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In the present study we have quantified the levels of organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) in commercially available brands of yogurt (17 conventional and 15 organic) present in the Spanish market, with the goal of estimating the daily intake of these contaminants by the Spanish population through the yogurt consumption. On the one hand, with respect to organochlorine contaminants (OCPs and PCBs) our results showed that hexachlorobenzene, *p,p′*-DDE, and all the PCBs congeners that are considered markers of environmental contamination were present in almost all the yogurt samples. The concentrations of these pollutants were found to be very low [well below the toxicological standard limits established by the European Union (http://ec.europa.eu/sanco_pesticides/public/?event=homepage)] in almost all the samples, but were even lower in organic than in conventionally produced yogurts. It is remarkable that in some of the samples (six conventional and three organic yogurts) the current maximum level for dioxin-like PCBs (2.5pg WHO-TEQ/g^{-1} fat) was exceeded. On the other hand, with respect to PAHs, the mutagenic and carcinogenic compounds benzo[k]fluoranthene, benzo[b]fluoranthene and chrysene were frequently detected in yogurt. From these results we have estimated that the daily intake of these pollutants is in general low. However, it should be highlighted that if the consumer inadvertently choose certain yogurts they could be exposed to high amounts of certain pollutants, that could be even higher than the tolerated daily intakes established by the European Union.

Keywords: organochlorine pesticides; polychlorinated biphenyls; polycyclic aromatic hydrocarbons; yogurt; organic yogurt; exposure assessment

1. Introduction

Anthropogenic contaminants include a high number of chemicals that have been recognized as important toxic environmental pollutants. Among them persistent organic pollutants (POPs) are organic compounds characterized by their stable structures and lipophilicity that are resistant to the degradation in the environment and biota. Over the last 30 years a number of these substances have been highlighted as a cause for concern (Dorgan et al. 1999; Ribas-Fito et al. 2001; Samanta et al. 2002; Knerr & Schrenk 2006; Valerón et al. 2009; Casals-Casas & Desvergne 2011; Dickerson et al. 2011; Boada et al. 2012) and have been the subject of extensive study and international regulation. Among them the halogenated hydrocarbons such as organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) are especially relevant. Besides, although polycyclic aromatic hydrocarbons (PAHs) cannot be strictly considered POPs because they are efficiently metabolized, due to their lipophilicity and continuous emission to the environment, they are frequently classified as POPs and studied together with other pollutants of this group.

These substances often accumulate and magnify up to the food chain, particularly in fat sources, and it is well accepted that food consumption is the main source of non-occupational human exposures to these contaminants. Thus, the ingestion of food contributes more than 90% of total human exposure (Baars et al. 2004; Darnerud et al. 2006; Polder et al. 2010), and several studies have reported that dairy products, because of their high-fat content, are a dietary route of POPs and supply around 30% of the total dietary intake of these contaminants in Western populations (Focant et al. 2002; Almeida-González et al. 2012; Luzardo et al. 2012; Luzardo, Rodriguez-Hernandez, et al. 2013).

In the Spanish population, milk and dairy products are the second group of food consumption after non-alcoholic drinks (AECOSAN 2006, 2011). Thus, according to the most recent nutritional survey in Spain, 79.1% of the population consumes dairy products every day, with an average consumption of 304 g/day, which is one of the highest of the Western countries. In relation to yogurt, the average consumption is estimated in 53 g/b.w./day, representing 17.43% of the dairy products' intake (AECOSAN 2011).

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Because of the previous reasons the monitoring of these anthropogenic pollutants in yogurt is justified. Thus, in the present study, 32 brands of yogurt [17 conventionally produced and 15 from organic production (EC 2007)] from the Spanish market were purchased during 2013 and screened for the presence of 57 anthropogenic pollutants, including 23 OCPs, 18 PCBs and 16 PAHs. The double goal of this study was to determine the possible differences in POPs' concentrations between the two types of production of yogurt (conventional *vs.* organic), and to estimate the daily dietary exposure of the Spanish population to these substances through the yogurt consumption. To our knowledge, this is the first study to compare the intake of contaminants depending on the mode of production of yogurt. We believe that the results of such studies are very useful for health authorities and consumers, allowing the consumer to choose those products containing a lower burden of environmental pollutants.

2. Materials and methods

2.1. Sampling and collection

In this study, 32 commercial brands of yogurt (17 from conventional production and 15 from organic production) were randomly purchased between April and November 2013 from supermarkets and also stores specialized in organic food of the Canary Islands (Spain). In the Canary Islands, in spite of the fact that there some brands of yogurt which are locally produced, the great majority of the available brands are also marketed throughout the country, and therefore the sampling of this study can be considered as representative of the Spanish market. Each of the 32 selected brands was sampled by triplicate during this period of time. The three samples of each brand were pooled to homogenize the potential fluctuations in the concentrations of pollutants. The fat contents given in the label by the manufacturer were used to obtain the final lipid-corrected values (average fat content 4.63% for conventional yogurt and 4.45% for organic yogurt). All the yogurt samples were frozen at −80°C until analysis.

2.2. Chemicals and reagents

Dichloromethane, n-hexane, ethyl acetate and cyclohexane were of the highest purity available (>99.9%) and purchased from Fisher Scientific (Leicestershire, UK). Ultrapure water was produced from a Milli-Q Gradient A10 (Millipore, Molsheim, France). Diatomaceous earth was purchased from Sigma-Aldrich (St. Louis, USA). Bio-Beads SX-3 were purchased from BioRad Laboratories (Hercules, USA). Standards of OCPs, PCB congeners and internal standards (ISs, PCB 202, *p,p′*-DDE-d8 phenanthrene-d10, tetrachloro-m-xylene, and heptachlor epoxide cis) were purchased from Dr Ehrenstorfer, Reference Materials (Augsburg, Germany). Standards of PAHs were purchased from Absolute Standards, Inc (Connecticut, USA). All standards were neat compounds (purity from 97% to 99.5%). Stock solutions of each compound at 1 mg/mL were prepared in cyclohexane and stored at −20°C. Diluted solutions from 0.05 ng/mL to 100 ng/mL were used for calibration curves.

2.3. Analytes of interest

A total of 57 analytes belonging to three relevant groups of POPs were selected for this study. The 23 OCPs and metabolites included were the diphenyl-aliphatics (*p,p′*-DDT, *p,p′*-DDE, *p,p′*-DDD, *o,p′*-DDE, *o,p′*-DDT, *o,p′*-DDD, dicofol and methoxychlor); the persistent and bioaccumulative contaminant hexachlorobenzene (HCB); the four isomers of hexachlorocyclohexane (*α*-, *β*-, *δ*- and *γ*-HCH); the cyclodienes heptachlor, dieldrin, aldrin and endrin, chlordane (cis- and trans-isomers) and mirex; endosulfan (*α*- and *β*-isomers) and endosulfan sulphate. With respect to PCBs we decided to include a total of 18 congeners: the dioxin-like congeners (DL-PCBs, IUPAC numbers# 77, 81, 105, 114, 118,123, 126, 156, 157, 167, 169 and 189), and the six congeners that are considered markers of environmental contamination (M-PCBs, IUPAC numbers# 28, 52, 101, 138, 153 and 180). Finally, we also included in the suite of analytes the list of the 16 US-EPA priority PAHs that is often targeted for measurement in environmental samples (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenz[a,h]anthracene and benzo[ghi]perylene).

2.4. Sample preparation and analytical procedure

The yogurt samples, stored at −80°C, were defrozen at room temperature and homogenized by brands. A total of 50 g of this homogenate was lyophilized for 72 h. Six grams of lyophilized yogurt were spiked with the ISs mixture and used for the extraction OCPs, PCBs and PAHs, following the method of extraction and clean-up recommended by the European Standard for the determination of pesticides and PCBs in fatty foods (EN 1996a, 1996b). The validity of this method has been previously proven in our laboratory for fatty samples (Almeida-González et al. 2012; Luzardo et al. 2012; Luzardo, Rodriguez-Hernandez, et al. 2013, 2014; García-Álvarez et al. 2014). This method gives acceptable recoveries that ranged between 78.5% and 103.2%. Briefly, for the fat extraction we used a Soxtec™ 2055 Auto Fat Extraction (Foss® Analytical, Hillerød, Denmark) apparatus, which consisted of an extraction unit, a

control unit and a drive unit. The samples were placed into the extraction unit, 20 ml of dichloromethane was added to each of the extraction cups in a closed system and the cups were heated using an electric heating plate. The three-step extraction consisted of boiling, rinsing and solvent recovery. The solvent was evaporated in a rotary evaporator (Hei-VAP Advantage™, Heidolph Instruments®, Schwabach, Germany) at 40ºC to prevent analyte losses. The results were calculated as the total amount of fat (g) per 100 g yogurt. Using a precision balance, the fat obtained was carefully weighted into a zeroed glass tube. The weighted fat was dissolved in 2 ml of cyclohexane/ethyl acetate (1:1) and subjected to purification by gel permeation chromatography (GPC; Bio-Beads) using cyclohexane/ethyl acetate (1:1) at a constant flow of 2 ml/min as the eluent. The first 25-minute elution volume, which contained the great majority of the lipids (>98%), was discarded. The 25–85 minute elution volume (120 ml), which contained all the analytes that were co-extracted with the fat, was collected. The sample was concentrated using a rotary evaporator, and finally, the solvent was evaporated to dryness under a gentle nitrogen stream. The residue was then reconstituted in 1 mL cyclohexane the sample was transferred to a gas chromatography (GC) vial that was used for the chromatographic analysis. The amount of pollutants per gram of fat was obtained multiplying by the correspondent correction factor. No additional clean-up steps were needed, and thus the 1 mL-extracts in cyclohexane obtained after the GPC were used for the detection and quantification by GC/triple quadrupole mass spectrometry analysis.

2.5. Procedure of chemical analysis

GC analyses of 57 compounds, plus ISs were performed in a single run on a Thermo Trace GC Ultra equipped with a TriPlus Autosampler and coupled to a Triple Quadrupole Mass Spectrometer Quantum XL (Thermo Fisher Scientific Inc., Waltham, MA, USA), using appropriate ISs as previously described (Camacho et al. 2012; Luzardo, Rodriguez-Hernandez, et al. 2013, Luzardo, Ruiz-Suarez, 2013, 2014; Luzardo, Ruiz-Suarez, et al. 2013). Briefly, for the chromatographic separation we used a 30 m × 0.25-mm i.d., 0.25 µm film thickness column (BPX5, SGE Inc., Austin, TX, USA) as the stationary phase. Helium (99.999%) was used as the carrier gas at a constant flow of 1 ml/min. The 61-min oven temperature programme was: 60°C held for 1 min, ramped to 210°C at 12°C/min and then to 320°C at 8°C/min and held for 6 min. The injector temperature was set at 270ºC and the transfer line was heated to 310°C. The injection volume was 1 µl in the splitless mode.

The GC was tandem-coupled to a TSQ XLS QqQ mass spectrometer, which was used for the detection and quantification of the 57 pollutants plus ISs. An electron ionization (EI)-MS/MS library was specially created for the target analytes under our experimental conditions. We calibrated the mass spectrometer scale with perfluorotributylamine on a weekly basis to ensure an optimal response over time and proper mass assignments. The instrument control, data acquisition and data analysis was performed using the Thermo Fisher Xcalibur software (Ver. 2.0.1). We constructed a timed SRM method for the simultaneous analysis of 57 pollutants plus ISs in a single run. Calibration curves contained all of the target compounds except for the ISs at each level (0.5–500 ng/mL). The operation conditions of the mass spectrometer were: electron impact ionization (70 eV) in SRM; emission current, 50 µA; ionization source temperature, 220°C; electron multiplier voltage, 1500 V; scan width, 0.15; scan time, 0.05 s; peak width, *m/z* 0.7 Da. Argon (99.99%) was used as the collision gas at 0.2 Pa.

2.6. Dietary intake estimates and calculations (exposure assessment)

The exposure assessment was calculated by multiplying the respective concentrations of contaminants in yogurts (mean values) by the amount of fat contained in the average daily yogurt consumption by adults (17–70 years old, average weight 68.5 kg, mean daily yogurt consumption 52.6 g) and children (7–12 years old, average weight 34.5 kg, mean daily yogurt consumption 64.8 g). Consumption data of yogurt by the Spanish population were obtained from the Spanish Diet Model for the Determination of the Consumer's Exposure to Chemicals (AECOSAN 2006, 2011). Exposures were assessed for all the contaminants, individually considered and also grouped in different forms. For calculations, we estimated on the one hand the lower bound approach, which assigned the zero value to all those results that were below the limit of detection (LOD) of the method. On the other hand, as recommended by the European Agency for Food Safety (EFSA), the upper bound approach was also calculated. In this approach a value equal to the LOD is assigned to all the non-detected results.

In this research we expressed the total value of OCPs residues (ΣOCPs) as the sum of the 23 measured OCPs and metabolites; the total value of DDTs (∑DDTs) as the sum of the measured values of *p,p′*-DDT, *p,p′*-DDE, *p,p′*-DDD, *o,p′*-DDE, *o,p′*-DDT, *o,p′*-DDD; the total value of HCH residues (ΣHCH) as the sum of the four HCH isomers (*α*-, *β*-, *δ*-, and *γ*-HCH); and the total value of cyclodiene residues (∑cyclodienes) as the sum of aldrin, dieldrin, endrin, cis-chlordane, trans-chlordane and heptachlor. Similarly we expressed the total value of PCB residues (∑PCBs) as the sum of the 18 PCB congeners measured. In addition the six congeners considered as

markers of environmental contamination by PCBs (#28, 52, 101, 138, 153 and 180) were also considered as a group (∑M-PCBs), and total value of dioxin-like PCBs (∑DL-PCBs) was also considered as the sum of the measurements of the 12 individual dioxin-like congeners (#77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189). Additionally, we estimated the potential toxicity (in terms of toxic equivalence to dioxins; dioxin toxic equivalents [TEQs]) for the DL-PCBs using the toxic equivalency factors as revised by the World Health Organization (WHO) in 2005 (Van den Berg et al. 2006). We expressed the total TEQs (∑TEQs) as the sum of TEQs individually obtained from the DL-PCBs. With regard to PAHs we considered the sum of the values of the 16 US-EPA compounds included in this study as the total content of PAHs (∑PAHs). Finally, according to the EFSA recommendations (EFSA 2008), we also considered as a group the sum of the eight compounds for which there are evidences of carcinogenicity (∑PAH8): benz[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, chrysene, dibenz[a,h]anthracene and indeno[1,2,3-cd]pyrene.

2.7. Statistical analyses

Database management and statistical analyses were performed using PASW Statistics v. 19.0 (SPSS Inc., Chicago, USA). The concentration of the contaminants included in this study did not follow a normal distribution; therefore, the results are expressed in terms of the median, and range (values minimum and maximum). Differences of contaminants among 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-tailed) was considered to be statistically significant.

2.8. Quality control

The recoveries of the 57 analytes and surrogates were acceptable with this method since in all the cases were above of 72%. All the individual measurements were corrected by the recovery efficiency for each analyte. All the measurements were done in triplicate and the values used for calculations were the mean of the three data. In each batch of samples, two controls were included every 12 samples: a reagent blank consisting on a vial containing only cyclohexane; and an internal laboratory quality control (QC) consisting on melted butter spiked at 20 ng g^{-1} of each of the analytes, which was processed with the same method than the samples. The batch analyses were considered valid when the values of the analytes in the QC were within a 10% of deviation of the theoretical value.

3. Results and discussion

3.1. Occurrence of OCP residues in yogurt samples

Our results showed that 100% of the samples presented quantifiable amounts of OCP residues. The average number of residues per sample from this group of pollutants was four OCPs, regardless the production method (within a range 2–7 residues in conventional and 2–6 residues in organic yogurts).

Fourteen of the analyzed OCPs (α-HCH, δ-HCH, *o,p′*-DDE, methoxychlor, heptachlor, endrin, aldrin, heptachlor epoxide, trans-chlordane, alpha endosulfan, beta endosulfan, sulfate endosulfan and mirex) were not detected in any of the samples.

The most frequently detected compound of this group of pollutants was *p,p′*-DDE, which was found in 100% of the samples (median = 6.17 ng g^{-1} fat in conventional samples *vs.* 2.43 ng g^{-1} fat in organic yogurts, $P < 0.05$), whereas its parent compound *p,p′*-DDT (banned in 1978) was also frequently detected: six samples of conventional yogurt (35.3%), and seven samples from organic production (46.7%; Table 1). These results are in agreement with other studies performed in dairy products and yogurt in China, Jordan or USA, where also *p,p′*-DDE was the most predominant OCP (Zhang et al. 2006; Salem et al. 2009; Schecter et al. 2010). The ∑DDT was higher in the group of conventional yogurts, although no statistical significance difference was reached.

With regard to the rest of OCP residues that were detected in yogurt samples it is remarkable that HCB was also frequently detected in yogurt samples of both methods of production (88.2% of conventional and 93.3% of organic yogurts). This is not a surprising result since HCB is a known by-product of many industrial processes (Nasir et al. 1998) that is frequently detected in foodstuffs. The high frequency of this compound in yogurt samples was also in accordance with our previous studies in other dairy products (Almeida-González et al. 2012; Luzardo et al. 2012). Hexachlorocyclohexane (β- and γ-HCH) isomers were also detected with a similar distribution between organic and conventional yogurts (median values of 0.59 *vs.* 0.74 ng g^{-1} fat, respectively). These concentrations were lower than those reported in other studies in dairy products, where the average values ranged between 1.2 ng g^{-1} and 12.8 ng g^{-1} (Zhang et al. 2006; Salem et al. 2009; Polder et al. 2010; Tornkvist et al. 2011; Gutiérrez et al. 2012; Weiss et al. 2013). We also found residues of dicofol in some of the samples (29.4% of conventional brands and 13.3% of organics), as also described for bovine or human breast milk from Brazil, China, Korea and Japan (Fujii et al. 2011; Avancini et al. 2013). Finally, with

Table 1. Levels of OCPs detected in conventional and organic yogurt samples (ng g^{-1} fat) from the Spanish market.

	CONVENTIONAL YOGURT			ORGANIC YOGURT			
No. of OCPs residues per sample	Mean ± SD 4.47 ± 1.46			Mean ± SD 4.13 ± 1.25			*P* NS
Compound	Median	Detection range	Frequency	Median	Detection range	Frequency	P^a/P^b
HCB	1.07	n.d.–4.01	88.2%	0.94	n.d–4.25	93.3%	NS
p,p′-DDE	6.17	1.49–26.65	100%	2.43	1.27–20.05	100%	0.047/NS
p,p′-DDT	0	n.d.–33.19	35.3%	0	0.00–33.79	46.7	NS
p,p′-DDD	0	n.d.–1.18	11.8%	n.d.	n.d.	n.d.	NS
o,p′-DDE	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
o,p′-DDT	0	n.d.–0.8	11.8%	0	n.d.–0.65	20%	NS
o,p′-DDD	n.d.	n.d.	n.d.	0	n.d.–0.19	13.3%	NS
∑DDTs	7.63	1.66–53.01	100%	5.65	1.33–38.66	100%	NS
Dicofol	0	n.d.–5.11	29.4%	0	n.d.–4.65	13.3%	NS
Mirex	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
Metoxychlor	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
α-HCH	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
B + *γ*-HCH	0.59	n.d.–16.41	82.4%	0.74	n.d.–1.82	80%	NS
δ-HCH	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
∑HCH	0.59	n.d.–16.41	82.4%	0.74	n.d.–1.82	80%	NS
Aldrin	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
Dieldrin	7.98	n.d.–30.58	70.6%	0	n.d.–16.28	40%	0.083/0.079
Endrin	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Chlordane (trans)	0	0.00–4.32	17.6%	0	0.00–40.52	6.7%	NS
Chlordane (cis)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
Heptachlor	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
∑cyclodienes	7.98	n.d.–30.58	70.6%	0	n.d.–56.79	40%	0.083/0.079
∑endosulfan	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
∑OCPs	23.85	3.66–85.59	100%	13.62	2.16–69.70	100%	0.062/NS

[a]Values resulting from the comparison between the medians, referred to the figures with an asteric (Mann-Whitney test).
[b]Values resulting from the comparison between the frequencies, referred to the figures with an asteric (Chi-square test).
n.d., non-detectable.

respect to cyclodienes, the only detected compound was dieldrin. The presence of this contaminant in dairy products has been also reported by other authors (Darko & Acquaah 2008; Schecter et al. 2010; Almeida-González et al. 2012; Luzardo et al. 2012). However it is remarkable that both, dieldrin frequency and median concentration were statistically different between conventional and organic yogurts (Table 1). Finally, it should be noted that, unlike other reports for dairy products (Darko & Acquaah 2008; Weiss et al. 2013), none of the isomers of endosulfan were detected.

From these results we can conclude that the total burden of OCPs (∑OCPs) was higher in conventionally produced yogurts than in organically produced yogurts (median values 23.85 *vs.* 13.62 ng g^{-1} fat, respectively). However, the median levels of the OCPs quantified in this series of samples were relatively low and all the quantified amounts were well below the maximum residue levels (MRLs) established by the European legislation (Table 2). However, our results indicate that, although in recent years there has been an evident decline in these residue levels, these pesticides remain present in the environment and therefore present in the food chain. It is a known fact that the levels of contamination by POPs of the different areas may greatly vary, depending on factors, such as the geographical and regulatory variations in their use and restrictions. This fact explains the differences of levels and patterns between the different studies.

3.2. Occurrence of PCB residues in yogurt samples

We found that all the yogurt samples showed quantifiable levels of PCBs. The number of PCB residues in both production methods was similar (conventional yogurt, average = 9 PCB congeners per sample, range 8–12; organic yogurt, average = 8 PCB congeners per sample, range 7–11). As shown in Table 3 the total content of PCBs of these samples was also very similar between both groups (organic *vs.* conventional), but some differences were found when we studied the content of M-PCBs and DL-PCBs separately.

Table 2. Comparison between the median concentrations determined in the yogurt samples and the legal limits established by the European Authorities.

Compound	MRL (mg/kg fat)	Conventional yogurt	Organic yogurt
OCPs[a]			
Aldrin and dieldrin	0.006	0.00037	0
∑Chlordanes	0.002	0	0
∑DDT	0.04	0.00035	0.00025
∑Endosulfans	0.05	n.d.	n.d.
Endrin	0.0008	n.d.	n.d.
Heptachlor	0.004	n.d.	n.d.
HCB	0.01	0.000049	0.000042
α-HCH	0.004	n.d.	n.d.
γ-HCH (lindane)	0.001	0.000027	0.000033
Methoxychlor	0.01	n.d.	n.d.
PCBs[b]			
EQT DL-PCBs-OMS	2.5 (pg/g fat)	0.16	0.08
∑M-PCBs	40 (ng/g fat)	1255	13.21
PAHs[c]			
Benzo(a)pyrene	1.0 (µg/kg fresh product)	0	0
∑PAH4	1.0 (µg/kg fresh product)	0.05	0

[a]Maximum residue levels (MRL) related to OCPs were obtained from Commission Regulation (EC) No 299/2008 (EU).
[b]Maximum levels related to DL-PCBs and M-PCBs were obtained from Commission Regulation (EC) No 1259/2011 (EU).
[c]Maximum levels related PAHs: benzo[a]pyrene and PAH4 (sum of benzo[a]pyrene, benzo[a]anthracene, benzo[b]fluoranthene and chrysene) were obtained from Commission Regulation (EC) No 835/2011 (EU).

Firstly, in relation to the six M-PCBs, all the congeners were detected in all the samples, with the exception of PCB 101 that was below the LOD in two samples. M_PCBs accounted for more than 90% of the total content of PCBs in these samples. The similarity between groups of yogurt was maintained in M-PCBs, being the concentrations and frequencies similar in conventional and organic yogurts (around 12 ng g^{-1}, Table 3). These values were also similar to those values reported by EFSA and other authors in European dairy products (EFSA 2008; Polder et al. 2010; Schecter et al. 2010).

However, in relation to the more worrying congeners, the dioxin-like PCBs, we found that the median values of concentration were higher in conventional yogurts than in organic ones (2.73 ng g^{-1} fat *vs.* 1.64 ng g^{-1} fat). When we used the TEQ approach as defined by the WHO in 2006 (Van den Berg et al. 2006) the median values obtained were 0.16 pg TEQ g^{-1} fat and 0.08 pg TEQ g^{-1} fat in conventional and organic brands, respectively. These values can be considered as low, or at least much lower that those described by EFSA (2012) or other independent studies (Bordajandi et al. 2004; Malisch & Dilara 2007; Esposito et al. 2009). Although the median values of PCBs reported in our study are well below the established limits (40 ng g^{-1} fat for M-PCBs and 2.5 pg WHO-TEQ/g^{-1} fat for DL-PCBs, Table 2), it should be highlighted that the DL-PCB content of several samples widely overpassed these limits. Thus, in six samples of conventional yogurt (35.3%) we found a range from 6.7 to 116.40 pg WHO-TEQ/g^{-1} fat, and in three samples of organic yogurt (20.1%) a range from 6.7 to 36.7 WHO-TEQ/g^{-1} fat, whereas in a previous study on 1415 samples of dairy products it was reported that only 0.5% of the samples overpassed the legal limits (EFSA 2012). This is a worrisome result which suggests that yogurt could be a significant source of dioxins for the Spanish population if some of these brands are unintentionally consumed, and are in agreement with previous results of our research group in milk and cheese samples (Almeida-González et al. 2012; Luzardo et al. 2012).

3.3. Occurrence of PAH residues in yogurt samples

According to our results 100% of the yogurt samples presented contamination by any of the PAHs included in this research, without relevant differences in the number of detected compounds between conventionally and organically produced yogurts. The average number of residues was seven (range 8–12 and 7–11, in conventional and organic yogurts, respectively). Only three PAHs were not detected in any of the samples: dibenzo-[a,h]anthracene, anthracene and benzo[a]anthracene. At the opposite end phenanthrene, fluoranthene and pyrene were detected in 100% of the samples, and these compounds were also found at the highest concentrations in yogurts of both types of production, with similar median values (Table 4). Our results were in agreement with the scarce data that are currently available for PAHs

Table 3. Levels of PCBs (ng g^{-1} fat) and 2005 WHO TEQs (pg g^{-1} fat) detected in conventional and organic yogurt samples from the Spanish market.

Compound	CONVENTIONAL YOGURT			ORGANIC YOGURT			
	Mean ± SD 9.35 ± 1.37			Mean ± SD 8.87 ± 1.25			*p* NS
No. of PCBs residues per sample	Median	Detection range	Frequency	Median	Detection range	Frequency	P^a/P^b
Marker PCBs							
PCB 28	5.87	3.89–16.77	100%	5.48	2.55–12	100%	NS
PCB 52	2.03	1.60–6.13	100%	1.92	0.7–4.1	100%	NS
PCB 101	0.79	n.d.–2.82	94.1%	0.53	n.d–2.31	93.3%	NS
PCB 138	2.58	1.07–8.92	100%	2.33	0.34–9.39	100%	NS
PCB 153	1.3	0.70–4.94	100%	1.12	0.38–5.32	100%	NS
PCB 180	0.65	0.11–4.42	100%	1.03	0.19–4.08	100%	NS
∑M-PCBs	12.55	10.71–38.88	100%	13.21	4.99–36.99	100%	NS
Dioxin-like PCBs							
PCB 77	n.d.	n.d.	n.d.	0	n.d.–0.14	6.7%	NS
PCB 81	0	n.d.–0.91	41.2%	0	n.d.–0.52	33.3%	NS
PCB 105	0.76	n.d.–2.35	94.1%	0.41	n.d.–2.19	80%	NS
PCB 114	n.d.	n.d.	n.d.	0	n.d.–0.24	6.7%	NS
PCB 118	1.32	0.31–4.27	100%	0.97	0.18–4.52	100%	NS
PCB 123	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
PCB 126	0	n.d.–0.63	17.6%	0	n.d.–0.36	6.7%	NS
PCB 156	0	n.d.–0.43	17.6%	0	n.d.–1.21	33.3%	NS
PCB 157	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
PCB 167	0	0.00–1.1	29.4%	n.d.	n.d.	n.d.	0.030/0.022
PCB 169	0	n.d.–1.98	29.4%	0	n.d.–0.37	13.3%	NS
PCB 189	0	n.d.–2.52	11.8%	0	n.d.–0.18	6.7%	NS
∑DL-PCBs	2.73	0.66–11.12º	100%	1.64	0.24–7.13	100%	0.074/NS
$\sum TEQs_{DL\text{-}PCBs}$ (pg/g)	0.16	0.19–116.38	100%	0.08	0.007–36.32	100%	0.074/NS
∑PCBs	14.72	11.43–47.38	100%	14.86	5.64–43.91	100%	NS

[a]Values resulting from the comparison between the medians, referred to the figures with an asteric (Mann-Whitney test).
[b]Values resulting from the comparison between the frequencies, referred to the figures with an asteric (Chi-square test).
n.d., non-detectable.

content in dairy products (Bordajandi et al. 2004; Falco et al. 2003; Kim et al. 2008; Çok et al. 2012; Veyrand et al. 2013) or in similar matrices such as human breast milk (Kim et al. 2008; Çok et al. 2012).

We considered very interesting to focus our study on the group of eight PAHs that have been signalled by the EFSA as the most worrying (PAH8; (benzo[a]pyrene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[g,h,i]perylene, chrysene, dibenzo[a,h]anthracene and indeno[1,2,3-c,d]pyrene) due to their mutagenic and carcinogenic properties (EFSA 2008). Thus, we found that benzo[a]pyrene (a group I-carcinogen according the International Agency of Research of Cancer) was detected in 5.9% of conventional yogurts and in 13.3% of organic samples. Other contaminants from this subgroup were also detected with a relatively high frequency: benzo[k]fluoranthene, benzo[b]fluoranthene and chrysene (Table 4), which appeared with similar frequencies between samples of both production methods. High concentrations of chrysene and benzo[b]fluoranthene were also detected in the foodstuffs that were analyzed in the second French Total Diet Study (Veyrand et al. 2013). However, the results that we report in this study (Table 4) are higher than those from the above-mentioned study (Veyrand et al. 2013), and also than those reported by the EFSA (2008). Nevertheless, it should be noted that the median levels of PAHs in yogurt did not exceed the legal limits (Table 2), as also occurred with the other groups of pollutants of this study.

3.4. Assessment of yogurt-related dietary intakes of OCPs, PCBs and PAHs residues

Dietary exposure calculations are done by combining data on consumption habits with the concentrations of contaminants in food samples. In Table 5 we have summarized the dietary intakes of all the substances included in this study arranged by two groups of age: children (6–10 years) and adults (18–64 years), and considering two possible scenarios: (1) the consumers choose yogurts from conventional production, and (2)

Table 4. Levels of PAHs (ng g^{-1} fat) detected in conventional and organic yogurt samples from the Spanish market.

Compound	CONVENTIONAL YOGURT			ORGANIC YOGURT			
	Mean ± SD 7.12 ± 1.83			Mean ± SD 7.20 ± 2.01			*p* NS
No. of PAHs residues per sample	Median	Detection range	Frequency	Median	Detection range	Frequency	P^a/P^b
Naphthalene	0	n.d.–9.69	23.5%	0	n.d.–3.59	46.7%	NS
Acenaphthylene	5.54	n.d.–85.28	52.9%	5.71	n.d.–40.42	53.3%	NS
Acenaphthene	6.40	n.d.–55.05	94.1%	0.99	n.d.–14.72	66.7%	NS/0.047
Fluorene	2.47	n.d.–14.25	70.6%	3.66	n.d.–7.40	93.3%	NS
Phenanthrene	37.82	16.60–85.99	100%	35.65	13.47–88.87	100%	NS
Anthracene	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
Fluoranthene	11.69	6.48–18.60	100%	9.65	3.52–35.15	100%	NS
Pyrene	27.48	10.72–56.65	100%	30.78	8.11–144.70	100%	NS
Benzo[a]anthracene	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Chrysene	0	n.d.–17.52	47%	0	n.d.–20.04	33.3%	NS
Benzo[b]fluoranthene	0	n.d.–5.00	41.2%	0	n.d.–6.13	40%	NS
Benzo[k]fluoranthene	0	n.d.–1.64	47%	0	n.d.–1.62	46.7%	NS
Benzo[a]pyrene	0	n.d.–0.70	5.9%	0	n.d.–1.73	13.3%	NS
Indeno[1,2,3-cd] pyrene	0	n.d.–0.26	11.8%	0	n.d.–0.39	6.7%	NS
Dibenz[a,h]anthracene	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	NS
Benzo[ghi]perylene	0	n.d.–1.46	17.6%	0	n.d.–1.67	20%	NS
∑PAH8	1.03	n.d.–24.15	64.7%	0	n.d.–29.47	46.7%	NS
∑PAHs	110.05	60.33–306.59	100%	102.21	38.28–294.69	100%	NS

[a]Values resulting from the comparison between the medians, referred to the figures with an asteric (Mann-Whitney test).
[b]Values resulting from the comparison between the frequencies, referred to the figures with an asteric (Chi-square test).
n.d., non-detectable.

the consumers choose yogurts from organic production. It is important to note that, to our knowledge, this is the first study estimating the intake of pollutants through the consumption of organic yogurt.

As regards to the intake of OCPs through yogurt, this can be considered very low in both groups of age, but there are relevant differences depending of the type of yogurt chosen. Thus, those consumer that choose organic

Table 5. Median values of dietary intakes of POPs (ng kg^{-1} b.w. day^{-1}) in relation to yogurt consumption for children and adults living in Spain and depending on the production method chosen.

	CONVENTIONAL				ORGANIC			
	Children		Adults		Children		Adults	
	Lba	Uba	Lba	Uba	Lba	Uba	Lba	Uba
OCPs								
∑HCH	0.051	0.086	0.021	0.035	0.062	0.095	0.025	0.039
∑Cyclodienes	0.692	0.818	0.284	0.336	0	0.128	0	0.053
∑DDTs	0.662	0.681	0.272	0.280	0.490	0.510	0.201	0.209
∑OCPs	2.068	2.242	0.850	0.921	1.181	1.363	0.485	0.560
PCBs								
∑M-PCBs	1.230	1.489	0.506	0.402	1.240	1.500	0.509	0.405
∑DL-PCBs	0.236	0.273	0.097	0.112	0.142	0.184	0.059	0.075
∑PCBs	1.277	1.316	0.525	0.541	1.288	1.329	0.529	0.546
$\sum TEQs_{PCBs}$[a]	0.014	0.514	0.006	0.211	0.007	0.507	0.208	0.166
PAHs								
∑PAH8	0.089	0.132	0.037	0.054	0	0.046	0	0.019
∑PAHs	9.544	9.594	3.921	3.942	8.863	8.922	3.642	3.666

[a]Values expressed in pg kg^{-1}b.w. day.
Lba, lower bound approach; Uba, upper bound approach.

yogurts are even less exposed to these contaminants (especially to DDTs and cyclodienes). This also occurred with the intake of equivalents of dioxins: the consumers of organic yogurts would consume the half of DL-PCBs than the consumers of conventionally produced ones. However, it is interesting to note that the results greatly vary depending on the approach chosen for the calculations. Thus, if we had chosen the lower bound approach we may say that the consumption of yogurt do not represent an important source of these contaminants, as our estimates of $\sum TEQs_{PCBs}$ indicate that in the highest mean value of intake (children that consume conventional yogurts= 0.017 ng kg^{-1} b.w. day^{-1}) the amount ingested would represent only 0.85% of the tolerable daily intake (TDI) of dioxins (2 pg kg^{-1} b.w.d^{-1}) recommended by the WHO (Fattore et al. 2008). However, according the recommendation of the European Commission (EFSA 2012), it is necessary to apply the upper bound approach in the calculations for the comparison with the TDI. Using this approach the intake of DL-PCBs through yogurt would represent as much as 31% of the TDI in children. Moreover, it is important to note that, independently of the approach used for the calculations, if 5 out of 32 samples (15.6%, four conventional and 1 organic yogurts) had been consumed, the TDI would be greatly overpassed, because of their high content of the most toxic congeners: PCB 126 and PCB 169 (Table 3). The intake of two of these yogurts would represent in children as much as the 608% and 503% of the TDI for dioxins. These findings are a matter of concern and further studies are needed that clarify which is the origin of this high contamination in certain yogurt samples, because some consumers could be highly exposed to dioxins through the intake of this food.

Finally, as regards to the intake of PAHs through the yogurt consumption, this can be considered as very low in the Spanish population. However, again it is remarkable that when consumers choose yogurts from organic production the intake of these pollutants is two times lower than if conventionally produced yogurts are consumed (Table 4).

Although according our estimates the intake of these anthropogenic pollutants through yogurt consumption is below the TDIs, it is necessary to emphasize that the daily intakes are based on 'acceptable' risk to human health and does not mean zero risk. If we take into account that many of these contaminants are endocrine disruptors and/or carcinogens, a completely 'safe' level cannot be established and therefore the efforts for diminishing their presence in the environment should continue until their complete elimination.

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