*Environ. Sci. Technol.* **2006**, *40,* 4394−4399

# Exposure to PBDEs and PCDEs Associated with the Consumption of Edible Marine Species

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In the present study, the concentrations of PBDEs and PCDEs were determined in 14 edible marine species widely consumed by the population of Catalonia (Spain). The daily intake of PBDEs and PCDEs associated with this consumption was also determined. A total of 42 composite samples were analyzed by HRGC/HRMS. The highest PBDE levels (ng/kg wet weight) were found in salmon (2015) followed by mackerel, swordfish, and red mullet (1124, 978, and 769, respectively), while those of PCDEs (ng/kg wet weight) were detected in red mullet (7088) followed by sardine (1829), anchovy (1606), tuna (1292), and mackerel (1031). For a standard male adult, total PBDE and PCDE intakes through edible marine species were 20.8 and 39.4 ng/day, respectively. The highest contributions to these intakes (ng/day) corresponded to the consumption of tuna (5.7), salmon (3.6), and hake (3.5) for PBDEs, and tuna (13.1), hake (7.3), and sardine (6.9) for PCDEs. Although currently there is not evidence of the dioxin-like behavior of PBDEs, further research is necessary to assess if long-term exposure to PBDEs, mainly through the diet, may mean adverse effects to humans. With respect to PCDE congeners, to establish TEF values would be of great value to evaluate human health risks.

## Introduction

The polybrominated diphenyl ethers (PBDEs) are a class of chemicals widely used as flame retardants in a number of different commercial and industrial products (electronic equipment, cars, textiles, insulation, etc.) (*1−3*). PBDEs are recognized as ubiquitous environmental pollutants, which have been identified in various media from around the world (*4−11*). PBDEs have some physicochemical properties similar to various other persistent organic pollutants (POPs), which are known to bioaccumulate in the environment (*3, 12−16*). The stability and lipophilicity of PBDEs causes them to biomagnify up the food chain, increasing in concentration at each successively higher trophic level (*13, 14, 17*). It is interesting to note that the PBDE technical product containing Tri-HxBDE is currently being considered as a potential candidate to be included under the 1998 United Nations Economic Commission for Europe (UNECE) POPs convention (*8*).

Nowadays, it is still unclear whether the current concentrations of PBDEs in human tissues have an adverse impact on human health (*17−19*). However, recent investigations have shown that in certain countries PBDE concentrations have increased in the tissues in which these pollutants were identified (*3, 17, 18, 20−22*). With respect to PBDEs, it was expected that, as for other POPs, the diet is the main pathway of human exposure. Notwithstanding, recent studies have demonstrated that house dust could be also a significant pathway of PBDE exposure for some individuals (*23−26*).

Recently, we determined the concentrations of PBDEs in a number of foodstuffs purchased from various cities of Catalonia (Spain). The dietary intake of these compounds by the population of this region was also estimated (*27*). The most important contribution to the dietary PBDE intake corresponded to fish and seafood, being approximately one-third of the total (*27*).

On the other hand, polychlorinated diphenyl ethers (PCDEs) are another group of halogenated aromatic compounds which are structurally related to PCBs. PCDEs mainly originate from byproducts of chlorophenols and chlorinated phenoxyacetic acids, as well as from incomplete combustions (e.g., waste incinerators) (*28*). The extensive use in recent decades of chlorinated phenols, especially pentachlorophenol as an herbicide and in wood preserving formulations, has led to a ubiquitous presence of PCDEs. These compounds are quite resistant to degradation, are persistent in the environment, and bioaccumulate in aquatic media (*28, 29*). PCDEs have been detected in a number of environmental samples (*28, 29*), as well as in human adipose tissue (*30*). The diet was also expected to be the main pathway of human exposure to PCDEs (*29*). However, data concerning PCDE levels in foodstuffs and dietary intake of these chemicals is currently limited to only one study (*31*). In that survey, PCDE concentrations could be detected only in fish and seafood samples. However, as for the PBDE survey (*27*), PCDE levels were only determined in three species of fresh fish and in two species of tinned fish. It is a limiting factor for establishing recommendations about human consumption (frequency and size of meals) of fish and seafood.

Taking the above into account, the purpose of the present study was to extend the data obtained in our previous surveys (*27, 31*). The levels of PBDEs and PCDEs were measured in 14 species of fish and seafood which are among the most consumed by the general population of Catalonia. The contribution of this food group to the dietary intake of PBDEs and PCDEs by this population was also determined.

## Materials and Methods

**Sampling.** In March−April 2005, edible marine species were randomly acquired in local fish markets, big supermarkets, and grocery stores from six important cities of Catalonia, Spain, whose inhabitants mean approximately 60% of the total population of Catalonia. The selected fish and seafood species were the following: sardine, tuna, anchovy, mackerel, swordfish, salmon, hake, red mullet, sole, cuttlefish, squid, clam, mussel, and shrimp. According to recent studies of the Spanish Ministry of Agriculture and Fishery (*32*), all these 14 species are included among the most consumed marine species by the Spanish population. A total of 42 composite samples (3 for each species) were analyzed for the levels of PBDEs and PCDEs. Each composite sample was made up of 20 individual samples of the same species.

**Analytical Methods and Instrumentation.** For the analyses of the groups of congeners of PBDEs and PCDEs,

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10.1021/es060484k CCC: $33.50

Published on Web 06/13/2006

methodological details including preparation of samples for analysis and cleanup were widely described in recent studies (*27, 31*). In the present investigation, detection limits for PBDEs varied from 0.4 to 4.2 ng/kg wet weight depending on the specific samples and the respective congeners. Detection limits for PCDEs varied between 0.2 and 0.4 ng/kg wet weight.

**Dietary Exposure Estimates.** Consumption data by the general population of Catalonia of the analyzed fish and seafood species were obtained from Serra-Majem et al. (*33*). When the PBDE or PCDE concentrations were under the respective limit of detection (LOD), daily intakes were calculated assuming the respective values would be equal to one-half of that LOD.

## Results

**PBDE Concentrations.** The concentrations of PBDEs (wet weight) in fish and seafood samples are summarized in Table 1. The highest levels of PBDEs (sum tetra- to octaBDEs) were found in salmon (2015 ng/kg), while the lowest PBDE concentrations were detected in cuttlefish (16 ng/kg). In most species, tetra-BDEs, followed by penta-BDEs, were the main contributors to total PBDEs.

The concentrations (wet weight) of six PBDE congeners (BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, and BDE-183) were also determined. These congeners were specifically analyzed taking into account that they are the most abundant and toxicological important PBDEs (*2, 3, 14*). The highest and lowest levels corresponded to BDE-47 and BDE-183, respectively. When the total sum of PBDEs was calculated including only these six congeners, the differences with the total sum of tetra- to octaBDE resulted notable for all species: 2015 vs 1620 ng/kg for salmon, or 16 vs 10.7 ng/kg for cuttlefish. The contribution of BDE-47 to the sum of the six BDE congeners individually analyzed was especially remarkable for all species.

In the current study, the mean total (tetra to octa) PBDEs was 564 ng/kg, which is higher than that found in our previous survey (*27*), 334 ng/kg. However, it must be noted that only 3 fresh marine species were then included: sardine, hake, and mussel. Among these, the sum of PBDE homologues concerning sardine decreased, while those corresponding to mussel and hake increased.

**Intake of PBDEs through Fish and Seafood.** Fish and seafood consumption and the mean intake of PBDEs for a standard male adult living in Catalonia are shown in Table 2. Total PBDE intake through edible marine species was estimated to be 20.8 ng/day. The highest contributions (ng/day) to this intake corresponded to tuna (5.7) and salmon (3.6), while the lowest PBDE intake (ng/day) corresponded to clam (0.02) and swordfish (0.06). It can be observed that the ranking of PBDE concentrations and that concerning the daily PBDE intake show important differences depending on the consumed amounts of each species. With respect to the current percentages of contribution of PBDE homologue groups to the intake of PBDEs through fish and seafood, the highest contribution corresponded to the sum of tetra-BDE (54%).

The intake of PBDEs by the general population of Catalonia was also estimated according to eight age/sex groups (children: boys and girls; and adolescents, adults, and seniors (> 65 years old), males and females in all cases). The results were calculated in ng/day and ng/kg body weight/day. Among the eight groups, the highest PBDE intake (ng/day) corresponded to adult females (21.8) and males (20.8), while the lowest intake corresponded to children (girls 8.2 ng/day and boys 12.2 ng/day). However, when the comparison was made according to the respective average body weight of each group, children (boys 0.51 ng/kg/day and girls 0.34 ng/kg/day), and adult females (0.40 ng/kg/day)

**TABLE 1. PBDE Concentrations (ng/kg Wet Weight) in Fish and Seafood Purchased from Markets in Catalonia, Spain (Mean, Median, and Range)**

| | ΣTeBDE | ΣPeBDE | ΣHxBDE | ΣHpBDE | ΣOBDE | ΣPBDEs (tetra to octa) |
|---|---|---|---|---|---|---|
| sardine | 373, 373 (324–423) | 208, 212 (197–214) | 113, 111 (107–120) | 3.5, 3.3 (3.2–4.0) | 13, 14 (11–14) | 710, 719 (646–765) |
| tuna | 282, 43 (33–772) | 141, 26 (17–381) | 116, 28 (19.3–300) | 6.2, 1.6 (1.6–16) | 13, 3.8 (0.9–34) | 558, 101 (71–1503) |
| anchovy | 320, 305 (274–381) | 169, 178 (150–180) | 104, 106 (86–121) | 3.9, 1.6 (1.6–8.5) | 13, 14 (9.8–15) | 610, 615 (546–669) |
| mackerel | 531, 519 (483–592) | 337, 337 (314–358) | 227, 217 (203–260) | 17, 18 (14–20) | 12, 12 (11–12) | 1124, 1166 (1027–1178) |
| swordfish | 374, 339 (147–635) | 264, 197 (64–530) | 328, 198 (39–745) | 7.0, 5.0 (4.8–11) | 6.0, 6.3 (5.0–6.8) | 978, 752 (260–1921) |
| salmon | 1300, 1310 (1140–1450) | 465, 445 (363–586) | 232, 206 (201–290) | 11, 9.1 (8.8–14) | 7.6, 7.2 (7.0–8.6) | 2015, 1983 (1720–2342) |
| hake | 129, 108 (66–213) | 48, 34 (32–77) | 36, 41 (23–44) | 4.3, 5.0 (1.6–6.2) | 4.1, 4.8 (1.9–5.7) | 221, 186 (132–345) |
| red mullet | 319, 153 (104–700) | 229, 133 (112–441) | 210, 171 (133–327) | 7.7, 9.2 (3.4–11) | 3.5, 3.5 (2.0–5.0) | 769, 471 (354–1482) |
| sole | 126, 151 (11–217) | 51, 68 (7.3–78) | 54, 74 (6.9–81) | 4.5, 4.5 (4.0–5.1) | 5.4, 4.0 (3.5–8.7) | 242, 316 (34–375) |
| cuttlefish | 4.3, 4.8 (2.1–6.1) | 4.5, 4.9 (2.1–6.6) | 2.8, 3.6 (1.0–3.7) | 2.5, 2.5 (1.6–3.4) | 1.8, 1.8 (0.8–2.8) | 16, 17 (5.9–23) |
| squid | 85, 74 (49–133) | 79, 48 (38–152) | 27, 31 (19–31) | 6.4, 5.5 (4.3–9.4) | 5.7, 5.8 (2.9–8.5) | 204, 164 (114–334) |
| clam | 28, 29 (26–30) | 25, 25 (21–27) | 11, 11 (8.4–13) | 6.8, 6.4 (5.4–8.7) | 8.1, 8.0 (6.9–9.4) | 79, 78 (72–86) |
| mussel | 170, 159 (72–279) | 136, 134 (47–227) | 26, 24 (6.7–49) | 11, 14 (4.4–14) | 6.2, 5.2 (2.5–11) | 350, 326 (143–580) |
| shrimp | 8.1, 7.2 (2.1–15) | 3.6, 2.1 (2.1–6.6) | 3.0, 2.3 (1.0–5.8) | 3.0, 1.6 (1.6–5.8) | 2.0, 0.9 (0.8–4.3) | 20, 14 (7.5–37) |

**TABLE 2. Estimated PBDE and PCDE Intake through Fish and Seafood Consumption by the Adult Population of Catalonia, Spain[a]**

| | daily consumption (g) | PBDE intake (ng/day) | PCDE intake (ng/day) |
|---|---|---|---|
| sardine | 3.8 | 2.68 | 6.91 |
| tuna | 10.1 | 5.66 | 13.1 |
| anchovy | 2.1 | 1.25 | 3.29 |
| mackerel | 1.1 | 1.27 | 1.16 |
| swordfish | 0.06 | 0.06 | 0.01 |
| salmon | 1.8 | 3.62 | 0.67 |
| hake | 15.8 | 3.49 | 7.26 |
| red mullet | 0.33 | 0.25 | 2.34 |
| sole | 5.5 | 1.32 | 1.07 |
| cuttlefish | 4.5 | 0.07 | 0.22 |
| squid | 3.2 | 0.65 | 3.10 |
| clam | 0.27 | 0.02 | 0.01 |
| mussel | 0.97 | 0.34 | 0.15 |
| shrimp | 3.5 | 0.07 | 0.10 |
| TOTAL | 52.9 | 20.8 | 39.4 |

[a] Results are given for a male adult of 70-kg body weight.

were the groups showing the highest PBDE intakes. The lowest PBDE intake corresponded to female adolescents and female seniors: 0.27 and 0.28 ng/kg/day, respectively.

**PCDE Concentrations.** The sum of tetra-, penta-, hexa-, hepta-, and octaCDE, as well as the total PCDE concentrations in fish and seafood samples are summarized in Table 3. The highest PCDE concentrations (ng/kg wet weight) were observed in red mullet (7088), while shrimp (*27*) was the species showing the lowest sum of PCDEs. It must be noted that in various shrimp and cuttlefish samples some PCDE congeners were under the analytical detection limit. With the exception of clam and mussel (for which tetraCDE was the predominant group), there was a predominance of the hexaCDE.

An important increase was noted when the current mean of total PCDEs (1095 ng/kg) was compared with that of our previous survey, 418 ng/kg (*31*). However, if the comparison is carried out with only the 3 species included in both studies, the differences between them are then scarce (small increases for sardine, mussel, and hake).

**Intake of PCDEs through Fish and Seafood.** Fish and seafood intake of PCDEs for a standard male adult living in Catalonia are given in Table 2. Total PCDE intake through fish and seafood was 39.4 ng/day. The highest contributions (ng/day) corresponded to tuna (13.1) and hake (7.3), while the lowest PCDE intake was due to clam (0.01) and shrimp (0.10). As for PBDEs, PCDE intakes from edible marine species showed important differences depending on the daily consumption of the different species. In relation to the percentages of contribution of PCDE homologue groups to the intake of PCDEs through fish and seafood, the sum of hexa- and heptaCDE reached the highest contributions: 35% and 30%, respectively.

In our previous PCDE survey (*31*), the intake of PCDEs through fish and seafood was 38.4 ng/day. The difference with the current 39.4 ng/day is irrelevant. However, a decrease can be observed if the comparison includes only the three species analyzed in both studies: 61.8 and 55.5 ng/day in the previous and current surveys, respectively. The intakes of PCDEs by the general population of Catalonia were also determined according to the eight above indicated age/sex groups. The results were calculated in ng/day and ng/kg body weight/day. The highest PCDE intakes (ng/day) through fish and seafood consumption corresponded to adult males (39.4) and adult seniors (37.4), while the lowest PCDE intake corresponded to children (girls 17.6 ng/day and boys 21.1

**TABLE 3. PCDE Concentrations (ng/kg Wet Weight) in Fish and Seafood Purchased from Markets of Catalonia, Spain (Mean, Median, and Range)**

| | ∑TeCDE | ∑PeCDE | ∑HxCDE | ∑HpCDE | ∑OCDE | ∑PCDEs(tetra to octa) |
|---|---|---|---|---|---|---|
| sardine | 113, 60 (57−221) | 304, 238 (190−485) | 635, 629 (455−822) | 523, 598 (366−604) | 254, 284 (193−285) | 1829, 1758 (1312−2417) |
| tuna | 20, 7.2 (1.8−51) | 114, 44 (5.8−292) | 439, 141 (4.8−1170) | 426, 153 (3.4−1120) | 294, 94 (2.3−785) | 1292, 439 (18−3418) |
| anchovy | 125, 68 (30−277) | 333, 249 (92−659) | 542, 471 (278−876) | 391, 368 (211−595) | 215, 202 (132−311) | 1606, 1358 (743−2718) |
| mackerel | 64, 63 (55−74) | 237, 249 (203−260) | 319, 320 (232−405) | 277, 315 (179−337) | 134, 144 (73−184) | 1031, 1113 (742−1238) |
| swordfish | 4.4, 4.0 (1.7−7.4) | 22, 14 (12−41) | 82, 93 (16−136) | 63, 83 (7.9−97) | 29, 35 (4.8−47) | 200, 226 (48−325) |
| salmon | 59, 52 (34−92) | 122, 109 (88−169) | 129, 122 (114−151) | 38, 43 (18−52) | 25, 24 (24−26) | 372, 362 (302−453) |
| hake | 9.6, 4.7 (4.0−20) | 50, 28 (22−99) | 176, 104 (74−351) | 153, 87 (60−312) | 71, 47 (33−134) | 460, 257 (208−916) |
| red mullet | 259, 273 (88−414) | 1297, 906 (594−2390) | 2317, 1950 (1110−3890) | 1867, 1630 (962−3010) | 1349, 1640 (627−1780) | 7088, 6399 (3382−11484) |
| sole | 17, 17 (3.1−31) | 35, 46 (3.9−55) | 70, 102 (3.7−103) | 46, 43 (2.9−91) | 27, 27 (3.1−52) | 195, 244 (17−323) |
| cuttlefish | 0.8, 0.2 (0.2−1.9) | 8.8, 9.5 (0.2−17) | 17, 20 (1.7−30) | 8.1, 2.4 (0.2−22) | 15, 1.7 (0.1−44) | 50, 41 (2.4−107) |
| squid | 71, 47 (4.9−160) | 176, 109 (33−385) | 334, 250 (124−628) | 258, 157 (130−486) | 140, 117 (60−244) | 978, 623 (409−1903) |
| clam | 21, 29 (2.0−34) | 13, 17 (1.5−20) | 7.5, 9.9 (2.5−10) | 3.0, 3.6 (1.1−4.3) | 2.6, 2.7 (1.1−4.1) | 48, 67 (8.2−67) |
| mussel | 111, 81 (54−197) | 14, 12 (11−20) | 14, 12 (12−19) | 6.6, 5.6 (4.6−9.5) | 3.9, 3.9 (2.8−5.1) | 150, 119 (95−236) |
| shrimp | 0.5, 0.2 (0.2−1.2) | 2.5, 0.5 (0.4−6.6) | 9.9, 5.3 (0.8−23.7) | 6.8, 2.2 (0.2−18) | 7.8, 2.7 (0.1−21) | 27, 11 (1.8−70) |

ng/day). However, when the comparison was made taking into account the respective average body weight of each group, children (boys 0.88 ng/kg/day and girls 0.73 ng/kg/day), and adult females (0.65 ng/kg/day) were the groups showing the highest PCDE intake.

## Discussion

In relation to POPs, one of the edible marine species to which in recent years a greater notable attention has been paid is salmon. In 2004, Hites and co-workers (*34*) reported that the levels of various POPs were significantly higher in farm-raised salmon than in wild Pacific salmon. Important differences in the concentrations of PBDEs (0.1−4.2 ng/g wet weight) were found depending on the origin of the salmon (*35*). In the current study, the sum of PBDE levels in salmon was about 2.0 ng/g (wet weight), and also within the range (0.14−5.5 ng/g wet weight) reported by Tittlemeier et al. (*36*) for salmon samples purchased from Canadian markets. However, it was lower than that recently found by Bethune et al. (*37*) in Norwegian farmed Atlantic salmon (3.9 ng/g wet weight), or by Otha et al. (*38*) in salmon filets obtained from Japanese markets (5.9−10.4 ng/g wet weight). Recently, in the raw muscle and skin of Atlantic salmons of Norway, Bayen et al. (*39*) reported ranges of PBDEs of 2.5−7.6 and 4.1−11.5 ng/g wet weight, respectively. They found that BDE-47 was the dominant PBDE congener, more than 70% of the total PBDE concentration, a percentage similar to that of the current study. A lower contribution of BDE-47, 48%, was reported by Tittlemeier et al. (*36*). Hites et al. (*35*) also found that BDE-47 was the most abundant congener in wild and farmed salmons.

With respect to previous data concerning other edible marine species, Christensen and Platz (*40*) reported a sum of 4 PBDEs in blue mussels collected in the Danish marine environment in the range of 0.08−0.81 ng/g wet weight (0.26 ng/g weight in the current study for the same 4 PBDE congeners, or 0.35 ng/g wet weight for the sum of total PBDEs). In a subsequent study, Christensen et al. (*41*) found PBDE levels (including the same previous 4 BDE congeners) of 0.11 ng/g wet weight in blue mussels from southern Greenland. In mussels and shrimps purchased from Canadian markets, Tittlemeier et al. (*36*) reported geometric means of the sum of 18 BDEs of 0.26 ng/g wet weight (range 0.05−0−63) and 0.05 ng/g wet weight (range 0.001−0.68), respectively, values of the same order of magnitude as those of the present survey. Recently, in bivalves from the San Francisco Estuary, the sum of PBDE concentrations ranged from 13 to 47 ng/g dry weight in mussels, and from 85 to 106 ng/g dry weight in clams (*42*). In turn, de Boer et al. (*43*) determined the levels of 6 PBDE congeners in marine mussels from various origins in The Netherlands. Only BDE-47 and BDE-99 could be detected in all samples. The concentrations ranked between 0.9 and 4.3 ng/g dry weight, and from 0.4 to 1.6 ng/g dry weight, for BDE-47 and BDE-99, respectively. In our study, BDE congeners 47, 99, and 100 also reached the highest concentrations in mussel and clam.

In a recent study in which the concentrations of PBDEs were determined in various marine products, Ashizuka et al. (*44*) reported the following sums (expressed in ng/g whole basis) of PBDEs for those edible species that were also included in the present study: sardine, 0.24; mackerel, 0.55; tuna, 0.009, and squid, 0.26. The most dominant congener present in these marine samples was again BDE-47. On the other hand, when various marine species from the Belgian North Sea (BNS) and the Western Scheldt Estuary (SE) were analyzed for selected PBDEs, the sums of 6 PBDE congeners in sole and shrimp were 0.37 (BNS) and 2.7 ng/g wet weight, and 0.04 (BNS) and 3.5 ng/g wet weight, respectively (*45*). Sole and shrimp were the only species included among those analyzed in the present study. The current values were lower than those of that survey. In turn, a wide range (<0.1−53 ng/g of lipid) was found in samples of skipjack tuna (used as a bioindicator) collected from a number of locations belonging to waters of various regions in the world (*46*).

Recently, Johansson et al. (*47*) determined levels and trends of PBDEs in archived freeze-dried samples of blue mussels from selected French coastal sites collected between 1981 and 2003. These authors corroborated data showing that the levels of PBDEs in the North American environment were higher than those in Europe (*3*). Brown et al. (*48*) found a mean concentration of the sum of BDEs 47, 99, 100, 153, and 154 of 302 ng/g lipid weight in 15 species of edible fish from California coastal waters.

In this discussion, we have summarized data from a number of previous studies carried out in various countries. However, the comparison among results is rather complicated. While PBDEs bioaccumulate in marine species, their levels increase with the age of the fish (*49*). Moreover, the concentrations of PBDEs depend basically on the environment in which the respective species are collected. An additional difficulty for comparison of results is that some studies report PBDE concentrations as the sum of homologues (tetra to octaBDE), while in other investigations PBDE congeners are individually analyzed and their sums reported. Other differences also concern the expression of the results: wet weight, dry weight, or lipid weight. Finally, some investigators analyzed only some parts of the fish and seafood samples: muscle, liver, skin, etc., which makes comparison of the data still more complicated. Anyhow, it seems that the PBDE concentrations found in the current study are of the same order of magnitude, or even lower, than those found in studies where data may be compared (*3, 14, 50*).

With respect to the contribution of fish and seafood consumption to the daily intake of PBDEs, there are notable differences depending on both the dietary habits and the levels of PBDEs in the respective edible species. In a food basket study, Ryan and Patry (*51*) estimated a dietary intake of PBDEs for Canadian adults of 44 ng/day. Fish contributed approximately with 4% to this amount. In a Swedish study, the daily intake of PBDEs was 31 ng, with fish being 58% of the contribution to this quantity (*52*). In a market basket study on dietary intake of PBDEs in Finland, Kiviranta et al. (*53*) found a daily intake of these compounds of 44 ng. Fish contribution was 55% of the total. In the United States, two surveys concerning concentrations of PBDEs in foodstuffs have been recently carried out (*54, 55*). In a market based study, Schecter et al. (*54*) determined the levels of PBDEs in foods of animal origin (meat, fish, and dairy products). Although PBDE exposure via dietary intake was not calculated, fish was the food group showing the highest PBDE levels. In the other survey, no fish and seafood samples were analyzed (*55*).

In a previous study of our research group, we estimated a dietary intake of PBDEs for a standard male adult (70 kg body weight) of Catalonia of 97 ng/day. The contribution of fish and seafood to that intake was 31 ng/day. In the present study, PBDE intake through 14 edible marine species was 21 ng/day. However, it must be noted that the daily consumption of fish and seafood by the population of Catalonia has decreased for standard adult males from the previous 92 g (*27*) to the current 53 g. Moreover, it is also important to remark that in the previous survey only 3 fresh marine species and 2 tinned species were analyzed versus the current 14 species.

In relation to the potential health risks derived from PBDE intake through fish and seafood by the general population of Catalonia, it must be taken into account that the database on toxicological concentrations of PBDEs is extremely limited for a full comprehensive risk assessment (*2, 14*). On the basis of the most sensitive endpoints for toxic effects of PBDEs, a LOAEL (lowest observed adverse effect level) of 1 mg/kg/day (based on thyroid hormone effects in rats) was suggested as reasonable for compounds or mixtures of PBDEs (*2,56*). The comparison of the current PBDE intake through fish and seafood by an adult male of 70 kg body weight (0.30 ng/kg/day) with the suggested LOAEL value results in a safety factor of various orders of magnitude.

In recent years, various investigations have shown that in contrast to dioxins and furans and dioxin-like PCBs, most PBDE congeners did not show any capacity to induce CYP1A by the Ah receptor mediated pathway (*57*−*59*). Relative induction potencies (REPs) of the most active PBDEs toward CYP1A1 are about 0.0001 compared with 2,3,7,8-TCDD. These values are similar to some mono-*ortho*-PCBs and 2 orders of magnitude less than those of coplanar PCBs (*57*). At the present time, PBDEs contribute negligibly to the TEQ. However, it must be noted that PBDE levels in the environment are rising while those of PCBs, and PCDD/Fs are falling (*18, 21, 22*). Consequently, although the dioxin-like behavior of PBDEs might be discarded, it does not exclude important toxic effects not mediated by the AhR such as reproductive and developmental toxicity, endocrine disruption, immunotoxicity and neurotoxicity (*2, 22, 56, 60*).

With respect to PCDEs, to date, the dietary intake of these compounds has been only estimated for the general population of Catalonia (*31*). When the concentrations of PCDEs were measured in 54 samples belonging to 11 groups of foodstuffs, all values were under the respective detection limits with the exception of the fish and seafood group, either fresh or tinned in vegetable oil. Dietary intake of PCDEs by a standard male adult of 70 kg body weight was 41 ng/day (*31*) (39 ng/day in the current study). Although both intakes are analogous, it must be noted that in the present study fish and seafood was the only food group assessed.

As with other POPs such as PCDD/Fs, PCBs, or PCNs, the mechanism of action for the toxicity of PCDEs could be related to bind and activate the aryl receptor (AhR). However, it has not been clearly shown yet. Therefore, further investigations focused on demonstrating the AhR-mediated PCDE effects are necessary before PCDEs can be included as dioxin-like compounds for assessment of human health risks. On the other hand, the lack of reported data on dietary intakes of PCDEs makes it quite impossible to carry out a comparison with those of the current study. Consequently, we encourage investigations on the human health effects of PCDEs and the pathways of exposure, including estimates of dietary intake in different countries.

## Acknowledgments

This study was supported by the Catalan Food Safety Agency, Generalitat de Catalunya, Spain. We are grateful to Dr. L. Serra-Majem and co-workers for providing their recent data about dietary habits of the population of Catalonia.

## Supporting Information Available

Information on the analytical methods, dietary exposure estimates, specific concentrations of six PBDE congeners, estimated intake of PBDEs and PCDEs through fish and seafood by various age/sex groups of the population of Catalonia, and the percentages of contribution of the different groups of homologues to the daily intake of PBDEs and PCDEs through fish and seafood. This material is available via the Internet at http://pubs.acs.org.

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*Received for review March 1, 2006. Revised manuscript received April 28, 2006. Accepted May 18, 2006.*

ES060484K