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## PBDES IN MEDITERRANEAN SEAFOOD: INTAKE ESTIMATES IN THE ITALIAN COASTAL POPULATION

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### Introduction

Polybrominated diphenyl ethers (PBDEs) are additive flame retardants widely used in plastics, textiles, electronic castings and circuitry. They are ubiquitous in the environment and likewise in biota, food and feed. According to the EFSA report (1) the most contaminated food category is “Fish and other seafood”, followed by “Meat and meat products”, “Animal and vegetable fats and oils”, “Milk and dairy products” and “Eggs and egg products”. Eight congeners appear to be of interest, BDE-28, -47, -99, -100, -153, -154, -183 and -209, but only four of them, BDE-47, -99, -153 and -209, were considered by the EFSA Panel to derive a benchmark dose. Recently, two projects, focused on the characterization of the chemical contamination of Mediterranean seafood, were granted by the Italian Ministry for Education, University and Research (MIUR), and the Italian Health Ministry (MINSAL) with the aim to give suggestions for risk reduction strategies(2,3,4,5). The two projects, globally, appear to address the scope of the Marine Strategy Framework Directive (MSFD) (Directive 2008/56/EC) setting a framework within which EU Member States shall take the necessary measures to achieve or maintain good environmental status (GES) in the marine environment by the year 2020 (6). However, their results can be considered also for the estimation of PBDE human intake from seafood in the coastal population in Italy (7).

### Materials and methods

The following 18, 19, and 10 geographical sub-areas of FAO fishery Zone 37, presumably unexposed to direct contamination sources, were selected: Southern Adriatic Sea (10–40 miles off the coastline close to Monopoli port); Ionian Sea (15 miles off Portopalo port); and southern Tyrrhenian Sea (15 miles off the Bagnara Calabria coastline). In the case of mussels the monitoring involved the coastal area of a low anthropized Italian region on the middle Adriatic Sea: the Marche region. The sampling design covered many of the fish and crustacean species available on the Italian market (8) selecting twelve wild and two farmed aquatic species. The PBDEs congeners no. BDE-28, -47, -49, -66, -77, -85, -99, -100, -138, -153, -154, -183, -197, -206 and -209 were analysed by HRGC-HRMS in fish and crustaceans (3) and by HRGC-LRMS/MS in mussels (5). To assess seafood consumption and the related chemical contaminant intake, a specific questionnaire targeted to record Seafood Frequency Consumption (SFQ) of Mediterranean coastal population was developed. The SFQ was adapted from guidelines of UNEP (2008) and of the US EPA.(9) The individual daily consumption related to a single seafood species was calculated by dividing the cumulative figure by the number of the species belonging to the group. The single seafood species contribution to the individual intake was calculated by multiplying the single BDE contamination to the pertinent individual consumption derived from the above mentioned elaboration. Intake estimates were expressed on daily basis (7).

### Results and Discussion

Table 1 shows the BDE-47, -99, -153 and -209 upperbound concentrations (ng/g fw) in fish and mussels from the Mediterranean Sea (3,5). The four selected congeners are the most relevant for the toxicological risk assessment as result of dietary intake. The upper section of the table shows the mean, median and standard deviation of the four relevant PBDE congeners in edible fish, the lower one describes the congeners determined at the highest frequency (135/135) in mussels. The highest bromine substituted congeners (eg BDE-153 and BDE-209) were determined only in very few mussels samples, possibly because of the species feeding habits. The mean and median concentration of the four main

congeners determined in fish do not show great discrepancies from the EFSA estimates (1). However, the comparability could be influenced by the sample number and the represented species.

The species-specific concentrations of the congeners are shown in Table 2 by the median estimates. These Mediterranean species are among the most consumed in Italy (7) and they represent the 45.0% of the seafood total consumption. BDE-47 concentration range is 0.064–1.38 ng/g fw with the highest values determined in crustaceans (e.g. Norway lobster) and predators (e.g. Atlantic Bonito). The BDE-99 concentration range is 0.001–0.193 ng/g fw with the highest levels determined in predators. BDE-153 and BDE-209 highest concentrations were measured in crustaceans and predators, while the lowest in mussels, but the species-specific levels do not seem to show consistent patterns.

Table 3 reports the species-specific and total mean daily consumptions and PBDE exposure rates derived for Italian coastal populations due to the Mediterranean fish and seafood consumption. The fish consumption of 0.307 g/kg b.w. day, accounting for approximately 45% of total fish and seafood, is well below the value 2.6 g/kg b.w. day reported by EFSA (1) for people at high risk. However, this last figure reflects a specific diet characterized by high fish consumption that might lead to higher PBDE dietary intake (people who consume fish every day like fishermen or fish sellers might be at higher risk). In the exposure assessment, the upper bound approach was considered. The BDE-47, BDE-99, BDE-153 and BDE-209 total intake estimates were 0.092, 0.011, 0.005, and 0.011 ng/kg b.w. day, respectively. The highest contributes came from BDE-47 and BDE-99 mainly due to the consumption of rose shrimp, gilthead seabream, swordfish, Atlantic bluefin tuna and European sea bass.

In conclusion the present study shows that the coastal population exposure to PBDEs as a result of Mediterranean seafood consumption does not highlight deviation from the EFSA intake estimates referred to general EU population, where toddlers/children up to 3 years and sensitive groups (i.e. high consumers) are not framed.

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**Table 1.** PBDE concentrations in selected Mediterranean seafood species (ng/g fw).

	<i>Chemical</i>	<i>mean</i>	<i>sd</i>	<i>median</i>	<i>EFSA mean</i>
<b>FISH</b>	<b>BDE47</b>	0.351	0.39	0.196	0.40
	<b>BDE 99</b>	0.059	0.076	0.023	0.09
	<b>BDE 153</b>	0.019	0.028	0.0069	0.02
	<b>BDE 209</b>	0.031	0.062	0.0156	—
<b>MUSSELS</b>	<b>BDE47</b>	0.072	0.029	0.071	—
	<b>BDE 99</b>	0.030	0.015	0.027	—

**Table 2.** Median occurrence of selected toxicologically relevant PBDEs in Mediterranean seafood species.

<b>Species</b> <i>(scientific name)</i>	<b>Occurrence (ng/g, fw)</b>			
	<b>BDE47</b>	<b>BDE99</b>	<b>BDE153</b>	<b>BDE209</b>
Bullet tuna ( <i>Auxis rochei</i> Risso)	0.470	0.169	0.038	0.034
European sea bass ( <i>Dicentrarchus labrax</i> L.)	0.278	0.001	0.003	0.048
European anchovy ( <i>Engraulis encrasicolus</i> L.)	0.129	0.024	0.003	0.014
European hake ( <i>Merluccius merluccius</i> L.)	0.096	0.011	0.002	0.006
Surmullet ( <i>Mullus surmuletus</i> L.)	0.064	0.048	0.011	0.013
Norway lobster ( <i>Nephrops norvegicus</i> L.)	1.38	0.068	0.119	0.013
Rose shrimp ( <i>Parapenaeus longirostris</i> L.)	0.849	0.043	0.084	0.009
Atlantic bonito ( <i>Sarda sarda</i> L.)	1.03	0.193	0.039	0.016
Atlantic mackerel ( <i>Scomber scombrus</i> L.)	0.265	0.120	0.024	0.016
European pilchard ( <i>Sardina pilchardus</i> Walb.)	0.521	0.057	0.018	0.013
Gilthead seabream ( <i>Sparus aurata</i> L.)	0.213	0.009	0.005	0.025
Atlantic bluefin tuna ( <i>Thunnus thynnus</i> L.)	0.573	0.096	0.027	0.231
Atlantic horse mackerel ( <i>Trachurus trachurus</i> L.)	0.295	0.023	0.007	0.006
Swordfish ( <i>Xiphias gladius</i> L.)	0.555	0.088	0.026	0.034
Mediterranean mussel ( <i>Mytilus galloprovincialis</i> )	0.071	0.028	<0.004	<0.071

**Table 3.** Mean daily consumption of Mediterranean seafood species and correlated average PBDE dietary exposure estimates of the Italian adult coastal population.

Species (scientific name)	Consumption (g/kg-bw day)	Exposure (ng/kg-bw day)			
		BDE47	BDE99	BDE153	BDE209
Bullet tuna ( <i>Auxis rochei</i> Risso)	0.001	0.00034	0.00012	0.000028	0.000025
European sea bass ( <i>Dicentrarchus labrax</i> L.)	0.033	0.0091	0.000049	0.00011	0.0016
European anchovy ( <i>Engraulis encrasicolus</i> L.)	0.047	0.0060	0.0011	0.00016	0.00066
European hake ( <i>Merluccius merluccius</i> L.)	0.021	0.0020	0.0002	0.0001	0.0001
Surmullet ( <i>Mullus surmuletus</i> L.)	0.016	0.0010	0.0008	0.0002	0.0002
Norway lobster ( <i>Nephrops norvegicus</i> L.)	0.004	0.0052	0.0003	0.0005	0.0001
Rose shrimp ( <i>Parapenaeus longirostris</i> L.)	0.024	0.0201	0.0010	0.0020	0.0002
Atlantic bonito ( <i>Sarda sarda</i> L.)	0.002	0.0022	0.0004	0.0001	0.0000
Atlantic mackerel ( <i>Scomber scombrus</i> L.)	0.015	0.0041	0.0018	0.0004	0.0003
European pilchard ( <i>Sardina pilchardus</i> Walb.)	0.013	0.0069	0.0008	0.0002	0.0002
Gilthead seabream ( <i>Sparus aurata</i> L.)	0.063	0.0134	0.0006	0.0003	0.0016
Atlantic bluefin tuna ( <i>Thunnus thynnus</i> L.)	0.014	0.0082	0.0014	0.0004	0.0033
Atlantic horse mackerel ( <i>Trachurus trachurus</i> L.)	0.002	0.0005	0.00004	0.00001	0.00001
Swordfish ( <i>Xiphias gladius</i> L.)	0.020	0.0109	0.0017	0.0005	0.0007
Mediterranean mussel ( <i>Mytilus galloprovincialis</i> )	0.033	0.0024	0.0009	0.0001	0.0024
<b>All Mediterranean fish and seafood</b>	<b>0.307</b>	<b>0.092</b>	<b>0.011</b>	<b>0.005</b>	<b>0.011</b>