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Brominated flame-retardants (PBDEs and HBCDs) in fish species from Italian market

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Introduction

PBDEs and HBCDs are used as additives, not chemically bonded, to reduce plastic polymers flammability. They can be released in the environment during polymer production, use and disposal and can accumulate in the food chain. Few data are available in Italy about their levels in the environment and in food [1,2]. This study aims to contribute to the PBDEs and HBCDs concentrations assessment in a selection of marine fish samples collected in Italian local markets.

Sampling

15 PBDEs (28, 47, 49, 66, 77, 85, 99, 100, 138, 153, 154, 183, 197, 206, 209) and **3 HBCDs** (α-, β-, γ-HBCD) were analysed in 18 marine fish samples (11 species) collected from local markets, in Central Italy.

N. SAMPLES	SPECIES	BINOMIAL NAME
1	Tuna	*Thunnus alalunga*
2	Sole	*Solea solea*
2	Grey mullet	*Mugil cephalus*
1	Spiny dogfish	*Squalus acanthias*
2	Hound shark	*Mustelus mustelus*
2	Mackerel	*Scomber scombrus*
2	Swordfish	*Xiphias gladius*
1	Atlantic horse mackerel	*Trachurus trachurus*
2	Cod	*Merluccius merluccius*
1	Anchovy	*Engraulis encrasicolus*
2	Mullet	*Mullus barbatus*

Table 1 *– Number, species and binomial name of the collected samples.*

Analytical method

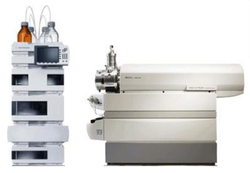
20 g of fish tissue spiked with 1.0 ng of PBDE-L (BDE-28-,-47-,-99-,-100-,-153-,-154-,-183-,-209- $^{13}C_{12}$) and 1.0 ng of α– and γ– HBCD-$^{13}C_{12}$)

5 mL H_2O, 15 mL EtAc, 6 g anhydrous $MgSO_4$, 3 g NaCl. Shaking and centrifugation

Withdrawal of the upper organic layer: 10 mL

1° Clean-up step: tandem H_2SO_4-Extrelut NT 3/silica SPE

2° Clean-up step: Gel Permeation Chromatography



Syringe Standards Addition (0.5 ng of β-HBCD-$^{13}C_{12}$)

Syringe Standards Addition (0.5 ng of BDE-77- and BDE-138-$^{13}C_{12}$)

LC-MS Triple Quad (AB SCIEX 3200 Q Trap)

GC-MS Triple Quad (Agilent 7890A/7000)

Figure 1 *– Sample preparation flow diagram [3].*

Results and Discussion

PBDEs and HBCDs were measured above MDL in most of the samples. Their concentrations were generally in the range of ppts. Only BDE-47, the dominant congener in all species, was measured in 16 out of 18 samples and reached ppb levels in one sample. In 12 samples were measured also BDEs 100> 99> 49 and 154.

Total **PBDEs** contamination was estimated as sum of the 15 congeners adopting a lower bound approach (concentrations lower than MDL were set to 0.0). The highest PBDE levels were measured in a **spiny dogfish** and **mackerel** (sum: **2837** and **642 pg/g respectively**), while **cod** and **hound shark** showed the lowest levels (sum: **26** and **39 pg/g respectively**).

The highest **HBCD** isomer was always the α, which was quantified in 13 out of 18 samples [4]. Only in one mackerel also γ-HBCD was identified. The total **HBCDs** content ranged from <MDL (cod and hound shark) to **274** and **309 pg/g** in **spiny dogfish** and **mackerel** respectively.

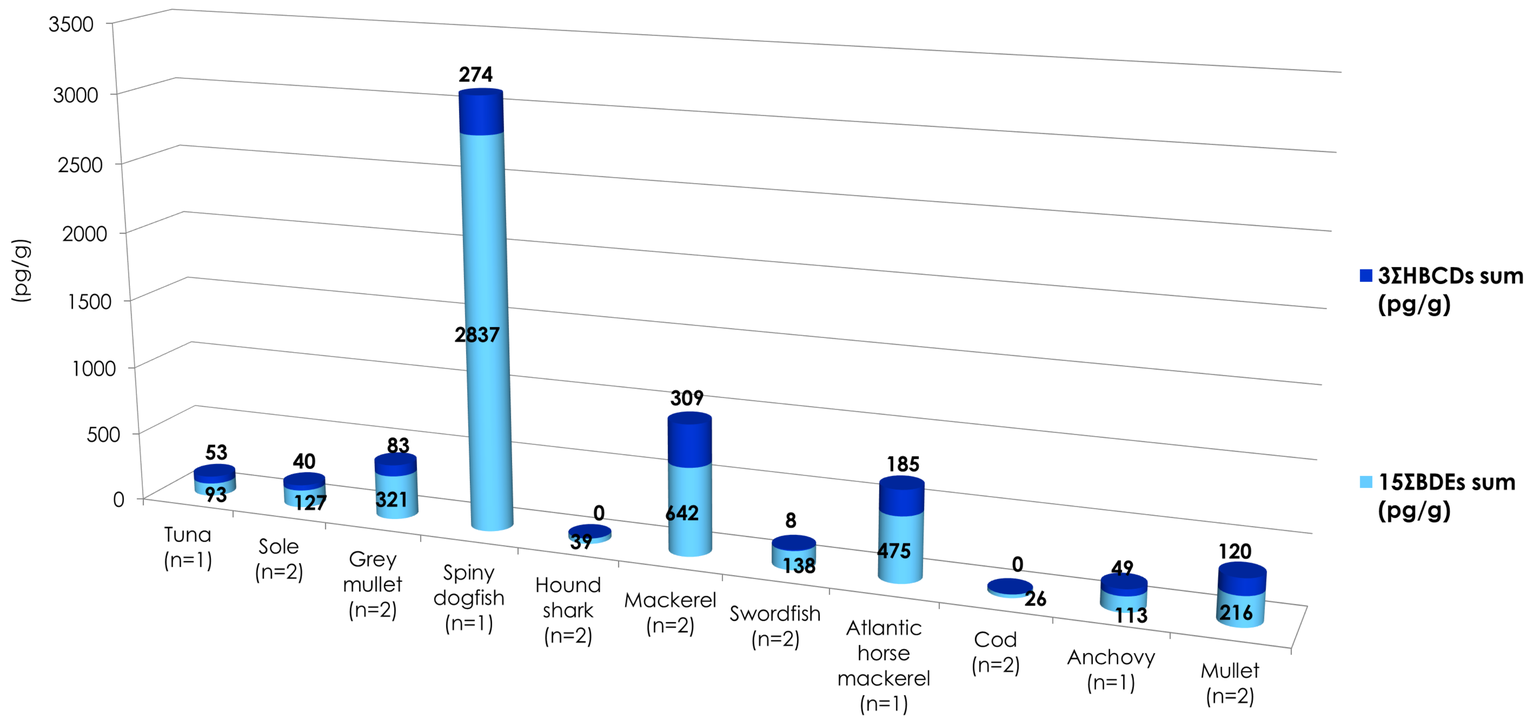


Figure 2 – *BDEs (light blue) and HBCDs (blue) level (sum of 15 and 3 congeners respectively) for each species analysed (n=number of samples). Spiny dogfish shown the highest PBDE level, while mackerel had the highest HBCD contamination.*

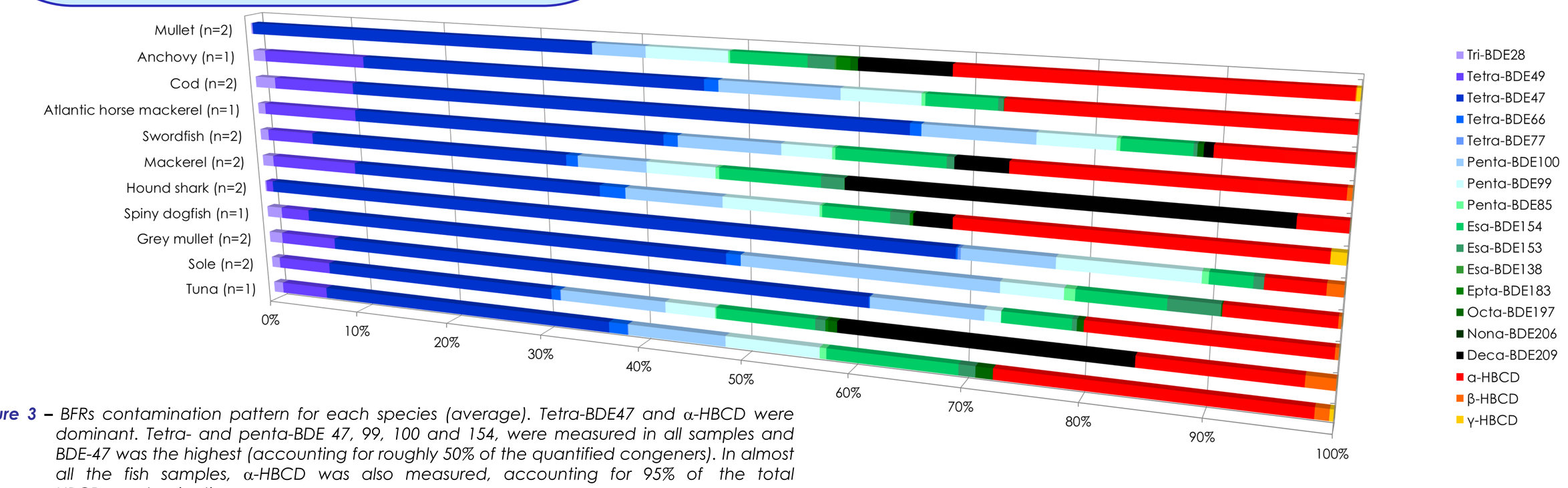


Figure 3 – *BFRs contamination pattern for each species (average). Tetra-BDE47 and α-HBCD were dominant. Tetra- and penta-BDE 47, 99, 100 and 154, were measured in all samples and BDE-47 was the highest (accounting for roughly 50% of the quantified congeners). In almost all the fish samples, α-HBCD was also measured, accounting for 95% of the total HBCDs contamination.*

Conclusions

The highest levels for ΣPBDEs and ΣHBCDs were measured in spiny dogfish (lipid 7.7%), confirming that fat species are generally the most contaminated while lean ones (cod and hound shark - lipid <1%) have lower concentration. No data are available in literature for spiny dogfish while comparable results are reported for all the other species [5,6].

References

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