese samples analyzed, thus, while the lowest contaminated samples showed low levels of DL-PCBs (nearly 0 or undetected pg WHO–TEQ/g fat for organic and conventional cheeses), the most contaminated brands of cheese (those included in the 75th percentile) reached levels as high as 76 (organically produced brands of cheese) or 3 pg WHO–TEQ/g fat (conventionally produced brands of cheese), (Table 2b). Ultimately, only the daily consumption of cheese could account for approximately half of the TDI established by International Agencies (Table 5) for the adult population consuming the most contaminated brands of conventionally produced cheese, while for adults consuming organically produced brands the percentage could reach as high as 125% of the TDI. Children who consume the most contaminated brands of cheese are in a worse situation (Table 5).

These results are extremely worrisome and further studies required to clarify the origin of cheese contamination, especially if we consider that the average contribution of DL-PCBs from dairy products to the total TEQ, account for no more than 60% (Durand et al., 2008), and that, fish and fishery products are the main contributors of PCBs to the diet (Fattore et al., 2006; Llobet et al., 2008; Marin et al., 2011). The toxicity of dioxins and dioxin-like compounds is related to the amount accumulated in the body during a lifetime. Toxicological properties of DL-PCBs are similar to those characteristic of polychlorodibenzodioxins (PCDDs) and polychlorodibenzofurans (PCDFs) (Van den Berg et al., 2006) and certain evidence suggests that even low doses of DL-PCBs, similar to those found in the background contamination of food, can cause subtle effects during prolonged exposure, especially in children's

Table 4a

Cheese-related estimated daily intakes (EDI) of organochlorine pesticides in adults (26.10 g cheese/day) and children (20.60 g cheese/day) from the Canary Islands, and percentage (%) that it means in relation to the tolerable daily intakes (TDI) established by International Agencies.

	Adults (10–75 years)						Children (>10 years)					
	Conventional cheese			Organic cheese			Conventional cheese			Organic cheese		
	Mean ng/g fat	EDI ng/kg bw	%TDI[a]	Mean ng/g fat	EDI ng/kg bw	%TDI[a]	Mean ng/g fat	EDI ng/kg bw	%TDI[a]	Mean ng/g fat	EDI ng/kg bw	%TDI[a]
HCB	6.95	0.55		2.27	0.25		6.95	0.99		2.27	0.45	
HCH												
α-HCH	1.65	0.13		0.23	0.02		1.65	0.23		0.23	0.05	
β-HCH	21.07	1.66		0.65	0.05		21.07	3.00		0.65	0.13	
δ-HCH	0.03	0.00		n.d.	n.d.		0.03	0.00		n.d.	n.d.	
Lindane	8.46	0.66	0.00	0.53	0.04	0.00	8.46	1.20	0.02	0.53	0.10	0.00
∑HCH	31.22	2.45		1.41	0.11		31.22	4.44		1.41	0.28	
Cyclodienes												
Aldrin	1.19	0.09	0.01	0.08	0.01	0.00	1.19	0.17	0.17	0.08	0.02	0.02
Dieldrin	10.47	0.82	0.95	3.01	0.24	0.27	10.47	1.49	1.49	3.01	0.59	0.59
Endrin	n.d.	n.d.		n.d.	n.d.		n.d.	n.d.		n.d.	n.d.	
cis-Chlordane	2.44	0.19		0.62	0.05		2.44	0.35		0.62	0.12	
trans-Chlordane	2.39	0.19	0.06	0.16	0.01	0.00	2.39	0.34	0.07	0.16	0.03	0.01
∑Chlordanes	4.83	0.38	0.02	0.77	0.06	0.00	4.83	0.69	0.14	0.77	0.15	0.03
Heptachlor	2.09	0.16	0.16	0.18	0.01	0.01	2.09	0.30	0.30	0.18	0.04	0.04
∑Cyclodienes	13.75	1.08	0.27	3.27	0.26	0.06	13.75	1.96	0.39	3.27	0.64	0.13

n.d.: non-detectable.

[a] Percentage of TDI provided by the cheese-associated EDI for these populations.

Table 4b
Cheese-associated estimated daily intakes (EDI) of organochlorine pesticides in adults (26.10 g cheese/day) and children (20.60 g cheese/day) from the Canary Islands, and percentage (%) that it means in relation to the tolerable daily intakes (TDI) established by International Agencies.

	Adults (10–75 years)						Children (>10 years)					
	Conventional cheese			Organic cheese			Conventional cheese			Organic cheese		
	Mean ng/g fat	EDI ng/kg bw	%TDI[a]	Mean ng/g fat	EDI ng/kg bw	%TDI[a]	Mean ng/g fat	EDI ng/kg bw	%TDI[a]	Mean ng/g fat	EDI ng/kg bw	%TDI[a]
Endosulfans												
α-Endosulfan	1.18	0.09	0.01	0.10	0.01	0.00	1.18	0.17	0.00	0.10	0.02	0.00
β-Endosulfan	0.55	0.04		n.d.	n.d.		0.55	0.08		n.d.	n.d.	
∑Endosulfans	1.73	0.14	0.03	0.10	0.01	0.00	1.73	0.25	0.00	0.10	0.02	0.00
Diphenyl-alyphatics												
4,4-DDE	26.21	2.06	2.10	4.81	0.38	0.39	26.21	3.73	0.04	4.81	0.95	0.01
4,4-DDT	0.66	0.05		0.10	0.01		0.66	0.09		0.10	0.02	
4,4-DDD	0.04	0.00		0.02	0.00		0.04	0.01		0.02	0.00	
Methoxychlor	0.32	0.03	0.01	0.13	0.01	0.00	0.32	0.05	0.00	0.13	0.03	0.00
∑DDTs	26.91	2.11	2.11	4.93	0.39	0.39	26.91	3.83	0.04	4.93	0.97	0.01
∑OC-compounds	85.16	6.69		12.88	1.01		85.16	12.12		12.88	2.53	

n.d.: non-detectable.

[a] Percentage of TDI provided by the cheese-associated EDI for these populations.

Table 5
Cheese-associated estimated daily intakes (EDI) of TEQ–PCBs (pg WHO–TEQ/kg bw/day) in adults (26.10 g cheese/day) and children (20.60 g cheese/day) from the Canary Islands and percentage (%) that it means in relation to the tolerable daily intakes (TDI) established by International Agencies.

	Adults (10–75 years)						Children (>10 years)					
	Conventional cheese			Organic cheese			Conventional cheese			Organic cheese		
	Mean pg/g fat	EDI pg/kg bw	%TDI[a]	Mean pg/g fat	EDI pg/kg bw	%TDI[a]	Mean pg/g fat	EDI pg/kg bw	%TDI[a]	Mean pg/g fat	EDI pg/kg bw	%TDI[a]
TEQ–PCB 77	0.03	0.00	0.12	n.d.			0.03	0.00	0.21	n.d.		
TEQ–PCB 81	0.61	0.05	2.38	n.d.			0.61	0.09	4.34	n.d.		
TEQ–PCB 105	n.d.			n.d.			n.d.			n.d.		
TEQ–PCB 114	n.d.			n.d.			n.d.			n.d.		
TEQ–PCB 118	0.12	0.01	0.47	n.d.			0.12	0.02	0.85	n.d.		
TEQ–PCB 123	n.d.			n.d.			n.d.			n.d.		
TEQ–PCB 126	8.87	0.69	34.68	23.32	2.52	125.90	8.87	1.26	63.11	23.32	4.58	229.13
TEQ–PCB 156	0.38	0.03	1.49	0.00	0.00	0.02	0.38	0.05	2.70	0.0	0.00	0.00
TEQ–PCB 157	n.d.			n.d.			n.d.			n.d.	0.00	0.00
TEQ–PCB 167	0.22	0.02	0.86	0.00	0.00	0.00	0.22	0.03	1.57	0.00	0.00	0.00
TEQ–PCB 169	0.13	0.01	0.51	n.d.			0.13	0.02	0.92	n.d.		
TEQ–PCB 189	n.d.			n.d.			n.d.			n.d.		
∑TEQ–PCBs	10.37	0.81	40.54	23.33	2.52	125.95	10.37	1.48	73.78	23.33	4.58	229.23

n.d.: non-detectable.

[a] Percentage of TDI provided by the cheese-associated EDI for these populations.

neurological development (Park et al., 2010). For these reasons, the European Commission's Scientific Committee on Food (2001), established a TDI of 2 pg WHO–TEQ/kg bw/day for WHO–TEQ, including all food items containing PCDDs, PCDFs, and DL-PCBs (Fattore et al., 2006). The European Union, through the Scientific Committee on Food (SCF), has implemented a strategy (SCF, 2001) to reduce human exposure to toxicants (mainly dioxin-like compounds) present in food items of animal origin.

In conclusion, we have developed an independent survey in the Spanish archipelago of the Canary Islands to analyze the relevance of cheese (a dairy product frequently consumed by the population of these islands) as an active source of organochlorine contaminants and to evaluate its impact on consumers. We observed that cheese consumption could be a major exposure route for DL-PCBs and also, on a much smaller scale, for OCPs. Our results are worrisome because to the deleterious health effects have been attributed to organochlorine contaminants exposure (ATSDR, 2001; Bilau et al., 2008; Park et al., 2010; Holtcamp, 2012; Everett et al., 2007; Wolff et al., 2000); however, our results should be taken with caution because, (a) as showed in Table 1a, 1b, and 3, there are cheese brands that are highly contaminated by organochlorine contaminants, but there were a number of cheese brands that showed undetectable levels of these toxicants; and (b) in the Nutritional Survey of the Canary Islands the consumption of organic cheese was not specifically recorded, and it cannot be assumed a high consumption of cheese among consumers of organic products. Nevertheless, because all organically produced cheeses are from European countries and approximately 30% of the conventionally produced cheeses are from mainland Spain or for other European countries, our results can be extrapolated to the rest of the Spanish population, and consequently, should be considered by the Spanish Public Health Authorities.

Conflict of Interest

The authors declare that there are no conflicts of interest.

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