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An assessment of contaminant concentrations in toothed whale species of the NW Iberian Peninsula: Part I. Persistent organic pollutants

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ABSTRACT

Concentrations and patterns of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in the blubber of the five most common toothed whales off the Northwest Iberian Peninsula (NWIP), specifically common dolphin, long-finned pilot whale, harbour porpoise, striped dolphin and bottlenose dolphin, were investigated. The study revealed that differences in PCB and PBDE concentrations among the species are highly dependent on age and sex but also on ecological factors such as trophic level, prey type or habitat. Of the five species studied, bottlenose dolphin and harbour porpoise showed the greatest concentrations of PCBs. Both species exceed the toxic threshold of $17 \mu\text{g g}^{-1}$ lipid weight (PCBs Aroclor equivalent) for health effects on marine mammals, for 100% and 75% of the individuals tested, respectively. Overall, the PCB and PBDE levels observed in the NWIP toothed whales were of the same order of magnitude or lower than those reported by previous studies in areas of the NE Atlantic. However, they are often higher than those for toothed whales from the southern Atlantic and Pacific Ocean.

Keywords: PCBs; PBDEs; bioaccumulation; patterns; toothed whales; Northwest Iberian Peninsula

1. Introduction

Human activities in marine and coastal environments have intensified since the 1950s. Furthermore, reliance of human populations on coastal areas for urban development and exploitation of marine resources is predicted to keep increasing in the near future. The Northwest Iberian Peninsula (NWIP), situated at the northern limit of the NW African upwelling system (Figueiras et al., 2002), is a good example of such processes. Over the last fifty to sixty years, industrial development and an increase in other human activities in the area have increased the pressures on the marine environment. In this context, and to realise the ambition of clean, productive and biologically diverse seas, the European Community developed the Marine Strategy Framework Directive (MSFD, Directive, 2008/56/EC of the European Parliament and of the Council of 17 June 2008) the main objective of which is to deliver “Good Environmental Status” of European marine ecosystems by 2020. To achieve this, better knowledge of the contamination status of marine populations is needed, specifically in connection with both Descriptor 8 and Descriptor 9 of the MSFD.

The persistent organic pollutants (POPs), including polychlorinated biphenyls (PCBs) and pesticides (e.g. dichlorodiphenyltrichloroethane, DDT) are among the primary pollutants of concern in marine ecosystems cited on the OSPAR list of Chemicals for Priority Action (OSPAR, 2010); they are lipophilic synthetic organic compounds that have been produced for industrial and agricultural purposes since the 1940s, or are by-products of other industrial processes developed over a similar period of time. Although their production has been banned since the end of the 1970s, PCBs can still be found in wildlife and other environmental components e.g. sediments (OSPAR, 2010). Other classes of organic chemicals are also of concern nowadays, notably the brominated diphenyl ether formulations (PBDEs) (de Boer et al., 1998) and the hexabromocyclododecanes (HBCD) another brominated flame retardant (e.g. Zegers et al., 2005). Marine mammals, as long-lived apex predators, are at risk from these toxic compounds, since they have a high bioaccumulation potential and biomagnify through food webs (Aguilar et al., 1999). Due to their lipophilic nature, POPs reach their highest concentrations in fatty tissues and, particularly, in the hypodermic fat or blubber. Compared to most terrestrial mammals, marine mammals appear to have a lower capacity to metabolize and excrete lipophilic organochlorine compounds (Tanabe et al., 1988; Duinker et al., 1989; Boon et al., 1992). This capacity is lower in toothed whales than in pinnipeds (seals and sea lions) (Tanabe et al., 1988), which makes them especially vulnerable to POPs. Although information on the actual effect of POPs on the health of marine mammals is scarce (Reijnders et al., 1999), results from laboratory feeding studies and field investigations have

allowed the determination of several threshold values for adverse effects (e.g. Kannan et al., 2000). The concentration of contaminants in marine mammal tissues primarily varies in relation to prey consumption, but is also a function of their specific capacity to transform these compounds to metabolised forms and/or ultimately excrete the native form or the associated metabolites (Aguilar et al., 1999). Other biological factors have also been found to be responsible for variation in POP concentrations in marine mammals. These include body size and composition, nutritive condition, age, sex, health status, duration of lactation, transfer from mother to offspring during both pregnancy and lactation (Aguilar et al., 1999). Thus, since the uptake of contaminants in marine mammals depends on the diet, feeding habitat and biological factors, any interpretation of concentrations or comparison between species would be incomplete without considering as many of the factors as possible.

For many years, the concentration of contaminants in the NWIP has been routinely monitored through the analysis of samples of sediments, seawater and commercial species such as shellfish (e.g. Carro et al., 2002; Prego et al., 2003). Potentially toxic substances have also occasionally been investigated in marine mammals since the 1980s, as part of the European funded BIO CET project (Zegers et al., 2005; Pierce et al., 2008; Murphy et al., 2010) among others (Borrell et al., 2001, 2006; Tornero et al., 2006), although to a lesser extent than in other marine organisms from this area.

The overall objective of this study is to assess the contamination status of the five most common marine mammals in the NWIP: the common dolphin (*Delphinus delphis*), the long-finned pilot whale (*Globicephala melas*), the harbour porpoise (*Phocoena phocoena*), the striped dolphin (*Stenella coeruleoalba*) and the bottlenose dolphin (*Tursiops truncatus*). This paper constitutes the first of a two part study. In this first part we report on the PCB and PBDE concentrations and patterns in these species, and evaluate their contamination status in comparison with threshold values for health effects on marine mammals as well as making comparisons with concentrations found in other geographical areas. In Part II of this study, that will be subsequently reported (Méndez-Fernandez et al., submitted), we investigate the concentrations of trace elements, which is another group of potential contaminants in the NWIP, in the context of biological and ecological factors.

2. Materials and methods

2.1. Sampling and study area

Sampling was carried out in the NWIP, from the northern limit of the Galician coast in Spain (43°3'N, 7°2'W) to Nazaré on the Portuguese coast (39°36'N, 9°3'W) (Fig. 1). Experienced

members of the Spanish (*Coordinadora para o Estudo dos Mamíferos Mariños*, CEMMA) and Portuguese (*Sociedade Portuguesa de Vida Salvagem*, SPVS) stranding networks have been attending stranded and by-caught cetaceans for over twenty years and over ten years, respectively. Animals were identified to species, measured, sexed and, if the decomposition state of the carcass allowed, full necropsies were performed and samples collected whenever possible. All procedures followed the standard protocol defined by the European Cetacean Society (ECS), as did the coding of decomposition state and condition (Kuiken and Garcia Hartmann, 1991).

A total of 172 stranded and by-caught individuals was selected for this study, covering five toothed whale species (common dolphin, $n = 114$; long-finned pilot whale, $n = 9$; harbour porpoise, $n = 19$; striped dolphin, $n = 21$ and bottlenose dolphin, $n = 9$) over the period 2004 to 2008. The common dolphin is the cetacean species stranded in the greatest numbers; this is believed to reflect the relatively high abundance in the area (Santos et al., 2013c) and the large number of individuals being by-caught in NWIP fisheries (López et al., 2002, 2003). The animals recovered in a “fresh” state (a score of 1 to 3 from the ECS protocol, i.e. originally stranded alive, freshly dead or mildly decomposed) were selected. Teeth were extracted for age determination, gonads collected for determination of reproductive status and blubber samples for POP analyses. All blubber samples were taken from the left side in front of the dorsal fin. Samples were entire vertical cross-sections of the blubber so as to prevent any possible effects of stratification of the blubber. The samples were wrapped separately in aluminium foil and after the necropsies, all samples were stored frozen at -20°C until required for analysis.

Analysis of POPs is costly and the present study was budget-limited. As such, effort was focused on the best sample sets (i.e. individuals for which most data on other variables were available). Thus, in this part of the study 120 blubber samples, out of a possible 172, were analysed for PCBs (common dolphin, $n = 81$; long-finned pilot whale, $n = 3$; harbour porpoise, $n = 12$; striped dolphin, $n = 15$ and bottlenose dolphin, $n = 7$) and 20 for PBDEs (common dolphin, $n = 19$; harbour porpoise, $n = 1$).

2.2. Age estimation and reproductive status

Age was estimated by analysing growth layer groups (GLGs) in the dentine and cementum of teeth, following adapted methods based on Lockyer (1993) and Hohn and Lockyer (1995). Teeth were decalcified and sectioned at $25\ \mu\text{m}$ using a cryostat. The most central and complete sections (including the whole pulp cavity) were selected from each tooth, stained with Mayer’s haematoxylin (modified by Grue) and ‘blued’ in a weak ammonia solution,

mounted on glass slides and allowed to dry. GLGs were counted under a binocular microscope. All readings were made blind (without access to individual biological data), and replicate counts were made by two readers. If the age estimates obtained by the two readers differed by more than 1 year, readings were repeated. If the increments were difficult to count, both readers discussed the interpretation and either reached an agreed age or judged the tooth to be unreadable.

Female reproductive status

Females were examined at the time of necropsy for evidence of pregnancy and/or lactation. Formalin-fixed ovaries were weighed, measured and sectioned at 2 mm intervals along the broad ligament. The presence of mature follicles, *corpora lutea* and *corpora albicantia* were recorded. Microscopic examination was conducted to confirm macroscopic findings. Portions of ovary were paraffin-embedded and sectioned at 5-8 μm , sections were then stained with Mayer's hematoxylin and eosin and examined by microscopy. Females were classified as adults (mature) and juveniles (immature) based on the presence/absence of ovarian structures.

Male reproductive status

Testes with attached epididymis were weighed, and a central cross-section was formalin-fixed. Standard histological analysis of paraffin-embedded sections was conducted. Sections were cut at 5-8 μm and stained with Mayer's haematoxylin and eosin. Microscopic analysis was conducted to measure the diameter of seminiferous tubules and to record cell activity (sertoli cells, interstitial tissue, and germinal cells such as spermatogonia, spermatocytes, spermatids and spermatozoa). Males were classified as adults (mature) and juveniles (immature) based on seminiferous tubule diameter and cell activity.

2.3. Determination of persistent organic pollutants

2.3.1. Lipid determination

The total lipid content was determined using a modified Folch et al. (1975) method. The samples were weighed (100-150 mg), homogenized, and then extracted three times with a mixture of chloroform:methanol (1:2, 2:1 and 4:1, v/v). A volume of 6.5 mL of 1% sodium chloride was added and the mixture separated into two phases. The lower layer, containing the lipid and lipophilic compounds, was collected and traces of water removed by addition of dry sodium sulphate. These extracts were shaken and stored at 4°C for 1h. Centrifugation for 10 minutes at 3000 g was used to separate the organic extract from the particulate material and the solvent was removed under a stream of nitrogen in a water bath at 40°C. When all solvent had evaporated, the weight of residue was determined and the lipid content calculated by gravimetry.

2.3.2. Extraction and clean-up for PCB and PBDE analyses

Samples were extracted by Pressurised Liquid Extraction (PLE) (Walsham et al., 2006). For each extraction, approximately 200 mg of blubber was cut (in vertical sections), homogenised, and mixed with sodium sulphate (~ 20 g). This mixture was spiked with appropriate internal standards (PCBs by GC-MS: ^{13}C -CB28, ^{13}C -CB52, ^{13}C -CB101, ^{13}C -CB153, ^{13}C -CB138, ^{13}C -CB156, ^{13}C -CB180, ^{13}C -CB189, ^{13}C -CB194 and ^{13}C -CB209; PBDEs: FBDE160¹). Samples were then refrigerated overnight before being ground to a fine powder using a mortar and pestle. Solvent-washed PLE cells (100 mL) were packed as follows: solvent-washed filter paper, pre-washed sodium sulphate (10 g), 5% deactivated alumina (30 g), solvent-washed filter paper and the samples/sodium sulphate mixture prepared as above.

Samples were extracted by PLE using an ASE 300 (Dionex Ltd., Camberley, Surrey, UK) under elevated temperature (100°C) and pressure (1500 psi). Five minutes of heating was followed by 2 x 5 min static cycles. The cell flush was 50% total cell volume (i.e. 25% of the cell volume for each flush = 25 mL per flush) with a 120 s purge (using nitrogen) at the end of each sample extraction. The extraction solvent was *iso*-hexane.

Special precautions were required when analyzing PBDEs due to their sensitivity to UV light. Specifically, incoming light was minimized in the laboratory by placing UV filters over the windows.

Following PLE, the extract for PCB analyses was concentrated by Syncore Analyst R-12 (fitted with flushback module) (Buchi UK Ltd, Oldham, UK) to ~ 0.5 mL and passed through silica columns, before transferring with washing to amber glass GC vials. For 20 samples the extract was split in two, before being concentrated, one half for PBDE analysis and the other one for PCB analysis. The concentrated extracts were analysed for PCBs by Gas Chromatography Electron Impact Mass Spectrometry (GC-EIMS) and for PBDEs by Gas Chromatography Electron Capture Negative Ionization Mass Spectrometry (GC-ECNIMS).

2.3.3. Determination of PCBs by GC-EIMS

The concentrations of 32 PCB congeners (IUPAC PCB numbers 28, 31, 52, 49, 44, 74, 70, 101, 99, 97, 110, 123, 118, 105, 114, 149, 153, 132, 137, 138, 158, 128, 156, 167, 157, 187, 183, 180, 170, 189, 194, 209) were determined by GC-EIMS using a HP6890 series gas chromatograph interfaced with an HP5975 Mass Selective Detector, fitted with a cool on-column injector and a 50 m x 0.22 mm x 25 μm SGE HT-8 column (SGE, Milton Keynes, UK). The initial oven temperature was 80°C, which was held for 1 minute. The temperature was raised by 20°Cmin⁻¹ up to 170°C and held at this temperature for 7.5 minutes. This was followed by a ramp of 3°Cmin⁻¹ up to a final temperature of 290°C which was maintained for

10 minutes. The MSD was set for selective ion monitoring (SIM) with a dwell time of 50 ms. Calibration standards containing all 32 PCB congeners and covering the concentration range 0.6-500 ng mL⁻¹ were analysed in triplicate, and the average response used to compute the calibration curve. Correlation coefficients of at least 0.99 were achieved for all PCBs.

2.3.4. Determination of PBDE by GC-ECNIMS

PBDEs were analysed and the concentrations of nine congeners, specifically BDE 28, 47, 66, 85, 99, 100, 153, 154 and 183, were determined by GC-ECNIMS using an HP6890 Series gas chromatograph interfaced with an HP5973N MSD, fitted with a cool on-column injector.

A Thames Restek STX-500 column (STX-500, 30 m × 0.25 mm i.d., 0.15 µm film thickness, Thames Restek, Buckinghamshire, UK) was utilised, fitted with a Thames Restek Siltek (0.53 mm i.d.) 5 m guard column. The injector temperature was initially 120°C and after 2 min the temperature was elevated by 100°C min⁻¹ up to 300°C at which it was maintained until the end of the run. The carrier gas was helium, set at a constant pressure of 15 psi. Methane was used as the reagent gas at a pressure of 1.6 bar. The transfer line was held at 280°C and the ion source at 150°C. Injections were made at 120°C and the oven temperature held constant for 2 minutes. Thereafter, the temperature was raised by 15°Cmin⁻¹ up to 205°C. This was followed by a ramp of 6°Cmin⁻¹ up to a final temperature of 330°C. The MSD was set for selective ion monitoring with a dwell time of 50 ms. The ions monitored were m/z 78.9 and 80.9 (ions equating to bromine) for all PBDEs.

2.3.5. Quality control

The methods employed were validated by the replicate analysis of standards and samples, and through spiking experiments or analysis of certified reference materials (CRMs). The limits of detection (LODs) were determined through the repeated analysis of a low spiked sample and calculated from 4.65 x SD (standard deviation) of the mean concentration (Cheeseman and Wilson, 1989). LODs were dependent on the sample size. The replicate analysis of standards on separate days gave coefficient of variation (CV%) of ~ 3% for PCBs analysed by GC-EIMS. Recoveries greater than 75% were achieved for PCBs and PBDEs spiked samples and CRMs. Internal quality control procedures incorporated the use of a laboratory reference material (cod liver oil; LRM) for all determinants, and also a CRM for PCBs, in each batch of samples. Procedural blanks were performed with each batch of samples, and the final concentration adjusted accordingly. The data obtained from the LRM were transferred onto NWA Quality Analyst. Thus Shewhart charts were produced with warning and action limits (i.e. ± 2× and ± 3× the standard deviation of the mean, respectively). CRM data were accepted if recoveries were between 70% and 120% of the certified concentration. Quality assurance

was further demonstrated through successful participation in the Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME) Laboratory Performance Studies. Finally, all POP concentrations were normalized to the lipid content (%) of the blubber.

2.4. Data treatment

All data submitted to statistical tests were first checked for normality (Shapiro-Wilk test) and for homogeneity of variances (Bartlett test). Non-parametric tests were applied since the distributions of response variables were found to be non-normal and/or the homogeneity criterion was not satisfied.

Prior to the treatment of the POP concentration data ($\mu\text{g g}^{-1}$ lipid weight), an age-gender classification of the individuals based upon their sexual maturity was carried out for each species. The individuals were then divided into four groups: adult male, adult female, juvenile male and juvenile female. Hence, differences in the sum of the 32 PCB congeners (ΣPCBs) were tested between species and age-gender groups using the Kruskal-Wallis test followed by pairwise comparison tests, with the exception of pilot whales and age-gender groups with less than 2 individuals, which were excluded from the statistical treatment. For the same reason, no statistical test was performed for the sum of the 9 PBDE (ΣPBDEs) congeners.

We also verified the number of animals exceeding the toxic threshold concentration ($17 \mu\text{g g}^{-1}$ lipid wt) for total PCBs determined for adverse health effects in marine mammals (Kannan et al., 2000). Since this value was based on comparison with the main peaks in the commercial PCB mixture Aroclor 1254, the PCB concentrations in the samples cannot be compared directly to this limit and had to be converted. Aroclor equivalent concentrations were estimated from the concentration of the seven ICES PCBs (i.e. CB28, 52, 101, 118, 138, 153 and 180 as recommended by the EU Community Bureau of Reference), by multiplying the sum of the concentrations for the seven ICES PCBs by 3 (i.e. total PCB concentration [as Aroclor 1254] = $3.0 \times$ sum of seven ICES congeners in lipid weight) (Jepson et al., 2005).

The levels of significance for statistical analyses were always set at $\alpha = 0.05$ and analysis were performed using R version 3.0.1 (R Development Core Team, 2010).

3. Results and discussion

3.1. Persistent organic pollutant concentrations and patterns

Persistent organic pollutants enter marine mammals tissues almost exclusively *via* their food and the amounts in tissues vary greatly with intake factors (Aguilar, 1989), i.e. trophic level, prey type, and with the local environmental pollution. Thus, all these factors must be taken

into consideration when interpreting POP concentrations in marine mammals. The five toothed whale species studied here have rather similar trophic levels in the NWIP, ranging from 4.3 to 5.3 (Méndez-Fernandez et al., 2012). However, stomach contents and stable isotope analyses revealed that these species feed on different prey types and forage in different habitats (Méndez-Fernandez et al., 2012, 2013; Santos et al., 2007, 2013a, 2013b, 2013c). In addition, marine mammal organochlorine loads tend to increase with age during the juvenile stage of both genders, because the uptake of pollutants usually exceeds metabolism and excretion. In adult males, this pattern continues throughout their life, while in adult females, the transfer of pollutants to offspring during gestation and lactation progressively reduces pollutant concentrations with age (e.g. Borrell et al., 1996; Covaci et al., 2002; Wolkers et al., 2004). In this study, PCBs were detected in all five species across age-gender groups and significant differences were found for Σ PCB mean concentrations ($\mu\text{g g}^{-1}$ lipid wt.) among them (Kruskal-Wallis, $H = 16.13$, $P < 0.01$), with bottlenose dolphin showing the highest Σ PCB concentrations followed by harbour porpoise. Pilot whale, common dolphins and striped dolphins exhibited very similar mean age/gender-specific PCB concentrations (Table 1), and as expected the adult males (not represented in bottlenose dolphins and pilot whales) exhibited the highest Σ PCB concentrations (Table 1). However, there was no significant between-species variation in their concentrations (Kruskal-Wallis, $H = 0.45$, $P = 0.5$). In contrast, for adult females (not represented in bottlenose dolphins) there were significantly higher concentrations in harbour porpoise than in common and striped dolphin (Post-hoc test, $P < 0.05$; Table 1). Significant differences were found between juvenile bottlenose dolphins and juveniles of all the other species (Post-hoc, all $P < 0.05$). The high concentrations found in juvenile bottlenose dolphin and adult harbour porpoise are in accordance with their coastal habitat, their proximity to areas with the highest anthropogenic impact from contaminants, and their mainly fish-feeding dietary habits in the NWIP (López et al., 2004; Pierce et al., 2010; Méndez-Fernandez et al., 2012). In addition, both species are more frequently seen in the southern part of the study area (López et al., 2004; Pierce et al., 2010), which is more populated and industrialized than the northern part. Thus, these ecological factors may be one of the reasons for the high Σ PCB concentrations found for these two species in the NWIP waters. Although pilot whales and striped dolphins are observed in neritic habitats in the NWIP, especially for feeding (Méndez-Fernandez et al., 2013), these cetacean species are mainly associated with oceanic habitats, remote from land-based sources of contamination (López et al., 2004; Pierce et al., 2010; Méndez-Fernandez et al., 2012). In addition, pilot whales mainly feed on cephalopods that generally, and depending

on species, contain lower concentrations of PCBs than fish from a similar geographic region (e.g. Storelli et al., 2008).

Their different ecological feeding patterns are also reflected in the relative contribution of the distinct PCB congeners (Fig. 2). The mainly cephalopod feeders and oceanic species, the pilot whale and the striped dolphin, had a higher proportion of less chlorinated congeners (i.e. tri-, tetra- and penta-chlorobiphenyls) than the other three species, which had a greater proportion of highly chlorinated PCB congeners such as hexa- and hepta-chlorobiphenyls. This finding can be explained by the more efficient long-range transport of low-chlorinated PCBs through both atmosphere and water (Beyer et al., 2000). Nevertheless, all species showed a predominance of PCBs containing 5 or more chlorines. Hexachlorobiphenyls (56.3%) accounting for the highest percentage across all 5 cetaceans, followed by heptachlorobiphenyls (26%) and pentachlorobiphenyls (12.7%) (Fig. 2). The predominant pentachlorinated congeners were CB153, followed by CB138, 187 and 180. These results are in accordance with the patterns observed in cetacean species from different regions (e.g. Covaci et al., 2002; Wafo et al., 2005; Lailson-Brito et al., 2012; Leonel et al., 2012).

Intra-specific differences were only found for common dolphin (Kruskal-Wallis, $H = 16.69$, $P < 0.05$), which was the best represented species ($n = 81$; Table 1). Specifically, the adult males showed significantly higher concentrations than females (38.9 ± 22.2 and $8.7 \pm 8.1 \mu\text{g g}^{-1}$ lipid wt, respectively; Table 1). This suggests that adult females have a different accumulation pattern, which is consistent with the well-described transfer of POPs from mother to offspring during gestation and lactation discussed above.

The PBDE concentrations were almost 10 times lower than PCBs in blubber for common dolphin and harbour porpoise and for all age-gender groups (Table 2). This agrees with several previous studies on toothed whales (Nyman et al., 2002; Pierce et al., 2008; Yogui et al., 2011; Dorneles et al., 2010; Leonel et al., 2012). Of the 9 congeners analysed, congener BDE85 was not detected in any sample and congeners BDE28 and BDE183 were detected only in juvenile female and in adult male, adult female and juvenile male of common dolphin, respectively (Table 2). On average, BDE47 showed the highest concentrations in both species and in all age-gender groups, following the same pattern of variation as Σ PBDE concentrations (Table 2). Similar profiles were reported in other cetacean species from around the world (e.g. Weijs et al., 2009; Dorneles et al., 2010; Leonel et al., 2012).

The concentrations of Σ PBDE of the juvenile males of both species were similar, being slightly higher for harbour porpoise with $0.57 \mu\text{g g}^{-1}$ lipid wt. ($n = 1$) than for common dolphin with $0.31 \pm 0.18 \mu\text{g g}^{-1}$ lipid wt. ($n = 11$). For common dolphin, adult males were

more contaminated than adult females with the adult male group having the highest ($0.71 \pm 0.17 \mu\text{g g}^{-1}$ lipid wt.) and the adult female the lowest ($0.08 \mu\text{g g}^{-1}$ lipid wt.) mean concentrations. Similar to PCBs, this result supports the hypothesis that adult female animals reduce their PBDE concentrations through gestation and lactation. Besides differences of Σ PBDE concentrations found between juvenile males of both species, their patterns were also slightly different, namely $\text{BDE47} > \text{BDE100} > \text{BDE154} > \text{BDE99} > \text{BDE153} > \text{BDE66} > \text{BDE183}$ for common dolphin and $\text{BDE47} > \text{BDE100} > \text{BDE99} > \text{BDE154} > \text{BDE153} > \text{BDE66}$ (BDE183 was not detected) for harbour porpoise. The profiles found in the different age-gender groups of common dolphin were similar. The pattern reported in harbour porpoise is similar with those from other regions and from other cetacean species (e.g. Boon et al., 2002; Weijs et al., 2009; Leonel et al., 2012). Thus, in general, the Σ PBDE concentrations were higher in common dolphin than in harbour porpoise and congeners BDE28 and BDE 183 were detected only in common dolphins', indicating that common dolphin has difficulties with metabolizing PBDEs. However, we must consider these results as only indicative due to the low number of samples analysed for PBDEs.

3.2. Geographic comparison

The PCB concentrations of Iberian common dolphins from this study are in the range reported by previous published studies in the NWIP, in France and also in England. However these concentrations are much higher than in common dolphins from Ireland and those from the South Atlantic Ocean and from the Pacific Ocean (Table 3). It is important to note that the PCB concentration in the Irish common dolphin ($2.8 \mu\text{g g}^{-1}$ lipid weight) is the mean value of only 5 PCB congeners: CB118, 138, 153, 180 and 170. In the present study the sum of these 5 PCB congeners is $10.3 \mu\text{g g}^{-1}$ lipid weight. A different pattern was found when a comparison was made with common dolphins from the Mediterranean Sea and the east coast of the USA (NW Atlantic Ocean); common dolphins from these areas were more highly contaminated with PCBs than those from the NWIP (Table 3). The PBDE concentrations from male and female common dolphins in the NE Atlantic Ocean are lower than in other areas, especially when compared to dolphins from Korea in the Pacific Ocean.

Despite the small sample size for pilot whale, the PCB concentrations found in the NWIP are lower than in the rest of the NE Atlantic and Ligurian Sea, with the exception of one male specimen from Ireland. However, in this study carried out by Troisi et al. (1998) only 5 PCB congeners were analysed and the sum of these is also lower in Iberian pilot whales than in Ireland (8.3 and $10.2 \mu\text{g g}^{-1}$ lipid weight, respectively). In contrast, when the PCB

concentrations in pilot whales from the NW Atlantic and the Pacific Ocean are compared with the data from this study, with the exception of females from Massachusetts (Tilbury et al., 1999), the Iberian pilot whales contain higher concentrations (Table 3).

Harbour porpoise and bottlenose dolphin contained the highest PCB concentrations among the Iberian toothed whales. Comparing with other areas across the world we observed that both species have in general higher concentrations than seen in conspecifics from the Atlantic, Pacific and Indian Ocean. There were exceptions; the North, Baltic and Norwegian Seas for harbour porpoise and the Mediterranean Sea for bottlenose dolphin. This indicates that those seas are highly contaminated with PCBs, as has been demonstrated by previously published studies (Pierce et al., 2008; Weijs et al., 2009, 2010; Fossi et al., 2013). Borrell et al. (2006) reported mean PCB concentrations in male and female bottlenose dolphins stranded in the South of the Iberian Peninsula that were two times higher than in the present study (Table 3). These high values may be a result of the proximity of the study area used by Borrell et al to the Mediterranean Sea. PBDE concentrations in the only male harbour porpoise analysed in the present study were lower than for males from other areas and, as for the PCBs, North Sea porpoises.

Striped dolphins presented the lowest PCB concentrations among the Iberian toothed whales but also when we compare with specimens from other areas of the Northern Hemisphere (Table 3). Only one specimen stranded on the coast of England showed similar concentrations to those of the Iberian striped dolphins (Morris et al., 1989). Regarding data from the Southern Hemisphere, the concentrations found in the present study are higher than those reported in dolphins stranded on the Brazilian coast. This difference in PCB levels between the Northern and Southern Hemisphere is common to all five species studied here, reflecting the highly industrialized development of the Northern Hemisphere. PBDE concentrations were not analysed in striped dolphins from the NWIP. However, previously published studies showed similar values across different areas of the Atlantic and Pacific Ocean, with the exception of one female stranded on the coast of Japan which contained only $0.08 \mu\text{g g}^{-1}$ lipid wt of PBDEs and also only $3.2 \mu\text{g g}^{-1}$ lipid wt of PCBs (Isobe et al., 2009).

3.2. Toxicological aspects

Reliable toxicity data for predatory marine mammals are scarce. Instead, threshold levels are often extrapolated from terrestrial species, since the effects of toxic compounds cannot be tested in free-living animals because such experimental manipulations raise ethical considerations (Das et al., 2003). Thus, although the validity of these extrapolations could be

questionable, they can be justified by the current lack of better data. The harmful consequences of the bioaccumulation of POPs in marine mammals include depression of the immune system (e.g. Ross, 1995; de swart et al., 1996), increased risk of infection and reproductive failure. Specifically, a total PCB concentration of $17 \mu\text{g g}^{-1}$ lipid wt has been reported as a threshold level above which there are health effects in mammals (Kannan et al., 2000). This threshold was obtained in laboratory mammals (seals, European otters and mink) fed with field food items. In this study the threshold value was frequently exceeded for all species, often with more than 50% of the individuals (except pilot whale). However, this value was exceeded by all the bottlenose dolphins and 75% of the harbour porpoises. This result is even more important when it is considered in association with the previous study carried out in the Northeast Atlantic, including samples from the NWIP. This showed that almost half of the harbour porpoises for which cause of death was determined as being from pathological causes, had significantly higher concentrations of all classes of POPs than animals dying from other causes (Pierce et al., 2008). PBDE concentrations measured in common dolphins and harbour porpoises from this study are at least 10 times lower than those of PCBs, being slightly higher for harbour porpoise than common dolphin. There is still no information on a toxic threshold for PBDEs in marine mammals, although experimental exposure investigations revealed that PBDEs induce a wide variety of disorders in mammals (e.g. cancer, reproductive and developmental toxicity, endocrine disruption and central nervous system effects; Hana et al., 2004). As such it is not possible to say whether or not such concentrations are likely to impact on the toothed whales. However, what needs to be considered is that the PBDEs are present and have augmented the overall concentration of POPs in these whales. In future studies consideration should be given to the possible cumulative impacts of the range of contaminants found in these marine animals.

4. Conclusions

The inter-species differences found in the present study, covering the accumulation and patterns of POPs, illustrates the important influence of both the biological and ecological factors of each species in determining the contaminant loading. Overall, the POP concentrations observed in toothed whales from the NWIP were the same order of magnitude or lower than those reported by previous studies in areas of the NE Atlantic. However, they were higher than those of toothed whales from the southern Atlantic Ocean and Pacific Ocean.

The bottlenose dolphin and harbour porpoise are recorded in the EU Habitats Directive as Species of Special Interest (Directive 92/43/CEE), the protection of which requires the designation of Special Areas of Conservation (SACs) by EU Member States. In the present study both species seem to be the more contaminated with PCBs as shown by their higher concentrations when compared to the other three species. Their PCB concentrations also raise particular concern as they are much higher than the threshold level for PCB concentrations associated with adverse health effects on marine mammals. This study adds to our knowledge of POP concentrations in marine mammals that frequent the NWIP waters, complementing the existing database on these chemical contaminants in this area. However, there remains a lack of information on concentrations of other organochlorine compounds of special concern nowadays and this should be addressed through further studies. This is particularly relevant given that the European scientific community is developing research on large marine vertebrates as indicators of medium and long term marine environmental change for incorporation under the Marine Strategy Framework Directive. A complete database of chemical contaminants in marine mammals of the NWIP is an important step forward.

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Fig 1. Map of the study area including the 100 and 200 m isobaths. The 200 m isobath was taken as the limit for the shelf-break. The black line contouring the coast represents the length of the coastline which constituted the sampling area.

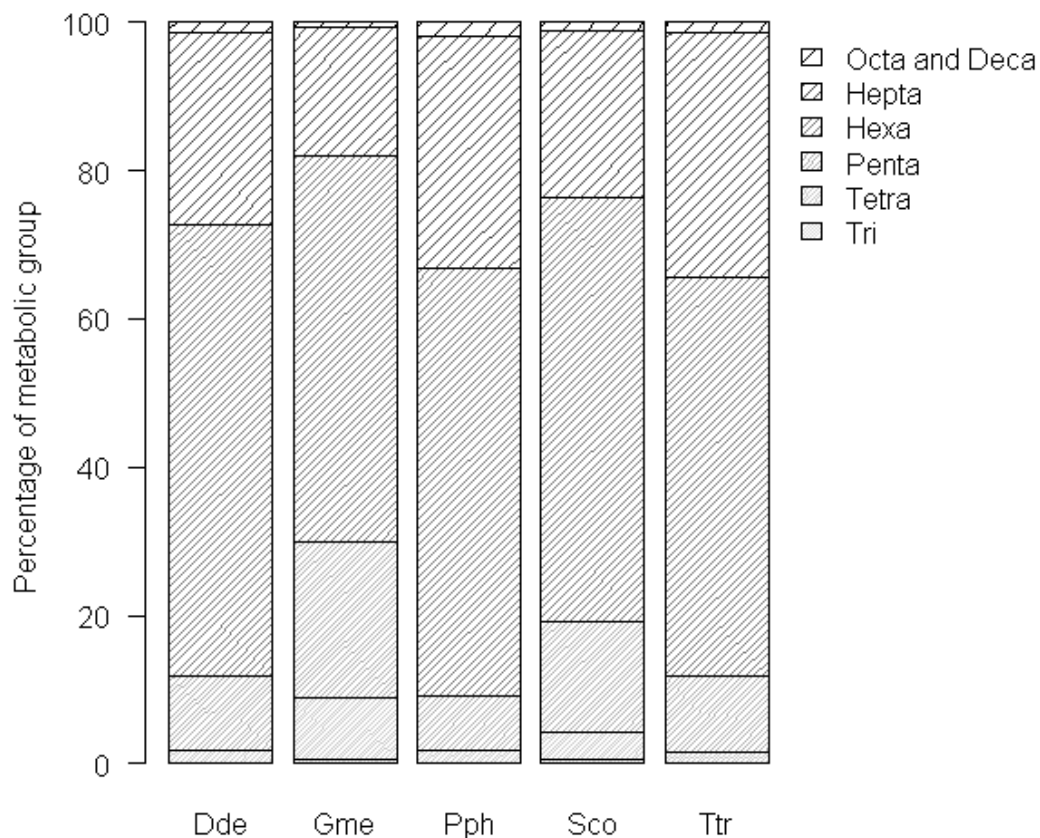


Fig. 2. Relative contribution of PCB congeners, grouped by the number of chlorine atoms in the molecule, to the Σ PCB in blubber of Dde: common dolphin (*Delphinus delphis*), Gme: pilot whale (*Globicephala melas*), Pph: harbour porpoise (*Phocoena phocoena*), Sco: striped dolphin (*Stenella coeruleoalba*) and Ttr: bottlenose dolphin (*Tursiops truncatus*) from the North West Iberian Peninsula.

Table 1. Age-/gender-specific PCB concentrations (mean \pm SD, $\mu\text{g g}^{-1}$ lipid weight), lipid content (%) and age values expressed in years (mean \pm SD) in blubber of common dolphin (*Delphinus delphis*), pilot whale (*Globicephala melas*), harbour porpoise (*Phocoena phocoena*), striped dolphin (*Stenella coeruleoalba*) and bottlenose dolphin (*Tursiops truncatus*) from the North West Iberian Peninsula. AF: adult female, AM: adult male, JF: juvenile female and JM: juvenile male, n = sample size.

Species and age-gender group	n	Age	Lipid content	CB153	ΣPCB^a
<i>Delphinus delphis</i>	81	6.6 \pm 5.4	60.6 \pm 16.2	5.2 \pm 5	17.2 \pm 14.1
AF	11	14.4 \pm 3.1	56.9 \pm 17.5	2.5 \pm 2.5	8.7 \pm 8.1
AM	8	14.3 \pm 3.8	40.5 \pm 11.9	13.1 \pm 8.2	38.9 \pm 22.2
JF	20	3.4 \pm 3.3	66.3 \pm 13.8	3.7 \pm 2.0	13.2 \pm 6.3
JM	42	4.4 \pm 3.1	62.5 \pm 14.1	5 \pm 4.3	16.9 \pm 12.1
<i>Globicephala melas</i>	3	5.2 \pm 5.5	65.2 \pm 10.9	3.6 \pm 4.1	16.2 \pm 19.7
AF	1	11.5	77.3	0.4	2
JF	1	2.0	62.3	2.2	7.9
JM	1	2.0	56	8.3	38.7
<i>Phocoena phocoena</i>	12	7.0 \pm 6.5	77.9 \pm 13.9	6.3 \pm 6.7	20.5 \pm 20.4
AF	3	13.7 \pm 6.2	76.8 \pm 8.1	12.0 \pm 9.7	37.5 \pm 30.8
AM	1	18.0	53.7	16.6 \pm 0	50.8
JF	5	2.8 \pm 1.6	87.0 \pm 13.7	2.9 \pm 0.8	10.8 \pm 2.8
JM	3	3.7 \pm 1.2	71.7 \pm 7.9	2.8 \pm 1	9.4 \pm 3
<i>Stenella coeruleoalba</i>	15	4.9 \pm 5.5	61.1 \pm 19.1	4.2 \pm 5.1	15.7 \pm 18.6
AF	2	14.0	65.6 \pm 8.7	0.3 \pm 0.01	1.8 \pm 0.3
AM	1	15.0	50.1	10.7	36.4
JF	5	3.8 \pm 3.5	65.7 \pm 5.5	2.6 \pm 1.6	9.9 \pm 5.4
JM	7	1.6 \pm 2.0	58.2 \pm 27.6	5.5 \pm 6.6	20.9 \pm 24.4
<i>Tursiops truncatus</i>	7	3.8 \pm 2.0	68.6 \pm 9.3	15.2 \pm 10.1	56.4 \pm 35.2
JF	3	2.3 \pm 1.6	71.8 \pm 0.8	13.3 \pm 8.8	48.9 \pm 30.9
JM	4	4.9 \pm 1.7	66.3 \pm 12.4	16.7 \pm 12.1	62.1 \pm 41.8

^a ΣPCB includes 32 congeners. See Materials and Methods section for full list.

Table 2. Age-/gender-specific PBDE concentrations (mean \pm SD, $\mu\text{g g}^{-1}$ lipid weight), lipid content (%) and age values expressed in years (mean \pm SD) in blubber of common dolphin (*Delphinus delphis*) and harbour porpoise (*Phocoena phocoena*) from the North West Iberian Peninsula. AF: adult female, AM: adult male, JF: juvenile female and JM: juvenile male, n = sample size.

Species/age-gender group	n	Age	Lipid content	BDE28	BDE47	BDE66	BDE100	BDE99	BDE154	BDE153	BDE183	ΣBDE
<i>Delphinus delphis</i>	19	7.4 \pm 5.1	59.9 \pm 13.8	0.01	0.23 \pm 0.19	0.004 \pm 0.003	0.05 \pm 0.04	0.02 \pm 0.01	0.03 \pm 0.01	0.012	0.001 \pm 0.0002	0.34 \pm 0.08
AF	1	16.0	63.4	nd	0.03	0.004	0.01	0.01	0.01	0.008	0.001	0.08
AM	2	15.5 \pm 0.7	42.3	nd	0.44 \pm 0.12	0.011	0.12 \pm 0.45	0.04 \pm 0.001	0.06 \pm 0.003	0.03 \pm 0.009	0.001 \pm 0.00005	0.71 \pm 0.12
JF	5	4.4 \pm 3.0	70.8 \pm 19.7	0.01	0.2 \pm 0.3	0.003 \pm 0.004	0.04 \pm 0.41	0.01 \pm 0.007	0.02 \pm 0.01	0.007 \pm 0.003	nd	0.33 \pm 0.05
JM	11	6.5 \pm 4.2	57.8 \pm 7.8	nd	0.2 \pm 0.13	0.003 \pm 0.001	0.05 \pm 0.03	0.02 \pm 0.01	0.02 \pm 0.008	0.01 \pm 0.004	0.001	0.31 \pm 0.05
<i>Phocoena phocoena</i>	1	3.0	40.3	nd	0.48	0.004	0.08	0.04	0.02	0.009	nd	0.57
JM	1	3.0	40.3	nd	0.48	0.004	0.08	0.04	0.02	0.009	nd	0.57

^a ΣBDE includes 9 congeners. See Material and Methods section for full list.

nd = values were less than the limit of detection (see Materials and Methods)

The congener BDE85 was not detected in any individual analysed.

Table 3. Mean \pm SD of PCB and PBDE concentrations ($\mu\text{g g}^{-1}$ lipid weight) in blubber of the five toothed whale species from all over the world. Sample size “n” of each species by sex and area is in brackets.

Species	Area	Sex	ΣPCB	ΣPBDE	References
<i>Delphinus delphis</i>					
	NE Atlantic Ocean				
	NWIP	M	20.4 \pm 16.1 (50)	0.51 \pm 0.17 (13)	This study
	NWIP	F	11.6 \pm 7.2 (31)	0.205 \pm 0.34 (6)	This study
	Spain	F	10.9 \pm 11.6 (23)	0.42 \pm 0.18 (23)	Pierce et al., 2008
	Spain	M	37.8 \pm 18.9 (33)		Borrell et al., 2001
	Spain	F	23.9 \pm 17.7 (23)		Borrell et al., 2001
	France	F	13.7 \pm 12.7 (36)	0.61 \pm 0.41 (36)	Pierce et al., 2008
	Ireland	M	2.8 ^a		Troisi et al., 1998
	Ireland	F	3.6 \pm 3.4 (11)	0.76 \pm 0.5 (11)	Pierce et al., 2008
	England	F	20.2 \pm 16.7 (43)		Law et al., 2013
	Mediterranean Sea				
	Spain	M	54.3 \pm 22.7 (11)		Borrell et al., 2001
	Spain	F	23.8 \pm 32.8 (11)		Borrell et al., 2001
	NW Atlantic Ocean				
	USA	M	36.5 \pm 4.0 (4)		Kuehl et al., 1991
	SE Atlantic Ocean				
	South Africa	M	5.9 \pm 5.05 (10)		de Kock et al., 1994
	South Africa	F	2.8 \pm 1.7 (7)		de Kock et al., 1994
	SW Atlantic Ocean				
	Brazil*	M	17.0 (1)		Kajiwara et al., 2004
	Brazil*	M	14.6 \pm 15.3 (2)		Lailson-Brito et al., 2012
	Brazil*	F	2.2 \pm 0.7 (2)		Lailson-Brito et al., 2012
	Pacific Ocean				
	NE Australia	F	0.6 (1)		Vetter et al., 2001

Table 3 (Continued)

	New Zealand	M	0.8 ± 0.5 (12)		Stockin et al., 2007
	New Zealand	F	0.14 ± 0.13 (7)		Stockin et al., 2007
	Korea	M	15.0 ± 7.6 (12)	1.7 ± 0.78 (12)	Moon et al., 2010
	Korea	F	15.0 ± 7.8 (10)	1.6 ± 0.68 (10)	Moon et al., 2010
<i>Globicephala melas</i>					
	NE Atlantic Ocean				
	NWIP	M	38.7 (1)		This study
	NWIP	F	4.9 ± 4.2 (2)		This study
	France	nd	189.0 ± 298.0 (7) ^b		Alzieu and Duguy, 1979
	Ireland	M	10.2 ^a		Troisi et al., 1998
	Faroe Islands	M	48.8 ± 23.1 (52)	2.4 (21)	Borrell, 1993; Lindström et al., 1999
	Faroe Islands	F	26.3 ± 23.1 (159)	1.6 (32)	Borrell, 1993; Lindström et al., 1999
	Ligurian Sea	nd	125.0 (1) ^b		Marsili et al., 1997
	NW Atlantic Ocean				
	Canada	M	9.0 ± 3.8 (5)		Muir et al., 1988
	Canada	F	3.5 ± 3.3 (9)		Muir et al., 1988
	Massachusetts	M	12 ± 2.7 (6) ^c		Tilbury et al., 1999
	Massachusetts	F	6.1 ± 1.1 (16) ^c		Tilbury et al., 1999
	Pacific Ocean				
	Tasmania	M	0.41 ± 0.04 (21)		Weijs et al., 2013
	Tasmania	F	0.36 ± 0.26 (33)		Weijs et al., 2013
<i>Phocoena phocoena</i>					
	NE Atlantic Ocean				
	NWIP	M	19.8 ± 20.8 (4)	0.57 (1)	This study
	NWIP	F	20.8 ± 21.6 (8)		This study
	NW Spain	F	5.3 ± 4.2 (3)	0.28 ± 0.04 (3)	Pierce et al., 2008
	France	F	13.8 ± 10.6 (2)	1.4 ± 0.94 (2)	Pierce et al., 2009

Table 3 (Continued)

	Ireland	M	6.2 (1) ^a		Troisi et al., 1998
	Ireland	F	0.53 ± 0.5 (12)	0.66 ± 0.49 (12)	Pierce et al. 2008
	Scotland	M	13.1 (21) ^d		Wells et al., 1994
	Scotland	F	10.5 ± 13.1 (31)	1.4 ± 1.3 (31)	Pierce et al. 2008
	South England	M	23.4 ± 21.6 (2)	2.1 ± 1.6 (21) ^d	Law et al., 2006, 2010
	East England	M	11.6 ± 9.7 (23)		Law et al., 2010
	Southern North Sea	M	46.4 ± 30.7 (21)		Law et al., 2010
	Southern North Sea	F	15 ± 8.6 (19)	1.06 ± 0.8 (19)	Pierce et al., 2008
	Baltic Sea	M	31 ± 18.5 (17)		Berggren et al., 1999
	Norwegian Sea	M	15 ± 11 (8)		Berggren et al., 1999
	Faroe Islands	M	13.4 ± 2.4 (3)		Borrell, 1993
	Faroe Islands	F	8.8 ± 1.05 (3)		Borrell, 1993
	Greenland	M	2.4 (32)		Borrell et al., 2004
	Greenland	F	1.7 (43)		Borrell et al., 2004
	NW Atlantic Ocean				
	Canada	M	13.2 ± 9.9 (45)		Westgate et al., 1997
	Canada	F	9.3 ± 5 (43)		Westgate et al., 1997
	Boston	M	33.6 (2)		Tilbury et al., 1997
	Maine (Canada)	F	15.8 (1)		Tilbury et al., 1997
	Pacific Ocean				
	North Pacific	M	18.2 ± 16.6 (17)		Calambokidis and Barlow, 1991
	North Pacific	F	11.7 ± 10.0 (26)		Calambokidis and Barlow, 1991
<i>Stenella coeruleoalba</i>	NE Atlantic Ocean				
	NWIP	M	22.8 ± 23.3 (8)		This study
	NWIP	F	7.6 ± 5.9 (7)		This study
	England and Wales		20.0 (1)	0.45 (1)	Morris et al., 1989; Law et al., 2003
	Mediterranean Sea				

Table 3 (Continued)

	Spain	M	65.6 ± 40.5 (15)		Borrell and Aguilar, 2005
	Spain	F	90.0 ± 70.7 (5)		Borrell and Aguilar, 2005
	France	M	57.7 ± 41.9 (19)		Wafo et al., 2012
	France	F	45.3 ± 45.7 (12)		Wafo et al., 2012
	Italy	M	215.3 (33) ^b		Marsili et al., 1997
	Italy	F	92.8 (26) ^b		Marsili et al., 1997
	NW Atlantic Ocean				
	USA	–	59.0 (3)	0.66 (1)	Taruski et al., 1975; Johnson-Restrepo et al., 2005
	SW Atlantic Ocean				
	Brazil*	M	12.7 ± 9.6 (3)	0.81 ± 0.47 (3)	Leonel et al., 2012
	Brazil*	F	5.3 ± 7.8 (6)	0.54 ± 0.45 (6)	Leonel et al., 2012
	Pacific Ocean				
	Japan	M	25.3 ± 7.2 (20)	0.36 ± 0.28 (20)	Isobe et al., 2009
	Japan	F	3.2 (1)	0.08 (1)	Isobe et al., 2009
<i>Tursiops truncatus</i>	NE Atlantic Ocean				
	NWIP	M	62.1 ± 41.8 (4)		This study
	NWIP	F	48.9 ± 30.9 (3)		This study
	South Spain	M-F	182.6 ± 90.8 (5)		Borrell et al., 2006
	South Portugal	M-F	75.3 ± 39.4 (7)		Borrell et al., 2006
	Canary Islands	M	12.7 ± 10.7 (6)		Carballo et al., 2008
	Canary Islands	F	2.9 ± 4.3 (3)		Carballo et al., 2008
	West Ireland		5.01 ± 6.0 (6)		Berrow et al., 2002
	Scotland	M	13.6 (1) ^d		SOAFD data ^e
	Scotland	F	11.0 (5) ^d		Wells et al., 1994
	Wales	M	290.0 (3)		Morris et al., 1989
	Wales	F	760.0 (1)		Morris et al., 1989

Table 3 (Continued)

Mediterranean Sea				
Spain	M-F	167.8 ± 99.5 (13)		Borrell et al., 2006
Balearic Islands	M-F	117.3 ± 104.1 (7)		Borrell et al., 2006
Italy	M	1192 (5) ^f		Corsolini et al., 1995
Italy	F	587 (2) ^f		Corsolini et al., 1995
NW Atlantic Ocean				
North Carolina	M	53.3 (17)		Hansen et al., 2004
North Carolina	F	11.6 (14)		Hansen et al., 2004
South Carolina	M	50.4 (5)	0.98 (35)	Hansen et al., 2004; Fair et al., 2010
South Carolina	F	8 (6)	5.9 (11)	Hansen et al., 2004; Fair et al., 2010
Florida	M	20 (9)	1.5 (31)	Hansen et al., 2004; Fair et al., 2010
Florida	F	12.6 (2)	0.58 (15)	Hansen et al., 2004; Fair et al., 2010
SE Atlantic Ocean				
Rio de Janeiro	M	11.8 ± 2.4 (2)		Lailson-Brito et al., 2012
São Paulo	M	5.9 (1)	0.06 (1)	Yogui et al., 2010, 2011
SW Atlantic Ocean				
South Africa	M	2.6 ± 3.3 (5)		de Kock et al., 1994
South Africa	F	1.6 (1)		de Kock et al., 1994
Pacific Ocean				
Australia	M-F	0.06 (6)		Kemper et al., 1994
Indian Ocean				
Bay of Bengal, India	M	1.2 (2)		Tanabe et al., 1993
Bay of Bengal, India	F	0.75 (2)		Tanabe et al., 1993

* = *Delphinus delphis* and *Stenella frontalis*

a = Sum of 5 PCB congeners: CB118, 138, 153, 180 and 170

b = Values expressed on a dry weight basis (from lyophilized tissue)

c = SEM (standard error of the mean)

d = As Aroclor 1254 formulation

e = SOAFD data (Scottish Office Agriculture and Fisheries Department)

f = Values expressed on a wet weight basis