Integrated assessment of infant exposure to persistent organic pollutants and mercury via dietary intake in a central western Mediterranean site (Menorca Island)

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Abstract

In this research the levels of organochlorine compounds (OCs) and mercury (Hg) in several food items from Menorca Island were presented. The dietary exposure assessment was performed in children population from the island. Finally, body burden of OCs and Hg in these infants were associated with their dietary intakes of the selected food items.

The dietary exposure to persistent pollutants by children population from Menorca Island was assessed. The concentrations of 11 organochlorine pesticides, 6 polychlorinated biphenils (PCBs) and 1 inorganic toxic element, Hg, were determined in 46 food samples that included fish, shellfish, meat, fruit, vegetables, cheese and eggs, which were acquired in local markets and department stores in the Menorca Island. The most contaminated food items were fish and shellfish, followed by meat and cheese products. OC levels were similar or lower than in other previous studies. However, 66 % of the analysed fish and shellfish species for Hg exceeded the human consumption safety limits according to the European Union Legislation. Pollutant data from food was combined with the pattern of consumption of these foodstuffs in order to calculate the estimated daily intake (EDI) of these contaminants. According to our results, fish and fruit were the main sources of OCs to the EDIs (contributing to 37 % and 29 %, respectively) while fish and shellfish were the main sources of Hg (76 % and 17 %). The estimated EDIs of OCs were well below to the reported FAO/WHO Tolerable Intakes. However, estimated weekly intake of Hg would exceed the Provisional Tolerable Weekly Intake indicated by EFSA in the case that the only fish and seafood source would be from the central western Mediterranean. Direct associations between fish/shellfish consumption and hair concentrations of Hg and fish and meat consumption and 4,4’-DDT concentrations in venous serum in the Menorcan children were observed.

Keywords: Organochlorine compounds, Mercury, Food, Dietary exposure, Menorca Island

1. Introduction

Anthropogenic release of organochlorine compounds (OCs) and mercury (Hg) into the environment has led to negative effects in human beings and ecosystems (Saeedi et al., 2016, Lamborg et al., 2014). The OCs are synthetic products widely used since the 1920s for several applications. Some organochlorine pesticides (OCPs) were extensively used in agriculture due to their insecticidal and fungicidal properties. Polychlorinated biphenyls (PCBs) were used for a large number of industrial applications, such as dielectrics in transformers, coatings, paints and insulating fluids (Wurl et al., 2005).

On the other hand, the environmental occurrence of Hg has been enhanced by mining and fossil fuel combustion (Lamborg et al., 2014). Hg is present in many chemical forms, being methylHg more toxic than the original metal. In the aquatic environment, Hg is readily transformed to methylHg by anaerobic bacteria. Most of the metal incorporated into aquatic organisms is in the methylated form (80-90%; Harris et al., 2003).

Human exposure to OCs has been related to several health effects including cancer, reproductive defects, neurobehavioral abnormalities and endocrine and immunological toxicity (Mrema et al., 2013). On the other hand, methylHg is a potent neurotoxic agent that can cause severe neurological damage to humans (Grandjean et al., 1997).

Despite their different origin and structure, OCs and Hg share common properties such as (I) strong chemical stability and environmental persistence, (II) bioconcentration in living organisms and biomagnification through the food chain due to their hydrophobic character and (III) toxicity for humans and wild animals. Accordingly, the production and use of OCs and Hg has been restricted and/or banned in many countries. However, these contaminants are still found in the environmental compartments (Arellano et al., 2011; Lamborg et al., 2014), foodstuff (Martí-Cid et al., 2010; Olmedo et al., 2010) and human tissues (Vizcaino et al., 2014; Garí et al., 2013).

Dietary intake is a major way of incorporation of these compounds into humans. This is particularly important for children since their organs and metabolism are still under formation and pollutant exposure may have stronger long term effects than in adults (Guxens et al., 2012; Vizcaino et al., 2014).

Assessment of the main routes of exposure to these chemicals in infant populations is needed for implementation of adequate strategies towards minimization of pollutant intake. Previous studies have investigated the main routes of exposure to OCs (Leontjew et al., 2016, Ahmad et al., 2010, Stefanelli et al., 2004) or Hg (Rubio et al., 2016, Olmedo et al., 2013, Millour et al., 2011) but to the best of our knowledge none of them has performed integrated assessments on both types of pollutants in the same cohort. Concerning the OCs, the present study encompasses both PCBs and OCPs.

Menorca Island is located in the central Western Mediterranean (39°47' - 40°05'N, 3°47' - 4°19'E) and has an extension of 702 Km^2. This small island represents a relatively isolated Mediterranean environment in which a large proportion of the food consumed by the inhabitants is generated locally. Furthermore, most of the fish consumption depends on the captures by local fishermen. The island has not any industry involving Hg use or production of OCs. Until the emergence of tourism, the main economic activity was agriculture. This island constitutes a representative case of a population living in a Mediterranean environment. High Hg levels are observed in the food-web of this semi-enclosed sea (Cossa et al., 1997; Gagnon et al., 1997; Pirrone et al., 2003; Heimburger et al., 2010; Zagar et al., 2007) despite its anti-estuarine circulation pattern that transfers nutrients and pollutants to the Atlantic Ocean. Identification of the routes of transport of this metal and other pollutants into the population living in sites under Mediterranean influence is important. Menorca can be taken as a model ecosystem of communities living in marine Mediterranean environments in which the main routes of dietary pollutant incorporation of OC and Hg can be assessed.

2. Materials and methods

2.1. Sampling

Food samples were acquired in local markets and department stores from the main two cities: Maó and Ciutadella (Fig 1; December 2014, n = 46). The selection was based on Food Frequency Questionnaires (FFQs) from 4 year old children from Menorca cohort (see next subsection below) and interviews with local experts.

The food samples included fish species of several trophic levels and fat contents (Table 1) and shellfish (Table 2), all them captured in the island surroundings. These fish species were representative of the local consumption, encompassing angler (*Lophius piscatorius*), European hake (*Merluccius merluccius*), black seabream (*Spondyliosoma cantharus*), common dentex (*Dentex dentex*), common pandora (*Pagellus erythrinus*), common seabream (*Pagrus pagrus*), dusky grouper (*Epinephelus marginatus*), forkbeard (*Phycis phycis*), Mediterranean moray (*Muraena helena*), red scorpionfish (*Scorpaena scrofa*), small-spotted catshark (*Scyliorhinus canicula*), thornback ray (*Raja clavata*), red mullet (*Mullus barbatus*), red sea bream (*Pagellus acarne*) and surmullet (*Mullus surmuletus*). The selected shellfish species were mussel (*Mytilus galloprovincialis*), squid (*Loligo vulgaris*), scampi (*Nephrops norvegicus*) and shrimp (*Aristeus antennatus*).

The food samples examined also included meat, e.g. beef, chicken and lamb (Table 2), fruits and vegetables (Table 3), cheese (Table 3) and chicken eggs (Table 3), all them produced locally.

After collection, each food item was dissected in two composite samples for OC and Hg analyses. The former were wrapped in aluminium foil and the second were sealed in plastic bags. The samples were then frozen at -23ºC until further analysis in the laboratory. Only the edible parts of each food item were analysed.

2.2. Study population

The Menorca cohort is part of the INMA (Spanish Children's Health and Environment) project, a research network that focuses on the effects of environmental contaminants during pregnancy and on fetal and child development (Ribas-Fitó et al., 2006; Guxens et al., 2012).

Between 2001 and 2002, 4-year old children from this cohort provided serum samples (n=285) and hair samples (n=302) for the determination of OCs and Hg, respectively. These data have been described in Carrizo et al. (2006) and Garí et al. (2013), respectively. Children's dietary intakes were assessed through FFQs (Vioque et al., 1991; Willet et al., 1985). Only the dietary intakes which concurred with the analysed food items were selected for study, as follows: fish, shellfish, meat (beef and chicken), fruit, vegetables, cheese (semi-skimmed cheese) and eggs. The rest of food items (*e.g.* dairy products, cereals, fats) were not included in the analyses.

All child's parents gave written permission for participation in the study, which was approved by the Ethics Committee of the Institut Municipal d'Investigació Mèdica (Barcelona).

2.3. Chemicals and standards

Solvents for residue analysis, *n*-hexane, dichloromethane, isooctane, acetone, concentrated 95-97% sulphuric acid, 65% nitric acid, 30% hydrogen peroxide, anhydrous sodium sulphate and silica gel were from Merck (Darmstadt, Germany). Sulphate and silica were activated overnight by heating at 400°C. The cellulose extraction cartridges (22 mm x 80 mm) were from Whatman International Ltd. (UK). Standard digestion teflon vessels (60 mL) were from Savillex (Minnesota, USA). Standards of OCPs and PCB congeners were from Dr. Ehrenstorfer (Wesel, Germany). 1,2,4,5-Tetrabromobenzene (TBB; Aldrich, Steinheim, Germany) and PCB 209 (Dr. Ehrenstorfer, Wesel, Germany) were used as recovery standards. PCB 142 (Dr. Ehrenstorfer) was used as internal standard. The standard mixtures of OCPs, PCB and a surrogate solution composed of TBB and PCB 209 were prepared in isooctane. Fish muscle certified reference material, ERM-BB422, was obtained from the Institute for Reference Materials and Measurements (Geel, Belgium).

The following pollutants were included in the analyses: pentachlorobenzene, hexachlorobenzene (HCB), 4 isomers of hexachlorocyclohexane (α-, β-, γ- and δ-HCH), 2,4′-DDT, 4,4′-DDT, 4,4′-DDE, 2,4′-DDE and 4,4′-DDD, the ICES PCB congeners (28, 52, 101, 118, 138, 153 and 180) and total Hg.

2.4. Extraction and clean-up

The methods of extraction and clean-up of OCs in fish, meat, vegetables and fruits were based on previous analytical procedures (Berdie et al., 1998, Vives et al., 2002). Briefly, each sample (3-4 g) was homogenized with activated sodium sulphate until a fine powder was obtained and then the mixtures were introduced into previously cleaned cellulose cartridges (6 h in Soxhlet). These mixtures were Soxhlet-extracted with 100 mL of *n*-hexane-dichloromethane (4:1 v/v) for 6 h. At this step, TBB and PCB 209 were added as recovery standards (50 ng/mL). The extract was concentrated with a rotary evaporator to 2 mL. Three mL of sulphuric acid were then added. After vigorous stirring in a Vortex-mixer (2 min), the mixture was centrifuged (4000 rpm, 5 min) to remove any foam in the interface and the sulphuric acid layer was discarded. This clean-up step was repeated until a colorless transparent acid layer was obtained (3-5 times). The solvent layer was introduced into a chromatographic column packed with 1 gr of sodium sulphate and silica gel (1:1 by weight). The extract was then evaporated to dryness under a gentle stream of nitrogen (10-20°C) and transferred to vials using 200 µl of isooctane. Before instrumental analysis, the sample was evaporated to nearly dryness under a gentle nitrogen flow and a solution of PCB 142 was added as internal standard (10 ng/mL).

Cheese and eggs samples were extracted following previous analytical methods, with some minor modifications (Kalantzi et al., 2001). Briefly, each sample (1-2 g) was homogenized with activated sodium sulphate (5 g) and then Soxhlet extracted with 50 mL of *n*-hexane. After cooling, the extract was spiked with TBB and PCB 209 as recovery standards (50 ng/mL) and concentrated with a rotary evaporator to 5 mL. The sample was then introduced into a 1.9 cm i.d. column containing 15 g of acidified silica gel (2:1 silica gel:acid by weight) and eluted with 60 mL of *n*-hexane. The extract was then evaporated to 1 mL under a gentle stream of nitrogen and introduced into a 0.6 cm i.d. column containing 3 g of activated silica. The sample was eluted with 33 mL of hexane (for PCBs) and 15 mL of hexane-dichloromethane (1:1 v/v; for OC pesticides). Before instrumental analysis, the sample was evaporated to nearly dryness under nitrogen and a solution of PCB 142 was added as internal standard (10 ng/mL).

For Hg determination each sample (5 g) was freeze-dried for 12 h and then introduced (0.1 g) into a digestion Teflon vessel, together with 3 mL of nitric acid and 1 mL of hydrogen peroxide. The mixture was heated in an oven (90°C) overnight. After cooling, the digested sample was dissolved with 40 mL of MilliQ water. Finally, samples were placed in 7 mL glass bottles and stored in a refrigerator until instrumental analysis.

2.5. Determination of persistent pollutants and quality assurance

The OCs were quantified using a gas chromatograph with electron capture detection (GC-ECD, Agilent Technologies 7890A, Palo Alto, California, USA) equipped with a HP-5MS capillary column of 60 m length, 0.25 mm internal diameter and 0.25 µm film thickness (J&W Scientific, Folsom, CA, USA), protected with a retention gap. The oven temperature was programmed from 90°C (holding time 2 min) to 130°C at 15°C/min and finally to 290°C at 4°C/min, keeping the final temperature for 15 min. Injector and detector temperatures were 250°C and 320°C, respectively. Injection (2 µl) was performed in the splitless mode, keeping the split valve closed for 30 s. Helium was the carrier gas (1.5 mL/min) and nitrogen was used as make-up gas (60 mL/min).

Structural confirmations were performed by GC (Agilent Technologies 7890A, Agilent Palo Alto, USA) coupled to mass spectrometry (MS, Agilent Technologies 5975C, Agilent Palo Alto, USA) operating in negative chemical ionization mode (GC-NICI-MS). The system was equipped with a HP-5MS capillary column of 60 m length, 0.25 mm internal diameter and 0.25 µm film thickness (J&W Scientific, Folsom, CA, USA), protected with a retention gap. Helium was used as carrier gas (1.2 mL/min). Ammonia was the reagent gas (2.5 mL/min). The oven temperature program started at 90°C which was held for 2 min, followed by a first increase to 130°C at 15°C/min and a final ramp to 310°C at 4°C/min with a final holding time of 10 min. Injector, transfer line and ion source temperatures were 280°C, 280°C and 176°C, respectively. The dwell time was 100 ms/channel.

One procedural blank was included in each batch of samples. Mean recoveries of spiked standards in the samples were 55% and 75% for TBB and PCB 209, respectively. Detection and quantification limits were determined as the average signal obtained from the blanks plus three and five times the standard deviation, respectively. Detection limits ranged between 0.0050 and 0.095 ng/g wet weight.

Total Hg was performed using inductively coupled plasma mass spectrometry (ICP-MS, Elan 6000) operating under standard conditions and using rhodium as internal standard. One procedural blank was included in each sample batch. The detection limit was 0.00010 mg/kg wet weight. Analyses of fish muscle certified reference material, ERM-BB422, containing 0.601 ± 0.03 mg/kg of Hg ensured that the instrument remained calibrated along the study. The mean recovery obtained was 94%.

2.6. Dietary exposure estimates

The estimation of the daily intake of pollutants through the consumption of selected food items was calculated by multiplying the mean pollutant concentration in the item (expressed in ng/g fresh product) by the average daily consumption rate of that item (expressed in grams/day). Then, total dietary intake was obtained by summing these products for each contaminant.

The daily consumption rates of the 4-year old children living in Menorca were obtained by combining the information from the FFQs (expressed in times/week) and the servings of each food item at the age of 4 years as defined by AESAN (expressed in grams; vegetables = 130 g, meat = 53 g, shellfish = 73 g, fish = 73 g, eggs = 50 g, cheese = 27 g and fruit = 87 g). The FFQs from INMA-Menorca were based on a representative sample encompassing 302 children (average body weight = 18.5 kg).

2.7. Statistical analysis

The statistical analyses were performed using R (R Core Team, 2015) software and Microsoft Excel (2010). The concentrations of OCs and Hg were expressed in ng/g wet weight (ww) and mg/kg wet weight, respectively. When OCs and Hg concentrations were under the limit of detection, the values were assumed to be one-half of the detection limits (ND=½ LOD). Either ww or dry weight (dw) were used for comparison with other studies.

Total HCHs were defined from the sum of α-, β- and γ-HCHs. Total DDT concentrations were the sum of 4,4’-DDT, 2,4’-DDT, 4,4’-DDE, 2,4’-DDE and 4,4’-DDD. Total PCB concentrations were the sum of 28, 52, 118, 138, 153 and 180. HCB and Hg were reported as individual compounds.

Serum OCs and hair Hg concentrations (Carrizo et al., 2006; Gari et al., 2013) were not normally distributed and were transformed into natural logarithms. Multivariate linear regression analyses were used to assess the association of children’s dietary intakes and other covariates (sex, place of birth, breastfeeding, number of siblings, maternal age, and maternal and paternal occupation and educational level), with each compound concentration at 4-year old children. The compound concentrations and food item variables included in the models were standardized.

3. Results

The descriptive data of the OC and Hg concentrations in the analysed foodstuffs are shown in Tables 1-3. Non-detected compounds (e.g. PeCB, β- and γ-HCH) or compounds presenting low detection frequencies (e.g. α-HCH, δ-HCH, 2,4'-DDT, PCB28 and PCB52) were not reported. The relative distributions of the main pollutant groups (HCB, ∑HCHs, ∑DDTs, ∑PCBs and Hg) to the total sum of persistent pollutants (∑PP) are shown in Fig. 2.

3.1 Fish and shellfish

All fish and seafood samples showed detectable levels of 4,4'-DDE and PCB congeners 118, 138, 153 and 180 (Tables 1 and 2). 4,4’-DDT and 4,4’-DDD were found in 87%

and 57% of the analysed samples, respectively. The other major compounds, e.g. HCB and 2,4’-DDE, were found in 50% of the samples (Tables 1 and 2). The patterns of DDTs and PCBs in fish and seafood were dominated by 4,4'-DDE and PCB153. The highest OC concentration in fish was found in European hake (6.1 ng/g ww of 4,4’-DDE). The highest concentration in shellfish was observed in mussel (1.8 ng/g ww of PCB153). These two samples were collected in Maó (Tables 1 and 2).

All fish and shellfish samples showed detectable Hg levels, ranging between 0.068 and 3.8 mg/kg ww (arithmetic means of 1.1 mg/kg ww and 0.92 mg/kg ww for fish and shellfish, respectively; Tables 1 and 2).

Small-spotted catshark caught in Ciutadella was the species showing highest concentrations within the seafood group (3.8 mg/kg ww; Table 1). As shown in Fig. 2 Hg was the major persistent pollutant in fish and shellfish, contributing 99% to the total load.

3.2 Meat

All samples showed detectable levels of 2,4'-DDE, 4,4'-DDD, PCB118, PCB138 and PCB153 (Table 2). The other compounds, e.g. HCB, 4,4’-DDT and 4,4’-DDE, were found in less than 60% of the samples and PCB180 was not found in any. The DDT and PCB patterns were mostly dominated by 4,4'-DDE and PCB118, respectively. The highest concentrations were found in lamb bought in Ciutadella (0.26 ng/g ww of 4,4’-DDE; Table 2).

Hg was also the major persistent pollutant in the meat foodstuff group, contributing to 98 % of ∑PP (Fig 2). However, it was only found in beef and lamb samples from Maó (Table 2).

3.3. Fruit and vegetables

All fruits and vegetables showed detectable levels of 2,4'-DDE, 4,4'-DDD, PCB118, PCB138 and PCB153 (Table 3). PCB180 was found in half of the analysed vegetable samples but not in any of the fruits. HCB, 4,4'-DDT and 4,4'-DDE were not found in any of the fruit or vegetable samples. The DDT and PCB patterns were dominated by 2,4'-DDE and PCB138 in most of the samples from both groups. The highest OC concentrations found in fruit and vegetables corresponded to PCB138, 0.13 and 0.22 ng/g ww, respectively. Hg was not found above detection limit in any sample (Table 3).

The PCBs were the major persistent pollutants in both groups of fruit and vegetables followed by DDTs (Fig 2). In the fruit group, PCBs and DDTs constituted 55% and 18% of the ∑PP, respectively. In the vegetables, these pollutants represented 67% and 15% of ∑PP, respectively.

3.4 Cheese

All samples contained 4,4’-DDT, PCB138, PCB153 and PCB180 above limit of detection (Table 3). PCB118 was found in 67% of the samples. HCB, 4,4’-DDE, 2,4’-DDE and 4,4’-DDD were below limit of detection in all cases. The DDT and PCB patterns were dominated by PCB138 and 4,4’-DDT, respectively. The highest concentration was found in cheese from Maó (0.3 ng/g ww of PCB138).

Hg was found above limit of detection in all samples, ranging between 0.097 and 0.24 mg/kg ww (arithmetic mean 0.16 mg/kg ww). The semicured cheese from Maó showed the highest level (0.24 mg/kg ww; Table 3). This metal was the major persistent pollutant in the cheese group, with a similar relative composition as observed in fish and shellfish (Fig. 2).

3.5 Eggs

4,4’-DDT was found in all analysed egg samples (Table 3). PCB118, PCB138, PCB153 and PCB180 were found in 50% of the samples. The PCB and DDT patterns were dominated by PCB153 and 4,4'-DDT, repectively. The highest OC concentrations were found in chicken eggs from Maó (0.24 ng/g ww of PCB153). PCBs and DDTs were the major persistent pollutant in this group (Fig 2), contributing to 62% and 15% to ∑PP, respectively. No Hg was found above limit of detection (Table 3).

3.6 Human biomonitoring

Hair Hg and serum OC concentrations from the studied infant population of Menorca are shown in Table 6. THg concentrations in hair ranged from 0.040 ug/g to 10 ug/g, with a geometric mean (GM) concentration of 0.99 ug/g (Garí et al., 2013), which represents 20% of the samples that were above the WHO recommended values in human hair. Concerning OCs, the most abundant was 4,4’-DDE, with a GM concentration of 0.87 ng/ml, followed by sum of PCBs (0.63 ng/ml), HCB (0.22 ng/ml) and 4,4’-DDT (0.069 ng/ml) (Carrizo et al., 2006).

4. Discussion

4.1 Occurrence of OCs in food items

The concentrations of OCs were considerably higher in fish and shellfish than in the other food groups (Tables 1-3) which is consistent with studies from other European countries, e.g. Sweden (Törnkvist et al., 2011). Fruits and vegetables showed the lowest concentrations of these pollutants.

The PCB congeners found in highest concentrations in all samples were those with highest molecular weights, PCB118, PCB138, PCB153 and PCB180, which involves those with highest hydrophobic properties (octanol-water coefficients, $logK_{ow} > 6.9$). The predominance of these compounds is consistent with their high hydrophobicity and bioaccumulation potential. 4,4'-DDE was the compound of the DDT group showing highest concentrations. The DDT/DDE ratios were low in all food samples, ranging between 0.013 and 1.2, which is indicative of old DDT exposure.

In general terms, the observed concentrations of HCB, total HCHs and total DDTs in meat, fruits, vegetables, eggs, cheese and dairy products from the island were low in comparison to those found in previous studies from other European regions, e.g. Croatia (Kljakovic-Gaspic et al., 2015), Austria (Mihats el al., 2015), Sweden (Törnkvist et al., 2011), Catalonia (Martí-Cid et al., 2010; Falcó et al., 2004; Llobet et al., 2003a) and Russia (Polder et al., 2010; Table 4). The concentrations of PCBs in the food items from the island were in the range of those described elsewhere although the number of cases for comparison was small (Table 4).

Concerning fish and seafood, the concentrations of HCB, total HCHs, DDTs and PCBs in the specimens captured nearby the island were small in comparison to the fish specimens consumed in these other European countries (Table 4). With a few exceptions, HCB from Sweden (Tornkvist et al., 2011) and DDTs and PCBs from the Canary Islands (Rodríguez-Hernández et al., 2016), the average concentrations of fish and seafood from Menorca were the lowest observed (Table 4). The differences were particularly significant for the DDT and the PCB groups since the average concentrations found in the fish/seafood group of Menorca were about 6-7 times lower than the average concentrations reported in Catalonia (Marti-Cid et al., 2010; Llobet et al., 2003a) or Russia (Polder et al., 2010) (Table 4). In general, decreasing concentrations in OCs have been observed (Krauthacker et al., 2009; Aguilar et al., 2005), probably as the result from the application of the Stockholm Convention.

The highest OCs concentrations were observed in the European hake from Maó (ΣOCs=11 ng/g ww; Table 1). This is a carnivore species that feeds on small fish and certain cephalopods, thus occupying a high level in the food chain. As mentioned above, most OCs are hydrophobic and have high $\log K_{ow}$ coefficients which increases their affinity for specimens with high fat content and for those located at high position in the food web. Accordingly, the lower OC levels in shellfish than fish (Tables 1 and 2) can be related to their lower fat content.

PCBs were the OCs found in highest concentrations in the meat samples, 0.43 ng/g ww (Table 4), followed by DDTs (0.15 ng/g ww), HCB (0.028 ng/g ww) and HCHs (<0.033 ng/g ww). The beef and lamb showed higher levels than chicken, probably due to the mammalian transference of these compounds through placenta and by breast feeding (Table 2; Shen et al., 2004).

Semicured cheese samples containing 15% fat showed lower OCs levels than semicured cheese with higher fat content (Table 3). The concentrations of these compounds in cheese and eggs from Menorca were lower than those found in Russia (Polder et al., 2010) and similar to those found in Catalonia (Llobet et al., 2003a; Table 4).

Finally, the fruit and vegetables analyzed in the present study showed the lowest OC concentrations which is consistent with the observed accumulation of these compounds in Catalonia (Martí-Cid et al., 2010; Falcó et al., 2004) and Russia (Polder et al., 2010; Table 2). This low accumulation pattern was not unexpected as these food items are in a low position of the food chain and their fat content is also low.

4.2 Occurrence of Hg in food items

Hg was present in all fish, shellfish and cheese samples and in 40 % of the meat samples. In contrast, it was not found in fruit, vegetables and eggs items (Tables 1-3). The highest Hg concentrations were found in seafood, ranging between 0.068 and 3.8 mg/kg ww. Sixty-six percent of the analysed species had Hg concentrations above the maximum level set forth by the European Union Maximum Residue Limits (MRL) for human consumption, 0.5-1.0 mg/kg ww depending on the species (EC, 2006).

The Hg concentrations observed in the present study were consistent with previous results reported in the same organisms from the Adriatic Sea (Storelli and

Barone, 2013; Storelli et al., 2003), Farwa Island (Lybian coast; Banana et al., 2016), the Gulf of Lion (Torres et al., 2015; Cresson et al., 2014), the Aegean Sea (Yabanli et al., 2015) and deep-sea sites from the Mediterranean basin (Koenig et al., 2013; Naccari et al., 2015).

A considerable variation was found in Hg concentrations among species. Biotic and abiotic factors may affect the accumulation of Hg in marine organisms. However, diet and position in the trophic web are determinant (Storelli et al., 2002).

The highest concentration was measured in small-spotted catshark collected in Ciutadella (average 3.8 mg/kg ww; maximum 14 mg/kg dw). In a recent study in fish from the Gulf of Lions (Northern Mediterranean), the same species recorded maximum average values, with an arithmetic mean of 7.1 mg/kg dw, and maximum concentration of 27 mg/kg dw (Cresson et al., 2014). In another study in the Mediterranean basin lower concentrations were observed (arithmetic mean 1.5 mg/kg ww, range 0.79-2.6 mg/kg ww; Storelli et al., 2002).

Small-spotted catshark is a predator with a diet mainly based on consumption of osteichthyes and crustaceans, cephalopods and gastropods. Its trophic level is high (3.93) since it is a top carnivorous predator (Mnasri et al., 2012). The observed high Hg concentrations are consistent with magnification processes due to their high trophic position and feeding behavior.

Concerning shellfish, the highest concentrations were recorded in shrimp from Maó (average 2.3 mg/kg ww). These results were in concordance with a previous study in the Mediterranean basin, with arithmetic mean and the maximum levels of 1.0 and 2.2 mg/kg ww, respectively (Koenig et al., 2013).

Semi-cured cheese was the second food group with highest average Hg concentrations (0.16 mg/kg ww) followed by meat (0.040 mg/kg ww; Table 1). Cheese with low fat content (15%) was the food item with lowest Hg concentration (0.097 mg/kg ww), which parallels the observations on OC concentrations. The Hg concentrations in these food items were higher than those found in other studies in the Canary Islands (Rubio et al., 2008) and Catalonia (Llobet et al., 2003b).

4.3 Dietary intake of OCs and Hg

The contributions of each food group to the total dietary intake of OCs and Hg in the cohort of four-year old children from Menorca are shown in Fig. 3. The most important OC sources were consumption of fish (37%) and fruit (29%). However, it is important to note the significant differences among consumption rates of these food items: 107 g/day in fruit and 20.2 g/day in fish (Table 5) involving 47 ng/d and 64 ng/d, respectively. The high significance of the fish contribution to the total dietary OC intake has also been observed in previous studies from other European populations (Mihats et al., 2015; Törnkvist et al.; 2011, Llobet et al., 2003a). Vegetables and shellfish were also identified as important contributors to the OC estimated daily intakes (EDIs; 13% and 7%, respectively).

Further understanding on the significance of each food item for the OC intake can be obtained by consideration of the Tolerable Daily Intakes (TDIs; JMPR, 2000). According to the results of the present study, the EDIs of OCs of the 4 year-old children from Menorca are much lower than the established TDI limits (TDI for HCB=160 ng/(kg bw·d), for ΣHCHs=5000 ng/(kg bw·d), for ΣDDTs=10,000 ng/(kg bw·d), for ΣPCBs=10 ng/(kg bw·d); Rodríguez-Hernández et al., 2016; JMPR, 2000; Mihats et al., 2015).

However, in comparison with previous studies, the PCBs EDIs of 4-year old children from Menorca was higher than children ingestion (6-15 years old) from Austria (ΣPCBs=3.37 ng kg^{-1}bw d^{-1}; Mihats et al., 2015). Focussing only on fish products, a recent study in children (average body weight=34.32 kg) from the Canary Islands found higher levels than those from the present study (Rodríguez-Hernández et al., 2016).

Fish and shellfish were the main contributors to the total Hg intake (76% and 17% of contribution, respectively). Because of its toxicity, the EFSA's CONTAM Panel established a Provisional Tolerable Weekly Intake (PTWI) of 4 µg/kg for this metal (EFSA, 2012). According to the present results, children from Menorca Island showed an estimated weekly intake (EWI) of 11 µg/kg, which exceeded more than twice the PTWI limits. This value represents the worst scenario, and taking into account that total consumption of fish and seafood is from local origin. The Menorca population also consumes fish and seafood from other Mediterranean areas, as well as from the Atlantic Ocean, and the consumption from local sites is supposed to be less than half of the total. The worst case scenario of a study in baby foods from the European Union market encompassed an EWI of 18.2 µg/kg in infants (7-8 months old) whose diet consisted on infant formulae of milk-based and fish-based infant solid foods (Pandelova et al., 2012). A study in women (25-44 years old) from Spain has shown that the Hg EWI largely exceeds the PTWI for methylHg as consequence of consumption of sword-fish and louvar (Herreros et al., 2008).

The specific fish and seafood EDI of Menorca (Mediterranean Sea) can be compared with one from the Canary Islands (Atlantic Ocean) based on the results reported in Rodriguez-Hernandez et al. (2016) from children having average body weight of 34.32 kg. The EDI identified in this case, 150 ng/(kg bw·d), is nearly ten times lower than 1500 ng/(kg bw·d) that was observed in the four-year old children from Menorca (Table 5).

4.4 Associations between pollutant concentrations in children and dietary intakes

The present dietary food analyses can be related to the hair Hg measurements in four-year old children from Menorca (Garí et al., 2013). The infant Hg concentrations show a positive and significant association with the fish and shellfish intake (Fig 4). This result is consistent with those obtained in a previous study in which the contribution of each food source to Hg intake was estimated from FFQs (Garí et al., 2013). The high Hg concentrations found in the analysed marine organisms (Table 1) and the dietary intake estimations show that 93% of the total dietary Hg intake originates from seafood consumption (Fig 3). Fish intake is highly beneficial although consumption of some kind of fishes such as dusky grouper and small-spotted catshark should be restricted during infancy.

There is also a significant dependence between the intake of 4,4’-DDT and fish and meat consumption (Fig. 4). Meat intakes were also found to be associated with DDT concentrations in another Mediterranean area (Valencia; Llop et al., 2010). In the analysed food items of Menorca, 87% of fish samples and 60% of meat samples presented detectable values for 4,4’-DDT. Moreover, egg intake is also related to high 4,4’-DDT levels, although without statistical significance (Fig. 4). All analysed chicken eggs samples showed detectable 4,4’-DDT levels.

The significant negative relationship between higher consumption of vegetables and concentrations of PCB138, PCB153 and PCB180, or fruit and concentrations of 4,4’-DDT in children suggests that high intake of these food items could be associated to lower consumption rates of other products, such as fish and meat, where PCBs and DDT tend to accumulate strongly (Fig. 4). As a result, lower intake of PCBs and DDT is observed. In this respect, PCB concentrations in the vegetable samples were above the limit of detection, and 4,4’-DDT was not found in any of the fruit samples analyzed. This last results is, however, not statistically significant.

5. Conclusions

In general terms, the concentrations of HCB, HCHs and DDTs in the food items produced in Menorca are low in comparison with other studies from other European regions. The concentrations of PCBs are in the range of those found in other European sites. However, the fish specimens from Menorca showed much lower PCB concentrations.

In contrast, Hg was present in all fish, shellfish and cheese samples and in 40 % of the meat samples of Menorca. However, it was not found in fruit, vegetables and eggs. Sixty-six percent of the analysed fish species had Hg concentrations above the maximum level set forth by the European Union MRL for human consumption.

The highest OC and Hg concentrations were observed in fish, presenting the maximum values in European hake and small-spotted catshark, respectively. These two species occupy high levels in the food chain where magnification processes are strongest for hydrophobic compounds with high bioaccumulation potential.

The most important sources for the intake of OCs in four-year old children from Menorca were consumption of fish (37%) and fruit (29%). However, the EDIs of these pollutants were low in comparison to the FAO/WHO TDIs. Fish (76%) and shellfish (17%) consumption were the main Hg sources. In the worst scenario, and assuming that total fish and seafood consumption is from local origin, the EWI of this metal in children from Menorca exceeded more than twice the EFSA PTWI.

A statistically significant and positive association was found between fish and shellfish consumption and hair Hg concentrations in 4 year-old children. Ninety-three percent of all dietary Hg intake originated from consumption of these food items. 4,4-DDT intake was also significantly directly associated to consumption of fish and meat. Hg and 4,4'-DDT are the only pollutants whose concentrations in children from the cohort were significantly associated to dietary inputs. Consumption of vegetables was protective against PCB accumulation.

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Table 1 OCs (ng/g wet weight) and Hg (mg/kg wet weight) in fish from markets of Menorca.

	Site of purchase	**HCB**	**4,4'-DDT**	**4,4'-DDE**	**2,4'-DDE**	**4,4'-DDD**	**PCB118**	**PCB138**	**PCB153**	**PCB180**	**DDT/DDE**	**Hg**
Detection Frequency (%)		52%	87%	100%	48%	57%	100%	100%	100%	100%		100%
Angler	Ciutadella	0.049	0.035	0.99	0.065	0.019	0.22	0.42	0.55	0.18	0.035	0.27
Angler	Maó	0.014	0.055	0.57	0.0064	0.0039	0.15	0.26	0.50	0.18	0.095	0.62
Black seabream	Ciutadella	0.014	0.031	0.39	0.010	0.0039	0.27	0.40	0.50	0.15	0.080	0.11
Black seabream	Ciutadella	0.019	0.010	0.79	0.037	0.0039	0.19	0.35	0.38	0.14	0.013	0.76
Black seabream	Maó	0.014	0.043	0.42	0.0064	0.0025	0.18	0.29	0.35	0.12	0.10	0.98
Common dentex	Maó	0.014	0.21	2.1	0.0064	0.017	0.36	0.94	1.3	0.42	0.10	2.0
Common pandora	Ciutadella	0.014	0.0079	0.15	0.047	0.011	0.18	0.24	0.25	0.076	0.054	0.16
Common seabream	Maó	0.083	0.071	0.95	0.0064	0.0025	0.27	0.50	0.77	0.22	0.075	0.69
Dusky grouper	Maó	0.014	0.039	0.54	0.0064	0.0025	0.25	0.49	0.64	0.28	0.071	2.5
European hake	Ciutadella	0.15	0.13	3.9	0.050	0.062	0.37	0.95	1.0	0.47	0.033	1.2
European hake	Maó	0.15	0.40	6.1	0.086	0.15	0.50	1.4	1.8	0.69	0.064	0.56
Forkbeard	Ciutadella	0.37	0.056	2.1	0.047	0.025	0.26	0.53	0.68	0.29	0.026	1.0
Forkbeard	Maó	0.28	0.090	1.7	0.0064	0.0025	0.32	0.64	0.87	0.37	0.052	1.4
Mediterranean moray	Maó	0.014	0.038	0.38	0.0064	0.0025	0.19	0.33	0.46	0.19	0.10	1.1
Red mullet	Ciutadella	0.014	0.010	0.26	0.040	0.0039	0.13	0.20	0.20	0.057	0.040	0.17
Red scorpionfish	Ciutadella	0.019	0.021	0.24	0.056	0.0079	0.27	0.52	0.72	0.26	0.088	1.4
Red scorpionfish	Maó	0.019	0.0079	0.57	0.0064	0.0025	0.17	0.33	0.50	0.17	0.014	0.42
Red sea bream	Ciutadella	0.019	0.033	0.36	0.045	0.0097	0.19	0.30	0.34	0.12	0.091	0.21
Small-spotted catshark	Ciutadella	0.014	0.12	2.3	0.0064	0.0025	0.47	1.6	1.3	1.1	0.050	3.8
Small-spotted catshark	Maó	0.014	0.035	0.36	0.0064	0.0025	0.19	0.36	0.50	0.16	0.095	1.1
Surmullet	Maó	0.044	0.040	0.23	0.0064	0.0025	0.16	0.23	0.26	0.073	0.17	0.39
Thornback ray	Ciutadella	0.014	0.0079	0.54	0.039	0.0039	0.20	0.33	0.37	0.17	0.015	1.7
Thornback ray	Maó	0.054	0.028	0.59	0.0064	0.0025	0.25	0.33	0.54	0.25	0.047	2.1
Mean		0.061	0.066	1.2	0.026	0.015	0.25	0.52	0.65	0.27	0.066	1.1
Min		0.014	0.0079	0.15	0.0064	0.0025	0.13	0.20	0.20	0.057	0.013	0.11
Max		0.37	0.40	6.1	0.086	0.15	0.50	1.6	1.8	1.1	0.17	3.8

Table 2 OCs (ng/g wet weight) and Hg (mg/kg wet weight) in seafood and meat from markets of Menorca.

	Site of purchase	**HCB**	**4,4'-DDT**	**4,4'-DDE**	**2,4'-DDE**	**4,4'-DDD**	**PCB118**	**PCB138**	**PCB153**	**PCB180**	**DDT/ DDE**	**Hg**
Shellfish												
Detection Frequencies (%)		43%	86%	100%	57%	57%	100%	100%	100%	100%		100%
Mussel	Maó	0.014	0.15	0.33	0.060	0.029	1.1	1.2	1.8	0.17	0.44	0.068
Scampi	Ciutadella	0.019	0.033	0.71	0.062	0.0089	0.26	0.35	0.40	0.15	0.046	0.74
Scampi	Maó	0.014	0.028	0.29	0.0064	0.0025	0.16	0.22	0.21	0.060	0.097	0.77
Shrimp	Ciutadella	0.014	0.0079	0.17	0.041	0.0083	0.14	0.15	0.12	0.016	0.046	1.8
Shrimp	Maó	0.014	0.028	0.20	0.0064	0.0025	0.18	0.21	0.16	0.045	0.14	2.3
Squid	Ciutadella	0.053	0.055	0.86	0.061	0.014	0.34	0.67	0.77	0.12	0.064	0.30
Squid	Maó	0.049	0.13	0.67	0.0064	0.0025	0.29	0.63	1.0	0.31	0.19	0.36
Mean		0.025	0.062	0.47	0.035	0.0097	0.35	0.48	0.64	0.12	0.15	0.92
Min		0.014	0.0079	0.17	0.0064	0.0025	0.14	0.15	0.12	0.016	0.046	0.068
Max		0.053	0.15	0.86	0.062	0.029	1.1	1.2	1.8	0.31	0.44	2.3
Meat												
Detection Frequencies (%)		40%	60%	20%	100%	100%	100%	100%	100%	ND		40%
Beef	Ciutadella	0.014	0.021	0.045	0.025	0.0039	0.19	0.17	0.11	0.011	0.46	0.00010
Beef	Maó	0.014	0.022	0.045	0.038	0.0085	0.19	0.21	0.13	0.011	0.50	0.093
Chicken	Ciutadella	0.014	0.0079	0.045	0.034	0.0084	0.10	0.13	0.088	0.011	0.18	0.00010
Lamb	Ciutadella	0.052	0.010	0.26	0.032	0.0082	0.12	0.060	0.0871	0.011	0.039	0.00010
Lamb	Maó	0.048	0.0079	0.045	0.027	0.0092	0.14	0.14	0.11	0.011	0.18	0.11
Mean		0.028	0.014	0.089	0.032	0.0076	0.15	0.14	0.10	0.011	0.27	0.040
Min		0.014	0.0079	0.045	0.025	0.0039	0.10	0.060	0.087	0.011	0.039	0.00010
Max		0.052	0.022	0.26	0.038	0.0092	0.19	0.21	0.13	0.011	0.50	0.11

Table 3 OCs (ng/g wet weight) and Hg (mg/kg wet weight) in fruit, vegetables, cheese and eggs from Maó (Menorca).

	HCB	**4,4'-DDT**	**4,4'-DDE**	**2,4'-DDE**	**4,4'-DDD**	**PCB118**	**PCB138**	**PCB153**	**PCB180**	**DDT/DDE**	**Hg**
Fruit											
Detection Frequencies (%)	ND	ND	ND	100%	100%	100%	100%	100%	ND		ND
Apple	0.014	0.0079	0.045	0.032	0.0039	0.12	0.13	0.041	0.011	0.18	0.00010
Apple	0.014	0.0079	0.045	0.039	0.0039	0.12	0.13	0.041	0.011	0.18	0.00010
Apple	0.014	0.0079	0.045	0.036	0.0039	0.12	0.13	0.041	0.011	0.18	0.00010
Apple	0.014	0.0079	0.045	0.041	0.0039	0.10	0.060	0.041	0.011	0.18	0.00010
Mean	0.014	0.0079	0.045	0.037	0.0039	0.12	0.11	0.041	0.011	0.18	0.00010
Min	0.014	0.0079	0.045	0.032	0.0039	0.10	0.060	0.041	0.011	0.18	0.00010
Max	0.014	0.0079	0.045	0.041	0.0039	0.12	0.13	0.041	0.011	0.18	0.00010
Vegetables											
Detection Frequencies (%)	ND	ND	ND	100%	100%	100%	100%	100%	50%		ND
Courgette	0.014	0.0079	0.045	0.065	0.0039	0.17	0.22	0.15	0.011	0.18	0.00010
Green bean	0.014	0.0079	0.045	0.052	0.0093	0.13	0.22	0.13	0.016	0.18	0.00010
Mean	0.014	0.0079	0.045	0.058	0.0066	0.15	0.22	0.14	0.014	0.18	0.00010
Min	0.014	0.0079	0.045	0.052	0.0039	0.13	0.22	0.13	0.011	0.18	0.00010
Max	0.014	0.0079	0.045	0.065	0.0093	0.17	0.22	0.15	0.016	0.18	0.00010
Cheese											
Detection Frequencies (%)	ND	100%	ND	ND	ND	67%	100%	100%	100%		100%
Semicured	0.014	0.037	0.045	0.0064	0.0025	0.12	0.30	0.24	0.094	0.82	0.239
Semicured	0.014	0.031	0.045	0.0064	0.0025	0.16	0.22	0.19	0.064	0.68	0.154
Semicured 15% fat	0.014	0.024	0.045	0.0064	0.0025	0.04	0.21	0.15	0.068	0.53	0.0966
Mean	0.014	0.030	0.045	0.0064	0.0025	0.10	0.25	0.19	0.075	0.68	0.163
Min	0.014	0.024	0.045	0.0064	0.0025	0.04	0.21	0.15	0.064	0.53	0.0966
Max	0.014	0.037	0.045	0.0064	0.0025	0.16	0.30	0.24	0.094	0.82	0.239
Eggs											
Detection Frequencies (%)	ND	100%	ND	ND	ND	50%	50%	50%	50%		ND
Chicken eggs	0.014	0.054	0.045	0.0064	0.0025	0.20	0.19	0.24	0.016	1.2	0.00010
Chicken eggs	0.014	0.031	0.045	0.0064	0.0025	0.040	0.048	0.033	0.011	0.70	0.00010
Mean	0.014	0.043	0.045	0.0064	0.0025	0.12	0.12	0.14	0.014	0.95	0.00010
Min	0.014	0.031	0.045	0.0064	0.0025	0.040	0.048	0.033	0.011	0.70	0.00010
Max	0.014	0.054	0.045	0.0064	0.0025	0.20	0.19	0.24	0.016	1.2	0.00010

Table 4. Arithmetic mean concentrations of OCs in food items of Menorca and in other European sites (ng/g ww).

	Fish and seafood	Meat	Fruit	Vegetables	Cheese and dairy products	Eggs	Location	Reference
HCB	0.053	0.028	0.014	0.014	0.014	0.014		*Present study*
	0.33	−	−	−	−	−	Canary Islands	Rodríguez-Hernández et al.,2016
	0.021	−	−	−	−	−	Croatia	Kljakovic-Gaspic et al., 2015
	0.45	0.090	−	−	0.070	0.040	Sweden	A. Törnkvist et al., 2011
	0.22	−	0.005	0.14	−	−	Catalonia	Martí-Cid et al., 2010
	0.56	0.82	−	0.020	0.603	0.38	Russia	Polder et al., 2010
	0.28	0.17	0.00093	0.0056	1.7	0.18	Catalonia	Falcó et al., 2004
∑HCHs	0.038	0.033	0.035	0.033	0.033	0.033		*Present study*
	0.13	−	−	−	−	−	Canary Islands	Rodríguez-Hernández et al., 2016
	0.32	−	−	−	−	−	Croatia	Kljakovic-Gaspic et al., 2015
	0.23	0.040	−	−	0.025	0.015	Sweden	Törnkvist et al., 2011
	0.20	−	0.004	0.16	−	−	Catalonia	Martí-Cid et al., 2010
	0.28	1.1	−	0.070	1.3	0.90	Russia	Polder et al., 2010
∑DDTs	1.1	0.15	0.098	0.12	0.088	0.10		*Present study*
	0.77	−	−	−	−	−	Canary Islands	Rodríguez-Hernández et al., 2016
	2.8	−	−	−	−	−	Croatia	Kljakovic-Gaspic et al., 2015
	3.3	0.24	−	−	0.16	0.10	Sweden	Törnkvist et al., 2011
	7.5	−	0.069	2.8	−	−	Catalonia	Martí-Cid et al., 2010
	6.4	11	−	0.11	0.81	9.7	Russia	Polder et al., 2010
∑PCBs	1.7	0.43	0.30	0.54	0.63	0.40		*Present study*
	1.3	−	−	−	−	−	Canary Islands	Rodríguez-Hernández et al., 2016
	4.3	−	−	−	−	−	Austria	Mihats et al., 2015
	11	−	−	−	−	−	Croatia	Kljakovic-Gaspic et al., 2015
	5.1	0.33	−	−	0.10	0.15	Sweden	Törnkvist et al., 2011
	11	−	0.036	0.21	−		Catalonia	Martí-Cid et al., 2010
	7.4	2.9	−	0.10	2.1	5.2	Russia	Polder et al., 2010
	12	0.37	0.0045	0.021	0.67	0.47	Catalonia	Llobet et al., 2003a

Table 5 Mean values of the daily intake of each food group by 4-year old children population from Menorca (ng contaminant/(infant·day (± standard deviation)) (average body weight = 18.5 kg).

Consumption rate	**Fish**	**Shellfish**	**Meat**	**Fruit**	**Vegetables**	**Cheese**	**Eggs**	**EWI[a]**	**EDI[b]**
	(20.2 g/day)	(5.30 g/day)	(13.7 g/day)	(107 g/day)	(30 g/day)	(7.82 g/day)	(15.1 g/day)	(µg/(kg bw·week)	(ng/(kg bw·d)
HCB	1.2 ± 1.9	0.13 ± 0.095	0.39 ± 0.27	1.5 ± 0.0	0.40 ± 0.0	0.11 ± 0.0	0.20 ± 0.0	0.0015	0.21
∑HCHs	0.75 ± 0.26	0.21 ± 0.088	0.45 ± 0.00	3.7 ± 0.36	0.97 ± 0.0	0.26 ± 0.0	0.50 ± 0.0	0.0026	0.37
∑DDTs	26 ± 32	3.1 ± 1.7	2.0 ± 1.3	10 ± 0.41	3.6 ± 0.18	0.69 ± 0.050	1.5 ± 0.24	0.018	2.5
∑PCBs	34 ± 21	8.7 ± 7.2	5.9 ± 1.5	32 ± 4. 8	16 ± 0.85	5.0 ± 1.1	6.1 ± 5.5	0.041	5.8
Hg	22000 ± 18000	4900 ±4500	550 ± 750	11 ± 0.0	3.0 ± 0.0	1300 ± 560	1.5 ± 0.0	11	1500

[a]EWI: Estimated weekly intake (EFSA, 2012). [b]EDI: Estimated daily intakes (JMPR, 2000).

Table 6. Human biomonitoring data from 4-year old children from Menorca population: OCs in serum (n=285; expressed in ng/ml) and Hg in hair (n=302; expressed in ug/g).

	Geometric Mean	Min	Max
HCB	0.32	0.030	4.5
4,4’-DDT	0.069	0.046	0.66
4,4’-DDE	0.87	0.041	43.9
PCB118	0.091	0.026	1.8
PCB-138	0.17	0.037	8.7
PCB-153	0.25	0.022	10.9
PCB-180	0.12	0.0091	7.2
Hg	0.99	0.040	10

Figure 1 Map of the Menorca Island.

Figure 2 Relative contributions of HCB, ∑HCHs, ∑DDTs, ∑PCBs and Hg to ∑PP in the food items from Menorca.

Figure 3 Contribution from each food group to the total dietary intake of HCB, ∑HCHs, ∑DDTs, ∑PCBs and Hg in the children living in Menorca.

Figure 4 Associations between dietary intakes of selected items and serum OC and hair Hg concentrations of four year-old children. Bars represent the 95% confidence intervals of each beta-coefficient. The models were adjusted for child’s sex, place of birth, number of siblings, breastfeeding, maternal age, maternal and paternal occupation and educational levels.